LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

NAA-SR-11450
Progress Reports

ANNUAL TECHNICAL PROGRESS REPORT AEC UNCLASSIFIED PROGRAMS FISCAL YEAR 1965

The preceding Quarterly Progress Reports during
GFY 1965 were:

NAA-SR-10501 July - September 1964

NAA-SR-10850 October - December 1964

NAA-SR-11200 January - March 1965

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

ATOMICS INTERNATIONAL

A DIVISION OF NORTH AMERICAN AVIATION, INC.

CONTRACT: AT(11-1)-GEN-8

ISSUED: NOV 10 1965

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

DISTRIBUTION

This report has been distributed according to the category "Progress Reports", UC-2, as given in "Standard Distribution Lists for Unclassified and Scientific Technical Reports" TID-4500 (43rd Ed.), February 1, 1965 by the AEC. A total of 475 copies was printed.

Printed in USA. Price \$7.00. Available from the Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Virginia

ANNUAL TECHNICAL PROGRESS REPORT
FISCAL YEAR 1965

CONTENTS

CONTENTS BY AEC CATEGORY	v
REPORTS PUBLISHED DURING FISCAL YEAR 1965	v
SODIUM-REACTOR EXPERIMENT	I
LARGE SODIUM-GRAPHITE REACTORS	II
FAST-REACTOR DEVELOPMENT	III
SODIUM-COMPONENTS DEVELOPMENT	IV
SCR ADVANCED DEVELOPMENT	V
HNPf-PNPf FUELS	VI
REACTOR TECHNOLOGY, SAFETY, AND RESEARCH	VII

ANNUAL TECHNICAL PROGRESS REPORT
FISCAL YEAR 1965

AEC Category No.	AI-GO-SA	Project Title	Page No.
58-e-11	7599-All }	Sodium Reactor Experiment Modifications	I-2
58-e-11	7626-All }		
58-e-11	7642-All	Sodium Reactor Experiment Modifications	I-19
04-01-04-21	7661-1310	SCCR Feasibility, Evaluation, and Design	III-7
04-01-04-21	7661-1331	Calculational Methods and Cross-Sections	III-20
04-01-04-21	7661-1332	Physics Experiments	III-30
04-01-04-21	7661-1341	Boiling Studies for Sodium Reactor Safety	III-35
04-01-04-21	7661-1342	Sodium Fires and Fission Product Release	III-41
04-01-04-21	7661-1343	Fuel Element Transient Heating	III-50
04-01-04-21	7617-1910	Hallam Alloy Fuel Element Surveillance and Examination	VI-2
04-01-04-21	7617-1912	HNPf Core-I UC Fuel Element Surveillance and Examination	VI-7
04-01-04-21	7617-1914	HNPf Core-II UC Fuel Element Surveillance	VI-9
04-01-04-21	7641-1322	Contaminant Removal Techniques	V-4
04-01-04-21	7641-1323	Large Component Test Loop	V-7
04-01-04-21	7641-1324	Evaluation of Long Term Operation on SRE Sodium Systems and Components	V-8
04-01-04-21	7641-1331	Improvement of Methods for Making Sodium-Cooled Reactor-Physics Calculations	V-16
04-01-04-21	7641-1332	SGR Critical Experiment	V-18
04-01-04-21	7519-1310	SGR Design and Evaluation	II-4
04-01-04-21	7519-1951	Austenitic Stainless Steels for Service in a High Temperature Sodium Environment	II-6
04-01-04-21	7519-1953	UC Fuel Element Development	II-19
04-01-04-21	7519-1955	LSGR Fuel Element Development	II-33
04-01-04-21	7519-1957	Development and Evaluation of UC Alloy Fuel Materials	II-37
04-01-04-21	7519-1959	UC Process Development	II-48
04-01-04-21	7519-1961	SRE Core-III Test Elements	II-53
04-01-04-21	7519-3210	SGR Dry Control Rod Concept Development	II-55
04-01-04-21	7519-3220	SGR Dry Control Rod Prototype Development	II-58
04-01-04-21	7519-3230	SGR Wet Control Rod Conceptual Development	II-59
04-01-04-21	7519-3610	SGR Calandria Structural Tests	II-60
04-01-04-21	7519-5210	SGR Calandria Maintenance	II-68
04-01-04-21	7519-5220	SGR Core Maintenance and Prototype Development	II-77
04-01-04-22	7602-1310	SRE Operations	I-4
04-01-04-22	7602-1320	SRE Operational Support	I-7
04-01-04-22	7602-1330	SRE Engineering and Analysis	I-9
04-01-04-22	7602-1350	SRE Engineering Surveillance	I-14
04-01-04-22	7602-1360	Handling and Shipment of Radioactive Waste Materials to Burial Sites	I-16
04-01-04-22	7602-1370	Power Expansion Program (SRE-PEP) Safety Analysis	I-17
04-01-04-22	7602-1920	Handling and Shipment of Source and Special Nuclear Materials Scrap to Recovery Sites	I-18
04-01-04-22	7602-Some	SRE Power Expansion Program Engineering	I-2
04-01-04-31	7593-1322	Operation of Sodium Component Test Installation	IV-4
04-01-04-31	7593-1328	High-Temperature Strain Measurements	IV-14
04-01-04-31	7593-1329	Mechanisms in Sodium	IV-16
04-01-04-31	7593-4121	Fabrication of Modular Steam Generator	IV-7
04-01-04-31	7593-4125	Installation of ALCO Steam Generator	IV-6

(more)

AEC Category No.	AI-GO-SA	Project Title	Page No.
04-02-02-01	7615-1970	PNPF Fuel Element Examination	VI-11
04-02-02-01	7629-1910	PNPF Core-II Fuel Element Development	VI-18
04-02-02-01	7629-1930	PNPF Core-II Design	VI-22
04-40-01-01	7604-1311	Age Measurements	VII-14
04-40-01-01	7604-1312	Extrapolation Length Measurements	VII-18
04-40-01-01	7604-1313	Cross-Section Analysis and Applications Project	VII-21
04-40-01-01	7604-1330	Fast-Spectrum Doppler Measurements	VII-29
04-40-02-09	7551-4539	Structure, Properties, and Fission Effects in UC and Other Refractory, Interstitial Fuels	VII-36
04-40-02-09	7551-4549	Study of BeO as a Reactor Material	VII-56
04-40-02-09	7652-4537	Fission Effects in Metal Fuels	VII-1
04-40-02-09	7652-4552	Synthesis, Structure, and Behavior of Metal- Hydrogen Systems	VII-8
04-40-03-01	7616-4584	Continuous Neutron-Flux Monitor	VII-72
04-40-04-03	7522-4612	Multicycle Reprocessing	VII-80
04-60-01-09	7549-1351	Measurement of Doppler Coefficients	VII-98
04-60-10-01	7608-1331	Fission Product Retention by Reactor Coolants	VII-86
04-60-20-01	7603-1373	Study of Reactor Housing Installations	VII-85
04-60-40-04	7640-1371	Atmospheric Decontamination in Reactor Accidents	VII-110
05-02-02-01	9305-5302	High-Temperature Chemistry	VII-125
05-02-02-01	9305-5305	Radiation Chemistry	VII-131
05-03-02-01	9305-5401	Electronic Structure of Metals and Alloys	VII-137
05-03-03-01	9305-5404	Radiation Damage in Crystalline Solids	VII-144

TOPICAL REPORTS PUBLISHED DURING FISCAL YEAR 1965
IN THE REPORT DIVISIONS INDICATED

NAA-SR Number	Title and Author	AEC Distribution
I. SODIUM REACTOR EXPERIMENT		
6463	Fuel Cladding Failure Monitor for Sodium Reactors, by R. J. Hall, R. E. Avery, and E. A. Moore	Reactor Technology
9430	Nuclear Parameters of the SRE Core II, by R. W. Keaten, R. D. Phelps, A. L. Gunby	Reactor Technology
9509	Measurement of the Reactivity Absorbed by Sodium in the SRE, by A. L. Gunby, J. M. Kallfelz	Reactor Technology
9510	Core II Physics Tests on the Sodium Reactor Experiment, by M. R. Pellet, W. J. Carlson, J. T. Dolan, A. L. Gunby, J. M. Kallfelz, R. D. Phelps	Reactor Technology
9511	On-Line Shim Rod Calibration, by E. A. Moore, C. L. Peckinpaugh	Reactor Technology
10006	Development of Special Viewing and Photographic Equipment for the SRE Reactor Core, by E. R. Meise	Engineering and Equipment
10010	Sodium Reactor Experiment Reactivity History Program, by L. M. Weaver	Reactor Technology
10151	Temperature Oscillations in the SRE, by C. L. Peckinpaugh	Reactor Technology
10152	Design of an Experimental Bowable Fuel Element for the SRE, by C. L. Peckinpaugh	Reactor Technology
10153	Temperature Distributions in the SRE Core II Fuel Element, by C. L. Peckinpaugh	Reactor Technology
10154	Analysis of Temperature Oscillations in the SRE, by C. L. Peckinpaugh and H. H. Cappel and R. W. Keaten	Reactor Technology
10186	An On-Line Reactivity Meter for the SRE, by M. R. Pellet	Reactor Technology
10379	Sodium Reactor Experiment Power Expansion Program, by W. J. Freede, J. K. Roberts	Engineering and Equipment
10721	Drilling Graphite Samples from Irradiated SRE Moderator Elements, by R. D. Phelps	Reactor Technology
10817	SRE Mark II Fuel Handling Machine, by H. B. Dietz	Engineering and Equipment
11234	Supplement to the SRE-PEP Safety Analysis Report, by A. V. Campise	Special

TOPICAL REPORTS PUBLISHED DURING FISCAL YEAR 1965
IN THE REPORT DIVISIONS INDICATED

NAA-SR Number	Title and Author	AEC Distribution
II. LARGE SODIUM GRAPHITE REACTORS		
7502	Compatibility of Uranium Carbide Fuels Cladding Materials, by P. E. Elkins	Metals, Ceramics, & Materials
7892	Thermal Neutron Spectrum Effects on Calculations for Graphite Lattices, by F. L. Fillmore	Physics
8887	Uranium Carbide Process Development for the Advanced Sodium-Cooled Reactor, by D. H. Turner	Metals, Ceramics, & Materials
9488	Steam Cycle Optimization Study for Large Sodium Graphite Nuclear Power Generating Stations, by D. F. Casey, G. A. Schneider	Reactor Technology
9501	1000-Mwe SGR Core Optimization Studies, by H. Rood	Reactor Technology
9604	Process Development for Bonding Zircaloy 2 Cladding to Graphite Moderator, by S. K. Lee	Metals, Ceramics, & Materials
9967	Flux Control for Irradiation Experiments, by W. K. McCarty	Metals, Ceramics, & Materials
11047	Some High Temperature Mechanical Properties of Zircaloy-2, Type 304 Stainless Steel and AM-350 Stainless Steel, by D. H. Stone	Metals, Ceramics, & Materials
11123	Hydraulic Studies of the LSGR Process Channel, by J. H. Brindley	Reactor Technology
III. FAST REACTOR DEVELOPMENT		
9477	Boiling Studies for Sodium Reactor Safety, Part II, by H. Lurie and R. C. Noyes	Engineering and Equipment
9508	Uranium Carbide Transient Heating Experiment Phase I, by S. J. Stachura, M. Silberberg, R. N. Cordy	Reactor Technology
11359	TR-SRE Fast-Flux Irradiation Facility	Special
11378	Feasibility Study of 1000 Mwe Sodium Cooled Fast Reactor, Vol I and II, by J. C. Cochran, D. T. Eggen and A. S. Gibson	Special

TOPICAL REPORTS PUBLISHED DURING FISCAL YEAR 1965
IN THE REPORT DIVISIONS INDICATED

NAA-SR Number	Title and Author	AEC Distribution
IV. SODIUM COMPONENTS DEVELOPMENT		
4527	Analysis of Stresses in Bellows, Part I, Design Criteria and Test Results, by W. F. Anderson	Engineering and Equipment
4527	Analysis of Stresses in Bellows, Part II-Mathematical, by W. F. Anderson	Engineering and Equipment
8119	Study of SCTI Control System, by C. W. Griffin, R. F. Audette, C. H. Bridges Jr., D. F. Casey, D. Logan, R. K. Seferian, and R. Schmidt	Reactor Technology
8541	A Device for Internal T. I. G. Arc Welding of Tube to Tube-Sheet Joints, by G. Kazlauskas	Engineering and Equipment
9762	Stress and Elevated Temperature Fatigue Characteristics of Large Bellows, by R. A. Winborne	Engineering and Equipment
9826	Final Design of Sodium-Heated, Modular, Steam Generators for the SCTI, by D. F. Casey, J. J. Hammer, D. Logan, and M. C. Williams	Engineering and Equipment
9827	Fatigue Characteristics of 37 Tube Modular Steam Generator Head, by M. M. Lemcoe	Engineering and Equipment
9848	Simplified Formulas and Curves for Bellows Analysis, by R. A. Winborne	Engineering and Equipment
V. SCR ADVANCED DEVELOPMENT		
9771	Material Bucklings of Critical and Subcritical Uranium Carbide Fueled Graphite Assemblies (Part I Experiment), by R. W. Campbell, H. C. Field, O. R. Hillig, E. J. Johnson, J. F. Johnson, and D. E. Latham	Physics
9772	Material Bucklings of Critical and Subcritical Uranium Carbide Fueled Graphite Assemblies - Part II - Theoretical Interpretation, by E. R. Specht	Physics
9803	Measurements of Thermal Neutron Absorption Cross Section and Diffusion Length of Using an Sb-Be Source, by J. A. DeJuren, T. E. Stewart, and E. R. Specht	Physics
9889	Experimental Study of Inelastic Strains in Hollow Cylinder of 6061 Aluminum Subject to a Bank of Internal Pressure, by V. W. Anderson	Engineering and Equipment
10531	Multigroup Calculation Methods with Application to the SGR Critical Experiment, by F. L. Fillmore	Physics

TOPICAL REPORTS PUBLISHED DURING FISCAL YEAR 1965
IN THE REPORT DIVISIONS INDICATED

NAA-SR Number	Title and Author	AEC Distribution
VI. HNPf-PNPf FUELS		
7785	Experimental Testing of Core Component Handling Equipment and Handling Techniques for Hallam Nuclear Power Facility, by W. A. Kaspic	Reactor Technology
8955	The NAA-47 U-10 Mo Fuel Irradiation Program for HNPf Core I, by A. Schmidt, R. Willard, and D. Magnus	Metals, Ceramics, & Materials
8956	Irradiation Swelling, Phase Reversion and Intergranular Cracking of U - 10 wt % Mo Fuel Alloy, by R. Willard and A. R. Schmidt	Metals, Ceramics, & Materials
9924	The Design Basis for Pin-Hole Gamma-Ray Cameras, by M. A. Greenfield and R. L. Koontz	Engineering and Equipment
9988	UC Fuel Element Design and Fabrication, by W. Draganchuk	Reactor Technology
10078	Nuclear Startup Experiments for HNPf, by M. P. Aronchick	Reactor Technology
VII. REACTOR TECHNOLOGY, SAFETY AND RESEARCH, AND OTHER		
8213	Refabrication and Encapsulation of Highly Irradiated Uranium Dioxide, by J. Guon, J. Bodine, and R. Sullivan	Metals, Ceramics, & Materials
8880	Conformal Ionic Mixture, by S. Yosim and M. Blander	Chemistry
9161	Reprocessing of Uranium Carbide by a Nitride-Carbide Cycle II, Kinetics of Nitride Conversion to Carbide, by L. A. Hanson	Metals, Ceramics, & Materials
9260	Crystallographic Expansion of BeO at the Alpha-Beta BeO Transformation Temperature, by S. B. Austerman	Metals, Ceramics, & Materials
9278	Reprocessing of Uranium Carbide Cycle III, Complete Cycle and Fission Product Study, by L. A. Hanson	Chemical Separations Processes for Plutonium and Uranium
9287	High Temperature Experiments on Fission Product Retention in Sodium, by W. P. Kunkel, D. M. Elliott, and A. S. Gibson	Reactor Technology
9335	Comparative Fuel Cycle Evaluations, Low Decontamination Pyroprocessing, and Aqueous Reprocessing, Part II UC Fuel in a Thermal Reactor, by R. Mattern and L. Colby, Jr.	Metals, Ceramics, & Materials

TOPICAL REPORTS PUBLISHED DURING FISCAL YEAR 1965
IN THE REPORT DIVISIONS INDICATED

NAA-SR Number	Title and Author	AEC Distribution
VII. REACTOR TECHNOLOGY, SAFETY AND RESEARCH, AND OTHER (Continued)		
9033	Boiling Depressurization Transients, by F. J. Halfen, F. Berganzoli	Reactor Technology
9503	The Age of Fission Neutrons to Indium Resonance Energy in Zirconium-Water Mixtures, by R. K. Paschall	Physics
9745	Argon Mass Flowrate Computer, by K. H. Kline	Instruments
9748	The Continuous Neutron Flux Monitor Project - An Interim Report, by C. R. F. Smith, T. J. Springer	Instruments
9982	AISITE II - A Digital Computer Program for Investigation of Reactor Siting, by R. A. Blaine and E. L. Bramblett	Mathematics and Computers
9943	AIROS - A Digital Simulator for Power Reactor Dynamics, by R. A. Blaine and R. F. Berland	Mathematics and Computers
5128	Vapor Pressure of Polyphenyl Coolants, by H. Mandel, F. Silvey, and K. Koda	Chemistry
5198	The Effect of Additives on Radiolytic Decomposition Rate of Terphenyl Coolants, by M. A. Rotheram and R. H. J. Gercke	Chemistry
8277	Radiolytic and Pyrolytic Decomposition of Ortho Terphenyl at High Temperature, by J. M. Scarborough	Chemistry
8448	Chromatographic Analysis of Gases Over Liquid Sodium, by D. L. Johnson	Metals, Ceramics, & Materials
8474	Evaluation of Coolant Impurity Removal Equipment at the OMRE, by P. Barbour and W. W. Davis	Reactor Technology
9473	Piqua Nuclear Power Facility Operations Analysis Program Progress Report No. 3, by J. J. Auleta, et al.	Reactor Technology
9637	Post-Irradiation Examination of Surface Film Formation on OMRE Driver Element No. 219, by R. J. Sullivan, et al.	Chemistry
9731	Radiolysis Products of Polyphenyl Coolants, Part III, OMRE High Boilers, by R. T. Keen, W. L. Orr, L. J. Miller, R. J. Sullivan, and R. C. Shepard	Chemistry
9733	Radiolysis Products of Polyphenyl Coolants, Part II, Low and Intermediate Boilers, by R. A. Baxter, R. C. Shepard, W. L. Orr, L. J. Miller, R. T. Keen, and M. A. Rotheram	Chemistry
9734	Determination of the Water Content of Polyphenyl Coolants, by M. A. Rotheram	Chemistry

TOPICAL REPORTS PUBLISHED DURING FISCAL YEAR 1965
IN THE REPORT DIVISIONS INDICATED

NAA-SR Number	Title and Author	AEC Distribution
VII. REACTOR TECHNOLOGY, SAFETY AND RESEARCH, AND OTHER (Continued)		
9744	Escape Probability Approximations in Lumped Resonance Absorbers, by John M. Otter	Reactor Technology
9796	Performance of Structural Materials Exposed to the Coolant in the OMRE, by R. S. Neymark	Metals, Ceramics, & Materials
9799	Hallam Nuclear Power Facility Reactor Operations Analysis Program Semiannual Progress Report No. 3 - September 1, 1963 to February 29, 1964, by D. K. Darley, et al.	Reactor Technology
10307	Piqua Nuclear Power Facility Operations Analysis Program, Progress Report No. 4, January 1, 1964 - June 30, 1964, by H. J. Rubinstein, et al.	Reactor Technology
10743	Hallam Nuclear Power Facility Reactor Operations Analysis Program Semiannual Progress Report No. 4, February-September, 1964, by H. J. Rubinstein, et al.	Reactor Technology
11142	Piqua Nuclear Power Facility Reactor Operations Analysis Program Semiannual Progress Report No. 5, July-December, 1964, by H. J. Rubinstein, et al.	Reactor Technology

SECTION I
SODIUM-REACTOR EXPERIMENT
CONTENTS

<u>Title</u>	<u>Identification</u>	<u>Page</u>
SODIUM REACTOR EXPERIMENT	Summary	I-1
SRE POWER EXPANSION PROGRAM (PEP) ENGINEERING	7599 - All 7602 - Some 7626 - All	I-2
SRE OPERATIONS	7602 - 1310	I-4
SRE OPERATIONAL SUPPORT	7602 - 1320	I-7
SRE ENGINEERING AND ANALYSIS	7602 - 1330	I-9
SRE ENGINEERING SURVEILLANCE	7602 - 1350	I-14
HANDLING AND SHIPMENT OF RADIO- ACTIVE WASTE MATERIALS TO BURIAL SITES	7602 - 1360	I-16
POWER EXPANSION PROGRAM (SRE-PEP) SAFETY ANALYSIS	7602 - 1370	I-17
HANDLING AND SHIPMENT OF SOURCE AND SPECIAL NUCLEAR MATERIALS SCRAP TO RECOVERY SITES	7602 - 1920	I-18
MISCELLANEOUS MODIFICATIONS	7642 - All	I-19

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Sodium Reactor Experiment
Program Manager: L. E. Glasgow
Reporting Period: FY 1965

I. PROGRAM OBJECTIVES

Objectives of the SRE Power Expansion Program are to: (a) modify the SRE core to permit operation at reactor outlet temperatures up to 1200°F, (b) increase power density in the SRE core sufficiently to permit irradiation of fuel elements at heat fluxes near 10^6 Btu/hr-ft², and (c) improve overall SRE reliability to achieve a 90% plant availability factor, or better.

The purpose of the SRE Power Expansion Program is to permit evaluation of high-performance sodium-graphite reactor fuels and other components under environmental conditions to which they would be exposed in a large, advanced, sodium-cooled power reactor.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

The heat transfer systems were completed, filled with sodium, and flow tested at 370°F. Three of the four pumps worked perfectly from the outset. The main primary pump, however, exhibited excessive internal flow leakage. This overloaded the overflow system and caused gas entrainment. The difficulty has been corrected and the pump now functions normally. The sodium systems are ready for operation.

All of the moderator and reflector elements were installed in the reactor without incident and slightly ahead of schedule. The maximum installation rate was 6 units per day or about twice the expected rate.

The full-scale test moderator element completed the accelerated test equivalent of 58 years of SRE service. The unit was sectioned and found to be completely sound and free of defects.

All of the fuel has been delivered to the site and assembled into complete units and is ready for loading.

The nuclear and heat transfer instrumentation was installed and tested. In all cases, the equipment performance equalled or exceeded the specification requirements.

The overall project was completed on schedule and within budget. The plant is ready to be placed in operation as soon as authorization is received.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Advanced Sodium Cooled Reactors

Project: SRE Power Expansion Program (PEP) Engineering

Project Engineer: W. J. Freede

Reporting Period: Fiscal Year 1965

AEC Category: 58-e-11 and
04-01-04-22

General Order: 7599/7602/7626

I. PROJECT OBJECTIVES

Objectives of the SRE Power Expansion Program are to: (a) modify the SRE core to permit operation at reactor outlet temperatures up to 1200°F, (b) increase power density in the SRE core sufficiently to permit irradiation of fuel elements at heat fluxes near 10^6 Btu/hr-ft², and (c) improve overall SRE reliability to achieve a 90% plant availability factor, or better.

The purpose of the SRE Power Expansion Program is to permit evaluation of high-performance sodium-graphite reactor fuels and other components under environmental conditions to which they would be exposed in a large advanced sodium-cooled power reactor.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. HEAT TRANSFER SYSTEM MODIFICATION, PHASE I, ENGINEERING

Necessary engineering followup has been rendered for assistance to SRE-PEP construction activities which have been completed during FY 1965 by contracting firms. All engineering work has been completed and components purchased for modifications to the primary sodium service system; installation work pertaining to cold-trap and filter units has been finished, and this portion of the primary sodium service system has been placed into operation.

The final report covering SRE-PEP heat transfer system modifications has been published.

B. CORE MODIFICATIONS

Nuclear analyses, thermal and hydraulic studies, and design layouts pertaining to the SRE-PEP core have been completed. Detail design drawings and engineering specifications concerning stainless steel clad reflector cells, Zr-2 clad moderator and reflector cells, standard fuel elements, shim and safety rods, core coolant (sodium) level indicator, moderator cell flow adapter sleeves, and various grapples for core components have been finished and issued. All these listed components have been manufactured and installed.

Four short-test Zr-2 clad moderator cells have been constructed, and three of them subjected to various thermal and pressure tests. Fatigue bend tests have been conducted on 50-mil Zr-2 strips at 700 and 1200°F in excess of 40,000 cycles without a single failure. Each bending cycle exceeded the amount of motion which a moderator/reflector cell corner would experience during a full startup, shutdown cycle in the core.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Seventy-four thermocycles between 500 and 1300°F have been applied to a full-sized Zr-2 clad moderator cell at various external pressures. It has been calculated that the aggregate strain due to these thermal cycles exceeded by a factor larger than 10 the amount which such a cell would experience over 5 yr in the SRE-PEP core. The tested moderator cell did not rupture, and only negligible dimensional changes were produced.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The SRE-PEP Heat Transfer System Modification Report (NAA-SR-10379) which has been issued describes all modifications made to the four SRE heat-transfer loops (main primary, main secondary, auxiliary primary, auxiliary secondary). All contractor work has been completed on the heat-transfer facility modifications in accordance with established schedules.

Completion of hydraulic tests regarding the standard fuel element and variable orifice assembly has confirmed design objectives. Information provided has supplemented fluid flow and heat-transfer analyses for the SRE-PEP core.

By eliminating dummy fuel elements during initial power operations, it has been shown that the complement of safety rods may be reduced from six to four. This arrangement will reduce startup costs; however, at a later date, when reactivity has been reduced by 1.5%, dummy fuel elements may be added without increasing the number of safety rods. Studies concerning nuclear properties of dummy fuel elements have shown that stainless steel clad units would be of little reactivity worth. Zr-2 clad dummy fuel elements, however, can contribute reactivity worth to the core and permit longer operation for each loading of fuel elements.

Experimental test results obtained from shrink-fit operations on short (test) moderator cells, creep-strain runs on Zr-2 at various temperatures, elevated temperature fatigue bend tests, and extended thermal cycle experiments have verified the moderator-reflector cell design concept. This test information, when subjected to calculation studies, has indicated that creep-fitting of the Zr-2 cladding onto the graphite interior of each can would not be required prior to installation in the core to effect satisfactory performance.

All manufacturing problems concerning Zr-2 cladding for moderator-reflector cells have been resolved, and sufficient quantities of this material (Zr-2) have been received to fabricate the total production order.

Cell replacement operations involving the SRE core have been completed on schedule.

Poison ring drop-tests conducted for other ASCR programs have indicated that original SRE poison columns may be used in the SRE-PEP core without compromising safety requirements. Consequently, this equipment has been used for the SRE-PEP core to effect an additional cost saving to the program.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Advanced Sodium Cooled Reactors	
Project: Sodium Reactor Experiment Operations	
Project Engineer: J.E. Owens	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-22
General Order: 7602	Subaccount: 1310

I. PROJECT OBJECTIVES

The primary objectives are to: (a) safely maintain the plant in a shutdown condition, (b) operate the required plant facilities for completion of PEP construction, (c) prepare the reactor for restart utilizing new equipment and PEP conditions, (d) assume accountability for Core III fuel, and (e) improve reliability of new sodium systems.

These objectives will be in effect until the reactor is restarted. At the completion of plant modifications the objectives will be to: (a) operate the reactor at new design conditions, (b) evaluate long-term system characteristics, (c) achieve an 80% use factor, and (d) utilize the plant as a fuel irradiation facility.

The specific objectives which support the above are to: (a) operate the plant with adherence to approved methods and procedures, (b) assist in plant modifications and maintenance, (c) expand the approved technical limits, (d) generate, collect, and organize operational data, and (e) maximize efficiency and economy in the use of manpower and material through a well-organized operator training program.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

The reactor has been shutdown this fiscal year for plant modifications under the Power Expansion Program (PEP). By enlarging the heat transfer system and increasing the core ΔT , the power capability of the SRE will be increased from 20 to 30 Mwt. Replacing the moderator cladding with Zircaloy II will allow reliable operation with a maximum outlet temperature of 1200°F.

On completion of heat transfer system construction in February, 1965, replacement of the moderator and reflector region of the SRE core was initiated. Fifty-seven moderator and sixty-two reflector elements were replaced on schedule and within budget. This was accomplished by improving equipment and procedures. For example, much of the waste generated during the program was packed in the shipping containers containing the obsolete moderator elements.

All core components necessary for the loading of fuel have been received and installed in the reactor. Operational testing of control and safety rods, sodium temperature instrumentation, and source assembly have been completed. Prior to installation of each component in the reactor, a flow control device is inserted in the process channel. This device is necessary to partition flow between fuel channels and the moderator coolant plenum.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Fuel necessary for operational loading of the reactor has been received, inspected, and placed in fuel storage cells. Thirty-two standard driver elements (6.5 enriched UC), two standard test elements, and two special test elements (15% enriched UC) are on hand. These constitute the complete SRE-PEP fuel inventory.

Cold-watch surveillance of the shutdown plant was maintained during the PEP modification period to ensure proper levels of flow and pressures in auxiliary systems operated during the construction effort. In addition to construction support, cleanup of the "poor man's hot-cell" and the Mark-I fuel handling machine, shipment of Core II fuel, and disposal of radioactive waste were all completed during cold-watch operation.

Purification of primary sodium during the modification period was accomplished by means of a special bypass installed from the primary fill tank to the sodium service system. Sodium was circulated through a cold-trap to remove oxide and a micrometallic filter for the removal of particulates.

Installation of a parking trolley for the Mark-II fuel handling machine facilitated the PEP construction and disposal of Core-II components. Equipment for the trolley was salvaged from the Piqua fuel handling machine carriage and was modified for use at the SRE.

Techniques were developed for obtaining samples from irradiated moderator elements. Samples of moderator cell cladding and graphite cores were retrieved for analysis under S/A 4410 (G.O. 7519). A topical report describing this effort, NAA-SR-10721, by R.D. Phelps entitled "Drilling Graphite Samples From Irradiated SRE Moderator Elements" was published.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

A major milestone was achieved with the completion of SRE-PEP modifications and activation of all systems. Successful completion of this program reflects the result of careful preliminary planning, prior preparation and improvement of equipment, and development of and adherence to procedures. Operation of the SRE-PEP reactor will continue the exploration of, (1) sodium as a coolant for central-station power reactors, and (2) development of long-lived fuel for cost reductions in the operation of future nuclear power plants.

Replacement of the moderator and reflector region at a rate greater than 2.3 elements per day demonstrates the feasibility of using the SRE as a reactor test facility. Accessibility of the core and ease of maintenance indicates an adaptability to conversion from a thermal reactor to a two-region or fast reactor with a minimum effort.

Installation of necessary components in the reactor and acceptance and storage of Core-III fuel completed the preparations required for loading of fuel in the core. On receiving AEC permission to load fuel, the reactor will be taken to critical and then to full design conditions for testing and evaluation of new fuel and components.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Cold-watch operation made significant contributions for the improvement of plant operation and safety during SRE-PEP modifications. Core-I fuel slugs were removed from the "poor man's hot-cell," Mark-I fuel handling machine, and Washcell "B", thus making this equipment available for modification and use, and eliminating a potential hazard. Modifications made to the helium and kerosene systems removed obsolete valves and lines, and relocated the remaining systems for more efficient operation and maintenance.

Purification of primary sodium during reactor shutdown allowed new systems to be filled with sodium in the best possible condition and minimized cold-trapping time after filling. Filtering prior to filling eliminated the need for a full-flow filter in the primary system.

Installation of the fuel handling machine trolley increases operating efficiency by providing a safe storage facility removed from the normal work area. Its value will be most apparent during future gallery shield block handling, Mark-II FHM modifications, and manipulation of moderator handling equipment.

Samples taken from irradiated moderator elements demonstrated the ease with which components can be removed from the reactor and sampled at the SRE facility thus eliminating the need for special handling and transportation equipment normally required for this operation. In this manner, samples can be obtained and analyzed, and results can be determined economically in a short time.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Advanced Sodium Cooled Reactors

Project: SRE Operational Support

Project Engineer: J. E. Owens

Reporting Period: Fiscal Year 1965

AEC Category: 04-01-04-22

General Order: 7602

Subaccount: 1320

I. PROJECT OBJECTIVES

The primary objectives of this project are to: (a) provide a preventive maintenance program, (b) provide for breakdown maintenance as required, (c) formalize standard maintenance procedures, (d) continue to update standard operating procedures, (e) provide for radiological health-and-safety support for SRE personnel, (f) provide for hot-cell support as required, and (g) provide an updated operator training manual.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

Revisions to the SRE operations manual were completed. Fifty-six procedures were revised to reflect changes in the reactor, heat transfer systems, gas systems, service cooling systems, and fuel handling techniques.

Revision of the SRE operators training manual was completed. Six chapters of the manual were affected by changes made during the Power Expansion Program (PEP).

SRE preventive maintenance schedule revisions were completed, reflecting equipment changes made during plant modifications. Routine preventive maintenance was conducted on all equipment remaining in use during plant shutdown and scheduled for new equipment as it was placed in service. The only equipment failures have been the water circulation pumps for the kerosene cooling towers. Repeated failure of the seal between pump and motor allowed small amounts of water to penetrate into the drive motor. The pump has been replaced by a more reliable model.

Modification of the piping from the primary fill-tank to the sodium service system permitted purification of primary sodium during plant shutdown for SRE-PEP construction. Sodium was circulated from the primary fill-tank through a cold-trap and micrometallic filter for removal of oxide and particulates.

The parking carriage for the Mark II fuel-handling machine was modified to provide 4-wheel drive. This was necessary to prevent loss of traction on the transfer links from the parking bay to the 75-ton bridge.

Modifications to the helium system were completed. Portions of the system made obsolete by PEP were removed, and the remaining valves and controllers moved from pipe trenches to more accessible areas. Removal of the Nak bubbler towers from the system eliminates a potential hazard and simplifies the helium system.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Health and Safety support was provided during removal of fuel and cleanup of the Mark I fuel-handling machine, "poor man's hot cell", wash cell "B," and for disposal of obsolete control and safety elements, thimbles, and shield plugs.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

Revision of the SRE Operations Manual to include PEP modifications assured a smooth integration of new equipment, reduced operating errors, and established uniform operating techniques. Continual revision of the manual to include the best operating technology is an important factor in maintaining a high level of operator confidence.

Completion of revisions to the SRE Operator Training Manual prior to placing new equipment in service insured adequate time for training and qualifying operators on modifications made during PEP construction. Significant reduction in operating errors and a better understanding of the plant characteristics by operations personnel are direct results of this program.

Routine preventive maintenance has continued to reduce the amount of breakdown maintenance required. Replacement of the water-circulation pumps for the kerosene cooling towers with a model in which the pump and drive motor elements are separate units connected by a shaft will eliminate a major source of breakdown maintenance. Expansion of the preventive maintenance program to incorporate new equipment and increase service to existing equipment will result in significant reduction in plant outage.

Purification of primary sodium during plant shutdown permitted new SRE-PEP systems to be filled with the best possible sodium. The ability to cold-trap and filter the sodium without circulation through vital components contributes to longer equipment life.

Four-wheel drive on the fuel-handling machine parking carriage assures its reliable operation. It was previously necessary to manually move the carriage a few inches when the drive wheels were on the transfer links. This was a hazardous and time-consuming task.

Removal of the NaK bubblers from the helium system and relocation of controllers and valves to more accessible areas were other steps in the continuing effort to simplify and thus reduce the cost of auxiliary systems associated with the SGR concept. Modifications to the system eliminated many fittings, valves, regulators, and gauges which are potential sources of leakage, thus reducing the use-rate of helium.

Removal of fuel from the Mark I FHM, poor man's hot cell, wash cell "B" and the subsequent decontamination of these areas eliminated a serious potential contamination hazard from the high bay area. Extremely high contamination levels were encountered under the machine during fuel removal and decontamination operations. The fact that personnel performing the work received but insignificant amounts of contamination reflects the safety aspects of the procedures used, and close cooperation between Health and Safety and the performing personnel.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Advanced Sodium-Cooled Reactors

Project: SRE Engineering and Analysis

Project Engineer: J. E. Owens

Reporting Period: Fiscal Year 1965

AEC Category: 04-01-04-22

General Order: 7602

Subaccount: 1330

I. PROJECT OBJECTIVES

The objective of this project is to provide engineering and technical support necessary to operate SRE-PEP. This specifically includes (a) performance and evaluation of all tests, (b) determination and analysis of plant performance, (c) design engineering studies for plant improvements, and (d) development of advanced testing and analysis techniques.

The purpose of the project is to provide engineering information required to determine and improve SRE safety, reliability, and performance.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

Detail and final planning, including step-by-step procedures, for the SRE-PEP startup testing program were completed. The program consists of seventeen individual tests which include preoperational, zero power, and rise-to-full-power tests. The program places emphasis on achieving full power as quickly as is consistent with safety requirements. Performance of all tests will require approved test procedures to ensure that the test program will be conducted in a safe and efficient manner. The test procedures consist of a master test procedure, test summaries to provide review personnel with concise and complete information, and detailed procedures for actual performance of the tests. The preparation and review of the master test procedure and test procedure summaries was completed on schedule and they are approved for use in the testing program. Preoperational testing is currently in progress. Test results and analysis will be reported under G.O. 7642.

A structural analysis of the SRE-PEP fuel element established a design value for centering spring stiffness of 0.15 to 0.20 lb/mil. Use of this spring rate will maintain possible temperature variations and cladding temperature below allowable limits. The analysis also indicated that the springs will not damage the process tube wall.

Detailed analysis of the temperature oscillations which occurred in SRE-Core II was completed. Final studies showed that the fuel element movement, which caused the oscillations, was a horizontal thermal bowing of the fuel cluster caused by temperature differences across the rod bundle. Based on this analysis, a theoretical model describing the temperature oscillations was developed. Calculated results from the model were in good agreement with measured results, and clearly indicated that an unstable condition existed and that sustained oscillations would result. This model has provided complete answers as to why the oscillations occurred, and how they may be prevented from recurring in future designs.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

Preparation of test procedures and summaries was completed on schedule. Detailed procedures, in conjunction with the test summaries provided performance personnel with an integrated and complete set of test procedures for the testing program. The approved testing program is based on current technical information and reflects the background of testing experience accumulated at the SRE.

A structural analysis of the SRE-PEP driver fuel element was done to (1) determine the element centering-spring stiffness required to minimize potential temperature oscillations and cladding hot-spots, and (2) evaluate the possibility that the centering springs may damage the process tube wall. Measurements of deflection versus load were made on a mockup of the PEP driver fuel element. The measurements show that the rod bundle acts structurally as a solid beam in the lower deflection region. This indicates that small temperature differences across the rod bundle may cause relatively large thermal bowing unless centering-spring stiffness is adequate to equalize the bowing forces.

Figure 1 shows the force required to constrain a bowing cluster as a function of eccentricity. Superimposed on this graph is the total equivalent restoring force supplied by the springs. The point where the two curves cross represents the equilibrium between spring and bowing forces, and indicates the equilibrium eccentricity that is expected.

7602-2501

Figure 1. Spring Restoring and Bowing Forces vs Eccentricity for SRE-PEP Standard Driver Fuel Element

It was also determined that cluster deflection should be limited to 0.025 in. to maintain potential temperature oscillations below 20°F peak-to-peak, and cladding temperature below 1300°F. The figure shows that the proposed spring stiffness in the range of 0.15 to 0.20 lb/mil will provide sufficient constraint.

The bearing stress on the process tube from the centering spring-loading was found to be twice the yield point, indicating that some deformation will occur. However, the magnitude of the deformation was found to be 0.0005 in., which is no more than expected in surface flaws of original moderator cans. Process tube damage due to fretting was not considered credible, since hydraulic forces which might cause spring vibrations are a hundred times smaller than the spring-loading against the tube wall. Therefore, the study indicated that the recommended spring stiffness will not damage the process tube.

Detailed studies and analysis of SRE-Core II temperature oscillations have

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

been completed. Previous reports indicated that movement of the fuel rod bundle in the coolant channel caused the oscillations. Final analysis shows that this movement was a horizontal thermal bowing of the fuel cluster caused by temperature differences and associated differential thermal expansion across the rod bundle. The driving force which caused this movement is described by the following empirical relationship.

$$F = 0.28 \Delta T \sin \pi t , \quad \dots(1)$$

where ΔT is the temperature difference across the fuel rod bundle.

Based on these facts, a theoretical model was developed which contains a central fuel region with isolated coolant regions on each side. The model simulated bowing of the element by moving the fuel region horizontally as a function of time and axial distance along the channel. Equation 2 describes the resulting change in flow area along the isolated channel:

$$\Delta A(z, t) = \Delta A_{\max} \sin \frac{\pi z}{L} \sin \beta t, \quad \dots(2)$$

where L is the length of the channel, and β is the frequency of oscillation.

The change in flow area has two effects on the coolant flow. First, since the overall pressure drop remains constant, the isolated coolant channel velocity will increase and decrease, thereby changing the channel mass flow. This is described as a flow change into the isolated channel inlet or

$$W_i = W_{i0} \left[1 + C \Delta A(L/2, t) \right]^{3/2} , \quad \dots(3)$$

where W_{i0} is the steady-state inlet mass flow rate and C is a constant relating flow change to an average hydraulic radius change. The second effect is the displacement of coolant as a result of fuel movement. The displaced coolant flows radially through the rod bundle and axially upward along its normal path. The axial coolant displacement causes a local acceleration and deceleration of the flow, i. e., "a pumping action". The flow at any time and axial position is expressed as a sum of the two effects, or

$$W(z, t) = W_i + \rho \int_0^z \epsilon \frac{\partial}{\partial t} \left[\Delta A(z, t) \right] dz , \quad \dots(4)$$

where ϵ is the "pumping" efficiency — about 10%.

The heat-transfer relationships of the model were expressed by standard heat-balance equations for the fuel and coolant. These equations were programmed for digital analysis and calculations performed on an IBM 7094.

The amplitudes of oscillations calculated with the model are shown in Figure 1. The calculated amplitudes are compared with measured results over the frequency

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

range where oscillations occurred. The measured amplitudes of 50°F at 0.5 cps and 100°F at 0.1 cps fall close to the calculated curve, and represent the maximum amplitude of oscillations observed during Core II operation. Further analysis of the calculated results indicated that the model has a gain of 300°F/in. for oscillation frequencies of 0.5 cps. In addition, it was found that the coolant temperature lagged the fuel position by 180° at the 0.5 cps frequency.

In the calculations, the fuel movement was not affected by the oscillating temperature difference across the fuel region, i. e., no feedback existed. However, in the physical case a definite feedback in the form of thermal bowing does exist. The gain of this bowing feedback was calculated to be 0.005 in./°F. The bowing feedback was included in the model, and the open-loop transfer function calculated for the system. The results of this calculation, shown in Figure 2, indicate that at a frequency of 0.5 cps, the model has a transfer function with a gain greater than one and a phase lag of 180 degrees. This is the necessary and sufficient condition for instability and sustained oscillation.

Figure 2. Calculated Amplitude of Exit Temperature Oscillations

A number of unexplained oscillation characteristics were matched and explained by the model. For example, the oscillations exhibited a threshold behavior where the amplitude suddenly increased at some critical channel delta-T. This threshold phenomenon existed because a minimum axial channel delta-T is required to produce a corresponding radial temperature difference which will result in a gain greater than one. Below this critical delta-T, only damped oscillations triggered by flow-induced vibration can occur, and these were actually observed. Above the critical delta-T, the oscillations are divergent, and the amplitude increased until element motion was limited by the channel wall. Therefore, the sustained oscillations observed above the critical delta-T were a limit-cycle oscillation.

Other experimental measurements indicated that the temperature oscillation amplitude along the cluster was not directly proportional to the axial temperature difference, but was much larger in the upper element region. The model pre-

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

dicts this measured characteristic and explains it by the axial acceleration or pumping of coolant. The pumping action created a buildup of flow along the channel with the flow reaching a maximum at the top. This flow buildup created the large temperature oscillations observed at the element exit.

A topical report on the above work, "Analysis of Temperature Oscillations in SRE-Core II" by C. L. Peckinpaugh, NAA-SR-10154, has been published.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Advanced Sodium-Cooled Reactors

Project: SRE Engineering Surveillance

Project Engineer: J. E. Owens

Reporting Period: Fiscal Year 1965

AEC Category: 04-01-04-22

General Order: 7602

Subaccount: 1350

I. PROJECT OBJECTIVES

The objective of this project is to study and evaluate the design and performances of the SRE for the purpose of (a) assuring safe, steady operation, and (b) making recommendations to improve the operating characteristics of the plant.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

Exposure to primary system sodium during Core II operation has had little effect on the mechanical and chemical properties of Zircaloy-clad moderator cans. These results were summarized in NAA-SR-TDR-10266,* "Metallurgical Examination of Zircaloy-2 Skin from SRE Moderator Can G-12," by J. Stearns.

Initial examination of fuel element cladding indicated a maximum carbon content of 0.24%. This low level compared with the maximum allowable of 0.5% attests to the reasonable approach for power and temperature increase of SRE Core II which was presented in NAA-SR-MEMO-7804.*

The necessary criteria were formulated for a carburization sampling station for the SRE-PEP service system.

Those working on G. O. 7641, S. A. 1324, Components Evaluation in the examination of cold traps and heat exchanger, were given technical assistance through this project.

Evaluation of Zircaloy-2 specimens exposed to 1200°F sodium indicates the need for further effort on the effect of low oxygen levels in sodium on Zircaloy.

Results of a study of the carburization problems at the SRE, covering the operation with the second core loading, indicate that most of the carbon inadvertently introduced into the primary system in 1959 has been removed. The limited amount left still served as a carburization source; but with reactor inlet/outlet temperature not exceeding 550 to 960°F, existing hot-traps were able to control the carburization potential of the sodium to the extent that the lifetime of the fuel cladding was not jeopardized.

With large carbon concentration present in the sodium, correlation was found between the carburization rate measured in the reactor sodium and the

*An intracompany report

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

calculated carburization rate. After removal of major carbon sources and after a low carburizing potential in sodium had been obtained, no evidence was found of an equilibrium condition between dissolved carbon in sodium and surface carbon concentration in SS 304. Neither was evidence found of decarburization of SS 304. This indicates that carburization of the SS 304 could continue until the available carbon supply is depleted; and points out the need for a continuing surveillance and sampling program. Wet chemistry analysis of sodium samples gave no meaningful information about the carburization potential in the sodium. Analysis of tabs exposed to the sodium coolant in the hot-trap side stream, however, served as the most reliable tool for determination of the carburization potential.

During the last three months of power operation, graphite in failed moderator elements was exposed directly to sodium at reactor outlet temperatures of 550 to 960°F. No measurable increase in carburization potential was found in the sodium.

A Fortran program has been written that will predict the heat transfer coefficient for the SRE steam generator, and compare it with the measured heat transfer coefficient during future operation. Deviation of the measured from the predicted coefficient will indicate anomalous behavior and instrumentation malfunction.

A Fortran code has also been developed that will perform routine analysis of the SRE AIHX heat transfer coefficient during forthcoming Core III operation.

The SRE reactivity computer program has been revised to conform to the Core III configuration, and has been checked out using predicted values of nuclear parameters. Some minor changes in the nuclear constants used in the reactivity code will be made when results of the initial nuclear testing of Core III are made available.

Efforts are continuing on development of the radial power distribution code. This computer program is similar to the one used on G. O. 7598, S. A. 1310, HNPF-OAP Analysis of Reactor Performance.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The development of operational analysis techniques by this project before operation of SRE Core III will provide the necessary tools to evaluate the reactor and its systems. This will ensure that at the time the plant begins operation, evaluation can be performed without delay. Also, the evaluation by this project of the significance of data obtained from the examination of SRE core components by other projects will be used in the overall surveillance activities. Finally, the continued evaluation of the results of material tests and examination of components and sample specimens that have been exposed in the SRE provides the overall program with information needed for operation of the systems.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Sodium Reactor Experiment and Power Expansion Program	
Project: Handling and Shipment of Radioactive Waste Materials to Burial Sites	
Project Engineer: L. E. Glasgow	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-22
General Order: 7602	Subaccount: 1360

I. PROJECT OBJECTIVES

To provide support for the SRE and Power Expansion Programs by the collection, processing, packaging, storage, shipping and disposition of all radioactive waste materials generated by the project.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

All of the low level radioactive wastes from program activities during the year, were received for disposal. Approximately 15,000 weighted gallons of SRE and PEP wastes were processed and sent to land burial at the Nuclear Engineering Company (NEC) site at Beatty, Nevada. No known back-log of such materials remain for disposal.

A total of 86 moderator elements and 33 reflector elements, after encapsulation in "cans" as they were removed from the reactor, were moved to RMDF vault storage in the SRE moderator shipping cask. These elements are presently scheduled for shipment to land burial during GFY 66.

In order to ship the elements over public roads, it was necessary to modify the cask to comply with ICC and AEC regulations. The modifications were completed ahead of schedule.

A low level, radioactive liquid waste processing system became fully operable during the last quarter. The system is designed to materially reduce future costs to project for the disposal of liquid wastes.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

All program objectives were attained within budget and on schedule.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Advanced Sodium-Cooled Reactors	
Project: Power Expansion Program (SRE-PEP) Safety Analysis	
Project Engineer: W. J. Freede	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-22
General Order: 7602	Subaccount: 1370

I. PROJECT OBJECTIVES

The objectives of this project are to compile and publish a final Safety Analysis Report for the SRE-PEP program, and to obtain approval for operation.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

The Safety Analysis Report for SRE-PEP has been printed and distributed during the first week of the report period, but it had been prepared during the previous fiscal year. Subsequent review by DRL and Chicago Health-Safety contributed to improvement changes in the document. Two chapters have been reorganized, and other clarifications have been made by text rephrasing or by discussion in an addendum. A series of review questions from DRL have been answered, and a second addendum to the report has been prepared to emphasize certain design conditions.

Much of the original transient analyses has been reviewed, and new analyses made of cases as defined within the questions received from DRL. The maximum credible accident (MCA) has remained the release of cover gas at the rate of 45% per day during the melting of one equivalent fuel element. Seismic analyses have been made of the primary containment components in accordance with criteria applied recently to other reactors under review.

Based on the material already submitted, it is expected that approval for operation will be obtained; however, direct presentation of the cases to DRL probably will be required for timely review.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

Structural analysis of the reactor and primary system using the seismic accelerations of a recorded earthquake confirm the containment integrity of the vessels. Minor damage can occur to gallery walls and pipe systems in a strong earthquake, but such members are adequately backed up by other structural members. The plant appears to be within generally accepted design criteria for earthquakes.

Review of the original analysis and treatment of additional specific cases, as posed by DRL, confirmed both the original classification of accident cases and selection of the maximum credible accident for SRE-PEP.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Sodium Reactor Experiment and Power Expansion Program	
Project: Handling and Shipment of Source and Special Nuclear Materials Scrap to Recovery Sites	
Project Engineer: L. E. Glasgow	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-22
General Order: 7602	Subaccount: 1920

I. PROJECT OBJECTIVES

The objectives of this project are to provide support to the SRE and Power Expansion Programs by collection, processing, packaging, storage, and shipping of both unirradiated and irradiated source and special nuclear scrap materials.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

The scheduled transfer of 64 SRE Core II fuel elements was completed. The fuel was encapsulated at the SRE, shipped to Radioactive Materials Disposal Facility (RMDF), and is now stored in Vault Building 022.

Completion of the Core II transfers brought to 122 the total units of Core I, Core II, and associated hardware items stored at the Building 022 vault.

SRE Core III unirradiated fuel scrap was transferred to Nuclear Materials Management Unit vaults for storage, pending eventual ultimate utilization in fabricating Core IV driver rods. No RMDF handling was required for this task.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

All program objectives were attained.

A building to house a uranium oxidation unit was completed, and the associated oxidation equipment installed. Preliminary tests and engineering evaluation of the installation indicated that further study and modifications to the equipment would be necessary before the unit could become operational. Operation of the equipment was deferred until GFY 1966.

A slightly different approach toward oxidizing uranium carbide was devised. A bench-scale prototype was fabricated at the RMDF to demonstrate the method. Experimental operation of the unit with uranium feed stock was inconclusive. Further work in this area will be recommended, but has been deferred pending programming.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: SRE-PEP	
Project: Miscellaneous Modifications	
Project Engineer: J. E. Owens	
Reporting Period: Fiscal Year 1965	AEC Category: 58-e-11
General Order: 7642	Subaccount: All

I. PROJECT OBJECTIVES

The objective of this project is to provide miscellaneous modifications to the SRE which are not included as part of the major construction work. Examples are: (a) new control room instrumentation, (b) sealing of the galleries and vaults, (c) modifications to the fuel-handling equipment, (d) new startup source, (e) new radiation monitoring equipment, and (f) preparation for and performance of preoperational tests on the system.

The purpose of this work is to ensure that the objectives of the SRE-PEP program are met in a safe and efficient manner.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

Construction and installation of the new nuclear and plant protective instrumentation were completed. Testing of the components and the installed system was also completed, and the system was placed into service on schedule. This new instrumentation consists of standard line components with proven reliability, and has the following design features: (1) solid-state construction using high quality components, (2) completely modular construction which facilitates maintenance, (3) triple redundancy in the logic system which meets the criterion that failure of any logic element can neither inhibit nor cause a scram, (4) a manual malfunction monitor which permits periodic checking of the operability of the logic system.

Rewiring of the control room was completed on schedule for SRE-PEP startup. This included replacement of the existing relay rack, modifications to the control console, and miscellaneous changes in the control room layout.

Sealing below the floor level in all galleries and vaults was completed. An epoxy elastic sealer (EC-801) was used to seal noticeable cracks in the concrete surface, concrete-to-metal edges, and separations around pipe penetrations. Obsolete pipe penetrations were welded or bolted closed, and bellows were inspected and rewelded where necessary. The concrete surface was then painted with two coats of primer and two coats of vinyl; the metal floor liner was covered with one coat of primer and two coats of vinyl. This vinyl coating (Amercoat No. 33) was selected after careful consideration of the required characteristics. It remains nonbrittle and thus easily follows the expansion or contraction of surfaces. It will not support combustion. Tests at Oak Ridge showed that this vinyl held up satisfactorily after exposure to 5×10^8 roentgens.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Engineering and fabrication of a carriage for the Mark II fuel handling machine (FHM) were completed ahead of schedule and within the allotted funding. A design was developed to utilize salvaged components from the Piqua mockup FHM carriage. By shortening and reinforcing the frame and by adding four additional wheels, the capacity was increased from 30 to 82 tons to meet anticipated requirements. The fabrication was carried out in AI shops. The new carriage was installed and is operating satisfactorily.

The safeguards analysis indicated that the control system for the Mark II FHM should be modified to eliminate the possibility of a fuel-handling accident. A new interlock sequence was incorporated, and the control schematic for the new control system approved. This new system insures that all operations with the machine will be performed in the proper sequence, and thus eliminates the possibility of operator error. A topical report (NAA-SR-10817 "SRE Mark II Fuel Handling Machine", by H. B. Dietz) which describes in detail the safety features of the new machine was issued. Installation of the new control system for the machine is in progress.

Design, fabrication, and irradiation of a new antimony-beryllium startup source were completed. Much of the existing hardware was reused to keep the cost to a minimum. The new antimony-oxide gamma source element consists of four separate capsules which may be used in combination for whatever source strength is necessary. Each capsule contains approximately 22 gm of Sb_2O_4 , which was converted from reagent grade Sb_2O_3 by high-temperature oxidation. The inner shield plug and hanger rod for the Sb_2O_4 source capsules slip inside an outer shield plug and hanger thimble for the beryllium collars. Design is such that the two hanger assemblies cannot be lifted from the reactor together, thus eliminating fast neutron shielding problems for the fuel-handling machine. The source has been installed in the reactor and is providing adequate neutron flux for startup instrumentation response.

A new stack radiation monitor and a number of remote area monitors (RAM) were installed. The stack radiation monitor uses a Tracerlab MAP-1/MGP-2 air particulate and gas monitor, with the gas sample taken at the stack exit. The RAM system includes monitors for the following areas: wash cell, liquid waste, vault cooling, gallery cooling, and fission product monitor ballast tank.

A sump well was drilled to about 30 ft below the high-bay floor in the trench just north of the storage cells. Its purpose is to detect any water which collects around the storage cells. Any ground water which might collect in the well will be pumped out if the water reaches a prescribed level. The system has been installed and tested.

A number of modifications were made to the helium system. These include moving pressure control stations from the outside trench areas to more accessible wall areas, removal of unused control stations, and installation of an integrating flowmeter in the helium makeup line to the reactor to measure leakage during operation.

Test summaries and step-by-step test procedures were written for the preoperational tests. These include: (1) fill and function check of sodium systems, (2) vibration check of heat exchanger and pumps, (3) function check of

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

N₂ cooling system, and (4) test and calibration of nuclear instrumentation and plant protective system. The test summaries are identical in format with those prepared for other tests under Project 7602-1330 (SRE Engineering). They contain all information necessary for a complete review of the tests and, with the addition of step-by-step procedures, were used to perform the tests. All preoperational tests except the function check of the N₂ cooling system were completed and the analysis of test results is in progress.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The new nuclear and plant protective instrumentation will greatly reduce the number and frequency of spurious scrams during future operation. Instrumentation of proven design and reliability was selected to ensure excellent performance at a reasonable cost and with a minimum procurement time.

Rewiring of the control room has produced a neat and logical cable arrangement which will greatly facilitate maintenance.

Sealing of the primary sodium system galleries will reduce the nitrogen cover gas consumption, thus reducing reactor operating costs.

Installation of the new carriage for the FHM has permitted expedient scheduling of construction work, FHM modifications, shipping of Core I fuel, and replacement of the gallery blocks. The cost of the work was significantly reduced by modifying the Piqua machine rather than by constructing a completely new carriage. At the same time, all desired objectives for the carriage were met.

The new neutron source provides excellent flexibility in source strength. Depending upon source-detector geometry, and time lapse between irradiation and startup, it may be necessary to have a stronger source to satisfy startup interlocks. The four separate Sb₂O₄ capsules may be chained together with simple hot-cell techniques to provide a gamma source of up to 400 curies after one irradiation cycle at the MTR.

Addition of the sump well next to the storage cells will ensure integrity of the cells against rust caused by groundwater collection. Thus, the possibility of radioactive contamination of ground water through cell leakage is eliminated.

Modifications to the helium system will reduce helium leakage by eliminating unnecessary piping and pressure-control stations. In addition, operation and maintenance will be simplified through relocation of equipment for easy access, resulting in large time-savings. The SRE-PEP Safeguards Analysis Report showed the need for monitoring the rate of cover gas leakage by measuring the helium makeup rate. Use of an integrating flowmeter on the makeup line to the core and pumps will provide an accurate record of core cover gas usage and leak rate.

The test procedure summaries have expedited both internal and external reviews by providing concise descriptions of the tests with the important precautions and prerequisites. The inclusion of definite, clearly stated criteria which must be met by the test has been especially helpful.

SECTION II LARGE SODIUM-GRAPHITE REACTORS CONTENTS

<u>Title</u>	<u>Identification</u>	<u>Page</u>
LARGE SODIUM-GRAPHITE REACTORS	Summary	II-1
SGR DESIGN AND EVALUATION	7519 - 1310	II-4
AUSTENITIC STAINLESS STEELS FOR SERVICE IN A HIGH TEMPERATURE SODIUM ENVIRONMENT	7519 - 1951	II-6
UC FUEL ELEMENT DEVELOPMENT	7519 - 1953	II-19
LSGR FUEL ELEMENT DEVELOPMENT	7519 - 1955	II-33
DEVELOPMENT AND EVALUATION OF UC ALLOY FUEL MATERIALS	7519 - 1957	II-37
UC PROCESS DEVELOPMENT	7519 - 1959	II-48
SRE CORE-III TEST ELEMENTS	7519 - 1961	II-53
SGR DRY CONTROL ROD CONCEPT DEVELOPMENT	7519 - 3210	II-55
SGR DRY CONTROL ROD PROTOTYPE DEVELOPMENT	7519 - 3220	II-58
SGR WET CONTROL ROD CONCEPTUAL DEVELOPMENT	7519 - 3230	II-59
SGR CALANDRIA STRUCTURAL TESTS	7519 - 3610	II-60
SGR CALANDRIA MAINTENANCE	7519 - 5210	II-68
SGR CORE MAINTENANCE, PROTO- TYPE DEVELOPMENT	7519 - 5220	II-77

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program:	Large Sodium Graphite Reactors
Program Manager:	K. W. Foster
Reporting Period:	Fiscal Year 1965

I. PROGRAM OBJECTIVES

The objective of this program is to provide most of the technical information on which to base the construction of a large SGR. The program is aimed at a 1000-Mwe plant, preceded by a prototype of about 200 Mwe. The design bases for these plants depart substantially from those for the SRE and HNPF, particularly in higher operating temperatures and increased specific power. These changes and the increase in power level are required to make the SGR competitive with other energy sources. The changes impose more stringent requirements on the reactor core structure and fuel elements. Development work on these components, together with expanded effort on UC fuel and continuation of work in the more general ASGR program on reactor physics, sodium components, and reactor safety, is expected to meet the objectives of this program.

II. PROJECT MANAGER'S SUMMARY AND EVALUATION

During the past quarter, steps were initiated to phase out the large SGR program in order to conserve funds and resources for support of the fast breeder reactor concept. Recent evaluations of fast reactor technology indicate that a concerted effort on this potentially more attractive reactor concept can result in a commercially available plant at an earlier date than was previously thought possible. In view of these considerations, a thorough examination of the LSGR program activities was made to determine those areas which are of continued interest to the AEC program for the fast breeder reactor. In the area of component development, activities were directed to confirm the adequacy of specific SGR design features. These efforts do not contribute in a substantial manner to the fast reactor concept; hence, activities of this nature were terminated. In the area of fuel work, there is a close relationship between much of the development of fuels for thermal and fast reactors. In general, many of the LSGR fuel activities were reoriented and incorporated in the fast reactor development program. At the close of FY 1965, the LSGR program will have been terminated, and those activities that relate to continuing projects will have been merged with the applicable program.

Efforts during the fourth quarter converged on terminating development activities and documenting results of LSGR systems and component studies. The conceptual design of a 1000-Mwe SGR and its 200-Mwe prototype was presented as a 2-volume report (NAA-SR-9213) in December 1963. Since that time, additional design and analyses of the prototype reactor structure were undertaken. This work is summarized in NAA-SR-11314, entitled "200-Mwe Prototype Large SGR Reactor Structure Design and Evaluation;" and follows the general outline of: (1) Reactor Structure Design Description, (2) Operating Environment, (3) Reactor Steady-State Studies, (4) Reactor Transient Studies, (5) Material Selection, (6) Mechanical-Structural Analyses, and (7) Calandria Maintenance.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

The results of these studies indicate a highly reliable reactor, conservatively designed to fulfill the operating requirements of the 200-Mwe SGR prototype. Existing technology is considered satisfactory in most areas, with some reservations pertaining to long-term exposure of stainless steel and graphite to fast neutron damage. Further analyses are required to complete the design studies, but to date no problems have been encountered that would require major design modifications. If an SGR prototype construction project was initiated, an associated development program would be advisable to confirm some of the design configurations and details established from the analyses.

Development activities on the LSGR control rod were completed with the fabrication of five prototypical LSGR absorber columns for the SRE-PEP control rods. These rods will provide information concerning the effect of long-term irradiation on this configuration of materials (Hastelloy-X, dysprosium oxide, and graphite). Based on the successful completion of an elevated temperature, full-scale absorber column test, which simulated approximately five years of reactor type operation, it appears that a thimble-enclosed concept will meet performance specifications of the LSGR. Laboratory wear tests of Hastelloy-X versus Type 304 stainless steel and compatibility tests of Hastelloy-X with dysprosium oxide provided corroborative evidence that this design configuration is adequate.

Reactor and calandria structural component tests were terminated, and final efforts were directed toward report preparations. Test results available before termination revealed no major areas of concern, but additional testing is required to confirm the adequacy of the structures.

Cutting and welding operations for remote replacement of process tubes were concluded, with satisfactory demonstrations that equipment and techniques will consistently produce good results. Testing of the graphite-handling machine proved its general operability for remote handling of graphite logs in the core. A final report of these development activities has been prepared. Data from experiments to determine dilation characteristics of irradiated graphite when contacted by liquid sodium indicate that graphite removal problems may develop unless adequate clearances are provided in the design between adjacent logs. Test results show that dimensional expansions up to 5% are possible after radiation exposure of approximately 1.1×10^{21} nvt (>0.18 Mev).

The FY 1965 LSGR fuels program continued to concentrate on the development of uranium carbide, sodium-bonded, 304 SS clad fuel elements. Emphasis during the first three quarters of FY 1965 was placed on: (a) 304 SS clad properties in an LSGR environment; (b) large-scale fuel rod irradiations at the NRX Canadian reactor; (c) long burnup capsule irradiations of hypo UC under LSGR conditions; (d) fabrication of full-scale UC test elements for SRE Core III; (e) improvements in UC processing to more closely control the carbon content; and (f) UC phase equilibria. Contributions to uranium carbide fuel element technology were made in each of these areas. During the last quarter of FY 1965, a major change in technical scope resulted in the early completion of all LSGR oriented tasks. Increased emphasis was initiated in March-April to implement fast reactor carbide fuel technology by transferring applicable tasks from the LSGR fuels program and reviewing the status of fast fuel at all sites. All LSGR fuel program tasks were completed by the end of the year.

The progress made on 304 SS showed the necessity for a continued high level effort on fuel clad materials. Only limited work is being performed at other sites at conditions of interest. Highlights of this year's effort showed: (a) importance of heat-to-heat composition and grain size on stress rupture properties; (b) importance of boron impurity content causing a loss of ductility in a reactor environment; (c) availability of seamless tubing with the same dimensional tolerance as for welded and drawn tubing; and (d) progress in the initiation of a large-scale irradiation test for in-pile stress rupture properties. These efforts directly apply to the fast reactor program and require a continued high level of investigation.

Two representative LSGR fuel rods, designated NRX-86-1 and -2, were fabricated and inserted in NRX. These rods have performed well at over 90% of the peak design conditions. A peak burnup of 11,000 Mwd/MTU will be completed by the end of FY 1965. These rods are recommended for continuation under the Hallam Fuel Surveillance Program to which they are most applicable.

Despite a few failures of UC materials irradiations, two highly successful tests at 1800°F and burnups of ~17,000 and 32,000 Mwd/MTU confirmed the high performance capability of this fuel for LSGR design conditions. In both cases, the fission gas release and density decreases were low. Thorough investigations of the failed capsules contributed to a greater understanding of the performance of this fuel at very high temperatures. These total data further confirmed the prediction that radiation stability increases with decreasing free uranium in hypo UC. The true capability of UC has not been tested. Higher temperature operation as well as longer burnup appear well within the capability of this fuel material. Design data on UC fuel performance were revised based on this information.

The reduced scope of the LSGR program resulted in a decrease of the test elements for SRE Core III. Four instrumented test elements were fabricated and assembled. These test elements are necessary to the reactivity requirements of the plant. SRE Core III represents the best short-term potential sodium reactor test facility for either thermal or fast fuel element test programs.

The performance requirement of near stoichiometric UC composition emphasized the need for UC processing feasibility and capability to produce this material. Two processes were studied on a small scale feasibility level: (a) hollow cathode discharge (HCD), and (b) plasma arc melting. Both processes omit the use of a graphite electrode and thus omit the uncontrolled source of carbon. Results of the HCD tests indicate an excellent potential for producing UC to ± 0.1 wt % C, whereas present processes control to ± 0.2 wt % C. Results of the plasma arc work are being evaluated. This process investigation has been proposed to be continued under the fast reactor program.

Further studies of the microstructure and physical properties of uranium carbide both in-pile and out-of-pile, with varying compositions, impurities such as O₂ and N₂, and heat treatments provided a greater understanding of the mechanisms of radiation stability and interpretation of radiation results. The solubility of uranium in UC versus temperature and impurity content received particular attention. Additional effort in this phase of work is recommended for a thorough understanding of both UC and UC + PuC.

As a result of the work performed on this and associated programs, hypo UC has been shown to be capable of high specific power operation to high burnup at competitive fuel cycle costs.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Large Sodium Graphite Reactors

Project: SGR Design and Evaluation

Project Engineer: K. W. Foster

Reporting Period: Fiscal Year 1965

AEC Category: 04-01-04-21

General Order: 7519

Subaccount: 1310

I. PROJECT OBJECTIVES

The project objectives are to evaluate mechanical arrangements, nuclear characteristics and economics of large SGR concepts in order to minimize power costs, increase reliability, and improve safety.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

All efforts during the fourth quarter converged on documentation of analytical and design studies of the 200-Mwe prototype reactor structure. The conceptual description of the 1000-Mwe large SGR and its 200-Mwe prototype was presented in Volumes I and II of NAA-SR-9213, issued in December 1963. Additional analysis and design since that time constitute the major portion of the 200-Mwe prototype reactor structure thermo-mechanical design. This work, summarized in NAA-SR-11314 entitled, "200-Mwe Prototype Large SGR Reactor Structure Design and Evaluation," issued in June 1965, follows the general outline of: (a) Reactor Structure Design Description, (b) Operating Environment, (c) Reactor Steady-State Studies, (d) Reactor Transient Studies, (e) Material Selection, (f) Mechanical-Structural Analysis, and (g) Calandria Maintenance.

The reactor structure design description covers all major components contained within the outer perimeter of the concrete cavity, and includes a listing of plant data on core performance parameters, core design values, and fuel element data. Some eight major design improvements incorporated in the prototype reactor structure since December 1963 are described in detail. Included in these are changing from brazed to welded process tube joints, increasing the calandria internal pressure to offset external static sodium pressure, relocating process tube bellows to the bottom end to reduce axial bowing loads, utilizing a thin-wall sleeve over the process tube to protect the graphite from inleaking sodium, leak detection and leak location systems, core graphite and reflector graphite log designs, and structural keys for seismic loads.

The operating environment section defines the operating requirements imposed on the reactor from the standpoint of the plant control system and the turbine-generator requirements. Load-following ranges are described along with load-change rates and plant operating limits. Setpoints for plant protective system actions of alarm, setback, fast shutdown, and scram are listed for all monitored operating parameters.

The reactor steady-state studies section summarizes all nuclear, hydraulic, and heat transfer work done to date. Graphic presentation of neutron flux

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

profiles for both fast and thermal flux in the core, reflector, unit cell, and control element cell are given. Reactor hydraulics studies include both the forced flow and convection cooling systems analyses. The capability of the reference orifice design to accommodate cores of mixed UC and UO₂ elements is shown. Heat generation rates and temperature profiles throughout all major reactor structure components are well documented. Fuel channel temperature extremes resulting from various design uncertainties establish justification for the 1200°F design temperature of the calandria structure. Methods for determining maximum operating temperatures of control elements and the methods for cooling are presented. Graphite temperature profiles throughout the core and reflector have been determined.

The section on transient studies gives a complete list of plant transients. The transients are divided into operational and accident transients, where the frequency of occurrence is defined and the resulting kinetic response is presented in the form of temperature history curves.

The materials selection section discusses the materials of construction, giving the basis for their selection and the design properties used in the analysis. Both graphite and stainless steel are covered. For the graphite irradiation, distortion and sodium dialation are treated. The stainless steel discussion includes creep, stress-rupture, irradiation effects, and corrosion rate estimates.

The mechanical-structural design summary includes the basic loading diagrams for the reactor structure complex. The stresses due to primary loads are determined and compared with ASME Nuclear Vessel Code Values. The effects of transient conditions on critical areas have been investigated and reported. The seismic analysis is summarized, indicating the ability of the structure to withstand lateral dynamic loading. The effects of creep buckling on the process tube and calandria shell are related to life expectancy and component reliability.

The maintenance section treats the calandria remote maintenance philosophy, giving the procedures for process tube replacement, core graphite replacement, and reflector graphite replacement. These features are utilized only if a sodium leak occurs, and are not a planned maintenance function.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The results of these analytical studies indicate a highly reliable reactor, conservatively designed to fulfill the operating requirements of the 200-Mwe SGR prototype. Existing technology is considered satisfactory in all areas with some reservations pertaining to long-term exposure of stainless steel and graphite to fast neutron damage. Further analysis is required in the area of the upper weld joint of the calandria process tube due to excessive fatigue stresses. Other than this, no problems have been encountered that would require major design modifications.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Large Sodium Graphite Reactors	
Project: Austenitic Stainless Steels for Service in a High Temperature Sodium Environment	
Project Engineer: B. R. Hayward	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-21
General Order: 7519	Subaccount: 1951

I. PROJECT OBJECTIVES

The major objective of this project has been the determination of the mechanical behavior of austenitic stainless steels for service in the core region of large sodium graphite reactors at operating temperatures of 1300°F maximum.

Specific objectives for this fiscal year were to: (1) obtain further information on the effect of simultaneous carburization and pressurization of tubular fuel cladding as compared with uniaxial properties of the same material; (2) continue studies on the influence of chemical composition upon stress-rupture properties; (3) begin studies on the effect of chemical composition and grain size on the biaxial low strain rate behavior of 304 stainless steel, and (4) initiate an investigation of the effect of radiation on 304 stainless steel in a large SGR environment.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. EFFECT OF CLAD CARBURIZATION ON STRESS-RUPTURE PROPERTIES (TASK 1)

Twelve 0.010-in.-wall tubular specimens of welded and drawn 304 stainless steel were subjected to internal pressure tests at elevated temperatures. Test conditions included study of the effect of carburization on the stress-rupture properties. Hyperstoichiometric UC was used as the carbon source. An outline of the test conditions and status is shown in Table 1. To date, rupture times have been obtained for nine specimens.

The rate of pressure increase, of course, has a definite effect on the strain behavior of the tubing. A comparison of the diametrical increase in fueled tubes loaded from 0 to 500 psig in 2 yr and from 0 to 500 psig in 90 days is shown in Figure 1. Dorn* has proposed that strain, time, and stress are related at high stress, by the equation

$$E = af(t) \exp(b\delta) ,$$

where

E = strain

t = time

δ = stress (a function of time in this case)

a, b = empirical constants (not material constants, per se)

*Dorn, J. E., "Some Fundamental Experiments on High Temperature Creep," J. Mech. Phys. Solids 3, 85 (1955)

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 1
STATUS OF PHASE-I TESTS ON JUNE 1, 1965

Specimen ^(a)	Internal Environment of Specimen	Test Temperature (°F)	Pressure (psig)	Time (hr)	Average Rate of Pressure Increase ^(b) (psig/day)	Maximum Diameter Strain (%)
1 ^(c)	Sodium Bonded Hyperstoichiometric Uranium Monocarbide	1,250	200	6,456	0.7	2
3 ^(d)		1,300	500	384	500	Not Measured
4 ^(d)		1,300	350	2,448	350	Not Measured
5 ^(c)		950	92	2,832	0.7	0
6 ^(d)		950	1,425 ^(f)	6,046	5.0	0.2
7 ^(c)		1,150	200	6,456	0.7	1
9 ^(d)		1,150	920 ^(f)	4,438	5.0	5
10 ^(d)		1,250	630 ^(f)	3,624	5.0	7
11 ^(d)	Sodium Only	1,250	500	2,992	500 ^(e)	4.5
2 ^(d)		1,250	405	13,608	5.0	Not Measured
12 ^(d)		1,250	500	888	500 ^(e)	7

- (a) All specimens are 12-in. long, 0.010-in. thick, and 0.540-in. ID of welded-and-drawn 304 stainless-steel tubing with welded 304 stainless-steel end caps.
(b) Pressure is increased during weekdays only and corrected monthly to a rate at which the 0.7 psig/day specimen will reach 500 psig after 720 days and the 5 psig/day specimen will reach an internal pressure of 500 psig after 90 days.
(c) Replaced on 6/1/64.
(d) Failures.
(e) Constant at 13,500 psi hoop stress.
(f) Pressure is increased at the noted rate until failure occurs.

It can be seen from Figure 1 that the tests at the higher pressurization rate may follow this relationship, while the tests at the lower pressurization rate tend to follow a linear relationship. It would be convenient to state that the lower pressurization rate curves represent the early portion of the higher pressurization rate curves; however, this does not appear to be the case since the diametrical strain achieved is approximately the same.

7519-2501
Figure 1. Effect of Rate-of-Loading on Strain

The strain-to-failure is relatively constant regardless of the mode of stressing. Table 1 shows the test conditions and failure ductility. Failure times are comparable to those determined by uniaxial tests, in that the biaxial points fall within the range of the uniaxial data spread. All failures to date have occurred in the weld area. The grain size (ASTM >10) in the weld area is two orders of magnitude smaller than the grain size (ASTM 4-5) of a normal area of the tube. High-temperature creep and stress rupture behavior are frequently better for coarse-grain than for fine-grained material.* A number of investigators have also found that grain-boundary sliding is strongly

*Deiter, G. E., Mechanical Metallurgy, McGraw-Hill (New York, 1961) p 363

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

related to crystal slip within the grains.* Since these two processes account for the greater portion of creep, it is easy to see why failures are occurring in the fine-grained section where the grain boundary concentration is highest. The fact that the failures occur with a high local strain and by grain boundary separation lends additional credence to the above explanation. The cracks formed radiate from the axis. Kennedy, Harms, and Douglas,[†] also found intergranular cracks in a normal direction to the principal stress using Inconel at 1500°F.

The results support a failure criterion based on the maximum-principal stress. That is, the maximum stress can be used to predict failure without any effects from the other principal stresses present. This has been substantiated by several investigators.^{†, §}

The carburization behavior of the stainless cladding is typical of the UC-stainless steel compatibility studies by Elkins.** The UC₂ phase of the fuel, present as a Widmanstätten phase, decomposes giving up carbon for transport to the cladding. Anderson and Sneesby,^{††} in previous work at AI have demonstrated the carburization characteristics of stainless steels in liquid sodium, and found the reaction between carbon, sodium, and stainless steel proceeds rapidly to equilibrium at elevated temperatures.

At these testing temperatures, sensitization of the steel also takes place, which allows the sodium to leach away the complex chromium carbides formed. This occurs at every grain boundary, thus reducing the cross-sectional area. This effectively increases the stress, and promotes creep.

Results of these experiments permit other conclusions to be drawn in addition to those involving the effect of stress on a sodium-stainless steel fuel element model using hyper-UC as a carbon source. (1) It was found that massive carburization of the cladding takes place in the 950 to 1350°F temperature range by decomposition of the hypostoichiometric uranium carbide fuel. (2) The most severe carburization takes place in the cladding adjacent to areas of maximum fuel surface, e.g., opposite fuel slug ends. (3) The areas of highest carbon concentration exhibit the greatest amount of deformation. (4) The strain-to-failure seems to be independent of temperature environment, or rate of pressure increase, as seen in Table 2.

*Kennedy, A. J., Processes of Creep and Fatigue in Metals, John Wiley & Sons (New York, 1963) p 193-212

†Kennedy, C. R., Harms, W. O., and Douglas, D. A., "Multiaxial Creep Studies on Inconel at 1500°F," J. Basic Engr. (December 1959) p 599

§Finnie, I., and Heller, W. R., Creep of Engineering Materials, McGraw-Hill (New York, 1959) p 199

**Elkins, P. E., "Compatibility of Uranium Carbide Fuels with Cladding Materials," NAA-SR-7502 (August 1, 1964)

††Anderson, W. J. and Sneesby, G. V., "Carburization of Austenitic Stainless Steel in Liquid Sodium," NAA-SR-5282 (September 1, 1960)

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 2
STRAIN TO FAILURE CHARACTERISTICS

Specimen	Temperature (°F)	Strain to Failure (%)	Environment	Failure Time (hr)	Rate of Pressure Increase (psig/d)
3	1300	6.5	Hyper*	384	0
9	1150	5.0	Hyper*	4428	5
10	1250	7.2	Hyper*	3624	5
11	1250	4.0	Hyper*	2992	0
12	1250	6.1	Sodium Only	888	0

*Hyperstoichiometric UC fuel

**B. EFFECT OF CHEMICAL COMPOSITION ON STRESS-RUPTURE BEHAVIOR
(TASK 2A)**

Thirty-nine uniaxial stress-rupture tests were performed at 1250°F to determine the effect of chemical composition on long-term mechanical behavior. The tests were performed on 304 SS 10-mil-wall by 5/16-in.-OD tubing in helium.

Type 304 austenitic stainless steel stress-rupture data display a wide variation as shown in Figure 2. Data usually exhibit failure times which vary by a factor of 5 to 10 at the same level of stress. An individual heat usually displays excellent linearity when log stress-to-failure is plotted against log time-to-failure. Therefore, as seen in Figure 2, roughly parallel plots of individual heats produce the wide band of stress-rupture data. The heats containing the greatest amount of carbon and nitrogen are the heats with the greatest strength.

7519-2502

Figure 2. Type 304 Stainless Steel Rupture-Stress at 1250°F

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

A plot of time-to-rupture at constant stress against total carbon and nitrogen content (see Figure 3) shows that time-to-failure increases exponentially with added amounts of carbon and nitrogen at 1250°F. Although carbon and nitrogen are believed to be of primary importance, additions of phosphorus and sulfur aid in yielding a closer data fit. The effect of total phosphorus, sulfur, carbon, and nitrogen is shown in Figure 4.

The strengthening is probably due to the fact that metallic carbide, nitrides, sulfides, and phosphides have structures which do not possess well-defined slip planes, and therefore, prohibit plastic flow in the matrix material. Additional work is under way to evaluate the role of phosphorus and sulfur, and to increase the reliability of strength predictions made on the basis of chemical analysis.

7519-2503

Figure 3. Effect of C and N
on Rupture Stress

7519-2504

Figure 4. Effect of C, N, P, and S
on Rupture Stress

C. EFFECT OF CHEMICAL COMPOSITION AND GRAIN SIZE IN THE FAILURE OF 304 SS SEAMLESS TUBING (TASK 2B)

Equipment was constructed for the simultaneous pressure-stress rupture testing of 96 specimens to improve the credence of the carbon-nitrogen strengthening, as discussed in the previous section. The tests are biaxially loaded (pressurized) at 1250 and 1350°F, and are performed with seamless tubing specimen material which will eliminate grain-size discontinuities present in weld-and-drawn tubing specimens.

The new test apparatus, consisting of creep frames, pressure system, furnaces, and controllers was installed. Each furnace is capable of testing four specimens simultaneously as seen in Figures 5 and 6. Each 4-specimen retort contains three thermocouples; one placed at the top of the specimens, one at the center, and one at the bottom of the specimens. These have a maximum temperature profile of $\pm 2^\circ\text{F}$. Each specimen is pressurized independently and has a pressure gage that indicates the loss of pressure that accompanies failure.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

— Figure 5. Task-2B Retort Cover
Showing Attachment and
Replacement

7519-2505

Figure 6. Two of 24 Task-2B Pressure
Stress-Rupture Frames Showing
Retorts in Furnaces and
Pressure Gages —

7519-2506

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Specimens were fabricated from six different heats of 304 stainless steel and two heats of 316 stainless steel. The analysis of the individual heats is shown in Table 3. One hundred and thirty-two specimens have been placed in test; to date, 49 have failed. The status of the tests is given in Table 4. Failures have occurred with between 16 and 30% plastic strain, and have been long 1- to 3-in. axial tears which are not characteristic of Task 1 failures where sodium and fuel are present on Task 2A welded and drawn specimens. Metallographic analysis of the region near the failures shows large amounts of plastic deformation of the grain boundaries as seen in Figures 7 and 8. Many intergranular voids are in evidence indicating a creep-type failure.

TABLE 3
CHEMICAL COMPOSITION OF TASK-2B SPECIMENS
(wt %)

Heat	Code	C	N	Mn	P	S	Si	Ni	Cr	Mo
27898X	A	0.06	0.060	1.67	0.011	0.011	0.50	10.41	18.93	-
06737	B	0.06	0.056	1.64	0.022	0.025	0.39	9.82	18.54	-
06820	C	0.06	0.047	1.84	0.023	0.009	0.44	9.79	18.54	-
00042	D	0.05	0.052	1.77	0.023	0.021	0.55	9.70	18.55	-
90213	E	0.05	0.037	1.68	0.018	0.021	0.45	9.67	18.47	-
26980X	F	0.06	0.059	1.68	0.027	0.013	0.66	10.27	18.71	-
96180	G	0.03	0.027	1.54	0.003	0.021	0.30	13.39	17.01	2.28
90106	H	0.04	0.043	1.68	0.022	0.017	0.51	13.63	17.33	2.32

7519-2507
Figure 7. Task-2 Specimen Fracture

7519-2508
Figure 8. Biaxial Test Creep-Voids and Plastic Deformation

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 4
STATUS OF TASK 2B TESTS

Heat	Temperature (°F)	Maximum Stress (psig x 10 ³)	Test (hr)	Maximum Strain at Failure (%)	Heat	Temperature (°F)	Maximum Stress (psig x 10 ³)	Test (hr)	Maximum Strain at Failure (%)
A	1350	20	~3	20.5	A	1250	5	1344	†
B	1350	20	~3	18.4	A	1250	5	1344	†
B	1350	20	~3	20.8	B	1250	5	1344	†
B	1350	20	~3	25.9	B	1250	5	1344	†
C	1350	20	~3	30.6	A	1350	5	1368	†
C	1350	20	~16	26.7	A	1350	5	1368	†
C	1350	20	~16	18.9	B	1350	5	1368	†
D	1350	20	~16	26.3	B	1350	5	1368	†
D	1350	20	~16	29.8	A	1250	10	1344	†
D	1350	20	~16	17.9	A	1250	10	1344	†
E	1350	20	~16	16.6	B	1250	10	1344	†
E	1350	20	~16	20.9	B	1250	10	1344	†
F	1350	20	~16	28.0	C	1250	10	1344	†
F	1350	20	~16	26.1	C	1250	10	1344	†
G	1350	20	~16	24.7	D	1250	10	1344	†
G	1350	20	~16	25.4	D	1250	10	1344	†
H	1350	20	193	23.2	E	1250	10	1344	†
H	1350	20	~16	25.7	E	1250	10	1344	†
A	1350	15	~84	19.4	F	1250	10	1344	†
A	1350	15	~108	21.0	F	1250	10	1344	†
B	1350	15	96	26.9	G	1250	10	1344	†
B	1350	15	~84	26.4	G	1250	10	1344	†
A	1350	15	92	22.7	H	1250	10	1344	†
A	1350	15	~108	22.9	H	1250	10	1344	†
B	1350	15	96	24.0	A	1250	10	1344	†
B	1350	15	90	24.8	B	1250	10	1344	†
A	1350	13.5	286	*	C	1250	10	1344	†
A	1350	13.5	~156	20.6	D	1250	10	1344	†
B	1350	13.5	~204	27.0	A	1250	10	1320	†
B	1350	13.5	~204	19.7	A	1250	10	1320	†
A	1350	13.5	~156	21.3	A	1250	10	1320	†
A	1350	13.5	~156	24.9	B	1250	10	1320	†
B	1350	13.5	~204	20.0	B	1250	10	1320	†
B	1350	13.5	144	*	B	1250	10	1320	†
C	1250	10	1320	†	C	1250	10	1320	†
C	1250	10	1320	†	D	1250	10	1320	†
D	1250	13.5	1320	†	B	1250	15	1072	*
D	1250	13.5	1320	†	C	1250	15	1320	†
E	1250	13.5	1320	†	C	1250	15	1320	†
E	1250	13.5	1320	†	C	1250	15	1320	†
E	1250	13.5	1320	†	D	1250	15	1056	*
F	1250	13.5	1320	†	D	1250	15	1188	*
F	1250	13.5	1320	†	D	1250	15	952	*
F	1250	13.5	1320	†	E	1250	15	720	*
G	1250	13.5	1320	†	E	1250	15	~804	*
G	1250	13.5	1320	†	E	1250	15	720	*
G	1250	13.5	1320	†	F	1250	15	1320	†
H	1250	13.5	1320	†	F	1250	15	1320	†
H	1250	13.5	1320	†	F	1250	15	1320	†
H	1250	13.5	1320	†	G	1250	15	1320	†
A	1250	15	1072	*	G	1250	15	1320	†
A	1250	15	1212	*	G	1250	15	1320	†
A	1250	15	1116	*	H	1250	15	1320	†
B	1250	15	952	*	H	1250	15	1320	†
B	1250	15	1188	*	H	1250	15	1320	†

*Failure data not yet complete
†Specimen in Test

NAA-SR-11450

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

7519-2509
Figure 9. Specimen and Bellows Train Assembly for NAA-123
Irradiation Experiment

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

D. EFFECTS OF RADIATION ON PROPERTIES OF AUSTENITIC STAINLESS STEEL (TASK 4)

The main cause for ductility loss of 304 SS in a thermal neutron flux at temperatures above 500°C is hypothesized to be helium gas entrapment.* The helium is formed by the transmutation of B¹⁰ and causes complete loss of ductility when present in amounts exceeding 400 parts per billion.† This helium coalesces at the grain boundaries and reduces the energy for grain boundary rupture.* This loss in ductility and associated loss in rupture life are of critical importance to ISGR cladding which is planned to operate at 500°C and above. This effect is quite well documented for short rupture time in nonliquid metal systems, but to quantitatively evaluate it for ISGR conditions the NAA 123 experiment was conceived.

NAA 123 is a pressure-stress rupture experiment to be tested at Hanford. It consists of four subcapsules, each containing five specimens. The specimens are NaK-filled and immersed in NaK (Figures 9 through 12). The subcapsules will be gamma-heated and individually controlled in temperature through use of a variable helium-argon mixture in an outer shroud tube located between the NaK and the reactor coolant (Figure 12).

The 20 specimens are annealed seamless tubing of 0.010-in. wall by 0.500 ID by 4 in. long, made from four heats of 304 SS with B¹⁰ contents varying from 47 to 935 ppb. Ten specimens will be tested at 1200°F, and the other 10 at 1300°F. The testing schedule is shown in Table 5.

An auxiliary (physics) experiment was run in the reactor to determine the gamma heating rate in the hole for which the NAA 123 experiment is scheduled. Results of this experiment showed very uniform temperatures for long periods of time at constant gas compositions. It also showed the gamma heating rate to be optimum for the experimental design.

An out-of-reactor test of the temperature control system is planned for before the end of the current fiscal year. All components for the experiment are on order for immediate delivery. No procurement problems are anticipated. The assembly of the capsule is 25% complete. A mid-October 1965 shipping date has been firmly committed to Hanford.

E. EFFECT OF COLD WORK ON STRESS-RUPTURE PROPERTIES

Tests were initiated to isolate the effect of sodium and cold-work on the pressure-stress rupture behavior of 304 stainless steel. The specimens and test conditions are shown in Table 6. The first test results (specifically Specimens 3 and 4 when compared with 5 and 6, and Specimen 15 when compared with 16) indicate that a small amount of cold-work (~12% RA) increases the rupture life at a given stress.

*Barnes, R. S., "Mechanisms of Radiation Induced Mechanical Property Changes ASTM Annual Meeting, Chicago, July 1964.

†Wier, J. R. and Martin, W. R., "Radiation Damage in Stainless Steels," 4th Meeting of the Reactor Structure Materials Committee, November 1964.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Figure 10. Thermal Transition Joint

7519-2510

Figure 11. Pressurization Bellows

7519-2511

Figure 12. Specimen Pressurization Scheme

7519-2512

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 5
PRESSURIZATION SCHEDULE

Subcapsule No.	1					2				
Temperature (°F)	1200					1300				
Heat*	A	B	C	D	E	A	B	C	D	E
Differential Pressure (psig)	600	600	600	600	250	400	400	400	400	250
Gas Bulb Pressure at 70° F (psia)	208	208	208	208	92	134	134	134	134	87

Subcapsule No.	3					4				
Temperature (°F)	1300					1200				
Heat*	A	B	C	D	E	A	B	C	D	E
Differential Pressure (psig)	185	250	250	250	250	350	350	350	350	250
Gas Bulb Pressure at 70° F (psia)	67	87	87	87	87	125	125	125	125	92

This schedule was predicated on the assumption that the controlling gas bulb thermometer in each subcapsule would be in Specimen E. All other specimens also have gas bulbs, but these bulbs are not connected to controlling transducers.

*Heat

A
B
C
D
E

Origin

Hallam cladding
Temescal Metallurgical electron beam ingot 1016
Temescal Metallurgical electron beam ingot 1017
U. S. Steel air induction melt
Same as "A" except that these specimens contain gas bulb thermometers for the control system

TABLE 6
EFFECT OF SODIUM ON 304 STAINLESS STEEL* AT 1250° F

Test No.	Hoop† Stress (psi)	Static Press (psig)	Heat Treat	Inside Element	Outside Element	Status
1	7,000	250	Annealed	Sodium Bonded Hypo-U-Mo	Sodium Only	Removed (2856 hr)
2	7,000	250	Annealed			Specimen Leak
3	13,500	490	Annealed			Failed (336 hr)
4	13,500	490	Annealed			Failed (360 hr)
5	13,500	490	1/8-hard			8616 hr
6	13,500	490	1/8-hard			8616 hr
7	27,000	980	Annealed			Failed (24 hr)
8	27,000	980	Annealed			Failed (1 hr)
9	7,000	250	Annealed	Sodium Gettered	Sodium Only	8616 hr
10	13,500	490	Annealed			8616 hr
11	13,500	490	1/8-hard			8616 hr
12	27,000	980	Annealed			Failed (2 hr)
13	7,000	250	1/8-hard	Helium Zr Gettered	Helium Only	7464 hr
14	7,000	250	Annealed			7464 hr
15	13,500	490	1/8-hard			Failed (1620 hr)
16	13,500	490	Annealed			Failed (518 hr)
39	13,500	490	Annealed	Normal-U	Sodium Only	3002 hr
40	13,500	490	Annealed	Normal-U		Failed (4 hr)
41	13,500	490	Annealed	Hyper-UC		Removed
42	13,500	490	Annealed	Hyper-UC		Removed

*All cladding used in this test is welded-and-drawn tubing from heat 99702 and has the following composition (wt %):

C	0.052	Mn	- 1.37	P	- 0.014	S	0.015	Si	- 0.40
Cr	- 18.34	Ni	- 9.67	Mo	- 0.12	Cu	- 0.077	Fe	- balance

†Hoop stress is based on original mean midwall diameter of 0.552-in. and 0.010-in. nominal wall thickness.

NAA-SR-11450

II-17

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

III. EVALUATION OF EFFORT TO DATE

This report is the final LSGR cladding summary. Much of the uncompleted SGR cladding work will be continued under the fast reactor program.

The NAA-123 experiment is proposed to continue under the fast reactor program and is currently under construction.

The correlation of stress-rupture life with variation in heat chemistry for 304 SS at 1250°F was quite successful when the interstitial elements C, N, P, and S were taken into account. The entire correlation is based on Task 2A data, and hopefully will be better if Task 2B can be completed.

No detrimental effects on stress-rupture life can be attributed to simultaneous carburization at this time. The two specimens still in test will be carried to failure.

Our work plus liaison with other sites indicate the following ultimate conclusions.

- 1) Rupture life at a given stress above 500°C is reduced by a factor of ~4 in a thermal neutron flux.
- 2) The cladding strain-to-failure is greatly reduced in reactor above 500°C, approximately by a factor of 10.
- 3) The above phenomena are probably a function of helium content as produced by the $B^{10}(n,\alpha)Li^7$ reaction.
- 4) The stress-rupture band for normal 304 stainless steel is approximately a decade wide on a log time versus log rupture stress plot.
- 5) The above spread is influenced by interstitial elements in austenitic stainless steels such as C, N, P, and S.
- 6) Preliminary test data suggest that a small amount of cold-work (~12% RA) will significantly increase creep strength and rupture life of 304 stainless steel.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Large Sodium Graphite Reactor

Project: UC Fuel Element Development

Responsible Engineer:

Project Engineer: B. R. Hayward

Reporting Period: Fiscal Year 1965

AEC Category: 04-01-04-21

General Order: 7519

Subaccount: 1953

I. PROJECT OBJECTIVES

The objectives of this project are to demonstrate the feasibility of UC fuel elements for large sodium-cooled reactors, to determine fuel element design criteria, and to set fuel element specifications. The reference design conditions are: maximum coolant temperature, 1200°F; maximum fuel surface heat flux, 10⁶ Btu/hr-ft²; and burnup, 8.61 x 10²⁰ fissions/cc (25,000 Mwd/MTU) average.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. GENERAL STATUS OF UC IRRADIATION PROGRAM

During the fiscal year two experiments containing hypostoichiometric uranium carbide, NaK bonded to 304 stainless steel cladding, were inserted in the General Electric Test Reactor (GETR). In addition, two instrumented long-rod demonstration tests containing hypostoichiometric UC of production quality, clad with Type 304 stainless steel, were fabricated and inserted in the NRX reactor in Chalk River, Ontario, Canada. During this same period, four capsule experiments were discharged and a detailed postirradiation examination was conducted. The details and status of these irradiations are shown in Table 1.

In general, the results continue to show that, where operating temperatures are maintained within the LSGR reference range, (1600 to 1800°F), the operational behavior of the hypostoichiometric UC-304 stainless steel clad system is excellent to peak burnups of at least 10.07 x 10²⁰ fissions/cc (31,000 Mwd/MTU). Under these conditions, fuel swelling and fission gas release were well within current LSGR design criteria. Postirradiation examination of four capsule experiments during the fiscal year substantiated this conclusion.

Capsule experiment AI-3-12 was successfully irradiated to a peak burnup of 6.14 x 10²⁰ fissions/cc (17,800 Mwd/MTU). The capsule was removed from the General Electric Test Reactor (GETR), where it occupied a position in a Radial Adjustable Facility Tube (RAFT), after a residence period of 6,265 hr at power. During its residence in the reactor, the capsule operated with fuel centerline temperatures in the 1600 to 1800°F range for 57% of its operating time with a maximum linear power output of 46 kw/ft. The fuel was 0.500-in. -diameter as-cast hypostoichiometric uranium carbide, 4.40 to 4.68 wt % C; and was sodium-bonded to 0.010-in. -thick 304 stainless steel cladding. The fuel underwent density changes which ranged from 3.5 to 7.2%. Metallographic examination showed that under the irradiation test conditions no apparent change in fuel structure occurred. Significant was the lack of intragranular fission gas void agglomeration, indicating that the temperature and burnup conditions were not sufficient to cause gross gas atom migration and coalescence, which supports the theory that at

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 1

LSGR UC FUEL IRRADIATION EXPERIMENTS CONDUCTED IN FY 1965

Test No.	Facility	UC (in.)		Burnup* Mwd/MTU		Maximum Fuel and Temperature (°F)		Date of		Gas Release (%)	Fuel Growth %Δρ
		Diameter	Length	Design	Current or Final			Insertion	Discharge		
						Design					
AI-3-11	GETR	0.500	18	35,000	31,000	1800	1900	5-63	2-65	0.65 Kr ⁸⁵	-5.2 to -8.6
AI-3-12	GETR	0.500	18	20,000	17,800	1800	1900	5-63	6-64	-	-3.5 to -7.2
NAA-81-2	MTR	0.500	18	25,000	18,000	1800	3000-4500	7-63	7-64	17.1 Kr ⁸⁵	-16.8 to 22.1
NAA-81-4	MTR	0.500	18	30,000	17,200	1800	3000-4500	7-63	7-64	4.25 Kr ⁸⁵	-2.8 to 18.5
NRX86-1	NRX	0.500	114	30,000	11,000	1825	1650-1700	8-64	9-66	-	-
NRX86-2	NRX	0.500	114	40,000	10,000	1825	1650-1700	9-64	5-67	-	-
AI-25-1	GETR	0.320	3.5†	~50,000	10,000	1800	1800-1850	1-65	4-67	-	-
AI-25-2	GETR	0.320	3.5†	~50,000	10,000	1800	1800-1850	1-65	4-67	-	-

*Please see text for equivalent "fissions per cc" values.
†3 subcapsules each.

temperatures below 1800°F and to burnup of ~18,000 Mwd/MTU fission products are accommodated within the lattice. Some transgranular cracking in the hypo UC was observed, but was more predominant in cooler regions of the capsule. The 304 stainless steel cladding operated as a satisfactory containment. Metallographic examination of the cladding showed that only slight carburization had occurred.

Capsule experiments NAA-81-2 and NAA-81-4, of similar design as Experiment AI-3-12, were removed from the Materials Test Reactor (MTR) due to anomalous thermocouple behavior indicative of cladding failure after residence periods of 6530 and 6770 hr at power, respectively. The experiments achieved peak burnups of 6.21×10^{20} and 5.94×10^{20} fissions/cc (18,000 and 17,200 Mwd/MTU) based on mass spectrographic analysis, respectively. Due to a loss of sodium bond in both capsules as a result of pinhole cladding defects, the design fuel temperature conditions were exceeded. The in-fuel thermocouples pegged off-scale at 2200°F. The sodium bond was completely lost in Capsule NAA-81-2 and partially lost in NAA-81-4, uncovering the top 8 in. of fuel in the column. The estimated fuel centerline temperature range was 3000 to 4500°F for the top 8 in. of NAA-81-4, as estimated from the cladding thermocouple behavior, and assuming radiation heat transfer in this region. It is believed all fuel slugs in NAA-81-2 underwent peak centerline temperatures equivalent to the top 8 in. of fuel in NAA-81-4. Both capsules were at peak temperature for less than 12 hr. The fuel swelled diametrically from 8.0 to 10.5% in NAA-81-2, and from 1.3 to 8.4% in NAA-81-4. Density decreases ranged from 16.8 to 22.1% for NAA-81-2, and from 2.8 to 18.5% for NAA-81-4. Metallographic examination of the in-fuel tantalum well and UC fuel showed a UC fuel-Ta well interaction indicative of incipient melting of the phases formed, and substantiates that excessive temperatures were achieved. Based on an average burnup of 5.66×10^{20} and 5.06×10^{20} fissions/cc (16,400 and 14,700 Mwd/MTU), the fractional Kr⁸⁵ release was 17.1% Kr⁸⁵ for NAA-81-2 and 4.25% Kr⁸⁵ for NAA-81-4.

Metallographic examination of fuel from the top of the column in experiment NAA-81-4 appeared, in general, similar to all of the fuel observed in capsule NAA-81-2. Interpretation of the microstructures leads to a number of significant conclusions, as follows.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

1) Evidence of grain boundary migration was observed in specimens taken from the top of the fuel column. Out-of-pile studies have indicated that the minimum temperature at which grain growth is observed is greater than 2400°F (1315°C). This adds further support to the supposition that this fuel was subjected to high temperatures, particularly since the fuel experienced the high temperatures for only an eight hour period at the end of the test. It should be pointed out that, in the case of fuel in Capsules NAA-81-2 and 81-4, the terminology grain boundary migration is used and not grain growth. This terminology was used in the case of the irradiated fuel to differentiate between an actual grain volume change and the movement of boundaries at the expense of other grains. Due to a lack of preirradiation grain size characterization data, it is impossible to determine whether an actual grain volume change occurred. It is highly improbable, however, that grain boundary migration has occurred without grain volume changes. It will be noted that the movement of grain boundaries swept fission gas bubbles, free intragranular uranium precipitates, and possible fission product precipitates before them and collected these at the new grain boundary. It would be expected that such behavior would increase fission gas release and fuel instability. The uneven grain boundary movement prevalent in these structures is of particular significance. Similar structures have been observed out-of-pile in heat-treated tungsten and molybdenum-doped uranium carbide. Therefore, it is suggested that the uneven grain boundary movement occurring here was caused by the pinning of fission product molybdenum and other refractory nuclides by dislocations. The pinning phenomenon strongly suggests that "doping" uranium carbide with refractory metals may stabilize the fuel at these temperatures.

The uneven grain boundary movement has not been observed in previous uranium carbide irradiations, where similar high temperatures have been experienced. It is to be recognized, however, that in all these tests the burnup was lower.

2) The operation of fuel at LSGR reference temperatures, 1800°F, caused no apparent change in structure. Significant is the lack of fission gas void agglomeration, indicating that the temperature and burnup conditions were not sufficient to cause gross gas atom migration and coalescence; and supports the theory that at temperatures <1800°F fission products are accommodated within the lattice. Transgranular microcracking was observed, but no evidence of grain boundary movement was apparent. As previously observed in fuel in AI-3-12, these data are similar.

3) Anisotropic diametral fuel growth was observed in samples from Capsule NAA-81-4, and is attributable to a peripheral columnar type grain structure which has a high degree of preferred orientation. A longitudinal section of the slug exhibited transilient swelling behavior where the peripheral grain structure changed from the equiaxed to the columnar type. These structures were observed in pretest photomicrographs and are attributed to variable cooling rates over the length of the fuel slug during solidification of the melt. The fractional growth increase due to the anisotropy was a minimum 2-1/2%.

Efforts to determine the origin of fission gas leakage in the fueled rod in experiment NAA-81-2 were successful. After removal from the outer capsule, the top endplug was pressurized in 250°F (120°C) oil at 40 psi (3 kg/cm²). Escaping bubbles revealed a pinhole type leak in the endplug weld. This pinhole is believed to have been the means of fission gas escape to the outer capsule. The fuel rod containing both UC fuel and sodium bond was pressurized with the bottom endplug in place; and again, with the endplug removed in 250°F (120°C) oil at 160 psi, no pinhole was found. It is believed that during the rod

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

cool-down, a sodium carbonate formed in possible pinholes and solidified, plugging these holes. Hence, the pinhole went undetected at postirradiation pressurization temperatures. It is suspected that the sudden increase in the fuel centerline temperatures of both NAA-81-2 and 81-4 was caused by a loss of the sodium bond through a pinhole leak near the bottom endplug similar to that detected in the top weld of NAA-81-2.

During the fiscal year fuel swelling, fission gas release and temperature data from previous irradiation experiments were reevaluated and used in updating design curves for uranium carbide. Fuel swelling and fission gas release versus temperature curves are shown in Figures 1 and 2. The mean

Figure 1. UC Fission Gas Release

7519-2513

Figure 2. UC Fuel Growth and Swelling

7519-2514

7519-2515
Figure 3. Appearance of Irradiated
Hypostoichiometric Uranium
Carbide Fuel Slugs

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

curves were established by statistically evaluating the data; the boundaries of the data scatter band fall within 95% confidence limits.

B. AI-3-11

The AI-3-11 experiment was designed to provide irradiation performance data for hypostoichiometric uranium carbide (UC) at fuel centerline temperatures of 1800°F. The design capsule power output was 32.5 kw/ft. The capsule fuel column consisted of 10% enriched UC slugs 0.500-in. diameter x 18 in. long, and was sodium-bonded (20-mil annulus) to 10-mil wall 304 stainless steel cladding.

The experiment was irradiated in the General Electric Test Reactor (GETR) in a Radial Adjustment Facility Tube (RAFT). The irradiation was terminated at AI's request on February 15, 1965 after achieving an estimated peak burnup of 10.07×10^{20} fissions/cc (31,000 Mwd/MTU) or 3.27 at. % U²³⁵. The fuel underwent peak centerline temperatures of -1900°F, both at the beginning and end of irradiation. The time-weighted average fuel centerline temperature was 1678°; temperatures less than 1600°F were neglected in the weighted average.

Results of the postirradiation examination are as follows. (Figures 3 through 7)

1) The capsule had maintained structural integrity, was in excellent condition, and had operated satisfactorily as a pressure vessel. No detectable increase in external rod diameter was noted.

2) Gamma activity scans over the length of the fuel column showed a relatively flat burnup profile, and indicated fuel slugs were broken at the top of the fuel column.

3) Kr⁸⁵ fission gas release from the fuel column was 0.65% of that theoretically produced based on an average burnup of 10.03×10^{20} fissions/cc (30,000 Mwd/MTU).

4) Disassembly of the fuel rod revealed cracked and broken fuel slugs. The in-fuel thermocouple and tantalum well appeared in good condition and showed no visual evidence of reaction with the UC fuel. The inner diameter of the 304 SS cladding appeared clean, with a light discoloration over most of the fuel column area.

5) Fuel density changes varied from a minimum of -5.2% to a maximum of -8.6%, and are shown in Table 2.

6) Metallographic examination of the fuel structure indicated that: (a) the irradiation test conditions caused no apparent change in microstructure; (b) there was no evidence of free uranium migration or agglomeration; (c) no intragranular fission gas precipitation and agglomeration were observed, indicating that the temperature and burnup conditions were not sufficient to cause gross gas atom migration and coalescence. These observations add support to previous theories that, at temperatures below 1800°F, fuel swelling is not observed; (d) intergranular porosity was observed in the uranium grain boundary phase; and (e) intragranular cracking was observed in all slugs examined, but was most evident toward the bottom of the fuel column where peak temperatures were less than 1700°F.

7519-2516

Figure 4. Appearance of Irradiated 4.53 wt % C in UC
Fuel Slug No. V

FUEL METALLOGRAPHIC SAMPLE No. 3
 ~15-16 in. FROM THE TOP OF THE
 FUEL COLUMN

AUTORADIOGRAPHY

7519-2517

Figure 5. Appearance of Irradiated 4.52 wt % C in UC
 Fuel Slug No. OO

Figure 6. Appearance of Irradiated 4.58 wt % C in UC Fuel Slug No. XXX

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

7519-2519

Figure 7. Cladding Sample. (1, just above fuel column; 2, ~2 in. from top of column; 3, ~12 in. from top; and 4, ~16 in. from top)

7) Autoradiographs showed only a very slight depression of activity from fuel slug peripheries to slug centers, and suggest that with prolonged burnups the radial burnup profile tends to flatten out due to preferential peripheral burnout, as indicated by burnout calculations. The relatively flat activity profile is also significant from the standpoint that it suggests negligible migration of fission products under the test conditions.

8) Examination of the 304 SS cladding showed that only slight carburization had occurred. However, the cladding was sensitized in the fuel column area.

TABLE 2

Position	Slug No.	Length (in.)	Composition (wt % C)	% $\Delta\rho$
Top of Column	V	2	4.53	-5.7
	XX	4	4.55	-6.3
	VVV	2	4.53	-8.6
	XXX	4	4.58	-8.0
	X	1	4.53	-7.6
	0	2	4.59	-6.5
Bottom	00	3	4.52	-5.2

C. TESTS IN PROGRESS

1. NRX-86 Series

Two instrumented capsules (NRX-86-1 and 86-2) containing 1/2-in. -diameter by 9.5-ft-length columns of hypostoichiometric UC (4.5 to 4.75 wt % C) of production quality, NaK-bonded to 10-mil wall 304 stainless steel, have continued satisfactory operation in NRX at AECL, Chalk River, Ontario, Canada.

The objective of the NRX-86 series of test is to irradiate 4.9% enriched uranium monocarbide (carbon content 4.65 ± 0.15 wt %) fuel rods under conditions approaching the Large Sodium Graphite Reactor (LSGR) reference conditions. The rods are scheduled to achieve peak burnup levels of 10.03×10^{20} and 13.79×10^{20} fissions/cc (30,000 and 40,000 Mwd/MTU).

One departure from LSGR fuel element reference design is that the fuel column, which is 0.490-in. -diameter by 9-1/2-ft. long, is NaK-bonded rather than Na-bonded within the 304 stainless steel cladding tube (9/16-in. OD by 0.010-in. wall). This departure was necessary to preclude the possibility of local bond melting on startup in the water-cooled reactor which could result in local cladding deformation.

The NRX-86 fuel rod assembly was encapsulated within a 1-3/8-in. OD by 0.035-in. wall stainless steel capsule tube with NaK as the heat-transfer medium. Three concentric stainless steel tubes surrounding the fuel rod assemblies provide additional thermal resistance between the fuel and coolant.

The capsules utilize sealed tapered helium thermal barriers at each end of the fuel rods to maintain the cladding temperature over the full length within the LSGR reference clad temperature range. Both rods were inserted into NRX core positions on October 19, 1964. The rods operated during FY 1965 at approximately 90% of design power level, achieving peak fuel centerline temperatures of 1650-1700°F and peak clad temperatures of 1080 to 1120°F. Peak burnup in Capsule NRX-86-1 was approximately 3.79×10^{20} fissions/cc (11,000 Mwd/MTU), and in NRX-86-2, approximately 3.45×10^{20} fissions/cc (10,000 Mwd/MTU), at the close of the fiscal year.

Coolant channel plugging has continued to be a problem in both rods. After each reactor shutdown, the coolant channels between the aluminum flow tubes and the stainless steel capsules have been flushed in-reactor with a 200°F, 5% citric acid solution. This procedure has proved satisfactory in temporarily restoring full coolant flow.

Some results of the NRX-85 examinations during the fiscal year have shed light on the coolant flow reduction problem experienced in the NRX-86 rods. Upon disassembly of NRX-85-3, it was observed that the aluminum flow basket was rather severely pitted. Mass spectrometer analysis of crud deposit samples revealed that approximately 42% of the deposits were aluminum. It was observed that the crud deposit collected on the stainless steel or nickel located near aluminum parts. Since NRX reactor operating personnel are concerned that localized pitting of the aluminum flow tubes could result in leaks of the light water coolant into the heavy water moderator, the aluminum flow tubes will be replaced

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

by stainless steel flow tubes. Fabrication of the flow tubes has been completed and the tubes are awaiting insertion into the reactor.

2. AI-25 Series

This series of experiments is directed toward establishing the effects of carbon content, burnup, and operating temperature on the irradiation stability of hypostoichiometric UC (4.5 to 4.75 wt % C). Each capsule consists of three fuel compartments, each containing a 0.320-in. -diameter by 3-1/2-in. -length fuel slug clad with 10-mil wall 304 stainless steel. The cladding tube is open at the top to permit intercommunication between the NaK annulus around the fuel and the NaK annulus around the fuel rod. GETR V-RAFT facilities, providing both radial and vertical movements, are being used as the irradiation facilities.

The design and assembly of the multicompartmented capsules, AI-25-1 and 25-2 were completed. The capsules were installed in GETR on January 26, 1965 and the irradiations began on February 4, 1965. The estimated peak burnup at the end of the fiscal year was 3.45×10^{20} fissions/cc (10,000 Mwd/MTU).

Fuel centerline temperatures are in the range of 1800 to 1850°F, as verified by the initial startup data.

To date, five of the six in-fuel thermocouples have failed. This is not unexpected, since the operating temperatures are higher than the recommended long-term operating limits for chromel-alumel couples. None of the cladding thermocouples has failed. Data obtained during the initial startup were sufficient to verify the heat-transfer analysis and establish correlations between cladding temperatures and in-fuel temperatures, thereby establishing confidence in predicting subsequent in-fuel temperatures from cladding thermocouple readings only.

III. EVALUATION OF EFFORT TO DATE

Results of the examination of AI-3-12, NAA-81-2, NAA-81-4, and of the AI-3-11 experiments which achieved peak burnup ranging from 5.94×10^{20} to 10.07×10^{20} fissions/cc (17,200 to 31,000 Mwd/MTU) have shown that the reference LSGR fuel rod system is satisfactory if operation is maintained within LSGR reference conditions. However, these results also indicate that, if operating temperatures are significantly higher than LSGR reference conditions, the carbon specification for the hypostoichiometric UC fuel may need to be tightened to assure a minimum free uranium concentration. It has been observed that in fuel which has inadvertently operated at significantly higher temperatures, e.g. above 2500°F, (even for time periods as short as 8 hr), grain boundary movement operates to sweep fission gas bubbles, free uranium precipitates, and fission product precipitates to the grain boundaries. Further, it is suggested that at these burnup levels, fission product nuclides such as molybdenum are acting to pin grain boundaries locally resulting in uneven grain growth.

The successful operation of the AI 3-11 capsule, which attained a maximum burnup of more than 10.07×10^{20} fissions/cc (31,000 Mwd/MTU), adds significantly to the confidence level of the hypo UC-stainless steel clad fuel rod system for LSGR and Hallam Core II use.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

In addition, the continued successful operation of two long-rod tests (NRX86-1 and 86-2) demonstrates that such rods are capable of successful operation under LSGR conditions. These long-rod tests are recommended to be continued under the Hallam Fuel Surveillance Program.

The large density decreases observed in the NAA-81-2 and 81-4 UC fuel, believed to be due to higher than normal temperatures, add impetus to the need for tests to specifically determine the effects of carbon content, burnup, and operating temperatures on the irradiation stability of hypostoichiometric UC. To answer this need in as short a time-period as practical, the higher burnup rate AI-25 series capsules were designed, and the first two capsules were inserted in the GETR late in January, 1965. The irradiation data obtained to date are primarily <1800°F. Further information is required between 1800°F and 2200°F.

The observed high-temperature structural changes noted in the NAA-81-2 and 81-4 fuel have emphasized the need for a better understanding of phase equilibria in the uranium-carbon system, and especially of the effects of introducing minor concentrations of fission product nuclides on long burnup phase relationship.

Reevaluation of fission gas release data and fuel swelling data as a function of irradiation temperature indicates that, at the LSGR design operating temperatures, fuel swelling and fission gas release are well below the permissible limits for the system. After successful demonstration of large-scale results, the changes to less conservative design criteria and fuel specifications are expected to result in lower fuel cycle costs.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Large Sodium Graphite Reactor	
Project: LSGR Fuel Element Development	
Project Engineer: B. R. Hayward	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-21
General Order: 7519	Subaccount: 1955

I. PROJECT OBJECTIVES

The objective of this project is to develop and test high-performance fuel elements for the Large Sodium Graphite Reactor Concept. The short-range objective of the project is to demonstrate the reliable operation of sodium-bonded, stainless steel-clad, hypostoichiometric UC fuel elements at temperatures equivalent to and in excess of the design conditions for a 200-Mwe SGR while attaining an average burnup of 25,000 Mwd/MTU within 2 yr. Long-range objectives include: (a) demonstration of fuel element concepts for advanced core loadings of a 200-Mwe SGR and the initial core loading of a 100 Mwe SGR, and (b) accumulation of information on the mechanical behavior of the LSGR fuel element systems during reactor operation.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

Two test element configurations were designed for SRE-PEP Core III. The first, designated Standard Test Element, is identical in design to the SRE-PEP driver elements with the addition of in-fuel thermocouples and dimensional characterization of a number of the fuel slugs. The second, designated Special Test Element, is of the same general configuration as the Standard Test and driver elements (i.e., eight wire-wrapped fuel rods per element) but has smaller diameter fuel rods (0.570 versus 0.680 in.) and higher enriched fuel (15 versus 6.5%) to obtain higher burnup rates, specific power, and fuel centerline temperatures. The fuel column length is also shorter (60 versus 72 in.) to compensate for the fission gas release anticipated for an average burnup of 25,000 Mwd/MTU.

The irradiation program for the four test elements was established. One Special Test Element will occupy the center core position; the two Standard Test Elements and the second Special Test Element will occupy three of the four 1st-ring core positions. One Standard and one Special Test Element will be removed when the reactor is shut down for the first driver element replacement (approximately eleven 80% full power months). These elements will have attained average burnup levels of approximately 15,000 and 9,000 Mwd/MTU, respectively. The remaining two elements will be removed when the Special Test Element has attained the program objective burnup level of 25,000 Mwd/MTU (approximately eighteen 80% power months). At that time, the Standard Test Element will have an average burnup of approximately 15,000 Mwd/MTU.

The results of thermal and nuclear analyses for the test elements are presented in Table 1. Also included are the UC carbon compositions in each fuel rod. Seamless, 304 stainless steel is used as the cladding for the Special Test Elements; and welded and drawn, 304 SS is used as the Standard Test Element cladding.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 1
TEST ELEMENT DESIGN DATA

Parameters	Standard Test	Special Test
Maximum Permissible Design Power (Mw)	1.044	1.239
Fuel Bundle Design Life (Mwd)	802	505
Weight of U Per Bundle (kg)	32.07	20.22
Rods per Cluster	8	8
Fuel Diameter (in.)	0.600	0.500
Fuel Enrichment (%)	6.5	15.0
Fuel Element Specific Power (kw/kg U)		
Average	32.57	61.28
Maximum	40.52	74.46
Active Fuel Length (in.)	68	60
Area of Fuel Rods, Cross-Section, Cold (in. ²)	2.26	1.57
Flow Area, Cold (in. ²)	3.55	4.106
Maximum ΔP Across Element (psi)*	1.62	2.04
Maximum Flowrate (lb/sec)	6.159	8.023
Maximum Sodium Velocity (ft/sec)	4.83	5.65
Sodium Inlet T (°F)	650	650
Sodium Outlet T (°F)	1165	1114
Maximum Clad T (°F)		
(a) With Hot Channel Factors	1293	1297
(b) Without Hot Channel Factors (nominal)	1236	1244
Maximum Fuel Temperature (°F)		
(a) With H.C.F.	1758	1997
(b) Without H.C.F. (nominal)	1667	1882
Maximum Rod Thermal Power (kw/ft) at 94.15% of total power		
(a) With H.C.F.	30.64	39.04
(b) Without H.C.F. nominal	27.80	35.43
Maximum Heat Flux (Btu/hr-ft ² x 10 ⁻³)		
(a) With H.C.F.	588	886
(b) Without H.C.F. nominal	533	804
Axial Power Peak-to-Average Ratio		
(a) Hot Channel	1.269	1.239
(b) Nominal	1.244	1.215
Thermal Conductivity (Btu/hr-ft-°F)	13.3 ± 1.5	13.3 ± 1.5
Fuel Composition (wt % Carbon)		
Rod 1	4.7 - 4.8	4.7 - 4.8
Rod 2	4.4 - 4.8	4.5 - 4.8
Rod 3	4.6 - 4.7	4.6 - 4.7
Rod 4	4.4 - 4.8	4.5 - 4.8
Rod 5	4.5 - 4.6	4.5 - 4.6
Rod 6	4.4 - 4.8	4.5 - 4.8
Rod 7	4.4 - 4.8	4.5 - 4.8
Rod 8	4.4 - 4.8	4.5 - 4.8

*Does not include pressure drop across orifice.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Each fuel slug in the Special Test Elements was visually examined for surface defects, radiographed for internal defects, and sampled for carbon analysis. Those slugs with carbon contents between 4.70 and 4.80 wt % were examined metallographically for the presence of undesirable UC_2 platelets. From 2 to 6 slugs in each fuel rod were completely characterized (dimensions, metallography, chemical analysis, and density), and were positioned at specified locations in the fuel rods. Archive samples of the fuel and cladding were retained for future analysis.

A similar fuel evaluation was performed on 2 fuel rods in each Standard Test Element. The remaining 6 rods in each element are production driver element rods and were not characterized.

A full-scale model of a Special Test Element was fabricated to check out the fabrication process. Hydraulic testing of the model was originally planned; however, the test did not warrant the expense, since the uncertainty of the hydraulic calculation was within the coolant control limits of the variable orifice.

An analysis of the spent fuel-handling operations was performed. The results, based on the temperatures of the fuel rod cladding surface in the fuel-handling machine as a function of the maximum operating power of the elements are as follows.

Test Element	Cladding Temperature After 10 hour Decay Period (°F)	Decay Period Necessary to Reduce Cladding Temperature To 1200°F (hr)
Special	1529	105-146
Standard	1230	12

The effect of an early adoption of the TR-SRE Concept on attaining the burnup objectives for the SRE-PEP Test Elements was investigated. The proposed modifications to the SRE consist of the removal of the central seven moderator fuel assemblies of the thermal core (the location of the Test Elements) to provide space for the fast region. Dependent upon scheduling, the removal of the central region of the SRE-PEP core could affect the program objectives of the SRE-PEP test elements. For example, a changeover to TR-SRE after operating SRE-PEP for 1 yr would have an appreciable effect on the fuel element irradiation program planned for the SRE-PEP Test Elements. Assuming a 60% power plant factor, the Special Test Elements will have achieved an average burnup of only 18,000 Mwd/MTU after 1 yr. Fuel and cladding behavior data at this burnup would be useful, but would be of limited value compared with data at 25,000 Mwd/MTU. To attain the program objectives of achieving a burnup of 25,000 Mwd/MTU, an operational period of approximately 18 mo is now required. These operational planning schedules are being evaluated with the program objectives.

A preliminary investigation of the feasibility of reinserting one or more of the Special Test Elements in the thermal region of the TR-SRE was conducted.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Reinsertion appears to be feasible; however, detailed core physics and thermo-mechanical studies would be required for substantiation to determine if the elements would operate within the fuel centerline and cladding temperature ranges expected in SRE-PEP.

A digital-tape punch data retrieval system was designed, procured, and installed at the SRE.

III. EVALUATION OF EFFORT DURING FY 1965

The scope of effort on this project was reduced during the year to include only the analysis and design liaison of test elements for SRE-PEP Core III. The SRE-PEP test elements are designed to fulfill the short-range objectives of the project, i.e., to operate at temperatures in excess of the design conditions for a 200-Mwe SGR while attaining an average burnup of 25,000 Mwd/MTU within 2 yr. The longer-range objectives of this project have been cancelled due to the modification in program emphasis from thermal reactor systems to fast breeder reactor systems. Therefore, this project is completed. The SRE-PEP core provides an excellent irradiation test facility for any AEC fuels program requiring a high temperature sodium environment, high specific power fuel conditions, instrumented test components, and a homogeneous flux spectrum. The materials liaison and analysis provided by this project indicates the feasibility of achieving the test objectives within the facilities and schedule planned.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Large Sodium Graphite Reactor	
Project: Development and Evaluation of UC Alloy Fuel Materials	
Project Engineer: B. R. Hayward	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-21
General Order: 7519	Subaccount: 1957

I. PROJECT OBJECTIVES

The primary objective of this project is the development of carbide fuel materials capable of high power, high temperature, and high burnup operation in sodium-cooled reactors. The immediate objectives, to which most of the current efforts are directed, are the evaluation of hypostoichiometric UC fuel as specified for current sodium reactor designs including the determination of physical-mechanical properties, and the investigation of its in-pile and out-of-pile behavior.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

The technical program under this project is divided into several parts, each of which must be evaluated with respect to its individual and related effect on the operational capabilities of the LSGR fuel materials and fuel systems. While the ultimate capability of any nuclear fuel can be ascertained only by in-pile irradiation testing, out-of-pile physical and mechanical properties and behavior must be determined to provide a basis for understanding the results of in-pile tests. This program utilizes continuous in-pile monitoring and postirradiation annealing experiments to provide a basic understanding of fission gas release from irradiated UC fuel materials. The results of in-pile experiments have suggested a number of out-of-pile tests of the UC materials to establish modes of behavior indicated by the in-pile behavior. This project has also provided for the determination of contact compatibility between the UC fuel material and austenitic stainless steel.

The technical progress for FY 1965 in each part of the program is presented below.

A. IRRADIATIONS

1. EXP-NRX-85 Multicompartmented Capsule Irradiations

This experimental irradiation series consisted of three capsules, two with four tandemly arranged compartments and the third with six compartments. The fuel specimens were 0.490 in. in diameter by 2.50 in. long, with 0.25-in. -thick hafnium wafers at each end to minimize end-flux peaking effects. The hypostoichiometric UC fuels (less than 4.80 wt% C) were thermally bonded with sodium to 304 stainless steel cladding. One specimen in the second capsule and two in the third were hyperstoichiometric UC (greater than 4.8 wt% C), and were contained in Nb-1Zr cladding. An enrichment of 10% U²³⁵ was used to produce the design operating conditions of 35.6 kw/ft of fuel and fuel surface, and central temperatures of 1150 and 1800°F, respectively. Burnup levels of 15,000, 30,000, and 40,000 Mwd/MTU were planned for EXP-NRX-85-1, -2, and -3

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 1
SUMMARY OF RESULTS FROM EXP-NRX-85
SERIES OF CAPSULE EXPERIMENTS

Capsule No.	Compartment No.	Fuel Slug No.	Fuel (a) Composition wt % Eq C	Calculated Fuel Temperature (b) (°F)				Measured (c) Burnup	
				Average T _℄	Average T _{surf}	Average Clad ID	Peak T _℄	Mwd/MTU	atom % U
X-85-1	1 ⁽ⁱ⁾	26	4.51	1875	1200	1145	1990	18,100	1.9
-1	2 ⁽ⁱ⁾	25	4.45	2130	1355	1295	2300	19,200	2.0
-1	3 ⁽ⁱ⁾	32	4.77	2050 ^(g)	1300 ^(g)	1240 ^(g)	-	17,200	1.8
-1	4 ⁽ⁱ⁾	18	4.63	1985 ^(h)	1260 ^(h)	1205 ^(h)	2320	19,200	2.0
X-85-3	3 ^(j)	2	5.14	1925	1220	1170	<2000	20,800	2.2
-3	5 ^(k)	31	4.76	1915	1215	1165	<2000	20,600	2.2
-3	6 ^(l)	17	4.93	1950	1230	1180	<2000	19,200	2.0

Calculated Fuel Temperature (d) (°F)			Peak T _℄ ^(e)	Fission ^(f) Gas Release (%)	Fuel Density Change (% $\frac{\Delta \rho}{\rho}$)	Cladding Material
Average T _℄	Average T _s	Average Clad ID				
2460	1560	1480	≥3000	18.7	-26.1	304-SS
2610	1640	1560	≥3000		-30.1	304-SS
2360	1490	1420	≥3000		-27.8, -19.6	304-SS
2610	1640	1560	≥3000		-20.5	304-SS
2300	1460	1390	≥3000	<1	-11.7, -5.3	Nb-1Zr
2270	1440	1370	≥3000	~2.5	-25.6	304-SS
2140	1360	1290	≥3000	<1	-20.8, -5.6	Nb-1Zr

(a)Eq C = wt % C + 0.857 wt % N + 0.750 wt %

(b)From cladding surface thermocouple readings

(c)These values were calculated from pre- and postirradiation mass spectrometer analyses of the U²³⁵ isotope content in the fuel, assuming no U²³⁸ depletion, no contribution from plutonium isotope fissions, and an effective U²³⁵ capture to fission ratio of 0.190. It is estimated that the standard error in these calculations is ≤ ± 1.5%

(d)Based on measured burnup analyses, 185 Mev/fission available for usable heat, a fuel thermal conductivity of 13.3 But/hr-ft-°F (0.055 cal/cm-sec-°C), and uniform heat generation in the fuel

(e)Based on heat transfer calculations for temporary loss of thermal bond and on metallographic fuel examinations

(f)Based on xenon analysis

(g)Interpolated data. Thermocouple failed on initial reactor startup

(h)Extrapolated data. Thermocouple failed October 19, 1963

(i)All compartments failed due to fuel swelling which was caused by the loss of the sodium thermal bond

(j)Capsule intact, partial bond loss

(k)Pin-hole failure

(l)Capsule intact, partial bond loss

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

capsules. NaK was used to bond the fuel compartments to the nickel outer tube, around which an aluminum flow basket was placed to provide the required water velocity to prevent coolant boiling. Each capsule contained eight stainless steel clad, Type K thermocouples for measuring capsule temperatures. To avoid fuel behavior and evaluation complications, all thermocouples were placed in the NaK space externally adjacent to the fuel cladding surface. An automatic digital temperature logging system was utilized as a supplement to the recorder to facilitate the temperature and burnup data reduction processes.

Examination and evaluation of the X-85-1 were completed and reported (in NAA-SR-11134, 1965). The irradiation of X-85-1 appeared quite normal, although the calculated average fuel temperatures were about 10 to 12% above the design temperatures. During the postirradiation disassembly of the capsule, it was found that the cladding in all compartments had ruptured in the region of the fuel, and these regions were discolored (darker). The failures appeared to be the result of fuel swelling rather than of excessive pressure. The failure modes for all the X-85-1 fuel compartments were postulated to be loss of the sodium thermal bond. The postirradiation examination of the X-85-3 capsule revealed that two of the six fuel compartments were intact. It was ascertained that the fuel-sodium bond was lost in the intact compartments by evaporation of the sodium from around the fuel and condensation in the top of the fission gas plenum above the fuel. A summary of the data from examinations of the X-85-1 and X-85-3 is shown in Table 1.

2. AI-24-1 Pressure-Monitored Irradiation Experiment

The AI 24 irradiation series was designed for continuous measurement of the rate-of-pressure rise in a fuel rod due to combined effects of fuel swelling and fission gas release of hypostoichiometric UC as a function of temperature and burnup. The fuel system consisted of a 10-in. column of 1/2-in.-diameter hypostoichiometric (4.5 wt % C) UC fuel, sodium-bonded in 304 stainless steel cladding. In-fuel thermocouples were placed at the center, at the midradius, and at the surface of the fuel to determine the radial temperature distribution and to provide data for the evaluation of changes in thermal conductivity during irradiation.

The first experiment, AI 24-1, was inserted in the General Electric Test Reactor (GETR) in a Radial Adjustment Facility Tube (RAFT) position in mid-September 1964. It was removed in May 1965, after 8 mo of irradiation to an accumulated peak burnup of about 8000 Mwd/MTU. The fuel centerline temperature was controlled in the 1750 to 1800°F range with the peak cladding temperature being in the 1160 to 1180°F range. These conditions correspond to a lineal rod power of about 25 kw/ft. The experiment was designed to reach the assumed maximum internal rod pressure in 6 mo with a burnup of 6000 Mwd/MTU; but the actual fuel swelling and fission gas release were significantly less than originally predicted. The internal gas pressure increased by only about 10 psi during the irradiation. This pressure increase indicates that less than 0.5% of the fission gas was released from the fuel, and that the volumetric swelling of the fuel was less than 2%. The irradiation was terminated because small leaks developed in the fuel rod, making further pressure readings unreliable. A cursory hot-cell examination was performed; further examination is proposed for FY 1966 under the Fast Reactor Program. A cutaway drawing of the capsule is shown in Figure 1.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

7519-2520

Figure 1. Materials-Irradiation Test Capsule

B. MECHANICAL PROPERTIES

Out-of-reactor hot-hardness tests have been conducted as a measure of the UC fuel strength. The hardness of several as-cast UC compositions, as a function of temperature, is shown in Table 2.

The results for the hypostoichiometric UC (<4.8 wt % C) were unexpected, in that the near-stoichiometric specimen (4.77 wt % C) was found to be softer than other specimens with lower carbon content. It is noted that this specimen contained the lowest amount of total impurities. The data indicate that a small increase in impurities (<0.1 wt % total) was sufficient to counteract the carbon concentration effect, so that the 4.19 wt % C specimen is harder than that of 4.77 wt % C up to about 700°C. The variations in hardness among the four hyperstoichiometric samples correlate reasonably well with variations in impurity content.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 2
COMPOSITION AND HOT HARDNESS OF UC

Composition (wt % C)	Impurities (ppm)			Hot Hardness at 0.4 Kg Load (DPH-Kg/mm ²)									
	Nitrogen	Oxygen	Spectro- graphic Analyses for Approxi- mately 23 Elements	Temperature (°C)									
				25	200	350	500	600	700	800	900	1000	
5.28	320	160	1100	641	556	513	489	394	342	295	230	-	
5.16	310	60	1100	553	542	512	436	372	332	290	-	-	
5.15	220	55	600	550	532	504	429	338	315	270	-	-	
5.14	-	800	800	620	574	504	482	406	325	278	221	306	
4.77	130	50	500	720	-	-	400	-	136	97	72	50	
4.64	180	200	800	865	-	-	470	-	203	115	-	-	
4.19	250	100	1000	770	-	-	505	-	147	73	-	-	
3.97	770	100	-	713	-	-	405	-	123	53	-	-	

C. UC PHASE EQUILIBRIA STUDIES

An understanding of the uranium-carbon phase relationships and the effect of impurities on these relationships is necessary to account for some of the anomalous behavior in irradiated UC fuels, and to aid the preparation of product specifications that will meet performance objectives. It has been recognized that the solubility of uranium in UC at high temperatures may have an important bearing on (1) the fission gas movement through the UC lattice, especially during the reversible dissolution-precipitation process which may occur during long- or short-time thermal fluctuations, (2) the interpretation of pre- and postirradiation microstructure, and (3) composition specifications.

Hypostoichiometric UC (4.5 to 4.7 wt % C) specimens have been quenched in oil after 1-hr heat treatments at 1300, 1600, and 1900°C. Quenching from 1600 and 1900°C produced a 2-phase grain boundary structure composed of UC dendrites in a matrix of metallic uranium. The etching characteristics of the UC phase in the grain boundaries were different from those of the main UC phase, probably due to the differences in carbon and impurity concentrations (NAA-SR-10768, 1965). The structure is quantitatively consistent with the high-temperature region of current uranium-carbon phase diagrams. Isothermal heat treatments of the fuel specimens at 1000 and 650°C for up to 500 hr showed incomplete redistribution of the grain boundary UC phase to the main UC grains, and of the uranium phase in the UC grains to the grain boundaries.

A capability for quenching individually heated UC specimens in NaK was established to facilitate faster cooling rates. The specimens were supported by a

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

graphite pedestal and were centered in a water-cooled, induction current concentrator. When the specimens attained the desired test temperature (determined by an optical pyrometer), the pedestal was tipped remotely and the specimens were allowed to fall into a NaK reservoir. The tests were performed in vacuum ($\sim 10^{-5}$ torr), and in a Zr-gettered argon gas atmosphere.

Inductively heated hypostoichiometric UC specimens were successfully quenched in NaK from 2200°C . The microstructure revealed variations of cooling rate as indicated, to the extent of uranium precipitation within the UC grains. Cracks resulting from the quenching were evident but the specimens remained intact. The 2-phase grain boundary was formed, as in the previous oil-quenched UC tests.

The UC induction heating-quenching treatments were transferred to an inert atmosphere glove box to alleviate some of the NaK handling problems which were encountered, especially during postquenching cleanup operations. This change also resulted in an increased experimentation rate.

X-ray diffraction measurements were carried out on the first series of quenched hypostoichiometric UC (4.5 wt % C) specimens. The observed reduction in the lattice parameter of the carbide phase (from 4.961 to 4.949 Å) was the result of "uranium solution," which is believed to be retained by rapid quenching. Later experiments with a series of hypostoichiometric samples have shown values for the lattice parameter ranging from 4.949 to 4.952 Å, a strong reduction from 4.961 Å (lattice parameter for stoichiometric UC), but with no systematic dependence on quenching temperature. Prolonged heat treatment (~ 100 hr) at 1000°C of samples quenched from 1800 and 2000°C caused the lattice parameter to increase to 4.959 Å, about that of stoichiometric UC, which is in agreement with the results of Buckley (AERE-R-3872, 1961).

Oxygen contamination is a factor which very likely caused perturbations in these experiments, since it is known that in some cases oxygen has an effect similar to that of free uranium and lowers the lattice parameter of UC. Also, it is known from other indications that the atmosphere in which the quenching experiments were performed was to some degree contaminated with oxygen. Furthermore, it is not known to what extent oxygen may have affected the published work of Buckley and of Accary (4th Uranium Carbide Meeting, Conn., 1963), since these authors did not discuss the problem.

At moderate temperature, 900°C , it has been shown that oxygen at low pressure, $\sim 10^{-4}$ torr, reacts with UC to form a coating of UO_2 and UC_2 , rather than dissolving in UC as it does at higher temperatures. This was shown by an experiment in which a single crystal of UC was held at 900°C and 10^{-4} torr of oxygen in a microbalance. The sample gained weight at an accelerating rate for an hour, forming a layer ~ 6 microns thick in which both UO_2 and UC_2 had a preferred crystallographic orientation with respect to the UC. At 10^{-5} torr, the reaction rate was not measurable in a few hours.

Attempts to use high-temperature x-ray diffraction techniques to measure lattice constants in the UC-U system at temperatures of reported formation of solid solutions, and to look for a possible new phase, were delayed by trouble with oxygen pickup on the specimen surfaces. In one trial, at about 10^{-4} torr

and 900°-1000°C, such a thick coating formed that the pattern of the UC was obscured. In a later trial with an improved vacuum system, a coating formed but was not thick enough to obscure the UC. Some further improvements in the elimination of air and/or oxygen from the system are required to make it possible to perform meaningful diffraction experiments at the higher temperatures (1600 to 2000°C), where the phase changes are expected to occur.

D. UC THERMOPHYSICAL PROPERTIES

The objective of this task is to investigate the thermal conductivities of uranium carbide fuels as functions of composition and temperature. The composition range for the monocarbide is from 4.0 to 4.8 wt % C, and the temperature range is from room temperature to 1200°C. Within the scope of this task, an effort will be made to obtain information needed to explain the thermal transport behavior in uranium carbides.

Thermophysical properties measurements have been made on several uranium-carbon compositions. The data were obtained with hypostoichiometric UC, in the temperature range of 25 to 700°C. The measured quantities include thermal diffusivity, electrical resistivity, and specific heat, from which thermal conductivity values were calculated according to the equation

$$K = \alpha C_p \rho ,$$

where

α = thermal diffusivity

C_p = specific heat

ρ = specimen density.

The calculated values of thermal conductivity determined this year at Atomics International are shown in Figure 2, based on experimental data and theoretical considerations. These data agree with the data obtained at other laboratories. Additional measurements are being made to confirm earlier results and to extend the temperature range to about 1300°C.

7519-2521

Figure 2. Thermal Conductivity of UC

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

E. UC STAINLESS STEEL COMPATIBILITY

Compatibility tests of Types 316 and 347 stainless steels with hypostoichiometric UC (4.1 to 4.3 wt % C) fuel pellets, with and without sodium, have been conducted at temperatures of 1200, 1300, 1400, and 1600°F. Hastelloy-N contact capsules were used to contain the compatibility specimens. The differential expansion between specimens and the capsule provided the necessary contact pressure. A total of 48 capsules was treated for 500, 1000, and 5000 hr.

In addition to the compatibility capsules, 16 control specimens of Types 316 and 347 SS were encapsulated with and without sodium, and heat-treated for 2000 hr at 1200, 1300, 1400, and 1600°F. These specimens permitted preparation of a display of photomicrographs which will be used as a standard in differentiating the normal sensitization and cold-work-induced carbide precipitation in the steels from the UC-induced transformation being investigated. Structure alterations at the surface of the SS specimens encapsulated with sodium indicated that this standard was vitally needed for a precise evaluation of the UC-SS compatibility. The heat treatment and metallographic inspection of all samples has been completed.

III. EVALUATION OF EFFORT TO DATE

A. IRRADIATIONS

1. EXP-NRX-85 Series

The capsule fuel temperatures exceeded the design temperatures sufficiently to cause excessive fuel swelling and rupture of 8 of 10 of the claddings. Thus, the primary objectives of the experiments were not achieved. However, materials and systems data, which are of considerable value, were obtained on the high-temperature irradiation behavior of hypo and hyper-UC, sodium-bonded to stainless steel and Nb-1 Zr alloy. A summary follows of the information and conclusions from EXP-NRX-85-1 and -3 capsules (from NAA-SR-11134).

1) Evidence of fuel grain boundary movement and "sweeping" of fission gas bubbles was observed. Grain boundary separations and high porosity in the uranium grain boundary phase also indicated that fission gas release from the fuel slugs was promoted along these routes.

2) Radial redistribution of the uranium phase occurred in varying degrees in all the hypo-UC fuel specimens.

3) Hypo-UC can be decarburized in the UC-Na-304 SS system provided that: (1) the fuel temperature is greater than 2350 to 2550°F (1300 to 1400°C); (2) there is insufficient free uranium metal in the fuel structure to react with all the metastable carbon at temperature (>4.68 wt % C in UC); and (3) there exists a carbon sink in the system, such as Cr in 304 SS. These conditions allow the liquid metal to carry the carbon from the fuel to the chromium constituent of the stainless steel cladding.

4) It is possible that the reactions observed to have taken place between the 304 SS and the hypo-UC fuel in Compartment 2, X-85-1, were $U_6Fe-UFe_2$, $U_6Fe-UFe_2-UC$, and/or U_6Fe-UC ; and were caused by the large amount of free uranium redistributed to the surface of the fuel in that compartment.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

5) Postirradiation tensile tests on the cladding showed little or no reduction in the short-time ductility at room temperature after integrated thermal neutron exposures of 2.1×10^{21} nvt ($\sim 4.2 \times 10^{19}$ nvt fast, 7.1 Mev) at temperatures of about 300°F (150°C). A ductility decrease was noted at 900°F (480°C), which did not recover at 1200°F (650°C). At 1200°F, the cladding ductility was lower by approximately a factor of 3 than at room temperature.

6) The fracture modes in the fuel area of the failed claddings appeared to be both trans- and intergranular, with little or no exhibited ductility at the fracture point. The fractures observed in the 1200°F (650°C) postirradiation short-time tensile tests also appeared to be of a similar brittle nature.

2. AI-24-1 Series

The irradiation demonstrated that an experiment of the AI-24 series design can be used to reliably determine the fuel rod internal pressure buildup and can, by taking pressure-data at both reactor full power and shutdown conditions, allow separation of the amount of pressure buildup due to fuel swelling and the amount due to fission gas release.

The LVDT displacement transducer-Inconel diaphragm combination was found to be sensitive and responsive throughout the irradiation. Independent calibration checks indicated good accuracy. Useful data on the conductivity of irradiated UC were obtained with the five in-fuel thermocouples (refer to Section III-D-1).

B. MECHANICAL PROPERTIES

From a comparison of the UC microhardness data for the hyper-UC and the hypo-UC samples, it is seen that the hypo-UC material is harder at low temperature and softer at temperatures above about 600°C. It is to be expected that hypo-UC material will be softer because of softening effects due to the free uranium metal. An explanation of the reversal of this effect at low temperatures may be that internal stresses occur in hyper-UC material because of differential expansion around the UC₂ precipitates. Such stresses may also be related to cracking, which has been observed to occur around the hardness indents.

C. PHASE EQUILIBRIUM

Microstructural observations and x-ray diffraction methods indicated that the metastable distribution of phases in UC fuels at room temperature is dependent on prior heat treatment, and that fuel microstructural changes will occur at proposed LSGR temperatures. The 2-phase grain boundary structure observed in both as-cast and heat-treated fuel samples has been tentatively identified as alpha uranium containing small grains or dendrites of UC_(1-x), where x varies approximately linearly from 0.015 to 0 between 3630°F (2000°C) and 2050°F (1117°C), respectively. These data essentially confirm Buckley's work on the solid solubility of U in UC above 2050°F (1117°C).

D. UC THERMOPHYSICAL PROPERTIES

1. Out-of-Pile UC Thermal Properties

On the basis of the data on UC thermal diffusivity derived at AI, and comparing the data with those in the literature, the accuracy in thermal diffusivity values is

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

estimated to be $\pm 5\%$. The specific heat data are believed to be accurate to about 7% up to about 500°C, with somewhat greater uncertainty at the higher temperatures. Thus, the calculated thermal conductivity values are estimated to be accurate to about 10% over the entire temperature range reported.

2. In-Pile UC Thermal Properties

Analyses and statistical data reduction have been made on the thermal histories of several high temperature uranium carbide irradiation experiments, which have been conducted by AI over the past two years, to determine the thermal conductivity of irradiated UC. Preliminary observations indicate that the thermal conductivity decreases, at exposures of less than 10^{17} nvt, to about 75% of its unirradiated value.

Data from seven experiments were considered in the evaluation. All seven were well instrumented, containing thermocouples both in the UC fuel and surrounding it. The AI-24-1 experiment contained 5 in-fuel thermocouples which confirmed the temperature distribution predicted by uniform heat generation in the fuel considering self-shielding by the fuel sample. The UC (4.4-4.9 wt % C) fuel specimens were 1/2-in.-diameter rods, 10 to 18 in. long, and were 7 or 10 wt % U^{235} enriched.

The data from the different experiments are quite consistent, considering that the experiments were designed as demonstration fuel rod tests, and not to measure thermal conductivity. An average correlation coefficient of 0.95 for the experiments was obtained, which indicates that random errors in thermocouple signals, recording apparatus, and data transcribing reduction were small. Large systematic errors are unlikely since different capsule designs were involved, different test positions in different test reactors were used, and structural materials with well-known thermal properties were used.

The possibility exists that the out-of-pile conductivity values, which have been extensively measured at AI using a laser thermal-diffusivity technique, are incorrect. However, the out-of-pile thermal conductivity values are in agreement with measurements made at other laboratories on arc-cast UC.

One unfortunate aspect of the analyses is that the experiments have high fuel flux levels, so that a 5×10^{16} to 10^{17} nvt exposure is attained while the experiment is still rising to design temperature and before the test reactor attains equilibrium power. This transient power situation lends enough uncertainties so it can not be said definitely that the transient conductivity decrease can be seen on the temperature-recording charts.

Other laboratories have compared electrical resistivity and lattice parameter increases in carbide fuels, and found them to saturate in a neutron exposure of 10^{16} - 10^{18} nvt. Frost recently published a review of fuels technology in which he described resistivity changes with irradiation and postirradiation annealing. The postirradiation annealing curves shown by Frost indicate that short-term irradiation electrical resistivity increases do not anneal out entirely until 1292 to 1472°F (700 to 800°C) temperatures are reached.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

The question is how much of the irradiation damage that causes these apparent thermal conductivity changes at low temperatures will be annealed out at high temperatures and high flux levels. These observations require confirmation and additional effort before they are regarded as final.

E. UC-SS COMPATIBILITY

Definite zones of interaction can be observed and measured in the specimens heat-treated with and without sodium at 1400 and 1600°F; the reaction layers thicknesses were of the order of 0.001 in. in the samples heat-treated for 1000 hr. In some cases, contact areas of 316 SS treated with Na, the transformation layer shows a "cast" structure (Figure 3) indicating the presence of a low melting temperature phase. In addition to the discrete reaction band, a less well defined zone of several mills thickness was observed, where it is believed cold-work-enhanced carbide precipitation, similar to sensitization, took place in the stainless steel during the exposure. The reaction zones shown for the 347 SS diffusion couple are somewhat less well defined (Figure 4). The specimens treated at 1200 and 1300°F showed transformation zones in very localized areas, and the thickness of the new structure is only a fraction of a mil. The superficially enhanced carbide precipitation is also very marked and makes it difficult to differentiate this effect from an interaction with the UC.

However, considering the width of the reaction zones (maximum of ~0.002 in.) for 4.2 wt % C UC-SS couples in contact in sodium for 5000 hr at 1400°F, there appears to be little compatibility problems for LSGR conditions, i.e., T clad $\geq$ 1250°F. These data essentially confirm those reported by Elkins (NAA-SR-7502, 1964).

7519-2522
Figures 3 and 4. Reaction Zone in Type 347 Stainless Steel (right) and Carbide Precipitates in Type 316 (left)

NAA-SR-10450

II-47

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Large Sodium Graphite Reactor

Project: UC Process Development

Project Engineer: B. R. Hayward

Reporting Period: Fiscal Year 1965

AEC Category: 04-01-04-21

General Order: 7519

Subaccount: 1959

I. PROJECT OBJECTIVES

The general objective of this project is to develop a low-cost process to produce cast UC slugs to close composition specifications indicated to be required for long burnup high-temperature performance in the LSGR.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. ALTERNATE MELTING-AND-CASTING FEASIBILITY STUDY

The hollow cathode discharge (HCD) melting method, a plasma cathode electron beam process, has been evaluated as a melting process for UC sintered feed-stock. The HCD melting method does not require the use of graphite electrodes, thus eliminating a major factor in UC compositional control problems associated with conventional arc-melting techniques. It is related in operation to high-voltage electron beam melting, but differs specifically by operating at a low voltage and at a furnace pressure in the range of 20 to 200 microns, whereas the electron beam melting furnace operates under high vacuum in the 10^{-5} torr range. Melting of UC at the higher pressure of the HCD furnace was expected to reduce uranium volatilization losses and attendant composition control problems encountered in prior electron beam melting experiments.

Out of three 12-button melt series made with different lots of sintered UC melt stock, the best results were obtained with a melt stock analyzing initially 4.69 wt % C and 675 ppm oxygen. The average carbon content of the resulting 175-gm UC buttons was 4.72 wt % the carbon range for the 12-button series was 4.64 to 4.82 wt %, or ± 0.09 wt %. Carbon range in the other two melt series was ± 0.10 and ± 0.145 wt %, respectively.

The HCD melting experiment was evaluated by a multiple regression analysis of the run data to determine trends in the relation of melting furnace operating variables to carbon content of the UC product. The results of the statistical evaluation show that the initial melt stock carbon content and a number of furnace operating variables including pressure, voltage, amperage, and melt time are all significant factors in determining the final carbon content of the UC product. The relationship between significant process variables can be expressed in the form of a regression equation which will predict the final carbon content of the UC product to within 0.088 wt % C at the 95% confidence level. For a given sintered UC melt stock carbon analysis, control of cast UC composition within a carbon level range of ± 0.1 wt % should therefore be possible in the HCD melting method.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

A statistical comparison study was made to attempt to determine the relative potential of the HCD melting process to the presently used carbon arc melting process. The evaluation study included a comparison of the predicted ability of the two methods to meet a set of arbitrarily selected specification ranges. Results of this study indicate that the HCD process is potentially superior to the carbon arc melting process. The overall comparison showed that the HCD process possesses a significantly smaller variation from the regression surface than does the carbon arc melting process. This means that the final UC carbon content is probably more predictable when using the HCD process, as here shown, for example, for two representative UC product carbon ranges.

Product Carbon Range (wt %)	Predicted Percent of Material in Range	
	HCD Melting Process	Carbon Arc Melting Process
0.40 (± 0.20)	>99.9	90.1
0.30 (± 0.15)	>99.9	78.3
0.20 (± 0.10)	99.9	59.1

Prior to the reorientation of the LSGR program to the fast reactor program, a contract was issued to Battelle Memorial Institute for performance of UC melting experiments by plasma arc techniques. These experiments were intended to determine the effect of plasma arc melting on uranium monocarbide stoichiometry. At the time of LSGR program reorientation, this study was still in the equipment shakedown phase.

B. CARBON ANALYTICAL TECHNIQUE IMPROVEMENT

This task constitutes part of the overall project effort to achieve closer process control of UC stoichiometry at reduced cost. As an aid to the planning of subsequent experimental work leading to improved analytical techniques for UC processing, a study and report has been completed, summarizing the current state of the art of carbon analytical techniques relating to UC processing.

A Leco 50-sec carbon analyzer, which operates on the principle of thermal conductivity measurement of the gases produced by combustion of a UC sample, was modified to further improve its precision. Difficulties were encountered during the initial experimental runs with the modified instrument, due to non-linearity of the amplification system. Corrective action was taken, and a statistically designed test series was in progress, but incomplete, at the time of reorientation of the LSGR program. Purpose of the test series was to evaluate the accuracy and precision of the modified analyzer.

A feasibility study has been made on the use of an emission spectrograph to determine carbon in UC. Exploratory tests run on a Baird-Atomic 3-meter concave grating spectrograph showed the presence of the 2478.573 angstrom

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

carbon line. Subsequent feasibility study experiments were conducted with cast uranium-carbon compositions in the 2.0 to 5.5 wt % carbon range. Feasibility of the method was demonstrated for cast UC compositions below 4 wt % but reproducibility of results was inadequate for cast carbide of higher carbon content. Experimental difficulties at the higher carbon levels were due to (1) operation of the emission spectrograph in an air atmosphere, and (2) breakup of the brittle UC specimens upon arc impact. These difficulties are not expected to be a factor in the ultimate application envisioned for this analytical method, namely as an on-line process control tool applied to UC in the molten state under inert atmosphere conditions.

C. PROCESS ENGINEERING ANALYSIS

The objective of this task is to determine the capabilities and limitations of current UC fuel fabrication processes, for the purpose of providing engineering guidance for subsequent UC process development efforts.

An engineering analysis of the HNPF Core II UC fuel fabrication process has been completed. In addition to analyzing process yields and costs, a statistical evaluation was performed to determine which process variables have a significant effect on the final fuel slug carbon content, and their relative degree of importance. Of twenty process parameters considered, these six were found to have a significant effect:

- 1) total arc-melting time
- 2) graphite electrode weight
- 3) ending wattage
- 4) time-averaged amperage
- 5) total watts squared
- 6) $(\text{time-averaged watts})^2 \times \text{electrode weight}$.

Under the current UC fabrication process practice of adjusting the starting crucible melt charge carbon level down to 4.0 wt % C by adding uranium metal, variations in UC melt stock carbon content were not considered an important variable, even though UC melt stock yield to compositional specification is only ~ 60.2%. If fuel casting were done by a nongraphite electrode melting process (such as HCD) where tighter carbon control of the cast product can be achieved, improvements in UC melt stock carbon control would offer an economic incentive. In the carbon arc melting process, up to 0.8 wt % C is expected to be drawn into the UC melt from the graphite electrode.

The statistical evaluation developed a multiple regression equation which permits a prediction of process capability to meet selected carbon ranges, as shown on the following page.

The actual yield of fuel slugs, prior to machining, for the entire HNPF Core II fabrication effort was found to average 89.0% in the carbon range 4.4 to 4.8 wt % (84.2% after machining), and 62.1% in the range 4.5 to 4.7 wt %, based on header analysis. Thus, the actual yields obtained were in excellent agreement with the predicted yields shown.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Product Carbon Range (wt %)	Predicted UC Within Carbon Range By Carbon Arc Casting (%)
0.50 (± 0.25)	96.1
0.40 (± 0.20)	90.1
0.30 (± 0.15)	78.3
0.20 (± 0.10)	59.1

D. PROGRESS PARAMETER INVESTIGATION

A series of experiments to establish process parameters leading to improved composition control in UC melt stock fabrication has been partially completed. The objective of this task was to modify the current UO_2 carbothermic reduction process by changes in operating conditions and/or changes in raw material specifications, so that higher yields and more uniform UC melt stock meeting the LSGR compositional specification could be obtained.

The reference UO_2 carbothermic reduction process involves dry V-blending of ceramic grade UO_2 (a non-free-flowing powder), graphite powder, and a binder; the blend is subsequently formed into $\sim 1/4$ -in. agglomerates, then sintered in a graphite crucible in a vacuum induction furnace at a finishing temperature of 2050°C . Small-scale experiments utilizing free-flowing oxide powder or UO_2 -graphite mixtures which had been ball-mill blended resulted in no significant improvement of UC melt stock uniformity, but indicated that less carbon pickup from the crucible at finishing temperatures lower than the reference 2050°C might result in a more uniform sintered UC melt stock composition. The experimental results also indicated that the UC product generally contained combined carbon plus oxygen in the 4.70 to 4.75 wt % range when graphite input was calculated for a nominal 4.65 wt % C composition.

In subsequent scaleup tests at the 10-kg and 20-kg level, graphite input was adjusted to aim for a UC composition of 4.70 to 4.75 wt % C, and a 1950°C finishing temperature was used. In 8 experiments out of a total of 14, the UC product exhibited a variability of approximately ± 0.02 wt % C per batch. Results in the remainder of the experimental series were sufficiently random to preclude any firm conclusions to be made.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

An evaluation of UC buttons produced from sintered UC melt stock by hollow cathode discharge (HCD) melting, a nongraphite electrode melting process, has established that control of cast UC composition within a carbon level range of ± 0.1 wt % should be possible. Results of a statistical comparison study have shown that the composition variation in the HCD melting process is significantly smaller than in the carbon arc melting process. A series of UC slug casting experiments, will be required to reliably establish the validity of this preliminary evaluation.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

A review of carbon analytical techniques has confirmed that the Leco 50-sec carbon analyzer method is a relatively low cost method and produces control data of sufficient accuracy for the currently required 0.4 wt % carbon range in UC. For tighter LSGR compositional control requirements (± 0.1 wt % C), new equipment developments which can increase the precision of the Leco 50-sec Analyzer, should be incorporated into the carbon analysis method.

Feasibility of the application of emission spectrographic techniques to the analysis of carbon in UC has been demonstrated. The ultimate application of this analytical method is as an on-line process control tool, applied to UC in the molten state. Molten UC samples excited by dc arc under an argon atmosphere are expected to provide rapid, reliable, and reproducible carbon measurements. Inadequate reproducibility of analytical results encountered with cast UC slugs of >4 wt % C content, because of brittleness of the test specimens, will not be a factor in the ultimate application of this method of carbon analysis. Future work is warranted by the results to date.

Statistical analysis of data derived from the HNPF Core II UC fuel fabrication process has shown that current process yields cannot be improved without changes in equipment and process procedures. If yields of 90% or greater should be required at a cast-UC carbon range specification of less than 0.4 wt % a change from carbon arc casting to a nongraphite electrode melting process should be considered, in conjunction with process changes leading to tighter composition control of the UC melt stock.

Small-scale process parameter experiments on the UO_2 carbothermic reduction process have indicated that sintered UC melt stock of more uniform carbon composition results when a finishing temperature of 1950°C is used instead of the 2050°C reference temperature. A change in nominal carbon composition to the 4.70 - 4.75 wt % C range instead of the reference 4.65 wt % C level appears also to be required. Additional experimental data with production-size batches is required to confirm these tentative conclusions.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Large Sodium Graphite Reactor	
Project: SRE Core III Test Elements	
Project Engineer: B. R. Hayward	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-21
General Order: 7519	Subaccount: 1961

I. PROJECT OBJECTIVES

The objective of this project was to fabricate four test elements for irradiation in SRE Core III. Two Special Test Elements will be fabricated from 15% enriched hypostoichiometric UC fuel and seamless 304 stainless steel cladding. Two elements, designated as Standard Test Elements, will be fabricated with the 6.5% enriched hypostoichiometric UC fuel and 304 stainless steel cladding used for the Core III driver elements. All of the test elements will be instrumented to evaluate fuel performance during reactor operation.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

UO₂ powder, 15% enriched, was purchased, converted to UC, and cast into 0.500-in.-diameter fuel slugs for the two Special Test Elements. The 6.5% enriched UC fuel slugs for the two Standard Test Elements were obtained from the production run of UC fuel for the SRE Core III driver elements. Type 304 stainless steel in two conditions, seamless and welded-drawn, was purchased as the cladding for both the Special Test and Standard Test Elements. The seamless tubing was used for the Special Test Element cladding with welded and drawn tubing for the Standard Test elements. Procurement also included thermocouples, hanger rods, and shield plug assemblies for these elements.

Selected fuel slugs, both 6.5 and 15% enriched, were submitted for complete dimensional preirradiation characterization (length, diameter, metallography, chemical analysis, and density). Slugs from the thermocoupled rods were Eloxed and core-drilled for insertion of the in-fuel thermocouples.

The fuel rods were fabricated by loading the slugs into the cladding tubes, sodium-bonding, and fusion welding the end closures. All fuel rods were inspected by x-ray for sodium level, by hot eddy current tests for bond quality, and by dimensional and visual examination. After inspection, the rods were assembled into fuel element bundles, the bundles attached to the shield plug-hanger rod assemblies, and the completed elements placed in storage preparatory to reactor insertion. Two additional driver elements were fabricated under this project to replace the test elements deleted earlier in the year.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The scope of this project was reduced during FY 1965 to a 6-element fabrication effort. The revised technical effort was based on physics requirements

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

for the core, schedule and cost, and LSGR program objectives. The objectives of this project were accomplished. Delivery of the elements to SRE was about six weeks late; however, this was in accord with the overall SRE-PEP schedule.

These were a few minor fabrication problems encountered due to tight specifications on dimensional tolerances. The use of the mockup assembly prepared under Subaccount 1955 greatly reduced the fabrication problems on the actual fuel elements.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Large Sodium Graphite Reactors	
Project: SGR Dry Control Rod Concept Development	
Project Engineer: K. W. Foster	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-21
General Order: 7519	Subaccount: 3210

I. PROJECT OBJECTIVES

The objective of this project is to complete the design, development, and testing of a developmental, dry control rod model of a variation of the Hallam control rod design. Test results from this developmental model will provide the basis for the design of a complete prototype, thimble-enclosed control rod suitable for use in the high specific power core of the near-term large SGR prototype plant.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

The reference design for the SGR Mark IV control rod consists of dysprosium oxide (neutron absorber material) located in the annulus formed by an inner core of graphite and a tube of Hastelloy-X which is also the structural support member (see figure).

7519-2535

Cutaway View of SRE-PEP
Control Rod

Five poison columns (four operating and one spare) suitable for use as control elements in the SRE-PEP were designed and fabricated. Each poison column consists of a Hastelloy-X tube of 2.298-in.-OD by 0.049-in. wall thickness and a filler of graphite which provides an 0.184-in. annulus for powdered dysprosium oxide packed to a density of 5.5 g/cc. The poison columns will be actuated by standard SRE drive mechanisms and operated within a helium-filled stainless steel thimble. Utilizing LSGR absorber materials as control rods in the SRE-PEP allows not only a determination of the effects of long-term irradiation on these materials, but also presents the opportunity to accumulate operating experience with this design configuration.

All short-term elevated temperature tests were completed. These included wear tests of Hastelloy-X versus stainless steel Type 304 at 1450, 1650, and 1825°F. In the 1450 and 1650°F tests, grooves of about 0.012 in. deep formed as "wear paths" on the specimens. These paths were from

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

1/16 to 1/8 in. in width. Both the 304 SS and the Hastelloy-X had grooves as well as material "buildup" on each of the test specimens. Hardness tests made at room temperature showed that the "wear product" (built-up material) was harder than either parent metal. It is apparent that this wear product, once built up, wears both the 304 SS and the Hastelloy-X. Total differential travel between specimens at each test temperature was approximately 200,000 in. This amount is equivalent to about one year of wear under reactor operating conditions. The effect of oxygen on the wear test specimens appears to be a most significant finding in this experiment. It was shown that a lower oxygen content of the wearing environment yields much less wear than a higher oxygen content. This suggests that future efforts be made to assure the lowest oxygen content in the thimbles in which the LSGR control rod operates. A detailed summary of the results of the investigation of the wear characteristics of Hastelloy-X versus SS Type 304 has been published in NAA-SR-TDR-11330.

The compatibility of Hastelloy-X with dysprosium oxide was investigated by packing small Hastelloy-X tabs in dysprosium oxide inside thick-walled stainless steel capsules. These capsules were exposed to 1850°F continuously for 1000, 2575, 5000, and 10,000 hr. An additional capsule exposed to 1850°F for 7500 hr was not scheduled for opening and examination by the date of writing of this report. Compatibility of the reference materials was evaluated by chemical analyses of the dysprosia and by visual, metallurgical, and electronic determinations of compositional changes of the Hastelloy-X. All tests performed indicated excellent compatibility between the two materials up to 10,000 hr. A thin layer of dysprosia in each capsule was stained dark green where it contacted the Hastelloy-X. Spectrographic analysis showed this discoloration to be a minor (fractional ppm) buildup of both chromium and iron in the layer of dysprosia. Metallographic analysis of the Hastelloy-X revealed a surface layer where the Hastelloy alloy content may have been affected by the dysprosia. The depth of this layer proceeds to approximately 0.0005 to 0.001 in., and is then apparently saturated.

Although Hastelloy-X was found to have excellent compatibility with dysprosia, there was noted a change in microstructure of Hastelloy-X when exposed to 1850°F in excess of 5000 hr. This change is quite independent of the dysprosia environment. A new phase developed after several thousand hours of thermal exposure. This growth occurred primarily in the grain boundaries of the Hastelloy-X. The new phase contained almost all the chromium and molybdenum originally constituted in the alloy. This secondary phase occupies about 30% of the material cross-section and is extremely hard (800 to 1200 Knoop). The remaining 70% of material cross-section contains nearly all of the iron and most of the nickel, and has a hardness (200 to 300 Knoop) equal to about one-fourth of the chrome-moly region. The hardness tests indicate that very little ductility may be expected in Hastelloy-X after exposure to 1850°F in excess of 9000 hr and that reduction in ductility probably commences after 5000 hr. A detailed summary of these compatibility tests has been published in NAA-SR-TDR-11370.

Short-term compatibility test results between dysprosia and graphite at 2200°F were largely inconclusive. However, this information may be obtainable by hot-cell examination of the control rod absorber column after final removal from the SRE-PEP.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Full-scale scram and shim testing of the reference developmental control rod (Mark IV) at elevated temperatures proceeded to an accumulation of 1,000,000 in. of travel between the Hastelloy-X clad poison column and the stainless steel thimble. Ninety-four percent of the accumulated wear was accomplished at LSGR reactor profile temperatures (1150 to 750°F) and, based on the table below, approximates 5 yr of shim and scram service. During this period of testing, scram rates remained relatively unchanged at 0.7 g for 648 scrams. This rate is well in excess of the required minimum scram rate of 0.1 g.

TABLE
ANTICIPATED ROD TRAVEL IN LSGR

Period (yr)	Scrams		Shim Travel			Total Accumulated Travel (in.)
	Number	Travel (in.)	Rate (in. /hr)	Duration (days)	Distance (in.)	
1	200	60,000	30	100	72,000	132,000
2	100	30,000	30	200	144,000	306,000
3 to 5	150	45,000	30	300	648,000	999,000
6 to 10	125	37,500	30	1,500	1,080,000	2,116,500

An examination of the Mark IV absorber column at the conclusion of these tests showed nominal amounts of material pickup and considerable wear. However, the wear noted was not considered restrictive and the fact that no "binding" occurred during shimming operations, as well as the scram rate remaining at 0.7 g, was positive evidence that the design configuration was acceptable so far as accumulated effects of wearing are concerned.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

Based on the successful completion of the elevated temperature full-scale test program which simulated approximately 5 yr of reactor-type operation, it appears that a thimble-enclosed concept will meet performance specifications of the next large SGR. The laboratory wear tests of Hastelloy-X versus stainless steel Type 304 and the compatibility tests of Hastelloy-X with dysprosium oxide provided corroborative evidence that this design configuration is adequate. In-pile test data and an operating history of similar configuration shim rods will be obtained from the SRE-PEP. These data will provide information concerning the effects of long-term irradiation on this configuration of materials.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Large Sodium Graphite Reactors

Project: SGR Dry Control Rod Prototype Development

Project Engineer: K. W. Foster

Reporting Period: Fiscal Year 1965

AEC Category: 04-01-04-21

General Order: 7519

Subaccount: 3220

I. PROJECT OBJECTIVES

The objective of this project is to fabricate, assemble, and proof-test, under closely simulated reactor operating conditions, a prototype of the final production control rod design in the 200-Mwe prototype plant. Results of these tests will establish final control rod operating characteristics and demonstrate conformance to design and performance specifications. In addition, this project will provide detail and assembly drawings, and final analyses (nuclear, thermal, and structural) of the prototype control rod design.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

As-built drawings prepared during the Mark IV control rod test program (Project 7519-3210) were reviewed. A reference layout depicting the as-tested Mark IV design was partially completed. No further progress was made because a change of funding commitments led to termination of project activities.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

Little direct progress had been accomplished under this particular project when terminated. Manufacture and assembly of the SRE-PEP control rods, which are being funded under 7519-3210, SGR Dry Control Rod - Concept Development, will provide information on control element fabricating techniques. Other supporting efforts under Subaccount 3210, such as material wear and poison compatibility tests, will provide a firmer basis for a thimble-enclosed control rod design.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Large Sodium Graphite Reactors	
Project: SGR Wet Control Rod Conceptual Development	
Project Engineer: K. W. Foster	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-21
General Order: 7519	Subaccount: 3230

I. PROJECT OBJECTIVES

The objective of this project is to design, develop, fabricate, and test a reactor control element which is cooled directly by liquid sodium.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A reference drawing of an improved control rod actuator design utilizing a rack and pinion was prepared. This design, designated Mark V, features a "floating" rack and pinion concept and a sealing bellows having a 39-in. stroke.

Fabrication of the test stand and actuating mechanism for the telescoping rack and pinion drive mockup was completed. The rack and pinion gear box and stroke multiplying assembly were brought to 90% completion. This unit will provide basic information concerning the problem areas associated with rack and pinion drives having motion ratios of 1:4.

Completion of this assembly and preliminary checkout operations were performed under the SCCR program in connection with the mechanisms in sodium project, 7593-1329. Testing of a Bellmetrics, 316 stainless steel (3-in. stroke) bellows in sodium proceeded to 43,000 cycles at 1200°F, at which time a malfunction of the cycling mechanism caused a shutdown of the test. The cycles completed represent 430% of design life in shim mode. The stroke to compressed-length ratio was maintained at 4:1. Disassembly and examination of the bellows and cycling equipment indicated that a pinhole leak had developed in the bellows. The leak sodium entered the bellows, causing a jam in the cycling equipment.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The service so far demonstrated by the second Bellmetrics bellows closely parallels the success obtained in tests of a similar bellows in FY 1963. The performance of the Bellmetrics bellows alleviates to a great extent some of the initial reservations regarding the use of bellows in the wet rod design. Analysis of the mode and type of failure of the bellows and evaluation of the tests of the rack and pinion drive arrangement will have a significant influence on the ultimate design of the sodium-cooled control rod.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Large Sodium Graphite Reactors

Project: SGR Calandria Structural Tests

Project Engineer: K. W. Foster

Reporting Period: Fiscal Year 1965

AEC Category: 04-01-04-21

General Order: 7519

Subaccount: 3610

I. PROJECT OBJECTIVES

It is the objective of this project to demonstrate the overall long-term structural reliability of the calandria core for the large SGR via structural tests on full-size components (or portions thereof), or on small-scale hardware as deemed necessary. Because present-day analytical predictions of creep deformation, creep buckling, and stresses in critical locations cannot be relied upon, structural tests of the kind outlined herein are required.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. PROCESS TUBE RADIAL BUCKLING STUDY

Two additional creep-buckling tests on the 4-in. OD by 0.025 in., 304 stainless steel process tubes were initiated. One test was conducted at a lower bound radial pressure of 3.0 psi at 1200°F in the radial buckling test fixture described in earlier reports. The test was discontinued at the end of 1464 hours because of a power failure and subsequent overpressurization. The second creep-buckling test, still in progress after about 5700 hour is being conducted at a pressure of 4.5 psi at 1200°F. This pressure was estimated to be high enough to cause collapse in less than 10^4 hr, but low enough to determine whether the radial pressure vs time-to-collapse curve shows any signs of leveling off at the low end of the curve. See Figure 1.

A topical report (NAA-SR-11323) has been written which contains a description of the test apparatus, test techniques, test data, and other pertinent information obtained during the course of this investigation.

B. PROCESS TUBE AXIAL BUCKLING STUDY

Modification of the apparatus to permit testing of full-length process tubes with the bellows at the bottom was completed. The apparatus was originally designed to accommodate a process tube assembly with the bellows at the top. Installation of high-temperature electric resistance strain gages, thermocouples, and deflection probes was also completed, and installation of the entire assembly at the test site was virtually completed at the time all work was terminated. A technical development report (NAA-SR-TDR-11423) has been written which contains a description of the test apparatus and its capabilities.

C. CALANDRIA VESSEL STAYED HEAD STUDY

Installation of the room temperature electric resistance strain gages and linear variable differential transformers on the stayed head was completed. After checkout and calibration of the instrumentation, room temperature tests

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

7602-2523

Figure 1. Process Tube Collapse Pressure vs Time

TABLE 1
SUMMARY OF STAYED HEAT STRESSES AND DEFLECTIONS⁽¹⁾

Strain Gage or Dial Gage Location	Stress ⁽¹⁾			
	Normal Core Operating Condition (1 psi, Hangar Load 1200 lb) ⁽²⁾		Loss of Core Pressure Condition (5.5 psi, Hangar Load 3450 lb) ⁽²⁾	
	Maximum Principal Stress	Minimum Principal Stress	Maximum Principal Stress	Minimum Principal Stress
35	+1098	+ 248	-1925	-10,060
36	+4050	+1600	-1750	- 7,200
38	+3600	+2600	-4800	- 6,250
42	+8300	+3200	-3400	-12,800
43	+5600	+1350	-1500	- 9,900
	Deflection (in.)			
	1	+0.013	-0.029	
	2	+0.048	-0.101	
	3	+0.058	-0.115	
	4	+0.046	-0.098	
	5	+0.014	-0.028	

Note: (1) + indicates tensile stress or deflection towards hangers.
- indicates compressive stress or deflection away from hangers.
(2) Simulated process channel concentrated load of 360 lb applied at center of blanking cap for each gun barrel for Normal Core Operating and Loss of Core Pressure Conditions.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

7519-2524

Figure 2. Elevated Temperature, Stayed-Head Test Fixture

were conducted. Figure 2 is an overall photograph of the test setup. Figures 3 and 4 are closeup views from above and below the test grid plate. Table 1 is a summary of the measured stresses and deflections. Refer to Figure 5 for gage locations.

Under the normal core operating condition, 1-psi pressure differential, 1200-lb hangar load, the surface of the test grid plate (corresponding to the sodium side of the prototype) was found to be in tension, the maximum tensile stress being at strain gage location 42. Here, midway between the toe of the cruciform and the midpoint of a line between toes of the cruciforms, a maximum tensile stress of 8300 psi was measured. At the midpoint of the line between toes of the cruciform (gage location 43), a tensile stress of 5600 psi was measured.

For the loss of core pressure test condition, the surface of the grid plate (corresponding to the sodium side of the prototype) was found to be in compression, the maximum compression stress being 12,800 psi again at gage location 42. At an opposite point on the side corresponding to the process tube side of the upper grid plate, a tensile stress of a lesser amount would exist. Since this point is sufficiently removed from the toe of the cruciform, it is reasonable to assume that the stress at a corresponding point on the opposite surface of the grid plate would be of about the same magnitude, but tensile in nature. At the midpoint between

7519-2525

Figure 3. Close View from Above Stayed-Head, Test Setup

7519-2526

Figure 4. Close View from Above Stayed-Head, Test Setup

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

□ TWO AND ◻ THREE GAGE ROSETTES, RESPECTIVELY
⊗ DEFLECTION MEASURING GAGES

Figure 5. Stayed-Head
Grid-Plate Test
Gage Locations

as high as 12,490 psi were generated for a 1/32-in. differential settlement of the center hanger.

Technical development reports NAA-SR-TDR-11324 and 11433 have been written, which contain a description of the test apparatus, test techniques, test data, accelerated creep calculations, and other pertinent information obtained during the course of this investigation.

D. CALANDRIA VESSEL PERIPHERAL HANGER CONNECTION STUDY

Tests in a special fixture were conducted on a full-scale section of the reference design "calandria-vessel-peripheral-hanger-connection and calandria-vessel-wall combination," as shown in Figure 6. The width (circumferential) of the test article was enough to permit incorporation of three hanger connections, and its length was sufficient to minimize the effects of the lower boundary on the local stress distribution in the vicinity of the juncture of the connection to the calandria vessel wall.

To assist in the identification of points of high stress, a photoelastic coating was bonded to the test article on the regions of interest, and the stress patterns surveyed with the aid of a reflection polariscope. Nine electric resistance strain gage rosettes were subsequently installed at the points of interest and the stresses then measured at these points. A single gage was also installed on the tension side of one of the hanger bracket bars. The location of these gages is shown in Figure 7.

A hydraulic load of 33,000 lb (3.76 x design load) was applied to each of the three hanger connections. The highest stress measured (Gage 1) was 15,300 psi, corresponding to a stress of 4070 psi at design load. The point at which this

the two cruciforms (gage location 43), a maximum compression stress 9900 psi was measured. At a corresponding point on the opposite surface of the cruciform, a tensile stress of about this same amount would exist.

Because of time limitations, only room temperatures tests were conducted, these being limited to measurements on the test section representative of a typical interior region of the calandria head.

An examination of the stress and deflection data in Table 1 indicates that no inflection points were found during either of the two tests.

It should be noted that time did not permit (1) correcting the stresses and deflections summarized in Table 1 to account for variation in load from hanger-to-hanger or the inherent deformations in the fixture itself, or (2) determination of the stresses due to a prescribed differential settlement between adjacent hangers. Tests conducted in the room temperature single-hanger connection test fixture revealed that stresses

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

7519-2528

Figure 6. Peripheral-
Hanger-Connection-
Test Setup

7519-2529

Figure 7. Gage Location for
Calandria-Vessel
Peripheral-Hanger-
Connection Test

stress was measured was within 1/4 in of the top of the vertical fillet weld. The second highest stress measured (Gage 2) was 4400 psi, which corresponds to a stress of 1170 psi at design load. The point at which this stress was measured was on the shell, 3/4 in. below the lowest extremity of the connection. The single gage (Gage 3) mounted on a bar of the connection indicated a stress of 40,000 psi, which corresponds to a stress of 10,000 psi at design load. Table 2 is a summary of the stresses at design load.

TABLE 2
SUMMARY OF HANGER
CONNECTION STRESSES

Gage Location	Stress at Design Load (psi)
1	4,070
2	1,170
3	10,000
4	133
5	400
6	610
7	560
8	530
9	850
10	610

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

It should be noted that the thickness of the wall portion of the test article was 1-1/2 in. and that the above stresses do not include stresses arising from thermal expansion of the upper grid plate, or radial or axial temperature gradients in the vicinity of the connection.

A technical development report (NAA-SR-TDR-11415) has been written which contains a description of the test apparatus, test techniques, test data, and other pertinent information obtained during the course of this investigation.

E. PROCESS TUBE - GRAPHITE LOG STRUCTURAL INTERACTION STUDY

Testing was completed to determine whether the reference design slotted graphite log could withstand the predicted gross shrinkage and bowing anticipated from 30 yr of irradiation distortion in service. Because of the fast neutron flux gradients in the core, a differential shrinkage occurs which induces log bowing. The process tube restrains the radial shrinkage and bowing. The question therefore arises as to whether the slotted log could withstand this restraint without fracturing, and whether the contact pressures imposed on the process tube by the graphite logs are of sufficient intensity to cause local buckling or other damage to the process tube.

Because of time limitations, testing was restricted to room temperature tests of a prebowed log-process tube combination shown schematically in Figure 8.

Test results indicated that the graphite log can accommodate a maximum combined diametral change due to radial shrinkage and log bowing of at least 1-1/32 in. before log fracture occurs. The maximum combined diametral change was calculated to be 1.4 in. at the worst core location, over a 30-yr period, using the best available data on graphite distortion due to irradiation.

7519-2530

Figure 8. Prebowed Graphite-Log and Process-Tube Combination

gages having a gage length of 1/64 in. were used. Peak stresses of 13,700 psi in the longitudinal direction and 16,900 psi in the circumferential direction were determined on the basis of extrapolation of measured stresses corresponding to a combined diametral expansion of 0.27 in. to the maximum predicted value of 1.14 in.

A technical development report (NAA-SR-TDR-11416) has been written which contains a description of the test apparatus, test procedures, calculations, and all pertinent information obtained during the study.

NAA-SR-11450

II-65

Stresses in the process tube generated by graphite log contact pressures were also measured. Figure 9 shows the locations at which the contact pressures develop. Stresses were measured by means of electrical resistance strain gages bonded to the inner surface of the process tube in the vicinity of the contact areas. To minimize "averaging" effects,

7519-2531

Figure 9. Contact Pressure Zones on Process Tube

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

On the basis of information obtained thus far in the process tube radial buckling study, it is clear that the 4-in. OD by 0.025-in. wall 304 stainless steel process tubes tested should be capable of withstanding the 2-psi-net external radial pressure caused by a malfunction of the calandria vessel pressurization system for the 1000-hr period of time. It should be noted that this pressure would exist only along the upper portion of the process tube, where the internal pressure under static conditions could be as low as 5 psi. According to the present design criteria, the calandria vessel would never be pressurized in normal operation to a pressure high enough to create a net external pressure across the thin wall section (0.025-in. thick) of the process tube.

As to the behavior of the calandria vessel stayed head, the limited data obtained thus far does not permit general conclusions as to the overall structural performance of the head under combined mechanical and thermal loads. However, it can be said that, for the normal operating condition, the stresses in the interior regions due to a 1-psi pressure differential are within allowable limits of $3S_m = 12,500$ psi, as specified in Section III of the ASME Boiler and Pressure Vessel Code. Nevertheless, it remains to be demonstrated that these stresses, plus the stresses due to differential settlement and the thermal stresses are within allowable limits.

For the loss in core pressure condition, stresses as high as 10,060 psi were measured at points at the toes of the cruciforms. In the ligament areas of the grid plate, stresses between 9900 and 12,800 psi were measured. These stresses are sufficiently high to warrant further study before a final conclusion can be drawn regarding the performance of the grid plate for this condition.

Results from the calandria vessel peripheral hanger connection test indicate that the stresses at the attachment welds and between hanger connections are sufficiently low so that a change in hanger spacing or vessel wall thickness would not be required to resist the vertical design hanger loads. However, before a final conclusion can be drawn, it will be necessary to determine whether the combined stresses, due to the vertical loads, the discontinuity stresses, and thermal stresses, are low enough to preclude the possibility of a stress rupture mode of failure.

As to the hanger connection itself, an increase in the width and/or thickness of the bars comprising the connections will be necessary to reduce, by a factor of at least two, the combined direct stress and flexural stress due to vertical component of the hanger load, and any other loads.

Results from the graphite log-process tube structural interaction study indicated that the reference design process tube could withstand the maximum anticipated total diametral change due to radial shrinkage and bowing of 1.14 in. at room temperature. It has been observed that the tensile strength at 1200°F increases by about 1/3* and that the creep rupture strength increases up to ten-fold,† over the room temperature values. There is also a possibility that stress

*Industrial Graphite Engineering Handbook, National Carbon Co., Union Carbide Corp., Revision 4-64

†AERE-R-4566, W. N. Reynolds, "The Mechanical Properties of Reactor Graphite" (1964)

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

relaxation may moderate the stresses arising from neutron irradiation. Because of this enhancement in strength at elevated temperatures and stress relaxation, it is felt that the existing graphite log design should be able to accommodate the distortion due to irradiation, without fracture and with a sufficient margin of safety.

As to whether the process tube can sustain the contact pressures imposed by graphite log distortion, this is a difficult question to answer without conducting tests at elevated temperatures to determine whether local buckling or stress rupture occurs under sustained pressure for extended periods of time. If substantial stress relaxation does not take place, stresses of the magnitude measured in the process tube of nearly 17,000 psi could conceivably cause tube damage or affect flow characteristics due to local buckling or stress rupture cracking.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program:	Advanced Sodium Cooled Reactors		
Project:	SGR Calandria Maintenance		
Project Engineer:	K. W. Foster		
Reporting Period:	Fiscal Year 1965	AEC Category:	04-01-04-21
General Order:	7519	Subaccount:	5210

I. PROJECT OBJECTIVES

The objective of this project is to demonstrate the practicability of the concept of remote maintenance of a calandria vessel, which requires the replacement of process tubes and graphite in case of a sodium leak into the vessel. Instrumentation and techniques will be developed for detecting and locating a sodium leak in the calandria. The design basis for the tooling and equipment required for actual reactor maintenance operations will be provided as a result of experimental demonstration.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

The development of equipment for remote welding and cutting of the calandria process tube top and bottom joints was completed. Heliarc welding (tungsten inert gas) and cutting techniques for making and parting of calandria joints were extensively tested, and optimum setting for the equipment variables established. Welding and cutting operations were performed on two designs of bottom joints in a test vessel containing sodium with a helium cover gas where reactor maintenance temperature conditions were maintained - sodium-wetted joints were parted and replacement process tube sections positioned and welded. On the first design, the thin-wall (0.044 in. thick) joint section is an extension of the bottom grid plate. Twenty joint welds were satisfactorily made in the sodium test vessel in the last 20 attempts. It was suggested that by inverting the joint the heavy wall (approximately 1/4 in. thick) would become the extension of the fixed bottom grid plate, while the thin wall (0.040 in. thick) would be an extension of the removable process channel. A planned series of 20 inverted lower joint replacements were completed, with 19 helium-tight welds accomplished. Radiograph films of the joints revealed that the 19 successful welds complied with AI specifications (NA-0107-002) for reactor system welds. The one unsuccessful weld resulted from using a joint with a damaged (out-of-round) thin-wall section. Rework of the joint with a tube expander permitted successful rewelds. The inverted lower joint was chosen for the reference design because it permits greater simplification in the geometry at the lower end of the process channel, provides positive location of the weld and cutter head, and the more delicate thin-wall section of the joint is removed with the process tube. The general procedure used in the environmental tests includes these directions: (a) install the lower joint assembly in the test vessel, (b) soak the joint in 750 to 800°F sodium for 3 hr, (c) drain the sodium and cool the helium-sodium vapor atmosphere to 250 to 350°F, (d) part the joint with the cutting equipment, and (e) install the new process tube member and weld it to the remaining nozzle member. Figure 1 illustrates the sodium environmental test setup for the bottom joint tests. The

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

7519-2532
Figure 1. Bottom Process Tube
Weld Joint, Sodium-Environment
Test Fixture, and Test Vessel

cuts were made on the ten replacement tests performed. The initial welds were made using a welding current of 65 amp at a weld speed of 4 in./min. Some difficulty was experienced in removing the process tube section after cutting because of localized (0.010-in.-diametrical) shrinkage of the top grid plate extension, induced by prior welding. By providing a 0.020-in.-diametrical relief on the process tube joint part and increasing both the welding current to 75 amperes and the welding speed to 10 in./min, the shrinkage problem was eliminated.

The effect of a temperature environment on the top joint welding and cutting tools was demonstrated. A furnace was constructed to heat the welding and cutting tools to temperatures expected during reactor maintenance operations. A top joint was successfully welded and cut at approximately 300°F in the bench test stand.

weld and cutting sequence was repeated five times with each of four calandria sections to demonstrate the number of times a calandria process tube may be replaced.

Relocation of the bellows at the bottom of the process channel required that a modification be made in the lower joint welding tool. The tool must hold the lower end of the process tube assembly securely against the bottom grid plate extension during welding, since the spring action of the bellows could otherwise raise the lower end out of position. In addition to this requirement, it was found necessary to include a device for retracting the electrode to prevent its damage during the tool insertion and removal. Fabrication of the modified welding tool was completed, but assembly and demonstration of the tooling were deferred. Design changes to bottom joint and bellows portion of the process channel assembly made it necessary to redesign the process channel handling tool and the lower joint leak-checking tool. Both tools were redesigned and manufacture was completed.

Assembly and shakedown testing of the equipment, Figure 2, for welding and cutting the 7-1/2-in.-diameter top process tube joint was completed. Five complete replacements were made on each of two top joints in the bench test stand. Excellent welds and satisfactory

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

7519-2533

Figure 2. Welding and Cutting Equipment

Demonstration tests were performed with the graphite-handling machine in the 4-channel test vessel. The handling equipment was designed for installation and removal of graphite filler logs which surround the process tube assembly. Some difficulty was experienced in starting the first log course because the log support gussets were mislocated slightly. After flat plates were installed to provide a firm base for the logs, a full compliment of logs was stacked and removed from one channel of the test vessel. The stacked logs reached a height of approximately 15 ft.

Developmental activities on the remote maintenance tools, which were tested under this project, have been reported in NAA-SR-11202, "LSGR Remote Maintenance Development."

Two systems were being developed to provide information on a sodium leak into the calandria vessel. The first system involves circulating gas (helium) from the calandria atmosphere through a sodium vapor detector. This detector consists of a sodium lamp, which produces monochromatic light characteristic of excited sodium atoms, and a photometer. The circulating gas passes between the lamp and photometer. Any sodium vapor contained in the gas stream will result in a decrease in light intensity impinging on the photometer, because unexcited sodium atoms in the vapor phase will absorb light of their own frequency. This detection system will indicate if a leak exists, but will not specify its location. Development activities to date consisted of design and fabrication of the test apparatus.

The second leak-detection system not only confirms that a leak exists, but also indicates which process tube has failed. A recent calandria design change incorporates a thin-wall sleeve (tube) around each process tube. The space

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

between these two tubes provides a channel for directing leaking sodium to the bottom of the process channel, where a sodium collection cup is located. Several schemes were evaluated for monitoring the presence of liquid sodium in each collection cup. One technique involves connecting a tube to each cup and measuring the increase in liquid level by means of a manometer attached to the other end of the connecting tube (located outside the reactor). This scheme would provide an early indication of the rate of the leak, since the cross-sectional area of the cup would be known.

Seven different types of 0.002- and 0.003-in. wall sleeves with 3/4- to 2-in. vertical overlap tack welds, spiral tack welds, staggered vertical-horizontal welds, and stovepipe closures were fabricated and tested for leakage path. The sleeves with 0.010-in. radial clearance were placed over a "mocked-up" process tube having a 0.012-in.-diameter leak hole. The sleeve being tested was placed on the process tube at the most disadvantageous position for the seam closure (i.e., with the leak stream directly at one edge of the overlap or stovepipe closure). Water in the process tube was pressurized to 3, 5, 6, 10, and 14 psig and allowed to leak through to an atmosphere pressure region. The best seam for containing the leak fluid was the stovepipe closure with a 2-in. overlap retained by tack welds. Restrictions to downward leak flow of 50% of liner circumference (below leak) resulted in upward flow and consequent overflow instead of circumferential migration. Further work is required to assure circumferential flow of the leaking fluid.

Graphite in contact with liquid sodium is known to expand because of absorption of sodium. To obtain additional information on this phenomenon, tests were conducted on irradiated and unirradiated graphite specimens (approximately 3/4-in. cubes) immersed in 50 gm of sodium to determine dilation as a function of radiation exposure.

The dilation characteristics of unirradiated Speer Mold Graphite (HNPF moderator type) were established by exposing graphite cubes to liquid sodium (600 to 1200°F) for varying times, with the sodium oxide content held to less than 10 ppm. The graphite did not expand more than 1.2%, and in most cases was less than 1%. The tests indicate that saturation occurred rapidly (within a few hours) at temperatures exceeding 800°F and within 100 hr at 600°F. No increase in graphite expansion as a result of thermal cycling was observed in one test where the sodium temperature was varied between 500 and 1000°F. The effect of oxygen contamination on the dilation of graphite was investigated by exposing samples to 1000°F sodium containing 50 and 100 ppm oxygen for 100-hr periods. These tests indicate that there is no appreciable effect on graphite dilation if the sodium contains less than 100 ppm oxygen.

Dilation tests on unirradiated TSP graphite (SRE Core I type) at 1000 and 1200°F for 100 hr resulted in expansions of approximately 1%. These tests confirm the results of earlier work at Atomics International on unirradiated graphite.

Dilation tests (Table 1) were conducted on unirradiated graphite samples from various manufacturers. In all tests, the specimens were exposed to 1000°F sodium for 100 hr. Graphite Types TSGBF, TSX, and CSGBF are control

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 1
RESULTS OF DILATION TESTS ON UNIRRADIATED GRAPHITE

Type of Graphite	Manufacturer	Expansion (%)	Weight Gain (%)	Comments
TS-504	Union Carbide	2.0, 2.9		Corners chipped off. Numerous cracks.
TS-373	Union Carbide	1.0, 0.9, 5.0		Some chipping.
YBF	Union Carbide	0.6, 0.5, 0.9	11.5	General appearance good. No cracking or chipping.
YBR	Union Carbide	0.8, 0.3, 0.8	15.9	General appearance good. No cracking or chipping.
YAY	Union Carbide	0.6, 0.5, 0.9	13.3	General appearance good. No cracking or chipping.
YAX	Union Carbide	0.6, 0.7, 0.9	10.5	General appearance good. Cracks observed.
TSGBF	Union Carbide	3.6, 0.4, 5.4		All corners chipped off. Numerous cracks.
TSGBF	Union Carbide			Sample deteriorated.
CSGBF	Union Carbide	1.0, 0.8, 0.8*	10.8	General appearance excellent. No cracking or chipping.
TSX	Union Carbide	0.04, 0.09, 0.9*	9.4	General appearance excellent. No cracking or chipping.
EGCR	Great Lakes Carbon Co.	0.6, 0.7, 0.4*		Cracks present prior to exposure.
EGCR	Great Lakes Carbon Co.	0.8, 0.7, 0.4*		No large cracks or chipping after exposure.
H253	Great Lakes Carbon Co.	0.8, 0.9, 1.2	7.8	General appearance good.
GSXP	Basic Carbon Co.	1.6, 2.2, 1.2		Chipped.
GXX	Basic Carbon Co.	0.3, 1.3, 0.8	8.3	General appearance good. Cracks observed.
TPXX	Graphite Specialties, Inc.	1.5, 0.6, 2.3		Chipped and cracked.
Natural Ceylon Flake Graphite	Unknown	2.3, 0.6*	9.5	General appearance excellent. No cracking or chipping

*Parallel to extrusion axis.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

samples for Hanford reactor high irradiation (4 to 5×10^{21} nvt) graphite. High irradiation specimens of TSX and CSGBF will be tested to determine their dilation characteristics.

In many cases, postexposure dimensional or weight determinations were not feasible due to the incompatibility of the graphite with sodium.

Emission spectroscopy was performed on the sodium which was used in the dilation tests. The results were in good agreement with the data on sodium impurities in the Liquid Metals Handbook. The silicon content was a little higher; however, the potassium and calcium impurity levels fell within the range presented in the handbook.

Dilation tests on irradiated graphite samples (STP graphite) from an unfailed SRE Core I moderator can were conducted to establish a correlation between accumulated irradiation exposure and the dilation of sodium saturated graphite. The results are summarized in Table 2.

Figure 3 shows the effect of irradiation on the dilation of graphite in sodium at 1000°F . Although data on the effect of accumulated irradiation exposure on the dilation of graphite at one temperature are limited and somewhat inconsistent, a trend of increasing dilation with increasing accumulated irradiation exposure is discernible.

7519-2534

Figure 3. Effect of Irradiation on Graphite Dilation

The conclusions of the irradiated graphite-liquid sodium tests are:

- 1) Dimensional changes in the order of 2 to 5% were observed when samples were exposed to sodium; and
- 2) The dimensional changes were anisotropic, the maximum dimensional changes being in the direction perpendicular to the axis of extrusion.

Emission spectroscopy results on the SRE primary sodium for the period January 1962 to December 1963 were examined. In view of the early work showing a strong effect of potassium on graphite dilation, the relatively high potassium content (~ 0.1 wt %) during the period from April 1962 to June 1962

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 2
RESULTS OF DILATION TESTS ON TSP GRAPHITE

Distance of Sample From Top of Moderator Can (in.)	Accum. Fast Neutron Flux (nvt > 0.18 Mev)	Exposure Temp (°F)	Exposure Time (hr)	Expansion (%)		
				Axis		
				1	2	3*
15	6.7×10^{19}	1000	100	1.8	1.9	0.9
27	5.6×10^{20}	1000	100	1.6	1.6	1.3
27	5.6×10^{20}	1000	100	3.2	3.1	2.2
39	9.1×10^{20}	1000	100	4.2	4.0	2.8
51	1.09×10^{21}	600	100	2.2	2.0	1.2
51	1.09×10^{21}	1000	100	3.1	3.0	2.0
60	1.12×10^{21}	1000	500	3.8	4.3	2.6
60	1.12×10^{21}	1000	100	3.8	1.3	2.8
63	1.11×10^{21}	1000	100	2.9	5.2	1.7
63	1.11×10^{21}	800	100	2.8	4.0	2.0
63	1.11×10^{21}	1000	100	4.3	4.4	2.6
63	1.11×10^{21}	1200	100	3.9	3.9	2.0
91	5.5×10^{20}	1000	100	3.0	4.4	2.1
91	5.5×10^{20}	1000	100	2.4	2.8	2.0
105	7.3×10^{19}	1000	100	2.2	2.3	1.3
116	1.7×10^{19}	1000	100	2.4	2.5	1.4
116	1.7×10^{19}	1000	100	2.4	2.5	2.1

*Parallel to extrusion axis

is of interest. To aid in the evaluation of this point, a dilation test was conducted on a sample from unfailed moderator can in sodium containing 0.1 wt % potassium with the following result.

Distance of Sample From Top of Moderator Can (in.)	Accum. Fast Neutron Flux (nvt > 0.18 Mev)	Exposure Temp (°F)	Exposure Time (hr)	Expansion (%)		
				Axis		
				1	2	3*
63	1.11×10^{21}	1000	100	5.4	3.6	2.1

*Parallel to extrusion axis.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

The measured expansion along axis labeled No. 1 was the largest observed. However, additional tests would be required to determine the effect of potassium content (~ 0.1 wt %) on the dilation of graphite.

The test equipment to determine effects of sodium vapor on dilation of both irradiated and unirradiated graphite was assembled and checked out. An experimental program is in progress (at 1800°F and below) that will determine the equilibrium sodium content of irradiated and unirradiated graphite as a function of time and temperature.

Lattice constant measurements by x-ray diffraction were performed on irradiated graphite specimens, both from failed and unfailed SRE moderator cans, to establish a relationship between accumulated irradiation exposure and the dilation of graphite contacted with liquid sodium. The surface of the freshly fractured material in the failed can sample exhibited a distinct violet tinge while in an inert atmosphere, suggesting formation of a sodium-graphite lamellar compound. However, when the x-ray pattern was indexed there was no positive manifestation of the lamellar compound. There was swelling, as shown by broadening in the C-spacing of crystal lattice. The same general crystal distortions (increasing in C-spacing) took place in the irradiated sample from the unfailed moderator can when it was exposed to sodium. Results of the examination are as follows.

- 1) No uniform lamellar compound formation was observed. There did appear to be some random change in c-spacing.
- 2) There did not appear to be as much expansion due to sodium interaction after irradiation as during simultaneous soaking and irradiation.
- 3) One of the graphite samples from the failed moderator can showed an increase in c-spacing of approximately 12%.

Exposure of the sample to air returned the c-spacing of the lattice structure to nearly normal values. Air exposure of the entire sample did not return the macroscopic sample to normal.

Samples of unirradiated columbium-coated Type CS graphite were exposed to 1000°F sodium for 100 hr to determine if columbium protects graphite from sodium. The columbium coating was applied using a salt bath electroplating technique. Six graphite samples were prepared; two each were coated with 1/2 mil, 2 mils, and 4 mils of columbium. Dye-penetrant inspection of the samples indicated numerous imperfections in the coating. Although, after exposure to sodium, coatings on the 1/2- and 4-mil specimens were severely cracked (peeled), the 2-mil coated specimen (despite a 1.7% dimensional increase) retained its shape with no visible cracking. Sectioning of the specimen revealed that the sodium had penetrated into the graphite. It was concluded that the columbium coating did not serve as a barrier against sodium regardless of the barrier thickness.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The cutting and welding equipment and techniques for the remote replacement of calandria process tubes have been well established. These techniques have been standardized to the extent that the procedures outlined will consistently produce good results.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Testing of the graphite-handling machine on the 4-channel vessel has demonstrated its general operability, and revealed areas where minor improvements are required.

Results from the liquid sodium and graphite interaction show that graphite expansion up to 5% are possible after radiation exposure of approximately 1.1×10^{21} nvt (> 0.18 Mev). The expansion of graphite will have to be considered in the design of a calandria type core reactor.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Large Sodium Graphite Reactors	
Project: SGR Core Maintenance, Prototype Development	
Project Engineer: K. W. Foster	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-21
General Order: 7519	Subaccount: 5220

I. PROJECT OBJECTIVES

The objective of this project is to design, fabricate, and test the prototype tools required for the initial core assembly and for maintenance of the calandria vessel of the large SGR prototype plant. Two specific classes of tools will be developed, the initial assembly tools for the nonradioactive core and the maintenance tools for a radioactive core.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A document which describes the designs of the initial installation tools, remote maintenance tools, and expected maintenance operations for the core of the LSGR prototype plant was completed. A preliminary layout of a test stand for use in demonstrating the full-length calandria maintenance tools was completed. The test stand as proposed will be an annex to the control rod tower, Building 354.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The design description provides a record of the maintenance requirements as visualized for the LSGR calandria core. It includes detailed information concerning presently developed tools, and outlines the general requirements for accessory equipment. This document should serve as a reference in the maintenance considerations for existing and future SGR type power plants.

SECTION III
FAST-REACTOR DEVELOPMENT
CONTENTS

<u>Title</u>	<u>Identification</u>	<u>Page</u>
FAST-REACTOR DEVELOPMENT	Summary	III-1
SCCR FEASIBILITY, EVALUATION, AND DESIGN	7661 - 1310	III-7
CALCULATION METHODS AND CROSS-SECTIONS	7661 - 1331	III-20
PHYSICS EXPERIMENTS	7661 - 1332	III-30
BOILING STUDIES FOR SODIUM REACTOR SAFETY	7661 - 1341	III-35
SODIUM FIRES AND FISSION PRODUCT RELEASE	7661 - 1342	III-41
FUEL ELEMENT TRANSIENT HEATING	7661 - 1343	III-50

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Fast-Reactor Development
Program Manager: D. T. Eggen
Reporting Period: Fiscal Year 1965

I. PROGRAM OBJECTIVES

The general objective of this program is to develop economic, safe, and reliable nuclear central stations suitable for the United States commercial power reactor network, and to utilize the skills and technologies which have been developed at AI in reactor design, sodium technology, reactor fuels, fast reactor physics, and sodium plant construction. To attain this objective, it is necessary to determine the operational, safety, maintenance, and economic characteristics, and to demonstrate various design parameters for the fast system.

II. PROJECT MANAGER'S SUMMARY AND EVALUATION

A reference coupled, fast reactor was chosen which is referred to as the modular, flux-trap concept. The concept incorporates a flux trap, develops a significant fraction of its power in the thermal regions which improves the negative Doppler characteristics, has small sodium void effect, reasonably high breeding ratio, and promising fuel cycle economics. Calculation methods for parametric surveys, both static and dynamic, and improved cross-sections have been developed. Sodium fire experiments studying the effects of oxygen depletion on burning rates and iodine release were conducted and documented. Experiments were completed on the steady-state boiling heat transfer of sodium under forced convection. A mode of meltdown behavior of sodium-bonded carbide fuel was experimentally observed, which indicates random dispersal caused by large local sodium pressures. A fast reactor fuel effort was initiated with emphasis on carbide fuel development and cladding evaluation.

A. FAST REACTOR CONCEPT EVALUATION

The sodium-cooled coupled reactor (SCCR) concept was investigated in depth in order to determine its role in attaining the fast reactor objectives of the AEC. This study established the controllability and safety characteristics which provide a margin of safety to protect the investment of the customer and the safety to the public. At the same time, advantage of the superior fuel cycle economics potentially available in fast reactors through high specific power, extended burn-up, and high conversion ratio was maintained. Design flexibility which allows for optimization of the mechanical and process system without major compromise caused by nuclear, economic, or safety considerations was attained.

Survey calculations, made of various core arrangements in a consistent fashion, indicated that the modular, flux-trap core arrangements have large isothermal Doppler coefficients and slightly positive or negative coolant void reactivity effects. To optimize the flux-trap design, various modular split-blanket core arrangements were studied. The "small" modular arrangement with an average blanket enrichment of 3-1/2% was found to have the best safety characteristics and competitive economics with other fast reactor designs. A larger modular arrangement had better economics but a less conservative safety margin.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

The reactor is characterized by a strongly negative isothermal Doppler coefficient and a weakly positive sodium void maximum reactivity effect. As a result, the reactor responds to most postulated accident situations with an inherent corrective action. A reactivity insertion of \$1.65, regardless of insertion rate, can be tolerated without gross damage to the core provided that normal protective action is effective. For an accident in which rapid scram does not occur, insertions as large as \$1.20 can be tolerated. In hypothetical loss-of-flow accidents, limited protective system action is sufficient to restrict core damage to a minor portion of the hottest fuel element.

Mechanical design of the reference reactor emphasized flexibility of the core arrangement to permit the substitution of a fast, blanket, moderator, or control element into any position in the core. This allows modification of the initial arrangement as a result of safety research and development, and as operating experience is accumulated. Graphite was selected for the moderator elements after comparison with beryllium oxide and zirconium hydride.

Mixed-oxide fuel was selected for the reference core since it appears to have the best near-term potential for economic operation, due to the relative amount of experience with uranium oxide. Mixed-carbide fuel with a sodium bond appears to have the best long term potential for low fuel cycle costs and has the potential for improved safety compared with the oxide.

By operating at an enrichment of 3.5%, the radial blankets provide approximately 40% of the total core power with an average fuel burnup of approximately 49,000 Mwd/T. The breeding ratio is 1.3, resulting in a doubling time of 15 yr. Though the inventory is undesirably high, the net fuel cycle costs and has the potential for improved safety compared with the oxide. The total costs of an investor-owned public utility 1000-Mwe fast reactor of the modular flux-trap type indicated that the total power costs could range from a high of 4.4 mills/kw hr to 3.2 mills/kw hr.

B. FAST REACTOR PHYSICS

The calculational methods effort during the year has been devoted to improving the reliability of analysis of fast breeder reactor characteristics, particularly cross-sections, safety coefficients and long-term burnup behavior. Lack of reliable cross-section data is generally accepted as the major single area of uncertainty in fast reactor physics. Cross-section work, including evaluation of recent experimental data, provision of spectra generation methods, development of more accurate averaging techniques, and digital automation of these procedures, constitutes an important contribution to fast reactor physics technology. A 35-group cross-section library structure was developed, using group widths of half a lethargy unit above thermal energies, which covers adequately the entire energy range up to 10 Mev. In order to utilize efficiently a 35-group library, the CAESAR multigroup diffusion theory code was expanded from 18-group to 35-group capacity and rewritten in FORTRAN-IV.

As a step in improving the methods for generating multigroup libraries, a card-punch routine for the computed neutron flux and current spectra was added to the ELMOE code. This permits the ELMOE spectrum to readily be used as input for cross-section averaging codes used to produce the multigroup libraries.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Fifteen- and thirty-five group libraries for the analysis of selected ZPR-III fast critical assemblies were generated as part of a program for testing updated cross-section libraries, and for use in fast reactor analysis. A code was written for collapsing multigroup cross-sections to few group sets for use in two-dimensional calculations.

An improved code for calculating resonance integrals and Doppler coefficients has been made available for use in fast reactor calculations.

Graphical display of numerical results greatly facilitates qualitative comparison of different situations, and often results in immediate insights not available from scanning columns of numbers. A code has been written which provides computer-plotted pointwise radial traverses of Doppler coefficient (differential volume contribution), power density, and fluxes and adjoint fluxes by energy group.

Reactor burnup codes were evaluated for use in burnup calculations. Improvement in the data processing and search options is needed in order to substantially reduce the labor involved in making fuel cycle calculations. An updated multi-group library has been prepared and used to investigate methods of determining isotopic compositions, power shifts, and fuel economics in fast reactor cores. The lifetime variation of reactivity coefficients is being studied further.

Analysis of a group of "standard" sodium-cooled fast breeder reactor cores is being made for use in the ANL cross-section comparison effort.

A survey of reactivity worths of various poison materials has been made. Data from AETR, ZPR-III, ZEBRA, and other fast reactor criticals have been included. Interestingly, the worth of Eu_2O_3 is as great as B_4C on a per volume basis. The poison worth of Ta is approximately one-half that of boron carbide. Neither has the disadvantage of He gas evolution.

Preparations have been made for the study of sodium void coefficients in the AI split-table critical machine. Prototypic cans for sodium containment were designed, fabricated, filled with sodium, welded shut, and tested satisfactorily.

Preliminary work has begun in preparation for reactivity worth determinations and Doppler measurements on proposed fuel samples. Self-shielding and self-multiplication effects have been calculated using perturbation theory for various sample changes and compared with a wide variety of experimental data. In order to determine that reactivity changes could be accurately predicted, comparisons were made for a full density and a reduced density sample of fully enriched uranium in cores having a wide range of fission energies. The samples were made up of a series of foils. All uranium was used for the full density sample, and uranium was interspersed with stainless steel foils for the reduced density sample. The cross-section library used with the experimental measurements has been updated to be consistent with cross-sections used in the design effort.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

C. REACTOR SAFETY

The study of steady-state-boiling heat transfer under forced convection, initiated in FY 1964, was completed. Unlike the familiar boiling curves of other liquids, which show nucleate boiling over a wide range of heat fluxes, these data show that convection heat transfer dominates almost up to the critical heat flux. The subcooled-flow data were correlated by the method of superposition. The critical heat flux under two-phase flow, correlated as a function of the exit quality, demonstrates a definite parametric dependence on system pressure. The CHF increases with the pressure, but decreases rapidly with the vapor quality of the two-phase mixture. This agrees with the results reported for other liquids and with the sodium pool boiling results previously reported.

In preparation for the study of hydrodynamic instabilities and transient flow due to boiling, the two-phase sodium loop was redesigned and modified. The new loop has a 6-ft annular (1/2 by 1/2 in.) test section uniformly heated along its length. Heat will be generated along 4-1/2 ft of the test section (maximum heat flux of 200,000 Btu/hr ft²) using graphite cloth as a resistance heater. Adequate performance of the new loop has been demonstrated.

The digital computer code TRANSFUGUE I has been completed. This code is based on a homogeneous flow model in which the two-phase mixture is assigned an average velocity, temperature, and density, and is then treated as a single-phase, variable density fluid. A second code, BURP, was developed as part of the SCCR evaluation. Both these codes calculate the axial distribution of velocity, void fraction, pressure, coolant temperature, and fuel temperature for transients simulating power excursions, loss of flow, loss of pressure, and channel plugging. TRANSFUGUE is a "heat transfer limited" code (void growth is limited by heat transfer only). BURP is a "pressure limited" code (void growth is limited by self-pressurization). Both have been used in the analyses of loss of flow and power transients in fast reactor cores.

The large fires apparatus was modified to permit experiments in reduced oxygen atmospheres. Ten runs were made varying oxygen concentration, surface temperature, and flow. Within the range of investigation, the results of these experiments showed that a pool of liquid sodium burns at a rate roughly proportional to the oxygen concentration. The release rate of sodium vapor, some or all of which may eventually react, was strongly dependent on sodium surface temperature, and not particularly dependent upon oxygen concentration. The iodine, released from the sodium, was associated in approximately the same concentration in the airborne sodium as it was in the sodium collected in the scrubber; the iodine concentration of the released sodium is always less than it was in the pool; and concentration of iodine in released sodium was not affected by oxygen concentration or gas flow, but showed a dependence on surface temperature. The energy-release measurements indicated that more energy was released than could be accounted for by combustion and natural convection alone. At zero oxygen, additional heat release mechanisms (radiation and particle-to-air heat transfer) are present.

The transport phenomena which influence the sodium-air reaction were examined in order to provide a basis for modeling and extrapolating to full-scale systems, and in order to distinguish the relative importance of various

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

parameters. The reaction occurs predominantly in the gas phase with the gas phase reaction rate controlled by diffusion of the reactants to the reaction zone. The important parameters are: sodium surface temperature, geometry, oxygen concentration, pressure, humidity, and liquid surface condition.

The Phase II UC transient heat experiments were successfully conducted with a total of four transients at TREAT. Capsule 1 was subjected to two transients designed to show the effects of a rapid transient (melting within one second) where fuel meltdown occurs prior to coolant boiling. Capsule 2 was subjected to two transients. The final test with Capsule 2 attempted to produce coolant boiling prior to meltdown, thus simulating meltdown conditions in a choked channel (melting over a period of 10 to 20 sec).

In the final transient with Capsule 1, the peak power was 6000 kw/ft. A mode of meltdown behavior was experimentally observed which indicates that random dispersal of molten carbide fuel can be caused by large local sodium pressures, with the subsequent solidification of the molten fuel in cooler regions of the system. The meltdown of sodium-bonded, carbide fuel appears to follow the sequence: bond vaporization, central fuel melting, melting at the fuel surface and fuel clad contact, reaction and/or melting of cladding material and, finally, flow of molten fuel and cladding into the coolant with subsequent dispersion due to rapid coolant heating and pressure generation.

Examination of Capsule 2 indicated there was no gross melting of the fuel, no changes in microstructure, and no deformation of the cladding. The results obtained in the final transient with Capsule 2 establish a rather useful performance limit with respect to tolerable conditions during reactor transients for liquid-metal-bonded carbide fuel. Thus power levels of 200 kw/ft can be tolerated for periods as long as 10 sec without cladding failure.

It is evident that the series of experiments have been built logically on previous experience, and that a modular experiment to simulate thermokinetic core behavior can be conducted which will permit postirradiation analysis and evaluation.

D. REACTOR FUELS

The fast reactor fuels effort was initiated late in the third quarter of the fiscal year by reorienting applicable SGR fuel development work to fast reactor environment and performance requirements. The fuels program will emphasize all phases of carbide (UC and PuC) technology.

The fast reactor cladding effort consisted primarily of the final design and initial fabrication of the NAA-123 irradiation experiment to evaluate the stress rupture properties of 304 SS at elevated temperatures. One of the key factors involved is the 304 SS clad boron impurity content which yields helium in a thermal neutron flux and results in a loss of ductility. Materials specimens are being carefully characterized. Other tasks included further evaluation of experiments started under the SGR program: (a) the effect of carburization on the stress rupture properties, and (b) heat-to-heat composition effects on 304 SS properties. One of the main problems resulting from the survey applying to fast reactors is the extent of the (n, α) and (n, p) reactions in a fast flux on the clad

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

properties. Samples of SGR clad materials were prepared for helium determination by electron microscope analysis of ultra thin sections. Analyses are incomplete.

The fast reactor fuel rod development effort concentrated principally on evaluating available data from tests in fast reactors. Many of the important details from sites such as Dounreay are unavailable. It was recognized that the fuel rod design criteria and fuel rod specifications form the primary basis for the entire fuel development program. Decisions and assumptions are necessary to resolve some of the following examples of problems: to design for fission gas release or retention, vented or unvented design, original density to allow for solid fission product accumulation, allowable clad strain, fuel swelling versus burnup and temperature, and fission product migration.

The carbide processing effort conducted preliminary processing studies on UC + PuC fuel through a subcontract. It was the purpose of these plutonium material tests to evaluate the Pu volatility loss during carbide feed stock preparation. PuC has substantially different thermal properties from UC, and therefore composition control during fabrication must be evaluated. A survey of processing problems indicates three potential methods of fabrication, arc melting and casting, powder compaction and sintering, and vibratory compaction. All of these methods require an allowance for density decreases. The original density decrease can be provided by homogeneous low density or heterogeneous low density material. Process engineering analyses are required that also relate to the performance requirements.

The carbide fuel material effort consisted of a literature search and a preliminary analysis of problem areas. An understanding of the phase equilibria of the mixed carbide is essential to performance characteristics and reliable processing methods. Single-phase material appears to be preferable; however, the effect of substantial quantities of impurities on the microstructure during operation requires study. The type of microstructure together with potential additives such as tungsten as a finely divided second phase and gaseous fission product accumulation inhibitor needs to be investigated. The mechanical property data on the mixed carbide are very limited. It is also known that the conductivity of PuC is less than UC, but that it increases with temperature. The reliability of the materials information together with program objectives will be used to initiate scoping irradiations under specific power conditions of interest.

Although limited experimental progress was made in the short active period of FY 1965, substantial groundwork of existing information and creative ideas for problem solution were prepared for a strong contribution to fast reactor fuels development in FY 1966.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Fast Reactor Development	
Project: SCCR Feasibility, Evaluation, and Design	
Project Engineer: A. S. Gibson	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-21
General Order: 7661	Subaccount: 1310

I. PROJECT OBJECTIVES

The general objective of this project is to determine the performance characteristics, safety, and economics of the Sodium-Cooled Coupled Reactor (SCCR). Specific objectives are to quantitatively establish core characteristics and fuel cycle economics as a function of geometry and coupling arrangement. Additional objectives are to provide mechanical design information of possible core arrangements and core components, and to explore certain plant design problems.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

The major accomplishments in FY 1965 may be summarized by the following conclusions which result from the work completed.

- 1) By a suitable selection of geometric and nuclear coupling arrangements, the Doppler reactivity effect can be enhanced and the sodium void reactivity effect can be made more negative.
- 2) Of the different sodium-cooled, coupled-reactor configurations studied, a modular split blanket flux trap version has the best potential to be designed with improved safety, as compared with other current fast reactor designs.
- 3) The safety characteristics of the reactor may be measured in terms of the amount of fuel damaged for a given incident. For very severe incidents, they may be measured in terms of kinetic energy released during the incident. Both measures appear to be predominantly affected by the Doppler coefficient of reactivity and the sodium void reactivity effect. Only secondary effects are produced by varying prompt neutron lifetime.
- 4) Mixed-oxide fuel appears to have the best near-term potential for economic operation, due to the relatively large amount of experience with uranium oxide. However, mixed-carbide fuel with a sodium bond appears to have the best long-term potential for low fuel cycle costs, and has the potential for improved safety compared with the oxide.
- 5) The reference modular flux trap reactor requires a high inventory of fissile materials. Competitive economics are achieved by reason of a high breeding ratio (1.3) and a high fraction of the power generated in the internal blanket regions (approximately 40%).

A topical report covering the technical results of the feasibility study was completed. The report is entitled "Feasibility Study of a 1,000-Mwe Sodium-Cooled Fast Reactor," NAA-SR-11378.

A. CORE SURVEY STUDIES

Survey calculations were made of various core arrangements to examine these designs in terms of certain defined measures of merit. The SCCR geometries studied can be classified into three general categories: (1) cylindrical, (2) annular, and (3) modular. Reflector-moderated versions of the cylindrical, annular, and modular cores and integral-moderated types of the cylindrical and annular cores were studied. The basic core arrangements evaluated are shown in Figure 1.

5-12-65

7661-2501

Figure 1. SCCR Core Configurations

A comparison of the static characteristics of the various geometries is presented in Table 1. The results in this table have been determined in a consistent fashion using the same basic mathematical techniques and neutron cross-section data, thereby permitting a direct comparison of the various parameters. An examination of these indicates that the modular core arrangements have large isothermal Doppler coefficients, and slightly positive or negative coolant void reactivity effects. The flux-trap design is seen to have a maximum void effect of +17¢ and is more than \$2.00 subcritical for complete voiding of sodium. It was for these reasons that the modular flux-trap design was selected as the reference core for more extensive analysis.

To optimize the flux-trap design, various modular split-blanket core arrangements were studied. In particular, the reference module was compared with a larger one having an additional row of fast-region fuel elements. The blanket enrichment of the flux-trap modular reactor was also found to sensitively affect the safety and economic characteristics of the design, and it was therefore explored as a variable. The results of these optimization studies, as summarized

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 1
COMPARISON OF SCCR CHARACTERISTICS

Characteristic	Cylindrical SCCR		Annular SCCR		Modular SCCR	
	RM	IM	RM	IM	Single Blanket RM	Split Blanket RM
$-T\left(\frac{dk}{dT}\right)$ Doppler with Na	0.013	0.018	-	0.011	0.0213	0.0180
without Na	0.009	-	-	-	0.0202	0.0181
δk^D to point of fuel damage (\$)	-1.50	-	-	-	-2.00	-1.40
$\delta k/k$ sodium void (\$)						
Maximum effect	-	-	-	-	+3.0	+0.17
Fast section voided	+11.1	~+6.0	-	+3.1	+0.95	-2.36
Total core voided	-	-	-	-	+3.0	-2.45
Prompt neutron lifetime, (μ sec)	13.6	27.0	~8.0	6.4	~1.8	2.0
Delayed neutron fraction, β effective	0.0038	0.0044	~0.004	0.0042	~0.0035	0.0035
Fissile loading (kg)	1530 Pu + 670 U ²³⁵	825 Pu + 1385 U ²³⁵	~2500	2471	~2260 core + 670 blankets	2900 core + 1300 blankets
Breeding ratio	1.0	~0.70	~1.10	1.15	1.43	1.47
% power in fast section	75	31	~80	86	68	64
Radial peak-to-avg. pwr	-	-	~5.0 (in thermal regions)	-	~1.18	1.29

in Table 2, were used to select the reference design. The small modular arrangement with an average blanket enrichment of 3-1/2% was found to have the best safety characteristics and competitive economics with other fast reactor designs. By emphasizing the safety features of the reactor some compromise in economics was made, since the large module arrangement has a more attractive fuel cycle cost (by approximately 0.2 mills/kw-hr), but has a larger positive void reactivity effect.

B. REFERENCE DESIGN

The principal characteristics of the reference design are shown in Table 3, and a schematic picture of the core arrangement is shown in Figure 2. As noted from Table 3, the reactor is characterized by a strongly negative isothermal Doppler coefficient ($T \frac{dk}{dT} = -0.018$) and a weakly positive sodium void maximum reactivity effect of +53¢. As also noted from Table 3, a breeding ratio of 1.3 is obtained for the reference core, and the blanket average enrichment under equilibrium conditions is 3-1/2%. By operating at this enrichment, the internal radial blankets provide approximately 40% of the total core power with an average fuel burnup of approximately 49,000 Mwd/T. The net effect of these various factors is the favorable fuel cycle cost shown of 0.67 mills/kw-hr. Fuel cycle costs are based on 10% interest charges on inventory, \$10/gram plutonium, and 80% plant load factor.

TABLE 2
RESULTS OF OPTIMIZATION STUDY

	Breeding Ratio [†]	Enrichment Pu ²³⁹ + Pu ²⁴¹ (Fast Region) (%)	Power per Module (Mwth)	Number of Modules	Fast Region Fissile Loading (kg)	Power Split			Δk_{Na} Na Void Fast Region (\$)	α_D With Na in Fast Region				α_D Without Na in Fast Region				Δk_p Doppler Power Effect (-\$)
						Fast Region	Inner Blanket	Outer Blanket		Fast Region ($-T \frac{dk}{dT}$)	Inner Blanket ($-T \frac{dk}{dT}$)	Outer Blanket ($-T \frac{dk}{dT}$)	Total ($-T \frac{dk}{dT}$)	Fast Region ($-T \frac{dk}{dT}$)	Inner Blanket ($-T \frac{dk}{dT}$)	Outer Blanket ($-T \frac{dk}{dT}$)	Total ($-T \frac{dk}{dT}$)	
Core Diameter (in.)																		
12	1.37	42.4	70.8	35.30	3640	0.57	0.24	0.19	-1.08	0.00139	0.01173	0.00857	0.0225	0.00149	0.01194	0.00915	0.0226	1.18
21.4*	1.34	22.7	178	14.04	2690	0.66	0.20	0.14	0.80	0.00376	0.00891	0.00572	0.0182	0.00236	0.00921	0.00633	0.0178	1.53
36	1.31	15.9	394	6.34	2480	0.76	0.15	0.09	4.49	0.00657	0.00552	0.00320	0.0143	0.00349	0.00588	0.00369	0.0125	1.77
Inner Blanket Thickness (in.)																		
2	1.27	20.8	182	13.69	2410	0.69	0.14	0.17	1.92	0.00524	0.00800	0.00906	0.0220	0.00355	0.00803	0.00971	0.0213	2.29
4*	1.34	22.7	178	14.04	2690	0.66	0.20	0.14	0.80	0.00376	0.00891	0.00572	0.0182	0.00236	0.00921	0.00633	0.0178	1.53
6	1.39	23.9	175	14.26	2860	0.65	0.24	0.11	-0.01	0.00327	0.00798	0.00346	0.0146	0.00184	0.00835	0.00392	0.0141	1.08
Graphite Thickness (in.)																		
1	1.46	21.7	165	15.15	2800	0.72	0.18	0.10	1.83	0.00407	0.00716	0.00413	0.0154	0.00232	0.00715	0.00446	0.0141	1.28
2	1.43	22.5	166	15.06	2870	0.71	0.18	0.11	1.10	0.00393	0.00802	0.00493	0.0169	0.00233	0.00821	0.00541	0.0161	1.34
4*	1.34	22.7	178	14.04	2690	0.66	0.20	0.14	0.80	0.00376	0.00891	0.00572	0.0182	0.00236	0.00921	0.00633	0.0178	1.53
6	1.23	21.9	205	12.20	2270	0.58	0.23	0.19	1.19	0.00374	0.00970	0.00643	0.0199	0.00235	0.00993	0.00702	0.0193	1.89
8	1.12	20.6	246	10.16	1790	0.50	0.26	0.24	1.80	0.00367	0.01035	0.00735	0.0214	0.00230	0.01041	0.00780	0.0205	2.33
Graphite Density (4 in. thick)																		
0.42	1.31	23.8	154	16.23	3240	0.75	0.16	0.09	0.17	0.00356	0.00530	0.00307	0.0118	0.00205	0.00534	0.00339	0.0107	0.96
0.84	1.37	23.4	161	15.50	3050	0.72	0.17	0.11	0.41	0.00371	0.00701	0.00442	0.0150	0.00220	0.00720	0.00491	0.0143	1.17
1.68*	1.34	22.7	178	14.04	2690	0.66	0.20	0.14	0.80	0.00376	0.00891	0.00572	0.0182	0.00236	0.00921	0.00633	0.0178	1.52
Outer Blanket Thickness (in.)																		
0	1.15	16.7	237	10.55	1520	0.55	0.45	-	4.55	0.00449	0.01760	-	0.0216	0.00264	0.01735	-	0.0199	3.16
1	1.29	21.4	187	13.38	2430	0.65	0.23	0.12	1.80	0.00389	0.01086	0.00601	0.0206	0.00240	0.01112	0.00652	0.0200	2.01
2*	1.34	22.7	178	14.04	2690	0.66	0.20	0.14	0.80	0.00376	0.00891	0.00572	0.0182	0.00236	0.00921	0.00633	0.0178	1.53
4	1.38	23.9	174	14.40	2890	0.67	0.18	0.15	-0.17	0.00354	0.00733	0.00449	0.0153	0.00213	0.00763	0.00507	0.0148	1.15
Core Height (ft)																		
2	1.12	27.8	80	31.1	3360	0.72	0.17	0.11	-1.07	0.00281	0.00667	0.00383	0.01331	0.00178	0.00705	0.00436	0.0132	0.98
4*	1.34	22.7	178	14.04	2690	0.66	0.20	0.14	0.80	0.00376	0.00891	0.00572	0.0182	0.00236	0.00921	0.00633	0.0178	1.53
6	1.41	21.2	278	9.0	2260	0.64	0.21	0.15	1.85	0.00389	0.00978	0.00649	0.02016	0.00245	0.01001	0.00708	0.0195	1.85

*Base core used for comparison
†Axial blanket not included

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

NAA-SR-11450

III-10

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 3
PRINCIPAL REACTOR CHARACTERISTICS
Reference Core (SB*Mod 3)

1. Power	
a. Electrical (Mwe)	1000
b. Thermal (Mwt)	2500
1) Fast	1388
2) Blanket and other	1112
2. Breeding Ratio (ave)	1.31
3. Doubling Time, including ex-core needs (yrs)	15.3
4. Critical Mass (equilibrium)	
a. Fast regions (kg Pu ²³⁹ + Pu ²⁴¹)	2187
b. Blanket regions (kg Pu ²³⁹ + Pu ²⁴¹)	866
5. Size, diam x height (ft)	15 x 7.2
6. Configuration	10 split-blanket, flux-trap modules
7. Coolant	Na
a. Outlet temperature, mixed mean (°F)	1075
b. Inlet temperature (°F)	750
c. Flow (lb/hr)	87 x 10 ⁶
d. Velocity, core/blanket (ft/sec)	18.5/11.6
e. Pressure drop, core/blanket (psi)	50/15
8. Core	
a. Fast Fuel	(U, Pu)O ₂
1) Assemblies	18 hexagonal per module
2) Size, active length x diam across flats (in.)	50 x 4.875
3) Pins	168 per assy
4) Size, clad OD/pellet OD (in.)	0.250/0.214
5) Enrichment (% fissile)	25
6) Specific power, ave/max (kw/kg)	173/350
7) Linear heat, ave/max (kw/ft)	11.1/22.7
8) Peaking factor, total	2.04

(continued)

* Split blanket

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 3
PRINCIPAL REACTOR CHARACTERISTICS
Reference Core (SB*Mod 3)
(Continuation)

b. Blanket Fuel	UO ₂
1) Assemblies, inner + outer	18 + 1/2 (30) per module
2) Size, active length x diam across flats (in.)	50 x 4.875
overall length (in.)	86
3) Pins	126 per assy
4) Size, clad OD/pellet OD (in.)	0.340/0.320
5) Average enrichment (% Pu)	3.5
6) Linear power, ave/max (kw/ft)	13/18.8
7) Peaking factors, total	3.33
c. Moderator	graphite hexagons
1) Assemblies	21 per module
2) Size, length x diam across flats (in.)	86 x 4.875
d. Control	4 per module
1) No. per module location/material	1 center/B ₄ C 3 moderator/RE
9. Safety Characteristics	
a. Sodium worth full core void/max (\$)	-0.75/+0.53
b. Doppler (-T dk/dt x 10 ³)	18.1
fast/inner blkt/out blkt	2.3/9.0/7.8
c. Doppler worth to point of fuel damage (\$)	-1.52
10. Material of structure and cladding	304 stainless
11. Fuel Cycle Costs (mill/kwh)	0.668

1. Kinetics and Safety

Because of the emphasis in the reference design on the importance of a strong negative Doppler coefficient and a weak positive sodium void effect, the reactor responds to most postulated accident situations with an inherent corrective action. Results of the safety analysis indicate that a reactivity insertion of \$1.65 regardless of insertion rate can be tolerated without gross damage to the core, provided that normal protective action is effective. For an accident in which rapid scram does not occur, insertions as large as \$1.20 can be tolerated. The accident analysis also indicates that for hypothetical loss of flow accidents, limited protective system action is sufficient to restrict core damage to a minor portion of the hottest fuel element undergoing some melting. Figure 3 demonstrates typical reactivity insertion rates and levels for various classes of severe

*Split blanket

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

7661-2502

Figure 2. Modular Concept Flux Trap

5-14-65

7661-2503

Figure 3. Relationship of Severe Accidents to Tolerable Limits

accidents and the manner in which these relate to the limiting conditions for core damage.

2. Mechanical Design

Mechanical design of the reference reactor emphasized flexibility of the core arrangement and mechanical reliability. Figure 4 shows an isometric drawing of the reactor structure. Figures 5 and 6 show two separate methods for supporting the core which were studied. A design objective was to provide a mechanical arrangement which would permit the substitution of a fast, blanket, moderator, or control element into any position in the core. Achievement of this objective allows modification of the initial arrangement as safety research and development indicate that certain features become less important. For example, if a strong negative sodium void reactivity coefficient became less important after initial operation of the reactor, the large module design or a modification of it could be incorporated in the present reactor with improved economics. The multiple pedestal core support structure shown in Figure 6 permits this modification by allowing movement of different elements into alternative positions in the core.

The high- and low-pressure plenums shown in Figure 4 are to allow sodium flow to the fast and blanket regions of the modular core respectively. This design arrangement requires development and testing to demonstrate that leakage past the multiple pedestals is not excessive. The double grid plate structure in Figure 5 does not have the leakage problems associated with the multiple pedestal arrangement, but has other problems of thermal stress, close tolerance fabrication, and consistency of alignment.

Coolant flow through fuel channels is adjusted by means of fixed orifices. An alternative variable orifice design was studied but not used in the reference design because of the additional mechanical complexity and development required. Several alternative moderator elements were studied, including beryllium oxide, zirconium hydride, and graphite. Each had certain development problems. The graphite appeared to be the best understood of the materials studied, and for that reason was selected. The graphite element is designed for a 5-yr life span with a total expansion allowance of 7% in the can. Freestanding 304 stainless steel walls, 1/16 in. thick, are used in the moderator can.

The reactor vessel is fairly large, being 26 ft in diameter. This large size occurs for two reasons. First, all coolant inlet and outlet lines exit above the surface of the core to prevent uncovering the core if the primary system integrity is disturbed. This necessitates an internal flow area for the coolant to be directed from the inlet nozzles to the bottom of the vessel. Secondly, in order to simplify the fuel-handling system, fuel is stored in the peripheral regions of the reactor vessel.

3. Fuels and Economics

Both mixed-oxide and mixed-carbide fuel elements were studied. Mixed-carbide cores appear to have a potential for improved economics and safety over the oxide. This potential exists if it is assumed that the carbide fuel utilizes a sodium bond and the oxide core utilizes a gas bond. If a successful bonding

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

5-14-65

7661-2504

Figure 4. Isometric Drawing of Reactor Structure

arrangement can be developed for the oxide, then the picture may change. The mixed-oxide fuel (nonbonded) was selected as the reference case, since it appears to be in a more advanced state of development than the carbide.

An analysis of the total costs of an investor-owned, public utility 1000-Mwe fast reactor of the modular flux-trap type indicated that the total power costs could vary from a high of 4.4 mills/kw-hr to a low of 3.2 mills/kw-hr. The high value represents the probable cost for a 1st-generation plant with rather stringent safety requirements and with fuel costs which reflect compromises in terms of economic performance to achieve certain desirable safety characteristics. The low costs indicated are believed to be representative of values which could be obtained after a successful research and development program designed to provide a background of safety and economic information.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

5-14-65

7661-2505

Figure 5. Double Grid Plate Core Support Structure

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

5-14-65

7661-2506

Figure 6. Modular Pedestal Core Support Structure

NAA-SR-11450

III-18

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

Exploration of the various SCCR core arrangements has demonstrated the desirability of certain characteristics over others. For example, the work has illustrated the importance of strong Doppler and weak positive sodium-void reactivity coefficients, and has demonstrated that neutron lifetime plays a limited role in reactor safety. It has demonstrated that appropriate combinations of Doppler and void reactivity coefficients, breeding ratios, power splits between the fast and blanket regions, and peak-to-average power ratios can be achieved by suitable geometric and coupling arrangements.

The modular flux-trap version of the SCCR has been found a feasible method of obtaining improved safety characteristics compared with other fast reactor designs presently available and with competitive economics to these other designs. With a vigorous research and development program which supports research in the areas of safety and fuel and component development, the potential described for the modular flux-trap SCCR appears to be a realizable goal within the time scale of the AEC fast reactor development program.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Fast Reactor Development	
Project: Calculation Methods and Cross-Sections	
Project Engineer: H. A. Morowitz	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-21
General Order: 7661	Subaccount: 1331

I. PROJECT OBJECTIVES

The general objective of this project is to develop reactor physics calculational techniques and codes for reactivity, neutron economy, and critical mass for sodium-cooled reactors that are efficient, reliable, and accurate. Specific objectives include improvement in the methods for calculating multigroup cross-sections for use in multigroup codes, and the improvement of various features of the codes themselves. Special emphasis is placed on the treatment of resonance capture, inelastic and anisotropic scattering, and neutron leakage, emphasizing the neutron spectrum and the cross-sections in the kilovolt energy region and above. The influence of various reactor characteristics on the energy release from core meltdown accidents is of vital concern. Improvement will be sought in the calculation of reactivity coefficients, and in the determination of their dependence on core burnup. An important objective of this project for the fast reactor program is improvement of machine codes and neutron cross-section libraries for making reactor neutronic calculations. Comparison will be made between calculated results and experimental measurements.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

The effort under this project started in October 1964 to support evaluation of the SCCR concept.

A. MULTIGROUP CAESAR CODE

A 35-group cross-section library structure using group widths of half a lethargy unit above thermal energies was developed for making fast and coupled reactor calculations. Such a structure provides ample detail for description of threshold reactions, also covers adequately the entire energy range up to 10 Mev. Its structure also coincides with the group structure used by a number of other laboratories, thereby facilitating future intercomparison of results. Provision was made for 10 down-scattering groups, which is adequate for the description of practically all inelastic scattering.

In order to utilize efficiently a 35-group library, the CAESAR multigroup diffusion theory code, which had been developed at Atomics International, was expanded from 18- to 35-group capacity. To accomplish this, the code was reprogrammed since such an increase in the number of groups exceeded the 32K core storage of the IBM 7094. At the same time, the new code was written in FORTRAN-IV in preparation for use in the IBM System 360. As a step in this code development, the FORTRAN-II version of CAESAR was optimized, thereby reducing its running time by about a factor of 5. The PERT and ARES-II codes were converted to FORTRAN-IV for use in the new CAESAR code.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Programming and compilation of CAESAR-IV was finished; and most of the code options, such as searches and perturbation theory calculations, were debugged. Certain improvements in the output edits were initiated. A revised code description will be prepared.

B. RESONANCE ABSORPTION CROSS-SECTIONS

An improved code, **TRIX**, for calculating resonance integrals and Doppler coefficients, which has been developed in cooperation with the Doppler coefficient project, 7549-1351, has been made available for use. This code incorporates an extensive revision of the analytic methods used in the **ARES-II** code. Principal improvements include: (1) updated resonance parameters for U^{238} , Th^{232} , and the plutonium isotopes; (2) inclusion of a Doppler broadening function for the contribution from interference between resonance and potential scattering; (3) an improved calculation of the escape probability of neutrons from absorber lumps; (4) calculation of unresolved resonances by averaging the integrals over statistical distributions of neutron and fission level widths; and (5) a flux depression correction to the group lethargy width by which the resonance integrals in the group are divided to form the effective group cross-section. For energies below about 70 ev, resonance integrals and flux depression for resonances broad enough to contribute significantly to more than one group are split among 3 groups. The splitting is based on the natural line shape neglecting scattering interference.

Effective capture and fission multigroup cross-sections and temperature coefficients for U^{238} , Pu^{239} , Pu^{240} , and Pu^{241} were generated with **TRIX** for the self-shielding range encountered with the **SCCR** fast and blanket regions. Cross-sections were generated for both the **FAIM** 18-group library structure and the 35-group 0.5 lethargy width structure. Four **ZPR-III** critical assemblies were selected for testing cross-section libraries. U^{235} and U^{238} cross-sections were generated in the 35-group structure for these assemblies.

A preliminary document for internal use describing the operating procedures for **TRIX** was distributed. A report describing the code in detail is being written.

An investigation was started into the theory of resonance overlap and its effect on the Doppler coefficient. The purpose was to determine which features of this effect have a significant influence on the Doppler coefficient, and to make preparations for including such features in our methods for calculating effective multigroup cross-sections. It appears that a correction to the constant potential scattering cross-section used in the **TRIX** resonance integral calculation should be made in order to approximately account for energy dependent non-resonance contributions to the total cross-section.

C. CROSS-SECTION LIBRARY METHODS

As a step in improving the methods for generating multigroup libraries, a card-punch routine for the computed neutron flux and current spectra was added to the **ELMOE** code. This permits the **ELMOE** spectrum, which includes the effect of anisotropic scattering for light elements and is calculated with a very fine energy group structure, to readily be used as input for the cross-section averaging codes **GRAVE** and **PRISM**, which are used to produce the multigroup libraries. The spectrum inputs in the **GRAVE** and **PRISM** codes were enlarged

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

to accept the output punched by ELMOE. Cross-sections from the AI master cross-section tape are averaged over the input spectra.

In order to facilitate the construction of spectrum-weighted, multigroup cross-section libraries, the GRAVE, PRISM, and TRIX codes are being combined in a new code called GRAVE II. This work is being done in cooperation with the cross-section analysis and application project. Some errors in the integration scheme used in PRISM were found and corrected. The organization of the TRIX code was modified so as to permit its inclusion in GRAVE II. The ground rules for the structure of GRAVE II were developed, and the coding is well under way.

The procedures for forming group-averaged cross-sections were reexamined, and recommendations made for modifications to the procedures formerly employed. In the case of resonance absorption, the calculation of the group cross-section from the resonance integrals of resonances contained in the group usually includes a division by the group lethargy width. In order to obtain the correct ratio between resonance and smooth absorption in mixtures, the group lethargy width should be corrected by an amount related to the flux depression caused by the resonances. Also, each resonance in the group should be weighted by the flux per unit lethargy at the resonance. These changes are being incorporated in the new group-averaging code, GRAVE II, previously mentioned.

The group diffusion coefficient for a mixture can be written as

$$D = \frac{\int_{u_1}^{u_2} J(u) du}{3 \int_{u_1}^{u_2} \Sigma_{tr}(u) J(u) du} \quad \dots (1)$$

where $J(u)$ is the neutron current spectrum in the group bounded by lethargies u_1 and u_2 . The macroscopic transport cross-section for the mixture is

$$\Sigma_{tr}(u) = \sum_i N_i \sigma_{tr_i}(u) \quad ,$$

where N_i is the atom density of the i^{th} isotope in the mixture. If it is assumed that the neutron spectrum is separable in space and lethargy, then

$$J(u) = \frac{\phi(u)}{\Sigma_{tr}(u)} \quad .$$

With this assumption, Equation 1 may be rewritten as

$$D = \frac{1/3 \int_{u_1}^{u_2} \frac{\phi(u)}{\Sigma_{tr}(u)} du}{\int_{u_1}^{u_2} \phi(u) du} \quad \dots (2)$$

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

The order of summation (to form $\Sigma_{tr}(u)$) and integration may be interchanged in Equation 1 but not in Equation 2. Nevertheless, it was formerly the practice to interchange this order in Equation 2, and calculate the group diffusion coefficient as

$$D = \frac{1}{3 \sum_i N_i \langle \sigma_{tr} \rangle_i} , \quad \dots (3)$$

where the spectrum averaged transport cross-section for the i^{th} isotope is

$$\langle \sigma_{tr} \rangle_i = \frac{\int_{u_1}^{u_2} \phi(u) du}{\int_{u_1}^{u_2} \frac{\phi(u)}{\sigma_{tr}(u)} du} . \quad \dots (4)$$

An investigation of the error so introduced in Equation 3 showed in all cases that the value of D from Equation 3 was too large, and that the resulting effect on reactivity for typical cases was substantial. For the ZPR-III, Case 29 critical assembly being used as one of the cross-section test problems, it was found that the reactivity error was

$$\rho_{correct} - \rho_{eqn 3} = 0.026 .$$

A typical fast power reactor was also examined and the effect found to be nearly 50% larger than in the above case. This result can be ascribed to the large amounts of low mass scatterers (Na, Fe, etc.) in the power reactor examined. Further studies showed that a direct flux weighting of the transport cross-sections appear to give better results than the reciprocal flux weighting shown in Equation 4. However, current weighting as indicated by either Equation 1 or 2 is presently being used in fast reactor calculations.

Since the multigroup libraries being generated are based on data from one microscopic library (the AI master cross-section tape), while the neutron spectra weighting functions are based on a second microscopic library (ELMOE), possible differences in the light element scattering cross-sections in these libraries were investigated. Comparison between the scattering cross-sections of sodium, iron, and nickel in each library was made. There is little difference between the AI and ELMOE libraries for these cross-sections, and the few differences that do exist cannot account for more than a 1% difference between ELMOE and PRISM removal cross-sections for these isotopes. For sodium, the AI data show slightly smaller peak values and slight energy displacement for the 2.85 kev and 55 kev resonances. Above 100 kev, the agreement is good except that the AI data show four more resonances than the ELMOE data shown. For iron, the data are in excellent agreement below 600 kev, but the AI data show considerably more structure above 600 kev. For nickel, the agreement is excellent over the entire energy range.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

A report is being written which describes the procedures presently used in constructing group-averaged cross-section libraries.

D. CROSS-SECTION LIBRARY PRODUCTION AND TESTING

In cooperation with the cross-section analysis and application project, several multigroup libraries were produced.

Fifteen-group libraries were generated for use in SCCR analysis. Group constants were calculated by averaging the microscopic data over a spectrum calculated by the ELMOE code for the SCCR standard core. Transport cross-sections were averaged with the current spectrum, and other cross-sections averaged with the flux spectrum. The light element scattering cross-sections were taken from ELMOE, resonances cross-sections were calculated with TRIX, and the remaining data were taken from the AI master cross-section tape. The resonance cross-sections are not averaged over the ELMOE flux spectrum, since a constant background potential scattering cross-section is employed in the present TRIX calculation. Where available, inelastic scattering level data were used in calculating the total transfer matrices, whereas the evaporation model was used where no level data were available. The relation between energy loss and scattering angle for elastic scattering was accounted for by means of a 9th-order Legendre polynomial expansion of the differential cross-section.

Thirty-five-group libraries for the analysis of selected ZPR-III fast critical assemblies* were generated as part of a program for testing updated cross-section libraries. The first of these was for Assembly 29,* and some discrepancy in the results was anticipated. All cross-sections were taken from the AI master cross-section tape except for resonance cross-sections, which were calculated with ARES-II since this is the resonance code incorporated in GRAVE. The U²³⁵ cross-sections above 2.5 kev were the smooth data on the master tape. The neutron flux and current spectra for Assembly 29 were calculated with ELMOE. Transport cross-sections were weighted directly with the neutron current spectrum, and reciprocally with the flux spectra for comparison. The resulting reactivity calculated for this assembly was too high, the value obtained with current weighting of σ_{tr} being $k_{eff} = 1.058$, while the value obtained with σ_{+tr} reciprocally weighted with the flux being $k_{eff} = 1.032$. An investigation of the U²³⁵ fission cross-section values on the AI master tape showed that these were some 5% higher than data recently obtained from Aldermaston. Since other evidence also supports the belief that these AI cross-section values are too high, the basic data was revised downward in conformity with the Aldermaston data.

ZPR-III critical assemblies 31, 34, and 36 were also selected as a part of the cross-section testing program. Thirty-five-group libraries for all 4 assemblies were prepared, using the revised U²³⁵ cross-sections and resonance cross-sections generated with the TRIX code. The neutron spectra for each assembly were calculated with ELMOE. The transport cross-sections were weighted directly with the current spectrum and other cross-sections with the flux spectrum, except that the TRIX resonance cross-sections were not weighted with the ELMOE spectrum. However, the correction to the group lethargy width to account for

*W. G. Davey, Nuclear Science & Engineering, 19, 259 (1964)

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

flux depression by the resonances of the absorbing isotope was included for the resolved resonances. The highest resolved resonance for U^{238} was at 3,904 ev, while for U^{235} it was at 61 ev.

Preliminary results for critical assembly 29 gives a k_{eff} of 0.973, and for assembly 36 gives k_{eff} of 0.989. Since diffusion theory results appear to give reactivities which are 1 or 2% low compared with transport theory (see section F ahead), this indicates fairly good agreement for the basic cross-section data employed. The calculations for the other assemblies are in progress, with the results being analyzed.

E. ARGONNE CROSS-SECTION INTERCOMPARISON STUDIES

Analysis of a group of typical sodium-cooled fast breeder reactor cores is being made for use in the ANL cross-section comparison effort. Determination of Doppler and sodium void coefficients, breeding ratio, and critical mass will be made for each case. Eigenvalue calculations are being completed.

Composition-dependent multigroup cross-section libraries were generated for each case. Fundamental mode spectrum calculations made with ELMOE were used to weight the energy-dependent cross-section data in the AI master library. The TRIX code was used to calculate Doppler-broadened, self-shielded group cross-sections for fertile and fissionable isotopes. Other group-averaged cross-sections were calculated with GRAVE and PRISM. Transport cross-sections were weighted directly with the current spectrum. Inelastic transfer cross-sections were determined from discrete level data where available, supplemented by evaporation model calculations.

F. TRANSPORT THEORY CALCULATIONS

A comparison between diffusion theory and an S_4 transport theory calculation was made for the SCCR split-blanket reference module. The geometry, composition, and cross-section libraries used in both calculations were identical. The model of the module was a 5-region circular cylinder having an effective bare height of 171.07 cm, and a radius of 56.591 cm. Starting with the center region and going outward, the 5 homogeneous regions are designated as control, fast, inner blanket, graphite, and outer blanket, respectively.

Fifteen-group cross-sections were generated according to the procedures described earlier. The DTF code was used for the transport calculations, and the in-group contribution to the scattering cross-section was calculated from the relation

$$\sigma_{gg} = \sigma_{trg} - \sigma_{ag} - \sum_{i=g+1}^{g+5} \sigma_{g \rightarrow i}$$

Calculations were made for fuel temperatures of 1000 and 1500°K, and for a case where the sodium in the fast region was voided. A concentration search was made for the reference case, with the diffusion theory calculation in order to obtain a just-critical module.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

The results are compared in the table, and it is evident that aside from the fact that the S_4 calculations give values of k_{eff} which are about 1.5% greater than obtained with diffusion theory, the agreement is quite good.

COMPARISON OF S_4 AND DIFFUSION
THEORY RESULTS

Parameter	Calculation	
	S_4	Diffusion Theory
k_{eff} (1500°K)	1.015	1.000
Breeding Ratio	1.080	1.068
$\alpha = \left(T \frac{dk}{dT}\right) \text{Doppler}$	-0.0175	-0.0181
Na Void Coefficient	-0.0100	-0.0098

G. TWO-DIMENSIONAL 4-GROUP CALCULATIONS

A code CLAPZ* was written for collapsing multigroup cross-sections to few-group sets for use in 2-dimensional calculations. This code provides a flux weighting of the multigroup diffusion coefficients (rather than Σ_{tr} as is done in other codes) in making the group collapse. The code can accommodate 50 groups with 10 groups of down-scattering.

The PDQ diffusion theory code was used for 2-dimensional calculations on the SCCR modular concept. The 4 energy groups used were selected so that group skipping in the collapsed scheme could be neglected. A 2-dimensional mesh was prepared wherein the hexagonal elements were represented by rectangles having the same area. Since PDQ requires a rectangular mesh and module boundaries for a hexagonal lattice structure form a triangle, it was necessary to employ a fictitious material having a very small diffusion coefficient and zero absorption and group transfer cross-sections to represent the space between the module boundaries and the PDQ boundaries.

The results obtained with these calculations are given in the report on the SCCR concept, NAA-SR-11378.

H. REACTIVITY COEFFICIENTS

The PERT code (1st-order perturbation theory using multigroup fluxes and adjoints calculated with CAESAR or FAIM) has been used to study the spatial distribution of Doppler and sodium-void coefficients of reactivity in typical split-blanket and single-blanket fast reactors. As expected, most (70 to 80%) of the isothermal Doppler coefficient is contributed by the blanket. Doppler coefficients were also calculated with CAESAR, the coefficients being obtained by subtracting values of k_{eff} calculated at two different temperatures. The values obtained by these two methods agreed to within 10%. Unfortunately, sodium coefficients calculated with PERT were a factor of two or more larger in magnitude

*J. M. Otter, "CLAPZ, A Group Reduction Routine," NAA-SR-MEMO, to be published (1965)

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

than the corresponding CAESAR sodium coefficients, although the general shape of the curve of reactivity versus volume voided was similar. Attempts will be made to pinpoint the source of this discrepancy and remove or alleviate it, if possible, since the use of successive CAESAR cases to provide sodium coefficient maps is very laborious and time-consuming.

Graphical display of numerical results greatly facilitates qualitative comparisons of different situations, and often results in immediate insights not available from scanning columns of numbers. A code has been written (TR-5) which provides computer-plotted pointwise radial traverses of Doppler coefficient (differential volume contribution), power density, and fluxes and adjoint fluxes by energy group. Input data consist of one or more flux decks punched by CAESAR and a set of Doppler cross-sections supplied by the user.

A modified version of TR-5 provides radial plots of the contribution of each energy group to the Doppler coefficient. On one typical design, results of this code (GPT-1) have shown that more than half of the entire isothermal Doppler effect was contributed by one energy group (Group 8, 3.35 to 0.454 kev), and that the large reduction in this group's contribution upon removal of all sodium in the core accounted for nearly all of the total reduction in core Doppler coefficient.

Further examination has shown that the drastic reduction (factor of 3) in the Doppler contribution from Group 8 is due almost entirely to the decrease in the flux in Group 8. The change in importance for Group 8 is negligible. Nearly all of the down-scatter from Group 7 to Group 8 is due to scattering from sodium, so that sodium removal nearly wipes out the source for Group 8.

Once the core sodium was gone, removal of sodium from the inner and outer blankets (the design studied was of the split-blanket, flux-trap type) had practically no additional effect on the Doppler coefficient.

Some improvements in output format and considerable reduction in writing of input data have been made in the graphical-display code TR-5.

Work has started on a new code to calculate the Doppler effect resulting from a nonisothermal temperature distribution. The gross radial power distribution will be calculated from the PERT flux deck punched by CAESAR or FAIM; from this power shape, a gross average radial temperature distribution will be calculated. From specified initial and final conditions, the temperature rise at each point will be computed; then the Doppler contribution from each point will be calculated and radially integrated. The result will be a more realistic estimate of the Doppler effect than is provided by the present regionwise method.

A study was completed on the influence of various inherent reactor characteristics on the energy release from a core meltdown accident in fast and coupled reactor systems. It was demonstrated that in the absence of a Doppler effect, a longer prompt neutron lifetime system would exhibit a lower energy yield from this type accident. In the presence of a strong Doppler effect, typical of ceramic-fueled fast and coupled reactor systems, the energy release is independent of neutron lifetime. Uncertainties in the equation of state of melted core materials have a similar effect on energy release in systems with a strong Doppler feedback.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

I. FUEL CYCLE

The reactor burnup codes SIZZLE and TRANSBURN have been evaluated for use in SCCR burnup calculations. It was found that the thermal version of the SIZZLE code appears to be adequate for this purpose, but that improvement in the data processing and search options is needed in order to substantially reduce the labor involved in making fuel cycle calculations.

An updated multigroup library has been prepared and used in one burnup calculation for a modular coupled core. The results of this case indicate 4 to 5% Pu buildup in the blanket by the time the core has accumulated about 100,000 Mwd/Mt average exposure. This Pu buildup suggests an increase in positivity of the sodium void coefficient, and in negativity of the Doppler coefficient as burnup progresses. The lifetime variation of these reactivity coefficients is being studied further.

A set of burnup calculations has been carried out to investigate methods of determining isotopic compositions, power shifts, and fuel economics in a fast reactor core. The study was made for an idealized 62.5-Mwe module with a split-radial blanket.

Calculations have been carried out with a modified version of the SIZZLE burnup code. Two separate burnup chains are available, one with cross-sections for the core material and the other for the blanket material. This modification makes it possible to calculate details of the power shifts during the cycle. A slight disadvantage is the omission of Pu²⁴² and the intermediate Np²³⁹ step.

Equilibrium cycles have been obtained for two alternative reprocessing schemes. In the first, core and blanket elements are reprocessed separately, and the fuel for each new core is obtained by adding fresh Pu from the blanket to that recovered from the old core elements. The second choice is an integrated scheme in which the core and blanket fuel are mixed before reprocessing, and the surplus Pu is recovered and sold. The two choices lead to significantly different isotopic compositions and core loadings. Integrated reprocessing should be cheaper, and it also reduces the relative concentration of higher Pu isotopes by 40%, thus leading to a smaller sodium void effect. On the other hand, the core fissile inventory is 5% higher than with separate reprocessing, and the breeding ratio and doubling time suffer accordingly.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The calculational methods and cross-section effort during the year have been devoted to improving the reliability of analysis of fast breeder reactor characteristics, particularly cross-sections, safety coefficients, and long-term burnup behavior. This work will help to provide a sound basis upon which a reliable fast breeder reactor technology can develop and to provide confidence in analytical evaluation of the safety of these systems necessary to encourage the growth of a useful fast breeder reactor economy.

Lack of reliable cross-section data is generally accepted as the major single area of uncertainty in fast reactor physics. Project cross-section work included evaluation of recent experimental data, provision of spectra generation methods,

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

development of more accurate averaging techniques and digital automation of these procedures. The need for improved neutron cross-section libraries for fast breeder reactor calculations is being met. The mechanics of producing multigroup, spectrum-weighted, cross-section libraries is being streamlined. When completed, this will result in a considerable reduction in the time and labor involved in making new libraries. A convenient source of reliable cross-section data for intercomparison with data from other such sources has been developed.

Improvement is being made in the tools for calculating fast-neutron spectra and Doppler coefficients. These improvements will contribute toward the reliability of calculated reactivity coefficients. Studies of the parametric dependence of reactivity coefficients have been made easier by development of the IBM 7094 Code TR-5, which facilitates qualitative comparison of different situations through an analog display.

Evaluation of methods for burnup calculations has shown the need for improvements in methods and data-handling, which will be made.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Fast Reactor Development

Project: Physics Experiments

Project Engineer: H. A. Morowitz

Reporting Period: Fiscal Year 1965

General Order: 7661

AEC Category: 04-01-04-21

Subaccount: 1332

I. PROJECT OBJECTIVES

The objective of the reactor physics experimental program is to supply information pertinent to the safety and economic studies of the fast reactor system and to confirm calculational methods and parameters used in the concept evaluation and preliminary design. This program will study the core physics for U, Pu, and U-Pu mixtures with an adequate spatial distribution of the proper core materials to yield the desired neutron energy spectrum. The specific objectives of this project include the study of Doppler effects, sodium void effects, and the interaction between the two in reactor spectra resulting from core compositions of interest. Because limited information is available on Doppler coefficients, these studies must be made by using the exact fuel composition and configuration planned for prototypal cores. Physics measurements will be made on a variety of materials, composition, and spectra utilized in fast reactors, particularly sodium because of its large volumetric fraction in fast-reactor cores. In order to enhance or control reactivity coefficients for safety reasons, moderating materials have been utilized regionally or homogeneously in many fast-reactor designs. The spectral effects, energy, and spatial distributions resulting from the use of these materials will be determined and compared with theoretical predictions.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

The reactivity worth of possible control rod materials for fast reactors has been summarized from information obtained in the ECEL, and intercompared over a range of median fission energies from 8.5 ev to 422 kev. The control rod materials studied were B, B¹⁰, Eu₂O₃, Gd₂O₃, and Ta. The worths had been determined using a reactor oscillator with sample sizes 2 in. long by 1/2 in. diameter. The results are shown in Table 1. For samples of this size, self-shielding is negligible above 63 kev. This work has been reported in NAA-SR-11368, entitled "Reactivity Worths of Several Fast Reactor Control Materials" by R. W. Campbell and S. G. Carpenter.

Preparations have been made for the study of sodium void coefficients in the Epithermal Critical Experiments Laboratory (ECEL) split-table critical machine. These measurements will study only the spectral effects of sodium voiding, since the spherical assembly used precludes a study of the leakage effects in the center test region. Up to the present time, sodium has not been used as a core material in this machine. Leak-tight stainless steel cans for sodium containment were designed. Samples were made, filled with sodium, welded shut, and found to perform satisfactorily. The boxes have 15-mil walls and, for a test region requiring 50% sodium by volume, the stainless steel occupies only 5% additional

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 1
REACTIVITY WORTHS[†] OF CONTROL MATERIALS
SAMPLES 2 in. x 1/2 in. ID
(¢/sample)

Material	Density (gm/cm ³)	Worth						
		Median Fission Energy (kev)						
		422	190	140	127	63	4.2	8.5 ev
B ¹⁰	1.66	-3.58	-1.53	-1.03	-1.887	-2.38	-4.17	-4.06
B	1.66	-0.729	-	-0.340	-0.527	-0.698	-1.80	-3.30
B	1.375*	-0.605	-	-0.282	-0.437	-0.579	-1.50	-2.74
Eu ₂ O ₃	5.18	-	-	-0.348	-0.409	-0.661	-2.29	-4.83
Gd ₂ O ₃	5.187	-0.136	-	-0.094	-0.134	-0.160	-0.541	-1.18
Ta	16.6	-0.452	-0.258	-0.192	-0.340	-0.444	-0.928	-1.096

*This density corresponds to a B₄C sample at 70% of its theoretical density.

†Because of regional size variations from core to core, it is not possible to directly compare the reactivity of a given material in one core with the same material in another core; however, one can compare the ratio of reactivities of 2 materials in one core to the ratio of reactivities of the same materials in another core.

volume. Approximately 5000 such cans are needed, half of these being empty for a one-to-one replacement of sodium-filled cans in the core. The boxes have been purchased and will be ready for sodium fill early next fiscal year.

Preliminary work has begun in preparation for reactivity worth determinations and Doppler measurements on proposed fuel samples that will be made later in the program. In both of these measurements one must take account of self-shielding and self-multiplication that occurs in fuel samples. In the Doppler measurements, these effects must be subtracted out to obtain a true Doppler coefficient. These effects have been calculated using perturbation theory for various sample changes and compared with a wide variety of experimental data. The normal expression* from perturbation theory is modified as follows.

$$\Delta k/k(\text{¢/gm}) = \frac{N}{R_o} \left\{ \sum_i (v\sigma_f)_i \phi_i^* \phi_i (1 - p_{c,i}) \left(\frac{1 - p_c}{1 - np_c} \right) - \sum_i \sigma_{a,i} \phi_i^* \phi_i (1 - p_{c,i}) + \dots \right\} \quad \dots (1)$$

*PERT, A Perturbation Theory Code, by H. P. Flatt, AI-64-MEMO-102

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

where

$p_{c,i}$ = collision probability for incoming neutrons of energy i

p_c = fission spectrum averaged probability that a fission neutron born in the sample will undergo a collision before escaping

$$n = \sum_i v \frac{\sigma_{f,i}}{\sigma_{a,i}} \chi_i \quad \chi_i = \text{fission spectrum}$$

R_o = constant for the reactor core

N = number of atoms/gm.

The term $(1 - p_{c,i})$ accounts for the flux reduction in the sample due to absorption of incoming neutrons. The term $(1 - p_c)/(1 - np_c)$ is the self-multiplication of fission neutrons produced by the primary flux. The p 's are a function of the optical path length given by the product of the group cross-sections and the sample radius. There are two additional terms of less importance in Equation 1 which are used in the calculations, and which treat the down-scatter and diffusion coefficient.

In order to determine that reactivity changes could be accurately predicted, comparisons were made for a full density and a reduced density sample of fully enriched uranium in cores having a wide range of fission energies. The samples were made up of a series of foils. All uranium was used for the full density sample, and uranium interspersed with stainless steel foils for the reduced density sample. The calculated and measured worths/gm of U are given in Table 2 versus the median fission energy of the test region. The measured and calculated results for the full density sample are in good agreement, with the average difference between the two being only $\approx 10\%$. Agreement for the reduced density samples is not good, particularly at the lower energies. This may be explained by the fact that the heterogeneity effect of the stainless steel foils cannot be adequately taken into account analytically, particularly at the lower energies.

To insure that reactivity difference could indeed be calculated, further comparisons were made with two full density samples of fully enriched U having different diameters. The measurements were obtained in 2 cores only and the reactivity differences are listed in Table 3.

The excellence of these results as compared with the previous measurements could arise from the fact that in this case the samples were homogeneous.

A complete set of infinitely dilute 15-group GRAVE-PRISM cross-sections has been prepared for use with the ECEL data. This set is consistent with the cross-sections used in preliminary design. Self-shielding of appropriate isotopes will be done by TRI-X. This library will considerably improve the comparison of design calculations and work performed at the ECEL.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 2
CALCULATED AND MEASURED REACTIVITY WORTHS^a OF
0.5-in. -DIAMETER URANIUM SAMPLES VS THE MEDIAN
FISSION ENERGY

MFE (kev)	Full Density ^b		Reduced Density ^c	
	Calculated	Measured	Calculated	Measured
422	0.0270	0.0266	0.0254	0.0243
140	0.00669	0.00592	0.00643	0.00484
127	0.00664	0.00599	0.00631	0.00540
63	0.01069	0.00939	0.01037	0.00840
0.0047	0.01288	0.0155	0.01201	0.0111
0.0096	0.01121	0.0124	0.01011	0.0161
0.0085	0.01086	0.0127	0.0098	0.0166

^aWorth is expressed in cents per gm of uranium

^bFull density = 18.34 gm/cm³

^cReduced density = 6.96 gm/cm³ (of uranium)

TABLE 3
REACTIVITY CHANGES

Median Fission Energy (kev)	$\Delta(\rho/\text{gm U})/\Delta(\text{diameter})^*$	
	Measured	Calculated
156	0.00039	0.00036
63	0.00051	0.00054

*The diameter changed from 0.25 in. to 0.438 in.

Perturbation calculations have been made for Pu²⁴¹ to determine its worth as an oscillator sample in the present ECEL core. Approximately 9 gm of Pu²⁴¹ are available* for such a measurement, and should yield considerably more information about the higher isotopes of Pu than is presently known. The sample has been examined to determine how it must be packaged for the oscillator measurement.

*From Doppler measurements in the Reactor Physics Program.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The survey of reactivity worths of control materials for fast reactors produced some interesting results. For example, Eu_2O_3 is essentially as good a poison as natural boron over the entire energy range studied. Over the higher energy range, the poison worth of Ta is approximately half that of boron; however, it is available in metallic form rather than powder. In addition, both Eu_2O_3 and Ta absorb neutrons by the (n,γ) reactor; and thus neither produces helium as does boron.

Work has begun on experiments for the measurements of fuel reactivity worths, sodium void coefficients, and Doppler coefficients for actual fuel geometries. The latter two effects are the outstanding contributors to the safety of fast reactor systems and early determination of the magnitude of these parameters is essential to a satisfactory reactor design. The work performed to date has been of a preliminary nature, and will lead to direct measurement of these parameters at an early date.

The problem of sodium containment has been adequately solved using deep-drawn stainless steel boxes for which the only welding required is that necessary for a final seal after sodium filling. The amount of stainless steel used in the boxes is considerably less than is present in typical fast-reactor cores imposing no limitation on this account.

An adequate treatment of the self-shielding and self-multiplication of fissionable samples appears to have been obtained as long as sample configurations remain homogeneous. Further comparisons will be made as time permits, to further verify the technique although a considerable degree of confidence can now be placed in its application.

The cross-section library used with the ECEL measurements is now compatible with those used by the design effort for the fast-reactor program.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Fast Reactor Development	
Project: Boiling Studies for Sodium Reactor Safety	
Project Engineer: A. A. Jarrett	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-21
General Order: 7661	Subaccount: 1341

I. PROJECT OBJECTIVES

The general objective of this project is to develop information on 2-phase flow required in the safety evaluation of sodium-cooled reactors. This information is important because of the key role that sodium boiling plays in reactor dynamics, in fuel meltdown accidents, and in the ultimate shutdown mechanism of the reactor. Specific experimental objectives include the measurement of boiling heat transfer characteristics, critical heat flux, 2-phase pressure drop, void fractions, transient voids and pressures, and the study of hydrodynamic instabilities. Specific objectives of the theoretical study, in addition to any analyses required to support the experimental work, are the development of a digital computer code which will predict transient void fractions, flow rate, and heat transfer for single and multichannel sodium flow, and the incorporation of this code into a general reactor kinetics code.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

During FY 1965, major effort was expended on determining the steady-state boiling heat transfer characteristics, critical heat flux, and hydrodynamic characteristics of sodium under subcooled and 2-phase forced convection flow. Concurrently, a program was conducted to develop a digital-computer code to predict transient boiling in single channel flow.

The study of steady-state boiling heat transfer under forced convection, initiated in FY 1964, was completed; obtained were the boiling characteristics and critical heat flux for sodium under pressures of 1.5 to 10 psia, flow rates of 0.3 to 3 gpm, and average subcoolings of 0 to 116°F or qualities of 0-14%.

The heat-transfer characteristics, plotted as heat flux versus the wall-to-liquid temperature difference, can be represented for each set of conditions by two straight lines, the break at their intersection indicating the change from forced convection to boiling dominated heat transfer. Unlike the familiar boiling curves of other liquids, which show nucleate boiling over a wide range of heat fluxes, these data show that convection heat transfer dominates almost up to the critical heat flux. The surface temperature data confirm this conclusion, for only at these very high heat fluxes were fluctuations of the temperature observed. An extensive summary of the data and comparisons with expected results have been published.*

*H. Lurie and R. C. Noyes, "Boiling Studies for Sodium Reactor Safety," Part II, NAA-SR-9477 (October 1964)

7661-2507

Figure 1. CHF Under Subcooled Forced Convection

7661-2508

Figure 2. CHF for Two-Phase Sodium Flow

The forced convection critical heat flux (CHF) data for sodium are shown in Figures 1 and 2 for subcooled and 2-phase flows, respectively. The subcooled-flow data were correlated by the method of superposition where it is assumed that the CHF in forced convection boiling is equal to the sum of the saturated pool boiling CHF and the heat-transfer contributions of both subcooling and forced convection flow. The latter convection contribution should be proportional to $h_c \Delta T$, where h_c is the convective heat transfer coefficient and ΔT the overall temperature difference; a better correlation of the data, however, was obtained by using $h_c \Delta T_{sub}$, where ΔT_{sub} is the subcooling of the liquid. Straight-line correlations of the 2 and 5 psia data yielded the following dimensional equations:

$$\text{CHF (Btu/hr-ft}^2\text{)}$$

$$= 737,000 + 1.33 h_c \Delta T_{sub} \quad p = 2 \text{ psia}$$

$$\text{CHF (Btu/hr-ft}^2\text{)}$$

$$= 1075,000 + 2.1 h_c \Delta T_{sub} \quad p = 5 \text{ psia}$$

Several of the data points at 5 and 8 psia were considerably lower than the values at 2 psia. It was concluded that these lower CHF values, obtained in the early stages of experimentation, were premature; that is, the burnout detector indicated critical heat flux due to a surge or oscillation in temperature even though

actual CHF had not been reached. Due to power limitations of the experimental equipment, only a limited range of flow rates and subcoolings could be investigated at these pressures. The data are sufficient, however, to indicate that the CHF does increase with system pressure. This agrees with the results reported for other liquids and with the sodium pool boiling results previously reported.*

The critical heat flux under 2-phase flow, correlated as a function of the exit quality, demonstrates a definite parametric dependence on system pressure (Figure 2). The CHF again increases with the pressure but decreases rapidly

*H. Lurie and R. C. Noyes, "Boiling Studies for Sodium Reactor Safety," Part II, NAA-SR-9477 (October 1964)

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

with the vapor quality of the 2-phase mixture. Analysis is continuing on the effects of flow rate and also on the possibility of correlating the data in terms of dimensionless parameters. Figure 3 gives a summary of the results shown in Figures 1 and 2; the critical heat flux for both subcooled and 2-phase flow is plotted as a function of the dimensional group $\dot{m}(h-h_{sat})$, where $\dot{m}$ is the mass flow rate and h the total enthalpy.

7661-2509

Figure 3. CHF Under Forced Convection

A study of steady-state pressure drops and void fractions as a function of absolute pressure, flow rate, and quality, was performed for vertical upflow of sodium in an unheated annulus. The data obtained are shown in Figures 4 and 5, and cover flow rates of 0.3 to 1.8 gpm, pressures of 1.5 to 10 psia, and qualities of 0 to 14%. The data are presented as the pressure drop parameter, ϕ_L , and the liquid fraction, R_L , as a function of the Martinelli parameter, X_{tt} . ϕ_L^2 is defined as the ratio of the 2-phase frictional pressure drop to the pressure drop that would occur were only liquid flowing in the pipe. The parameter X_{tt} is defined as the ratio of the liquid pressure drop to the gas pressure drop, each flowing alone; for both flows in the turbulent regime, (assuming $f = C/Re^{0.2}$)

$$X_{tt} = \left(\frac{1}{x} - 1 \right)^{0.9} \left(\frac{\rho_g}{\rho_L} \right)^{0.5} \left(\frac{\mu_L}{\mu_g} \right)^{0.1}$$

where x = quality, $\rho_{L,g}$ = liquid or vapor density, $\mu_{L,g}$ = liquid or vapor viscosity, f = friction factor, and Re = Reynolds number. The total pressure drop as measured experimentally was corrected for momentum pressure drops due to sodium vaporization during flow, and the resulting frictional pressure drop was analyzed for parametric dependencies. Over the ranges of flow rates and pressures covered during the experiments no such parametric dependence could be discerned. It is concluded that the slight scatter in the data is due to experimental variations and errors. The scatter in the void fraction data is much more severe

7661-2510

Figure 4. Two-Phase Pressure Drop for Vertical Upflow of Na

7661-2511

Figure 5. Liquid Fraction in Vertical Upflow of Na

This code is based on a homogeneous flow model in which the 2-phase mixture is assigned an average velocity, temperature, and density, and is then treated as a single-phase, variable density fluid. A second code, BURP, while developed

and analysis for parametric dependence on pressure and/or flow rate is continuing. Such dependence has been observed with other fluids (primarily water) and may well explain this marked scatter. In addition, the accuracy of the void fraction measurements at high qualities (low X_{tt}) is being examined. The void fraction is determined from the output of two electromagnetic flowmeters, one reading liquid flow and the other 2-phase flow. At high qualities, the electrical path within the flowmeter may be sufficiently disrupted to give erroneous readings. In Figures 4 and 5 it can be seen that the scatter of data increases in the low range of quality (high X_{tt}); this increase is probably due to increased probable error in the calculations of the quality at these low values.

In preparation for the study of hydrodynamic instabilities and transient flow due to boiling, the 2-phase sodium loop was redesigned and modified. The new loop has a 6-ft annular (1/4 by 1/2 in.) test section uniformly heated along its length. Valves at the inlet and outlet of the test section enable simulation of variable orifices, and a surge tank in parallel with the test section permits boiling instabilities and transients to be induced. Heat will be generated along 4-1/2 ft of the test section (maximum heat flux of 200,000 Btu/hr-ft²) using graphite cloth as a resistance heater, insulated from the test section by a thin sprayed-on layer of aluminum oxide. Adequate performance of the new loop has been demonstrated, calibration of the instruments and a heat loss calibration have been performed, and the first series of tests has been initiated. Data are being obtained for the 2-phase steady-state pressure drop with uniform heating along the test section. These results are required for the analysis of the transient tests.

In addition to the experimental work covered above, the digital computer code TRANS-FUGUE I has been completed.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

under separate funding, is based on research done under this project. Both these codes calculate the axial distribution of velocity, void fraction, pressure, coolant temperature, and fuel temperature for transients simulating power excursions, loss of flow, loss of pressure, and channel plugging. TRANS-FUGUE is a "heat transfer-limited" code (void growth is limited by heat transfer only. BURP is a "pressure-limited" code (void growth is limited by self-pressurization). Both codes have been used in the analyses of loss of flow and power transients in fast reactor cores.

The new test section being installed in the 2-phase sodium loop was also analyzed on the digital code. Several transients simulated on the code indicated that the flow behavior in the test section is very similar to the flow in an actual reactor channel, thus showing the importance of the planned experiments under this project.

These data have also given important information needed in the development of the digital code TRANS-FUGUE. The first phase of this code, TRANS-FUGUE I, has been completed for transient flow in single channels. Even though based on the simplified homogeneous flow model, the code produces correct solutions in many cases and gives important trends in others. This model must now be modified for multichannel flow and real 2-phase flow.

An important conclusion drawn from the analytical studies is that boiling in fast reactors is very unstable. Void growth is very rapid and cannot be followed numerically far into the unstable region both because of the unstable boiling situation and because of instabilities in the numerical technique. An important question as yet unanswered is what happens in the coolant channel after initial voiding. Is there reentry of coolant? Do flow oscillations occur? As a result of these studies, the instability experiments planned under this project will attempt to supply some of the answers to these questions. A second conclusion of the above studies is that both types of transient void growths are pressure limited. Under these circumstances the critical heat flux, already determined experimentally under steady-state conditions, is an important parameter. The heat transfer characteristics and critical heat flux under unstable flow, as yet undetermined but scheduled for experimentation, would, thus, offer additional important information.

Two topical reports, "Boiling Studies for Sodium Reactor Safety," Part II* and "TRANS-FUGUE I,"† were published. These reports summarize the early results of the experimental program and the detail development of the digital code, respectively. Another report "Steady-State Sodium Boiling and Hydrodynamics" is nearing completion.

*H. Lurie and R. C. Noyes "Boiling Studies for Sodium Reactor Safety," Part II, Pool Boiling and Initial Forced Convection Tests and Analyses, NAA-SR-9477 (October 1964)

†R. C. Noyes, J. G. Morgan, and H. H. Cappel, "TRANS-FUGUE I," A Digital Code for Transient Two-Phase Flow and Heat Transfer, NAA-SR-11008

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

Overall progress in both the experimental and analytical phases of the project has been very satisfactory, though a bit slower than expected. Important heat transfer and hydrodynamic data for sodium under forced-convection boiling have been obtained providing useful information for the analyses of sodium reactor accidents. Completion of the steady-state boiling heat transfer and 2-phase flow experiments completes one of the primary objectives of this project. Completion of these tests for subcooled and 2-phase sodium flow gives more reliable information required to more accurately calculate the reactor "burnout margin," and permits better analyses of the hazards involved in severe uncontrolled reactor accidents. The "burnout margin" is the ratio of the local critical heat flux to the normal operating heat flux. In addition, these steady-state results will assist in the planning and analyses of future tests on hydrodynamic instabilities and transients under boiling conditions.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Fast Reactor Development	
Project: Sodium Fires and Fission Product Release	
Project Engineer: A. A. Jarrett	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-21
General Order: 7661	Subaccount: 1342

I. PROJECT OBJECTIVES

The objective of this project is to provide quantitative information necessary to predict the release and transport of matter and energy from postulated primary coolant sodium fires. Of concern is the potentially serious dispersion of radioactivity (Sodium-24 and certain fission products). The method of achieving this objective is to derive equations or operative statements from studies of fundamental mechanisms, experimentation, and modeling principles.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. ANALYSIS

In view of the complexity of obtaining even partial analytical solutions for sodium fires transport processes, the techniques of fractional analysis offer the best overall approach for accomplishing the project objectives. The analytical effort has thus become the foundation for the entire project. The important objectives for analysis have now become to establish, evaluate, and manipulate into dimensionless form the relevant parameters and equations. The analytical efforts this fiscal year have been in this direction.

The transport phenomena which influence the sodium air reaction were examined in order to provide a basis for modeling and extrapolating to fullscale systems, and in order to distinguish the relative importance of various parameters. Attention was given to the chemical reaction, heat transfer, air flow, and diffusion. The reaction occurs predominantly in the gas phase with the gas-phase reaction rate controlled by diffusion of the reactants to the reaction zone (and reaction kinetics in the reaction zone). If it is assumed that chemical reaction rates are too fast to exert any rate control, the flame reaction region would be a plane separating a region devoid of oxygen from another devoid of fuel.

A study of these transport phenomena in the reaction region led to formation of differential equations* describing the one-dimensional diffusion and reaction of sodium vapor and oxygen and the conduction of heat. Further consideration resulted in the inclusion of multidimensional terms for turbulent heat and mass transfer and bulk flow.

*Quarterly Technical Progress Report, NAA-SR-11200, p III-28

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

What remains to be considered is radiant energy transport and the influence of viscous, inertial, and bouyant forces on the bulk transport and eddy diffusivity. There is also the necessity for consideration of the existence of a heterogeneous surface reaction which would require additional equations.

This analytical effort led to the following consideration of the importance of certain parameters, and served as a basis for the large-fires experimental effort. The important factors are: sodium surface temperature, geometry, oxygen concentration, pressure, humidity, and liquid surface condition.

As a basis for the analysis of transport in the regions outside the reaction zone, a preliminary thermodynamic model of gas and energy transfer was developed for an enclosure with small opening in the top. Equations were formulated for flow through the opening and temperature rise in the gallery. The buoyancy equation developed also describes the steady-flow situation in a large enclosure or an open environment.

In the study of fundamental mechanisms, Burgess, Grumer, and Wolfard* have shown that the burning rate of large pools of liquids in quiescent air is:

$$B_x = k \cdot h_c / h_v$$

where

B_x = burning rate (cm/min)
 k = rate constant (cm/min)
 h_c = heat of combustion
 h_v = heat of vaporization.

It is stated that this arises from the fact that the heat transfer is predominantly radiative from the flame to the liquid surface. A test of this, comparing the burning rates of sodium with methanol, unsymmetrical dimethylhydrazine, liquid hydrogen, and four hydrocarbons shows good agreement.† The heat release rate of sodium (kcal/cm²-min) is the smallest of these eight substances.

B. LARGE FIRE - 1

Analysis of the thermal data from the experimental sodium fire, § Large Fire - 1 (LF-1), conducted in FY 1964 showed that only about 10% of the heat generated was conducted through the sodium, and that most of the heat was carried out in the air stream. Based on a flame temperature of 1800°F and a

*Committee on Fire Research, National Academy of Science, "Symposium on the Use of Models in Fire Research," National Academy of Sciences - National Research Council, Washington, D. C., 1961

†See Quarterly Technical Progress Report NAA-SR-10501, p III-26-27.

§"Annual Technical Progress Report - AEC Unclassified Programs, FY 1964," NAA-SR-9999 p. I-C-39

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

wall temperature of 300°F, an emissivity of only 0.13 would be required to transfer all of the heat to the walls by radiation. Since this does not happen, most of the outward radiant energy must be absorbed by particles which have already reached a lower temperature by conduction-convection, and by radiation in turn to still cooler particles. In the heat transfer from particles of about 1μ in size, or smaller (approximate size of newly created oxide particles), the energy redistribution time* is on the order of 10^{-7} sec; and it can be considered that an equilibrium exists almost instantaneously. Under such conditions, the temperature difference between particles and surrounding gas will not exceed 1 to 5°C.

The data obtained from LF-1 showed that the temperature reached at the surface of the pool was about 1300°F; immediately above the surface there was a sudden rise due to burning.

C. EXPERIMENTAL PLANNING

Several problems concerned with meaningful iodine measurements were encountered in LF-1 which have resulted in modifications of the experimental equipment and procedures.†

Examination of the hazards requirements for sodium reactors and the analytical studies showed clearly the need for including oxygen concentration as an experimental variable. Certain sodium spills could occur in cells with reduced oxygen atmospheres, or the combustion process itself would create a reduced oxygen atmosphere. It also became apparent during this examination that under circumstances where the sodium temperature is high (from causes other than heat of combustion), considerable matter and energy release might occur even if little or no oxygen were present.

In order to make best use of the large fires apparatus, it was decided to conduct so-called "equilibrium" experiments in reduced oxygen atmosphere; that is, the desired experimental conditions would be set and the system allowed to come to equilibrium with respect to temperatures and exit oxygen concentration, then the appropriate measurements would be made.

The parameters chosen were:

1) Oxygen concentration, since its reduction has a direct effect on diffusion and kinetics. Of primary interest were low concentrations; therefore 0, 1, 2, and 4% were chosen for immediate investigation. Later experiments will investigate the 8 to 16% range.

2) Sodium surface temperature, since a multitude of heat transfer and heat generation phenomena affect the surface temperature, including non-combustion heat sources. In addition, since presence of the surface oxide is temperature-dependent, it is desirable to make measurements below and above the temperature (~1225°F) at which oxide presumably resides on the surface. The temperatures chosen were 1000 and 1300°F, and were controlled by pot heaters.

*Schack, Z. Tech. Physik, 6, 530-40 (1925)

†For details, see op. cit. NAA-SR-10501, pp III-27-30.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

3) Flow. Although air flow is not an independent variable of concern in a postulated accident, an air flow was maintained in order to maintain a supply of air at the desired oxygen level. It was hoped that the turbulent transport phenomena were not influenced by induced air flow in the present burn chamber. In order to determine if the flow were influential, two widely divergent flows were tested at each condition. The high flow is the capacity of the equipment (~200 scfm). The low flow is 10 to 20 scfm.

To supply, monitor, and maintain "air" at oxygen concentrations ranging down to 0%, a number of major modifications and inclusions were made to the large fires experimental apparatus.

The "air" was made by mixing quantities of nitrogen and oxygen. For the runs just completed, the nitrogen gas was supplied from a system which contains a large (1800-gal.) liquid nitrogen tank, a gasoline fired vaporization system, and a high pressure pump. The nitrogen flow in the burning apparatus can be varied from 0 to 200 scfm. The oxygen was supplied from a 600-gal. trailer equipped with vaporizing coils; oxygen flow can be varied from 0 to 60 scfm. The gas was metered through 3 rotameters before entering the inlet manifold. A mixing fan was provided on the inlet manifold to assure homogeneity of the inlet gas. According to Longton,* combustion will not occur unless some hydrogen or water vapor is present. Because the nitrogen and oxygen have a cryogenic origin, the water-vapor content of the gases will be negligible. Therefore, a water-vaporization tube, sized to allow a small but sufficient amount of water vapor into the gas stream, has been added to the inlet manifold. The gas enters the burn chamber through 2-1/2-in.-diameter inlet pipes located concentrically around the burn chamber at four places. The exhaust rate was adjusted to balance the inlet rate and maintain a pressure of about 1 to 4 in. of water to minimize leakage of air.

The heating capability was increased in order to maintain desired surface temperatures at low oxygen concentrations, where combustion heat is insufficient for this purpose. An additional oxygen analyzer was obtained to measure inlet as well as outlet concentration. In order to minimize leakage in the analyzer train a sigmamotor pump was added. A special permanent soft gasket was applied to the chamber to eliminate leakage in that location. A humidity instrument was installed to monitor the moisture content of the inlet gas.

A test of the gas system showed that it could be balanced successfully for high and low flow, with zero pressure in the burn chamber. Several tests using the liquid nitrogen from the supply truck were made.

A total of 1260 lb of sodium were successfully transferred from three barrels to the heatup tank. The tank was heated to 1000°F, and 35 mc of I-131 in one gram of NaI crystals were injected successfully into the tank. Surveys around the bottom and sides of the tank showed that good dispersion had occurred within two hours after injection.

*P. B. Longton, "Alkali Metal - Gas Reactions Part X: The Influence of Hydrogen on the Reaction of Sodium with Oxygen," IGR-TN/C - 536 (April 1957)

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

D. LARGE FIRE 2 (LF-2) EXPERIMENT

After radiological surveys and analysis of two sodium samples from the pre-heat tank showed that the NaI was uniformly mixed, the burn pot was preheated, the scrubber-water circulating system and the exhaust blower were started, and a flow of nitrogen gas was established through the burn chamber. When the oxygen was purged from the system, 600 lb of sodium were transferred into the 23-in. -ID burn pot in the burn chamber from the preheat tank, and maintained at 1000°F.

Seven scheduled runs were made on three different days. Information was also obtained between scheduled runs. The standby condition which existed between experimental days was one in which no forced air flow was used in the chamber and only a static nitrogen atmosphere was present.

For each flow rate and oxygen concentration at a given bulk sodium temperature, the following observations were made:

1. Temperature profile of sodium in the burn pot.
2. Temperature profile above the burn pot and in the adjoining carryover piping.
3. Concentration of oxygen entering and leaving the burning region, and the amount consumed.
4. Humidity content.
5. Collection of Na in scrubber.
6. Collection rate of I^{131} activity in scrubber.
7. Specific activity of I^{131} in burn pot ($\mu\text{c/lb Na}$).
8. Concentration of I^{131} activity and sodium in gas being transported to the scrubber.*
9. Particle-size distributions, relative mass as a function of the aerodynamic diameter.*

The results for this experiment appear in Table 1.

E. APPLICATION OF EXPERIMENTAL RESULTS

The results of the recent sodium fires experiments were analyzed and tabulated in detail, and used to recommend modifications to the assumptions previously made in the accident analysis for the Fast Reactor Core Test Facility at Los Alamos.†

Zero oxygen runs in LF-2, Figure 1, showed that a considerable amount of heat and sodium are released without combustion, and that heat release must include contributions from sodium surface radiation, convection, and evaporation. Combustion appears to be oxygen-concentration dependent and not greatly affected by flow unless the forced flow is very low. At 1000°F and low oxygen concentrations (0 to 4%), a reasonable burning rate of 0.5 lb/hr-ft²-% O₂ was recommended for calculational purposes. It was also recommended, based on thermal

*Results tabulated in report for Project 7640-1371 "Atmospheric Decontamination in Reactor Accidents," see Section VIII.

†NAA-SR-Memo 11403 "Study of Sodium Fires and Particulate Release as Applied to the FRCTF (June 1965).

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 1
LF-2 RESULTS

Run Number	Gas Flow (scfm)	Oxygen (vol %)		Bulk Temperature (°F)	Burning Rate (lb/hr-ft ²)	Sodium Release Rate (lb/hr-ft ²)	Iodine Release Rate (μc/hr-ft ²)	Specific Activity Release Na (μc/lb)	Specific Activity Pool Na (μc/lb)	Specific Activity Ratio
		Inlet	Outlet							
A	12	0	0	1000		0.16	0.66	4.4	24.6	0.18
B	12	1.2	0.7	1000	0.3	0.25	1.53	6.1	24.6	0.25
X*	0	0		1000		0.03	0.20	6.0	24.6	0.24
C	20	2.0	1.1	1000	1.0	0.23	1.46	6.1	24.6	0.25
D	18	4.1	2.4	1000	1.6	0.31	1.80	5.6	24.6	0.23
E	200	0	0	1000		0.59	2.84	4.8	24.6	0.20
Y*	0	0		940		0.02	0.02	~1.0	25.0	~0.04
F	17	0	0	940		0.10†	0.66	7.0	25.0	0.28
G	192	0	0	940		0.26	1.63	7.0	25.0	0.28
Z*	0	0		1000		2 ± 1§		12.6	24.6	0.51

*Overnight

†An independent method of measurement yielded 0.06 lb/hr-ft². Both methods were subject to marked error.

§Based on burn pot volume loss.

- ① - ⑥ AIR SAMPLE POSITIONS
- * SODIUM SAMPLING TEMPERATURE PROFILES ISOTOPE INJECTION
- ** ROTAMETER FOR O₂ AND N₂ INLETS
- SP-1, SP-2 SAMPLE PORTS FOR O₂ (INLET-1 AND OUTLET-2) ANALYSIS

7661-2512

Figure 1. Large Fire No. 2. Test Apparatus

gradients measured in the gas space in LF-2, that all heat releases including combustion energy be to the gas mixture. The release rate of sodium from the surface was found to be most strongly dependent on surface temperature, which in LF-2 was within 5°F of the measured bulk temperatures. An oxygen concentration or burning rate dependence was not observed for release rate. The recommended empirical factor was 0.035 lb of Na/hr-ft²-mm Hg (Na vapor pressure at surface temperature). Figure 2 displays the sodium burning and release data.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

7661-2513

Figure 2. Sodium Burning Rate and Sodium Release Rate

The iodine was released from the sodium in such a way that the concentration of iodine relative to sodium in the released material ranged between 1/6 and 1/2 the iodine concentration in the burn pot (see Table 1, Col. 10).

F. METALLURGICAL TESTS*

Since the LF-2 was the first experiment to use a carbon steel vessel, it was deemed prudent to provide experimental information on the relative corrosion rates of stainless steel and carbon steel in environments which could be found in sodium fires. Specimens of 1020 steel and 304 stainless steel were exposed to sodium peroxide at 1000°F for 24 hr and a sodium-sodium oxide bath at 1600°F for 24 hr. Little or no corrosion was seen after exposure to the sodium-sodium oxide bath, but definite attack could be observed for the specimens that had been exposed to sodium peroxide. During the sodium peroxide exposure, up to 0.016 in. of metal was removed from the stainless steel specimen, while less than 1/3 of this amount of material was removed from the mild steel specimen. It was concluded that sodium peroxide is much more corrosive to steel than sodium oxide, and that mild steel is significantly more resistant to such attack than 304 stainless steel. Stainless steel tabs were exposed to the peroxide at 900°F for 24 hr. Although corrosion was clearly visible, it was not nearly so extensive as the 1000°F exposure. An 1100°F peroxide test was then performed with even more severe corrosion. These results may help to explain the eroded condition sometimes observed on the outside of sodium containers with small leaks.

*NAA-SR-TDR-11453, "Corrosion of 304 Stainless Steel in Sodium - Oxygen Compounds"

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The survey study of fundamental combustion mechanisms and transport phenomena coupled with early experimental results indicated the importance and existence of gas phase reaction. The importance of careful thermal measurements in this region was thus clearly established, if understanding of heat release and particle generation and initial motion is to be obtained.

Further studies of reaction region transport phenomena, including a combustion study of other liquids, showed the importance of radiation as a heat-transfer mechanism, which may be of primary concern when considering modeling of reaction rates or energy release rates. Thus the measurement and calculation of surface and particle cloud temperatures and emissivities become important.

The systematic study of reaction region forces and energy and mass transfer terms, by establishing transport equations and using other techniques of fractional analysis, showed that oxygen concentration and surface temperature are the most important parameters in describing these phenomena. Geometry is important in so far as it influences the convective flow in the region.

The thermodynamic model provides an excellent base from which further fruitful modeling studies can be built. A comparison of experimental low-flow determinations, from future experiments, and that predicted by the buoyancy equation can now be made. The importance of the experimental measurement of gas outflow from an enclosure opening was made clear.

Within the range of investigation, the results of recent low oxygen experiments showed that a pool of liquid sodium burns at a rate roughly proportional to the oxygen concentration. The release rate of sodium vapor, some or all of which may eventually react, was strongly dependent on sodium surface temperature, and not particularly dependent upon oxygen concentration. The iodine released from the sodium was associated in approximately the same concentration in the airborne sodium as it was in the sodium collected in the scrubber; the iodine concentration of the released sodium was always less than it was in the pool; and concentration of iodine in released sodium was not affected by oxygen concentration or gas flow, but showed a dependence on surface temperature. The energy-release measurements were not as complete as desired. More energy was released than could be accounted for by combustion and natural convection alone. At zero oxygen, the point was clearly made that additional heat release mechanisms (radiation, particle-to-air heat transfer) are present.

A formal evaluation of effort to date to determine future action was made. In the area of matter and energy release, clearly further experiments of the type of LF-2 are needed, with a wider choice of air flows and surface temperatures than previously proposed. Smaller scale experiments are certain to be a help in this area. In addition, the analytical study of fundamental release mechanisms should be continued. The only effective experimental study of matter and energy transport (especially particle transport) will be in environments with a

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

large gas volume capability. The precise physical arrangement of such an experimental facility must await the final results of studies in fractional analysis, modeling, and particle transport done as part of the formal evaluation.

The metallurgical tests showed that carbon-steel vessels should be perfectly adequate for sodium fire tests. Since the most severe corrosion in LF-1 occurred near the bottom, and since Na_2O_2 cannot exist in the presence of sodium metal, the severe corrosion cannot occur until near the end of a fire when localized areas near the bottom are free of sodium so that Na_2O_2 may form.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Fast Reactor Development

Project: Fuel Element Transient Heating

Project Engineer: A. A. Jarrett

Reporting Period: Fiscal Year 1965

AEC Category: 04-01-04-21

General Order: 7661

Subaccount: 1343

I. PROJECT OBJECTIVES

The objective of this project is to experimentally examine the events which occur under various conditions of nuclear transient heating of typical SCR uranium carbide fuel element configurations, and from such studies to develop the foundation upon which to base the understanding of SCR core behavior during power transients. The following information pertaining to the transient behavior of prototype, uranium carbide fuel rods, when subjected to transient irradiation in the TREAT reactor, will be obtained: (1) effects of transient-heating on gross fuel structure and cladding deformation; (2) effect of thermal stresses produced by high, transient heat fluxes on fuel rod cladding; (3) transient-heat-transfer characteristics of UC-sodium systems; and (4) characteristics of fuel meltdown. These studies will then permit assessment of accident limits for fuel damage and of reactor shutdown behavior.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

The effort during FY 1965 was an extension of the Phase I studies which had provided information regarding the behavior of sodium bonded uranium carbide fuel rods for energy generation levels up to the onset of fuel melting. The Phase I studies, in addition to indicating the gross characteristics of transient heating up to the melting region, also provided important information regarding behavior of the instrumentation over a wide range of conditions including bond boiling. The specific purpose of the Phase II testing was the generation of information regarding meltdown behavior. Two distinct types of severe transients planned for the Phase II studies are based on consideration of accidents warrant study in connection with safety analyses. The first of these simulated the reactor behavior for a large, rapid reactivity insertion. The transient consisted of a normal TREAT transient (temperature limited, tail clipped) sufficiently severe to completely melt the fuel. Because of the rapid heating of the fuel during this transient, initial fuel melting preceded boiling of the sodium coolant. The second transient provided a simulation of meltdown in a so-called "choked channel." The conditions for the second capsule were a controlled excursion in a steady-state, TREAT power level of approximately 50 Mw was maintained for approximately 10 sec. Under these conditions, final melting of the fuel occurs in an environment of boiling sodium.

At the time of this writing, analysis of the Phase II experiments was complete except for some detailed chemical analysis of the fuel and cladding specimens from the destructive burst. While this information should be useful in making description of the behavior more complete, the present picture provides a very consistent explanation of the course of events during the Phase II tests.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

A. EXPERIMENTAL PREPARATION

The Phase II in-pile capsule design resembled the Phase I design, the most notable differences being use of a graphite heat sink (rather than aluminum) and use of additional instrumentation. The two Phase II capsules each contained 6-in. lengths of 0.852-in. diameter uranium carbide fuel rods (hypostoichiometric 4.6% C, 4.9% U²³⁵), which were sodium-bonded to 0.952-in. OD by 0.020-in. -wall, 304 stainless steel cladding. The fuel rod was surrounded by stagnant sodium contained in a graphite crucible which also served as a heat sink. Capsule instrumentation included: tungsten/tungsten-26% rhenium fuel thermocouples; three bond and three coolant thermocouples (chromel-alumel); a fuel section pressure transducer; and thermocouples to monitor heat sink, inner and outer capsule and transducer temperatures. The fuel thermocouples were tantalum-sheathed and thoriated, and the chromel-alumel thermocouples were stainless steel-sheathed and MgO-insulated. The heat sink was wound with stainless steel-sheathed nichrome heater wire for purposes of preheating the capsule prior to the test transient. A schematic of the Phase II capsule design is presented in Figure 1. A detailed callout of the thermocouple nomenclature is shown in order to distinguish the various fuel, bond, and coolant thermocouples in the subsequent discussion. The pressure transducer was of the variable permeability type and proved to be somewhat sensitive to reactor power as well as to temperature. In order to compensate for the latter, calibrations were conducted over a temperature range. The temperature at the transducer inlet port was monitored by a thermocouple. Corrections for power pickup were made on the basis of the behavior of the transducers during the calibration transients. The output signal followed power very closely, and did not appear to have any appreciable sensitivity to energy.

During the large transients, the thermocouples sometimes exceeded the limits of calibration. Extrapolations were carried out for both the chromel-alumel and tungsten/tungsten-26% rhenium thermocouples. Data which exceed the normal limits of the calibrations were regarded as approximate. The capsules were accompanied by an instrumentation package which permitted all calibrations to be conducted prior to shipment to TREAT. These calibrations were then rechecked upon installation at TREAT. This approach permitted the elimination of all noise problems and allowed a greater number of instrumentation channels than could be carried by the TREAT channels from the reactor to the control building. Following irradiations at TREAT, the capsules were shipped to the AI hot cell where they were x-rayed, disassembled, and examined visually. Fuel, cladding, and bond samples were taken for metallographic and chemical analyses.

B. RESULTS OF PHASE II EXPERIMENTS

Two transients were conducted on each capsule. The table summarizes the results of the four transients.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Figure 1. Capsule Design

7661-2514

NAA-SR-11450

III-52

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

SUMMARY OF PHASE II TRANSIENTS

Capsule No.	Transient	Initial Period (sec)	Peak Power (Mw)	Integrated Power (Mw-sec)	Type
1 {	1	0.265	79	38.4	Temperature-limited Clipped Tail
	2	0.0704	1560	525	Temperature-limited Clipped Tail
2 {	3	0.25	120	83	Temperature-limited Clipped Tail
	4	0.52	55.8	598	Constant Power

1. Capsule 2 Experiments

a. Thermal Behavior

The first transient with Capsule 2 (Transient 3) was conducted for purposes of verifying the fuel sample energy release relative to TREAT energy. The results of this transient are useful in determining the accuracy of the thermal properties. In addition, because there are two radially positioned thermocouples in the fuel rod of the Phase II capsules, a good estimate of the flux depression within the fuel can be obtained.

The behavior of the fuel, bond, and coolant thermocouples is shown in Figure 2, together with the predicted behavior. The predicted curves are based on the analytical estimate of flux depression, but the average energy release value was adjusted upward by about 10% from the original calculation, in accordance with the Phase II data. It should be noted that the stated accuracy of the energy release estimates from physics calculations is $\pm 25\%$.

It can be seen from the comparison of the predicted fuel temperatures with the experimental data that the flux depression was overestimated; as a result, the centerline temperature is higher than predicted and the outer fuel temperature is lower than predicted. The coolant thermocouples were originally positioned so that the central and upper thermocouples (1 and 3) were at the surface of the heat sink and the lower thermocouple (2) was at the surface of the cladding. For this reason, the lower thermocouple (corresponding to the bottom of the 6-in. slug) reads higher temperatures than the others. However, it appears that the central coolant thermocouple (1) may not have retained its position against the heat sink as evidenced by the temperature dip between 4 and 5 sec when the upper and lower thermocouples are undisturbed.

The other important item of information obtained from the preliminary burst is the behavior of the pressure transducer. Figure 2 shows the response of the transducer to reactor power. On the basis of this behavior, the contribution due to power pickup was subtracted from the pressure transducer signal obtained during the final transient (II-3) with Capsule 2, in order to obtain the true pressure behavior of the fuel section.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

a. Fuel Temperatures

b. Bond Temperatures

c. Coolant Temperatures

d. Fuel Section Pressure

Figure 2. Phase II TREAT Experiments, Transient 3

7661-2515

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

In the large burst (Transient 4) conducted on Capsule 2, the TREAT energy release of 598 Mw-sec was inserted over a period of about 17 sec. This corresponds to 320 Btu/in.³ in the fuel, or an adiabatic temperature rise of 9000°F (neglecting the heat of fusion). Quite clearly, substantial heat transfer will occur, and the data show that over the duration of the burst about 50% of this energy was transferred out of the fuel (by virtue of the fuel temperature rise of 3800°F and fusion energy). The sequence of events during the transient is shown in Figure 3.

Bond thermocouple temperatures remained at the boiling temperature for almost 20 sec following the burst. While it is possible that bond boiling could have been sustained for this duration, it is more likely that a considerable portion of the bond condensed on the upper portions of the fuel section leaving the region around the fuel (at least partially) dry. In the 20-sec period following the burst, the bond and coolant temperatures remain relatively constant while the fuel temperature drops 2500°F. Here the heat transfer out of the fuel occurs at about the same rate as the transfer to the heat sink, so that sodium temperatures do not change appreciably.

b. Postirradiation Examination

Disassembly of the capsule indicated no gross melting of fuel, although fuel and bond thermocouples were fused to the fuel rod. However, the rod could be slipped out of the cladding and the dimensions of the cladding did not change (within sensitivity of 0.2 to 0.3%). It is possible that the stainless steel thermocouple sheath reacted with free uranium at the surface of the fuel slug or that contact of fuel with the sheath produced sheath melting. There was no evidence of segregation of free uranium at the surface (or at the center) of the slug. In some regions it appears that the tantalum sheath of the fuel thermocouples reacted with the fuel. Metallography of the fuel indicated that no changes in microstructure occurred when compared to the fuel prior to irradiation. This at first was taken to imply that substantial melting did not occur. However, the thermal data were too conclusive, and further investigation revealed that the freezing time (of a few seconds) in the experiment resembled that which results during the casting process. Metallography of the fuel before and after irradiation is presented in Figure 4, which shows no apparent differences (within the normal variations in the cast UC structure).

2. Capsule 1 Experiments

a. Thermal Behavior

The preliminary transient on Capsule 1 (Transient 1) was conducted for the same purposes as that for Capsule 2. The energy release was 38.4 Mw-sec, the initial period was 0.265 sec and the transient was sharply clipped. Figure 5 shows the predicted and experimental thermal data. As was the case in Capsule 2, the prediction of central temperature is somewhat low. There is some scatter in the data because of the difficulty in reading the oscillograph trace which had not been permanized. It does not appear that the central and lower coolant thermocouples (1 and 2) have maintained their positions against the heat sink and cladding, respectively, since the temperatures are quite similar. It

a. Power

d. Fuel Temperatures

b. Energy

e. Bond Temperatures

c. Pressure

f. Coolant Temperatures

Figure 3. Phase-II TREAT Experiments, Transient 4

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

<u>Time</u>	<u>Description</u>
8 sec	A small power dip occurs, causing an inflection in fuel temperature.
10-11 sec	The fuel temperature passes through the melting point of uranium, and there appears to be a small lag in the fuel temperature rise. This is seen more clearly by comparing the linearity of the energy curve between 10 and 14 sec with the linearity of the fuel temperature, and observing the deviation of the fuel temperature in the 10- to 11-sec region.
12 sec	The bond temperature appears to have reached its boiling point (approximately 1700°F). The fuel temperature rise slightly lags the calculated rate, which assumed that film boiling would occur immediately in the bond. It is also possible that an increased nucleate boiling coefficient took place for about 1 sec which is consistent with observed behavior of the coolant temperature (rate of rise increasing), and with the level of the heat flux which is below critical. It should also be noted that elimination of the power effect from the pressure curve indicates that during the 12- to 15-sec period, a pressure increase of about 20 psi was detected in the fuel section. Also notable is the behavior of bond thermocouple 3, which is placed at the very bottom of the fuel section between the base graphite plug and the cladding. This shows that although bond boiling occurs in the 6-in. fuel region at about 12 sec, the void does not communicate to the base until about 16 sec.
13 sec	The behavior of coolant thermocouple 1 shows the reduction in temperature rise, and therefore heat transfer, which occurs upon film boiling in the bond.
14-15 sec	At 3200°F for the central fuel thermocouple and at 3300°F for the outer fuel thermocouple, a distinct thermal arrest is observed for almost 1 sec. This effect has not been explained, although it could be due to a sharp reduction in the solid solution of U in UC (thus forming appreciable liquid phase). It is assumed to be real on the basis of the continuity of the thermocouple response (i. e., no indication of thermocouple failure).
17-22 sec	Although the central fuel temperature rose from 3100 to 4200°F while the energy input rose from 60 to 80% of the total, the temperature rise of the fuel for the final 20% of the energy input was less than 300°F. Consequently the data indicate a 2nd-order arrest or the formation of a second phase to a substantial degree. The heat absorbed corresponds to about an 800°F rise in fuel temperature, which indicates that melting was almost complete for the central regions of the fuel rod.

Figure 4. Metallography of Hypostoichiometric UC
(4.6 wt % C) Capsule-1 Fuel

7661-2525

NAA-SR-11450

III-58

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

is possible that the similarity is caused by the lower thermocouple being positioned near the heat sink. Once again the pressure transducer showed some sensitivity to reactor power, and this effect is subtracted from the pressure curve in the large transient.

The large transient (Transient 2) on Capsule 1 consisted of a 525 Mw-sec TREAT energy release on a temperature limited transient (tail clipped) having an initial period of 0.0704 sec. A peak power of 1560 Mw was reached. Reduced data are presented in Figure 6, however, much of the interpretation requires detailed examination of the oscillograph trace. Those details which are important will therefore be indicated by a description of the sequence of events in Figure 6.

The explanation of the behavior at 2 sec cannot be proven because of the complexity of the system, but consideration of the possible causes has produced a very consistent and plausible line of reasoning, as follows. Since the pressure pulses were observed following the termination of the transient, factors such as fuel vapor pressure, pressure due to dissolved gases within the fuel, and pressures due to rapid heating (in which the stresses are not relieved) can all be eliminated as causes of the fuel dispersion. Three other possibilities must then be considered: cladding vapor pressure; coolant vapor pressure, and the re-freezing of fuel in which coolant or cladding is trapped with the subsequent heating of the trapped material; and the generation of pressure pulses as the pressures exceed the (relatively weak) bursting limits of the resolidified fuel.

Cladding vapor pressures are not a likely source of the dispersion because of the relatively high boiling points of most of the constituents of stainless steel, coupled with the fact that the system pressure is greater than 100 psi. Furthermore, metallographic examination of the material after the test showed considerable amounts of fuel-cladding reaction and very little melted, unreacted cladding. This observation would not be expected if the contact of the molten fuel with cladding produced appreciable dispersion.

The possibility of refrozen fuel entrapping coolant cannot be eliminated conclusively. Such a hypothesis does not seem particularly reasonable in light of the timing of the pulses (which would be more random in this case), nor with the observation of a large pulse 20 msec after cladding failure (hardly sufficient time for solidification of a substantial fuel particle).

On the other hand, the generation of large local sodium vapor pressures with the consequent dispersion of fuel is completely consistent with all observations. First, the timing of the pulses could have resulted from a fairly thorough dispersion of the sodium following a pressure pulse, thus requiring a certain amount of time (on the order of gravity fall) before intimate fuel sodium contact would again develop substantial local pressures. The postirradiation examination of the capsule did not reveal any sodium in the lower portions of the graphite crucible, but rather a dispersion of sodium-fuel-clad mixtures over the area of the heat sink including upper portions of the capsule. Secondly, examination of the shape of the pressure pulses shows that the rise and fall rates resemble rather closely those observed during bond boiling. The magnitude of the pulses is difficult to estimate analytically because of the uncertainties in the geometry at the time of the dispersion, and because of the cooler regions that exist between the

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

7661-2518
a. Fuel Temperatures

7661-2519
b. Bond Temperatures

7661-2520
c. Coolant Temperatures

Figure 5. Transient-1, Phase II
TREAT Experiments

a. Reactor Power

b. Energy

c. Fuel-Section Pressure

d. Fuel Temperatures

e. Bond Temperatures

f. Coolant Temperatures

7661-2521

Figure 6. Phase-II TREAT Experiments, Transient 2

NAA-SR-11450

III-62

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

<u>Time</u>	<u>Description</u>
0.45 sec	Slightly less than one-half of the total energy has been released. The central fuel temperature is about 2200°F, although the predicted value is considerably higher (about 2500°F). There is a striking similarity in the temperature behavior as the fuel rises rapidly through 2000°F, which was observed in the fast, large transients in both the Phase I and Phase II studies. This behavior is being analyzed further, particularly to determine whether it is a problem in instrumentation response, local conditions in the region of the thermocouple, or a gross effect in the rapid melting behavior of the fuel.
0.5-0.6 sec	All bond thermocouples fail, as evidenced by their decrease to temperatures below 1500°F. Thermocouples functioning in the bond region during boiling usually read a constant temperature corresponding to the boiling point. The bond thermocouples at the center of the fuel rod gave indications of an arrest at about 1700°F. Although the one monitored by TREAT instrumentation had an arrest lasting about 0.1 sec, the thermocouple being monitored by the AI instrumentation had an arrest of about 0.01 sec, and rapidly rose to about 2400°F when failure occurred. The lower bond thermocouple appears to have failed before the temperature in that region reached the boiling point. It is notable that, as late as 0.6 sec, the lower bond thermocouple was reading less than 1700°F. Apparently, the developed pressures were being relieved at the top without any severe disturbance of the small annular region of sodium around the base graphite spacer (between the fuel rod and the bottom stainless steel end-plug). Rather surprisingly, the pressure transducer for the first time begins to detect repeated pulses which occur every few milliseconds and which have magnitudes of 10 to 20 psi. It is possible that the effect of the power disturbance shielded earlier fluctuations, or (more likely) that sodium vapor reaching the upper regions initially was rapidly condensed by the cool (700°F) surfaces. The fluctuations ceased after about 0.2 sec, and only one small disturbance was observed during the next 0.9 sec.
0.7 sec	More than 90% of the energy has been released, and the central fuel temperature reads about 4000°F.
1 sec	Center fuel temperature is 5200°F. The burst is complete at this time, although the center of the fuel has been receiving and continues to receive energy from outer regions of the fuel which have reached higher temperatures due to the flux shape in the rod. There did not appear to be any severe arrest in the 4000 to 4500°F temperature region, and the central fuel temperature takes a rapid jump from 5200 to 6200°F, falls off to 4500°F in a sporadic fashion during a 0.1-sec period, then remains fairly constant at about 4300°F for about 0.8 sec. At this time the coolant temperatures level off between 1200 and 1300°F for about 0.6 sec, and then rise more rapidly to the boiling point. It thus appears that after all of the energy is released in the fuel (the bond has previously voided), there is a short time (of about 1 sec) during which fuel and coolant temperatures remain relatively calm. As noted previously, pressure is also steady during this period. This period is associated with the time required to melt the surface of the fuel (which is kept cool during the early part of the burst by virtue of the good heat transfer from the slug), and to either react or melt with the cladding. Because this latter process absorbs energy, it is reasonable that the rate of increase of the coolant temperature will be arrested.
2 sec	At this point, the coolant temperatures (which have been extremely calm) oscillate severely, followed in 0.02 sec by a pressure pulse of about 50 psi. The central fuel temperature drops by more than 1000°F and the bond thermocouples also oscillate. This behavior is clearly due to the failure of the cladding and the movement of fuel and cladding into the stagnant coolant. This causes rapid heating of the sodium and the resulting pressure surge. Three additional pressure surges are observed, each about 0.2 sec apart, with the largest magnitude reaching about 100 psi.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Figure 7. X-ray of Capsule Before
and After Irradiation

7661-2522

Figure 8. Sectioned Inner Capsule

7661-2523

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

source of the pressures and the pressure transducer. An analog calculation of the behavior of hot fuel radiating across a sodium film with subsequent conduction of the heat through the sodium shows that sodium temperatures can reach the sodium critical point. Such a mechanism could readily produce pressure increases from 150 to 250 psi (the maximum pulse observed) in the space above the sodium.

b. Postirradiation Examination of Capsule 1

X-rays of the capsule before and after irradiation are presented in Figure 7. A photograph of the inner capsule after sectioning is shown in Figure 8. This shows most clearly the extent to which cladding and fuel were transported to the upper regions of the capsule. Photographs of the upper portion of the fuel rod and of the upper cap are shown in Figure 9. A composite photomicrograph taken of a section of the fuel 1-in. above the weld on the bottom end plug is given in Figure 10. This shows the considerable extent of fuel clad reaction and the dendritic structure which formed on resolidification. In addition, some large grains of UC are also evident. It is possible that certain portions of the fuel did not melt completely.

Figure 9. Upper Portion of Fuel Rod and Upper Cap

7661-2524

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

III. EVALUATION OF EFFORT

The results obtained in the final transient with Capsule 2 in effect established a rather useful performance limit with respect to tolerable conditions during reactor transients for liquid metal bonded carbide fuel. Relating the results to an SCR with central region power levels of 20 kw/ft, the experiment showed that power levels of about 10 times this magnitude could be tolerated for periods as long as 10 sec without gross cladding failure.

The experiment also indicated that the hypostoichiometric UC undergoes a thermal arrest in the region 4200 to 4500°F, consistent with phase diagrams which give a melting point of the stoichiometric UC of 4550°F. The heat of fusion corresponds to at least an 800°F temperature rise, which is fairly close to some estimates in the literature.

Bond boiling under slow transients may produce a short period of improved heat transfer but as boiling proceeds (and heating of the fuel), the eventual formation of a vapor film results in heat transfer which reflects the conductivity of sodium vapor. Examination of the change in the slope of the coolant temperature versus time indicates a reduction in heat transfer by about a factor of 10. Pressures observed due to bond vaporization were about 30 psi with some additional contribution (about 1 atm), due to heating of the inert gas in the fuel rod.

In the final transient with Capsule 1, a mode of meltdown behavior has been experimentally observed which indicates that random dispersal of molten ceramic fuel can be caused by large local sodium pressures with the subsequent solidification of the molten fuel in cooler regions of the system. The meltdown of sodium-bonded, ceramic fuel appears to follow the sequence: bond vaporization, central fuel melting, melting at the fuel surface and fuel clad contact, reaction and/or melting of cladding material and, finally, flow of molten fuel and cladding into the coolant with subsequent dispersion due to rapid coolant heating and pressure generation.

The course of meltdown could be altered by pressure within the fuel rod which might inhibit bond boiling and which might result in pressures which cause cladding failure by virtue of internal pressure rather than clad melting or fuel-clad reaction.

The results of this work can be extended further toward applicability to the behavior of power reactor systems if additional information regarding the likelihood of horizontal motion (through fuel element tubes or process tubes) was acquired, for example, by a multiple rod meltdown within an outer tube. In addition, the effect of other variables such as preirradiated fuel, fuel composition, cladding material, to name several, requires investigation.

In order to permit use of experimental meltdown information in the analysis of severe reactor accidents, modifications were made to the reactor kinetics code, AIREK, in order to permit a coupling of the thermal and neutronic behavior during various hypothetical accident conditions. Provisions were made to permit fuel melting and coolant vaporization and the simulation of the course of voiding which could be based on boiling codes such as TRANSFUGE and BURP or other analytical approaches. The dispersion mechanism observed in the

MATERIAL AT BOTTOM OF FUEL SECTION, (A) BEFORE IRRADIATION,
(B) AFTER IRRADIATION TREAT TRANSIENT

7661-2517
Figure 10. Metallograph of Fuel
Before and After Irradiation

NAA-SR-11450
III-67

OSTI ID: 00009406

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TREAT experiments was then employed in determining the reactivity changes associated with fuel motion and two models were considered. The first (referred to as the spherical model) assumed that horizontal displacement could occur and examined the reactivity effects associated with dispersal of material from the center of the core, uniformly distributing this material into the remainder of the core. The second (referred to as the cylindrical model) assumed that fuel could move only upward and downward, and examined the reactivity effects associated with movement of material out of the reactor from the melted portions. The application of these models consisted simply of determining the rate at which melting propagated through the core (by an AIREK calculation without the disassembly model) followed by the AIREK calculation which inserted the reactivity effect upon the initiation of fuel motion.

To determine the extent to which they might influence meltdown behavior and the disassembly model, several other items were considered: the rate of energy generation within the fuel; the rate at which dispersion due to sodium pressures occurs; the importance of fuel and cladding vapor pressures; the degree of inlet subcooling at the time of the meltdown; the temperature distribution within the core; the disposition of fuel regions above and below the melted portions; fuel rod geometry; clad material; the ejection and return of coolant to the melting regions of the core and the associated reactivity effects; the reactivity effect of transport of hot fuel to outer portions of the core; and the effect of burnup.

In many of the preceding areas, analytical approaches are quite limited and additional study will be necessary before a complete characterization of these phenomena can be established.

SECTION IV
SODIUM-COMPONENT DEVELOPMENT
CONTENTS

<u>Title</u>	<u>Identification</u>	<u>Page</u>
SODIUM-COMPONENT DEVELOPMENT . . Summary		IV-1
OPERATION OF SODIUM COMPONENT TEST INSTALLATION	7593 - 1322	IV-4
INSTALLATION OF ALCO STEAM GENERATOR	7593 - 4125	IV-6
FABRICATION OF MODULAR STEAM GENERATOR	7593 - 4121	IV-7
HIGH-TEMPERATURE STRAIN MEASUREMENTS	7593 - 1328	IV-14
MECHANISMS IN SODIUM	7593 - 1329	IV-16

ALCO & MODULAR STEAM GENERATORS INSTALLATION IN SCTI

MSG SUPERHEATER MODULES (2)

SODIUM INLET

STEAM OUTLET

FEEDWATER INLET

BLOWDOWN GAS DISCHARGE

TEST PIT

SODIUM OUTLET

MSG BLOWDOWN

RUPTURE MEMBRANES

BLOWDOWN TANKS

MSG EVAPORATOR MODULES (2)

ALCO (B-L-H) STEAM GENERATOR

MSG BLOWDOWN

SODIUM DUMP TANK

9-24-64

7641-1582

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program:	Sodium Component Development
Program Manager:	D. T. Eggen
Reporting Period:	Fiscal Year 1965

I. PROGRAM OBJECTIVES

The general objective of this work is to provide reliable and economic heat-transfer components for large, economically competitive sodium-cooled power reactors. The specific objectives of this program are to develop and test prototype sodium heat-transfer components, such as sodium-to-steam generators, sodium-to-sodium heat exchangers, pumps, valves, flowmeters, and instrumentation. This program also investigates fundamental sodium technological problems such as heat transfer on shell and tube sides, corrosion-induced or enhanced by heat-transfer conditions on both the water and sodium sides, material property changes induced by impurities in the coolants, and mass transfer in systems containing mixed materials and large temperature gradients.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. SODIUM COMPONENT TEST INSTALLATION

Performance of the entire sequence of preoperational tests was completed. This means that both primary and secondary sodium systems have been filled with sodium, and main pumps operated; pilot burners have been used to add heat in the main gas-fired heater; the steam system has been chemically cleaned; preheated, and made ready for the generation of steam; and individual equipment components have all been determined to be properly installed, calibrated, and made ready for proper service. A new and larger tank was purchased for the primary system, and the existing secondary tank was replaced with the original primary tank.

Performance of the preoperational testing program revealed a number of deficiencies in the plant, which have been corrected. Performance of the step-by-step preoperational test program made manifest a number of engineering and construction oversights which, if left uncorrected, would prevent proper operation or adequate control of the plant. Successful conclusion of all of the preoperational tests has rendered the plant ready for experimental operations. Procedures for the plant performance tests have been completed. Step-by-step procedures for steady-state and transient testing of the ALCO/B-L-H steam generator are approximately one-half done.

A number of additional standard operating procedures were written. A new plant description was written, and a table of the design specifications giving the operating limits of major equipment and of the various systems has been compiled.

All construction work required for placement of the ALCO steam generator, i. e., installation of steam and sodium piping, placement of heaters, thermocouples, and instruments and installation of associated electrical gear was

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

completed. Subsequent to the installation of the steam generator, the connecting piping and the steam generator were hydrostatically tested. A gas chromatograph was put into operation.

A larger (7500-gal) tank was purchased and installed for use as a primary fill/drain tank. The original primary tank was substituted for the secondary tank. The plant is finished. The project objectives have been satisfied.

B. FABRICATION OF THE MODULAR STEAM GENERATOR

Fabrication of the modules was completed with no major problems. The welding and x-ray inspection of the tube-to-tubesheet welds has presented little or no difficulty. Tube-to-tubesheet welding procedures which were developed during the design phase of this project were used in the fabrication of the modules. Of the first 37 tube-to-tubesheet welds made, two were considered questionable. Both were rerun and reinspected and found to be within the limits of the specification. The entire Cr-Mo tubesheet and welds were placed in a furnace for postheat treatment, then the tube bundle was assembled to the shell by placing the shell in pit. In making the tube-to-tubesheet welds on the superheaters, a short tube was accidentally installed in a tube bundle. It appeared advisable to cut off the short tube and replace it with a tube of correct length. Examination revealed that a guide "bullet" used in inserting the tube had not been removed, causing excessive internal gas pressure during the welding operation. A modified procedure was used to weld the tube to the splice piece. In any future tube-to-tubesheet welding operations of this nature, a pressure control device will be utilized.

Initial welding done on the upper shell required an unusual amount of rework. As the welders gained experience, the rework required became consistently less. Fewer difficulties were encountered in welding of the shell assemblies for the superheaters.

In the design of any future modular steam generators, consideration should be given to changing the cone reducers to standard concentric reducers. This would allow welding and radiography in straight sections of equal diametric with no change in section. Postheat treatment of the evaporator shell welds was done. Review of the assembly operations of the first evaporator and the cost of inserting the tube bundle in the shell showed that this operation should be changed. Assembly time was considerably reduced and radiography made easier with less possibility of damage to the tubes.

After completion of the entire unit, hydrostatic testing, rinsing, and drying were accomplished. Upon completion of the hydrostatic test, the outside of the evaporator modules was wire-brushed, given two coats of primer paint, and shipped to storage. The thermocouples which will be used to measure the individual tube outlet steam temperatures were clamped to the evaporator tubes. Calculations made indicated the thermocouple installation was adequate for determination of any flow instabilities. Fatigue tests on a module head were completed, and additional tests on standard bar type specimens showed that 5 Cr - 1/2 Mo is superior to 2-1/4 Cr - 1/2 Mo.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

C. INSTRUMENT DEVELOPMENT

Since inception of this project in March 1965, a detailed plan of action for the work effort has been prepared to perform a limited number of pilot tests of the more promising commercially available electric resistance strain gages, and to inspect the production facilities of strain gage manufacturers and vendors manufacturing special high-temperature strain gage wire. Pilot evaluations include the following.

- a) Apparent strain versus temperature characteristics at 900, 1000, 1100, and 1200°F
- b) Stability (drift) versus time characteristics at 900, 1000, 1100, and 1200°F (for periods up to 6 mo before and after thermal shocking)
- c) Repeatability characteristics at 1000 and 1200°F
- d) Resistance to thermal shock at rates up to 50°F per sec and temperatures up to 1200°F
- e) Variation in gage factor as a function in temperature.

The lack of strain gage reliability is probably often due to inadequate quality control, showing a need for improvement in techniques for casting the bars from which the wire is drawn and for maintaining more adequate quality control during the drawing operations. Too much lot-to-lot variation in commercial wire filament strain gages for accurate or meaningful strain measurement at temperatures above 750°F is evident. Development of procedures to make more reliable wire-filament strain gages, development of better weld joints, use of more stable alloys for 1200°F gages, and development of specifications for strain gages with considerably improved performance and reliability above 750°F, under steady-state and thermal shock conditions, is required. With such development, it should be possible to build high-temperature nuclear components and accessory equipment having far greater structural reliability with respect to creep, fatigue, stress-rupture, etc., than is possible today.

D. MECHANISMS IN SODIUM

Testing of bellows at a stroke to compressed length ratio of 4:1 in 1200°F liquid sodium was reinitiated using test equipment and candidate bellows reclaimed from previous bellows work. A Bellmetrics, Type 316 stainless steel bellows, previously exercised in 1200°F sodium, failed at 43,000 cycles, representing 430% of design life.

Bench testing of a motion-multiplying (4x) mechanism which consists of a 2-stage differential rack system was started. This mechanism contains no bearings as such, and shows great promise for applications where a long output stroke with a short input is desired, i. e., control elements and fuel handling. Approximately 4200 cycles have been logged by the test mechanism, with no sign of degradation.

Disassembly and inspection of a mechanism which utilized a sodium-vapor condensing and thermal barrier zone below a conventional rubber O-ring (rotary seal) in a pool of sodium at 1200°F for 5252 hr were completed. The condensing plates were very effective. Internal to the rotary shaft were two actuator shafts (one within the other) which could be moved axially through a 4-in. stroke. It was concluded that, in time, both shafts would have accumulated sufficient amount of sodium compound to induce binding.

The excellent performance of the Belmetrics bellows alleviates to a great extent reservations concerning the use of bellows as seals in liquid sodium. Moving parts in a sodium vapor environment will eventually become bound if a temperature gradient extending below the freezing point of sodium exists in the part-clearance spaces.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Sodium Component Development	
Project: Operation of Sodium Component Test Installation	
Project Engineer: J. A. Leppard	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-31
General Order: 7593	Subaccount: 1322

I. PROJECT OBJECTIVES

The project objective is to operate the Sodium Components Test Installation and to test, obtain data on, and evaluate the performance of prototype intermediate heat exchangers and prototype steam generators.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

Performance of the entire sequence of preoperational tests was completed. This means that both primary and secondary sodium systems have been filled with sodium, and main pumps operated; pilot burners have been used to add heat in the main gas-fired heater; the steam system has been chemically cleaned, preheated, and made ready for the generation of steam; and individual equipment components have all been determined to be properly installed, calibrated, and prepared for proper service.

During preparation of procedures for filling the primary sodium system, new calculations revealed that the fill and drain tanks installed for both the primary and secondary systems were too small. A new and larger tank was purchased for the primary system, and the existing secondary tank was replaced with the original primary tank. A total of 41,000 lb of sodium is contained in the primary system, and 36,600 lb in the secondary.

As a result of the tank capacity problem, the schedule for the completion of preoperational testing was extended to very nearly the end of the fiscal year. In order to optimize manpower utilization, the sequence of preoperational tests was altered to expedite testing of those parts of the plant which were not affected by tank construction work.

Performance of the preoperational testing program revealed a number of deficiencies in the plant which have been corrected. Typical of these problems was the addition of safety valves, sampling locations, and safety features.

Planned activities subsequent to the completion of the preoperational test program include plant performance tests and component tests. Procedures for the plant performance tests, the objective of which is to determine the operating characteristics of the SCTI as a plant, have been completed. Similar step-by-step procedures for steady-state and transient testing of the ALCO/BLH steam generator are approximately one-half done.

During this year, a number of additional standard operating procedures were written. A new plant description was written, and a table of the design specifications giving the operating limits of major equipment and of the various systems

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

has been compiled. All of these documents are part of the SCTI operations manual, which has been in preparation for two years, and is now essentially complete.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

Performance of the step-by-step preoperational test program made manifest a number of engineering and construction oversights which, if left uncorrected, would have prevented proper operation or adequate control of the plant. Action required to rectify these problems absorbed considerable time, but was considered necessary. It should be recognized that the purpose of the preoperational test program was to uncover these very conditions during system checkout and initial operation. Successful conclusion of all of the preoperational tests has rendered the plant ready for experimental operations.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Sodium Component Development	
Project: Installation of ALCO Steam Generator	
Project Engineer: J. A. Leppard	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-31
General Order: 7593	Subaccount: 4125

I. PROJECT OBJECTIVES

The project objective is to install the ALCO steam generator, design and effect plant modifications, and complete the modular steam generator installation design package.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

All construction work required for placement of the ALCO steam generator, i. e., installation of steam and sodium piping, placement of heaters, thermocouples, and instruments, and installation of associated electrical gear was completed. Subsequent to installation of the steam generator, the connecting piping and the steam generator were hydrostatically tested. A gas chromatograph, installed to monitor steam generator cover gas for hydrogen, was put into operation.

Concurrently with the preoperational test program (reported under 1322), a design and construction review was conducted to locate and correct plant discrepancies. As a result of these studies, several plant modifications were made, and all construction work connected with it has been finished. Included with these modifications were the following.

- 1) Provision of additional screen gates between the cooling tower and main circulating pump to permit cleaning the screens without shutting down the circulating water system
- 2) Installation of a jib crane adjacent to the cooling tower to facilitate handling of screens, pump parts, or other heavy equipment
- 3) Addition of sample holders for metallurgical determination of impurities in the sodium system
- 4) Change in location and in placement of additional steam and feed water sampling lines
- 5) Addition of several relief valves on the high-pressure steam piping required to meet ASME code
- 6) Addition of a cold-trap system with connections to both primary and secondary sodium systems completed with a plugging meter and an in-line sodium sampling station for each loop

A larger (7500-gal.) tank was purchased and installed for use as a primary fill/drain tank. The original primary tank was substituted for the secondary tank.

Design of the modular steam generator installation package was completed. Some of the structural steel required for this installation was placed in position, and much of the equipment required was purchased and placed in storage.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

With completion of the ALCO steam generator installation and plant modifications, and termination of all other construction work at the SCTI, the plant is finished. The project objectives have been satisfied.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Sodium Component Development

Project: Fabrication of Modular Steam Generator

Project Engineer: D. T. Eggen

Reporting Period: Fiscal Year 1965

AEC Category: 04-01-04-31

General Order: 7593

Subaccount: 4121

I. PROJECT OBJECTIVES

The project objective is to design and fabricate a 37-tube, low-temperature evaporator and high-temperature superheater module capable of providing 2400 psi, 1050°F steam at the turbine throttle. The modules will be fabricated for installation and test in the SCTI. The necessary design work for installation of the modules in the SCTI will be completed, and the necessary equipment for installation of the modules in the SCTI will be ordered.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. FABRICATION

Fabrication of the two evaporator and two superheater modules were completed. Details of the fabrication are reported in NAA-SR-11410, "Fabrication of Modular Steam Generator for the SCTI." Details of the design are reported in NAA-SR-9826, "Final Design of Sodium Heated Modular Steam Generator for the SCTI." Figure 1 shows the two completed superheater units on their storage rack.

1. Tube-to-Tube Sheet Welds

The tube-to-tube sheet welding procedures developed during the design phase of this project for 2.25% Cr - 1% Mo, 5% Cr - 0.5% Mo, Type 347 and Type 304 stainless steel were used in fabrication of the modules.

The procedures for the tube-to-tube sheet welding were: (1) the tube to be welded was brought to the tube stub on the tube sheet; (2) the copper chill and external gas manifold were positioned aligning the tube-to-tube stub; (3) a ball of stainless steel wool was inserted well below the weld to avoid intermixing of inert gas and atmosphere; (4) the weld head was positioned in the tube sheet bore to be welded; (5) the gas lines, power cables, and power supply were checked; (6) the weld cycle was initiated; (7) upon completion of the entire weld cycle, the weld head and chill were removed; and (8) the ball of stainless steel wool was removed. This completed the weld.

Tube-to-sheet welding was accomplished by use of a weld head similar to the one developed earlier for this program. This head was an improved model, in that the drive motor was integrally mounted and the locating plate was simplified. Power was provided by the same power supply and programmer as used in the early development efforts. All tube-to-tube sheet welding was performed with the units in a vertical position. This was accomplished by installing the units in a steel-lined pit approximately 50 ft deep, in the assembly area floor.

7593-2501 -
Figure 1. Superheater Units on
Storage Rack

The procedure for setup was the same for both the evaporators (5% Cr - 0.5% Mo) and the superheaters (Type 321 stainless steel). The welding parameters were determined and the welding procedure specified for each alloy. The weld procedures specified the equipment to be used, the type of shielding gas, gas flow, electrode description, all of the automatic programming parameters and detailed step-by-step setup instructions, plus a sketch of the weld joint setup.

During the initial tube bundle assembly 15 tube-to-tube sheet welds were made on a mockup of the tube-sheet configuration. One mockup was made from 5% Cr - 0.5% Mo steel, the other from Type 321 stainless steel. Welds were made using the same weld procedures, respectively, as those to be used on the modules. The samples made of chrome-moly steel were postheat-treated by the same procedure as intended for that tube sheet. These samples were then submitted to Quality Control for x-ray dye penetrant, helium leak check, and finally metallographic examination.

Upon acceptance by Quality Control, the tube-to-tube sheet welding operations were started. Porosity and inclusion allowances were established by using a specially prepared comparison plate allowing approximately one-third the inclusions and/or pores of that allowed by Section VIII of the ASME Boiler and Pressure Vessel Code, for 1/4-in. plate material. Thinning, undercut, offset, or tungsten inclusions were not permissible. By welding from the middle row of tubes outward, radiography was performed upon

completion of every second row of welds. This permitted access to both rows of tubes. It also permitted dye penetrant inspection of the outside and inside of the welds.

a. Evaporator Units

Of the first 37 tube-to-tube sheet welds made, two were considered questionable because of pores or inclusions upon evaluation of the x-ray film. Both welds were rerun and reinspected and found to be within the limits of the specification. X-ray of the tube-to-tube sheet welds was done by double wall exposure. Shooting at a vertical angle of approximately 30° shows the image of the weld as an ellipse on the film. Two shots were made from opposite horizontal

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

angles so that the weld could be fully exposed on film. When an area was questionable, additional shots were made to clearly define that area. The x-ray machine enclosures were fabricated of lead, thus providing shielding for personnel to within 13 ft when the equipment was in operation.

After welding, the tube bundle was removed from the pit and the entire Cr-Mo tube sheet and welds placed in a furnace for postheat treatment. Such treatment of the 5% Cr - 0.5% Mo evaporator module was accomplished by heating at a maximum rate of 400° per hr to 1325 to 1375° F, holding for 1 hr and cooling at a rate of 250 to 500° F per hr to below 600° F. Heating was monitored by clamping six thermocouples in the tubes at the weld joints and four additional thermocouples randomly placed around the tube sheet. Temperatures were recorded on tape for the record. During heat treatment, the entire heated area was purged with argon gas.

The welds were cleaned, helium-leak tested, and dye-penetrant inspected. The tube bundle was assembled to the shell by placing the shell in the pit, then lowering the tube bundle into the shell in a vertical position. The tube ends were faced to equal lengths by means of a spot facing tool in a drill press mounted over the end of the bundle. The tube sheet was mounted in position for welding of the lower end. Welding and inspection both proceeded much more rapidly and without difficulty. Since the tubes were now fixed at the opposite end, welding and inspection required lateral springing of the tubes for operator access. This proved to be no obstacle and caused no damage to the tubes. Three sets of spacers, the 32-in. shell section, a cone reducer, and the lower cross were left off the assembly for this purpose, thus allowing over 60 in. of free tube length.

The first major discrepancy occurred on the second evaporator module. The 35th weld on the second end had been completed; a setup was made for the 36th weld. The weld head was erroneously replaced in the 35th position. Without the protection of the combination external chill and gas backup, the weld was burned through.

After evaluation of the damage, it was decided to attempt a salvage operation. The tube stub on the tube sheet was backfaced to remove all cast metal, approximately 1/8 in. Two inches of the tube end were removed. The then existing end of the tube was prepared for welding and a 2-1/8-in. splice section also prepared for welding. The splice section was welded to the tube sheet using the original weld procedure. The tube was welded to the splice piece using a modified weld procedure.

b. Superheater Unit

Later in making the tube-to-tube sheet welds on the superheaters, a short tube was accidentally installed in a tube bundle. This tube was not discovered until after it had been welded. Rather than make a splice on the opposite tube sheet, it appeared advisable to cut off the short tube and replace it with a tube of correct length. A new tube was inserted in the tube bundle without removing it from the pit. The tube stub on the tube sheet was refaced and prepared for welding. In welding, the weld joint was "blown" into the gas annulus of the external chill. Examination revealed that a guide "bullet" used in inserting the tube had not been removed, causing excessive internal gas pressure during the welding operation.

Salvage required reshaping of the tube stub to nearly its original configuration, using specially ground cutters and preparing a new splice section. This section was welded to the tube sheet using the original weld procedure. A modified procedure was used to weld the tube to the splice piece. To assure control of the internal gas flow, a small loosely welded ball of stainless steel wool is inserted several inches below the weld joint. At this weld, a second ball was left in the tube and again caused excessive gas pressure resulting in a "blown" weld. This weld was also removed and a second splice section prepared and welded in place without incident (see Figure 2).

7593-2502
Figure 2. Welded Tube
Splice

While a slight thinning in wall thickness was expected at the weld joints, most welds showed a slight reinforcement on the outside of the tube. This reinforcement was apparently due to a slight restraint exerted by the tooling in welding. Boroscopic examination and also checks with a plug gage showed the welds to be generally flush with the ID of the tubes.

2. Shell Welds and Assembly

Prior to any shell welding, several welders were certified for welding both 5% Cr - 0.5% Mo and Type 321 stainless steel under Section IX of the ASME Boiler and Pressure Vessel Code. Weld samples were prepared for both the shell section and the tube sheet to nozzle section. The samples of 5% Cr - 0.5% Mo were postheat-treated in accordance with the recommended heat cycles for the sample thicknesses. Additional samples were sectioned and examined by Quality Control and found satisfactory.

Initial welding was done on the upper shell sections without consumable inserts. Early welds required an unusual amount of rework. Procedures were changed to use a consumable insert in making the root pass, which helped considerably in controlling the "drop through" in the inside of the shell sections. Several different weld rods were used as filler; 508-18 rod produced the cleanest weld deposit. Later, as the welders gained experience, the rework required became consistently less.

Fewer difficulties were encountered in welding of the shell assemblies for the superheaters. This material was Type 304 stainless steel, on which alloy all welders had years of experience. Due to the limited clearance between the inside diameter of the shell and the tube bundle, control of "drop through" caused some problems. Weld shrinkage, particularly on the superheater shells, while within the tolerance, caused interference in the insertion of the spacer and support rod assembly into the shell. However, once the spacers passed the girth welds they were free and without interference from the shell.

In both the evaporator and superheater the design of the cone reducer caused the most difficulty. The weld joint being located at the break point on both ends of the reducer. X-ray film showed a sharp line at the edge of the weld. Grinding to blend caused thinning in shell section. Alignment of the joint often proved

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

difficult. Again, interpretation of the film caused much disagreement. Often this line was interpreted to be lack of fusion or undercut. Visual examination of the weld was often impossible in final stages of assembly because the areas in question were hidden by the tube bundle. The only solution was to remove the area of weld in question and reweld.

Postheat-treatment of the evaporator shell welds was done by coiling the area with electrically insulated copper wire, and wrapping with an insulation blanket. Each joint was monitored with calibrated thermocouples. Power was supplied with conventional welding power supplies. The heat cycle was manually controlled by varying the power output of the power supplies as required by the time-temperature cycle.

Review of the assembly operations of the first evaporator and the cost of inserting the tube bundle in the shell showed that this operation should be changed. The spacers and support rods were assembled and inserted into the center shell section. The diffusers, cone reducers, and fittings were added, except for the lower cross and split shell section. The tubes were now inserted. Assembly time was considerably reduced and radiography made easier, with less possibility of damage to the tubes.

B. HYDROSTATIC TEST

Preliminary planning included a hydrostatic test immediately after completion of the tube-to-tube sheet weld. It was intended to hold a closure plate over the tube sheet end by clamping to a collar below the neck of the tube sheet. The extreme torque required to seal the closure plate caused the collar to indent the sheet surface in the neck area. Rather than risk damage to the tube sheet, this method of test was abandoned.

After completion of the entire unit, plugs were welded in the nozzle extensions, the shell sealed with closure plates and hydrostatic-tested. The units were then rinsed and dried. Test of the tube-to-tube sheet welds was monitored by observing shell side pressure while the tube side was under test pressure.

Upon completion of the hydrostatic test and acceptance by the State Inspector, the outside of the evaporator modules were wire-brushed and given two coats of primer paint.

The evaporators were originally sealed by clamping closure plates over the shell nozzles and then coating them with a fiberglass epoxy. Movement and temperature changes caused these seals to leak. Therefore, a plate was fitted over the nozzles and fused to the weld preparation of the nozzle. The units were then again purged with dry gas.

C. EVAPORATOR MODULE OUTLET STREAM TEMPERATURE MONITORING THERMOCOUPLES

In order to provide maximum integrity to the tubes in the evaporator modules, the thermocouples which will be used to measure the individual tube outlet steam temperatures were clamped to the evaporator tubes. Calculations made indicated that the temperature errors which would be experienced due to the clamping of

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

the thermocouples would be 8°F or less. Since temperature changes of the order of 20 to 50°F would be expected if unequal flow-of-significance occurred, the thermocouple installation was considered adequate for determination of any flow instabilities.

D. FATIGUE TESTS

Fatigue tests on a module head were completed, and additional tests on standard bar type specimens comparing 2.25% Cr - 1% Mo and 5% Cr - 0.5% Mo, showed the latter to be superior. These tests are reported in NAA-SR-9827, "Fatigue Testing of 37-Hole Modular Steam Generator Head."

E. CONTROL STUDY

The control study using a variable node length model for the once-through steam generator was terminated early in the fiscal year due to a stability problem in the variable node length model. This problem has since been corrected by using a different method of solution on the analog computer. No attempt was made to rerun the control study due to the nearness of the project completion date. A summary of the work done relative to the control study and variable node length was documented.

F. INSTALLATION OF MODULES IN THE SCTI

The design drawings necessary for installation of the modules in the SCTI (based on the Modular Steam Generator being installed first) were completed.

The necessary equipment for installation of the modules in the SCTI has been received and stored.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The fabrication of the modules was completed with no major problems. The welding and x-ray inspection of the tube-to-tube sheet welds presented little or no difficulty. In the design of any future modular steam generator, consideration should be given to changing the core reducers to standard concentric reducers. This would allow welding and radiography in straight sections of equal diameter with no change in section. In any future tube-to-tube sheet welding operations where the weld configuration is dependent on the internal and external pressure of the shielding gas, a pressure control device should be utilized.

The report NAA-SR-9828, "Test Program for 2400 psi, 37 Tube, 20 Mwt, Modular Steam Generator" has been deferred to take advantage of information to be gained in the ALCO/BLH Steam Generator Test Program.

The project has met its objective of the design and fabrication of a sodium heated steam generator for high-quality steam. The modular steam generator concept with the related special manufacturing processes developed, appears to be economically competitive with other designs while providing the following.

- 1) Minimum possibility of caustic stress corrosion
- 2) Generation of 950 to 1050°F steam consistent with reliable performance

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

- 3) Possibility of generation of high pressure steam to exploit associated lower plant heat rates
- 4) Isolation and removal from service without major outage
- 5) Steam generator fabrication and installation schedules which will not govern the plant construction schedule.

Proof of performance of the modular steam generator has yet to be demonstrated, since the present schedule is for the modules to be tested in the SCTI after the ALCO steam generator test. With the modular steam generator being the second installation in the SCTI, additional design work will be required to modify the installation drawings. This added work will be given a separate subaccount.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Sodium Component Development	
Project: High-Temperature Strain Measurements	
Project Engineer: W. J. Hallett	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-31
General Order: 7593	Subaccount: 1328

I. PROJECT OBJECTIVES

The objective of this project is to develop suitable, reliable, high-temperature strain gages for experimental stress or strain measurements, or for application in process instrumentation components. Commercially available gages will be tested to determine their limits, applicability, and reliability as analytic instruments for use on component test loops and systems, and as sensing elements in pressure transducer and force-arm applications. Suitable high-temperature strain gages will be developed, as well as application techniques for high temperature use. Methods of accomplishing temperature compensation and sealing of the sensitive element will be investigated to permit use in contact with sodium or sodium vapor.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

Since inception of this project in March 1965, effort has been directed towards preparation of a detailed plan of action for the work effort, during which preparation of a limited number of pilot tests were performed on the more promising, commercially available electric resistance strain gages. Production facilities of strain gage manufacturers and vendors manufacturing special high-temperature strain gage wire were inspected in order to identify gaps in strain gage technology and manufacturing. The pilot evaluation includes the following.

- 1) Apparent strain versus temperature characteristics at 900, 1000, 1100, and 1200°F.
- 2) Stability (drift) versus time characteristics at 900, 1000, 1100, and 1200°F (for periods up to 6 mo before and after thermal shocking)
- 3) Repeatability characteristics at 1000 and 1200°F.
- 4) Variation in gage factor as a function in temperature.

Results from the pilot evaluations are not complete. However, it was clearly ascertained that gages from one lot of commercially available 1200°F electric resistance strain gages were found defective due to improperly welded joints between the grid and the lead tabs. Their stability was extremely poor. Gages of this same type, from the same manufacturer but from a different lot evaluated earlier, showed excellent stability with respect to drift, even at 1200°F. This indicates that the lack of strain gage reliability is probably often due to inadequate quality control. Also, inspection of the facilities and procedures of the vendor furnishing strain gage wire, of the type used in manufacturing these gages,

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

indicated a need for improvement in techniques for casting the bars from which the wire is drawn, and for maintaining more adequate quality control during the drawing operations.

Additional information from the pilot investigation, including evaluation of a full-bridge, 900°F sheathed type commercial gage for use in steam, etc, is currently being completed.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

On the basis of information obtained during this brief period, it is plain that there is too much lot-to-lot variation in commercial wire filament strain gages with respect to electrical characteristics and reliability for accurate or meaningful strain measurement at temperatures above 750°F. In part, due to the lack of sufficient quality control during fabrication of the gage, and in using alloys developed for manufacture of potentiometers or other less critical electric components.

The so-called metal-film or etched-foil gages, which have gained considerable popularity in the past several years, do not require making critical weld joints between the 0.001-diameter grid wire and the heavier lead tabs, since the grid and lead tabs are etched as a unit. In this respect, they are more predictable and reliable than the wire-wound gages which do require critical weld joints. Unfortunately, metal-film or etched-foil gages cannot be flame-spray bonded to the structure without damage to the gage. They require the use of ceramic cements which are more electrically conductive and more fragile as a bonding material than the aluminum oxide used in the flame spray process, and thus have been found inferior to flame-spray bonded electric resistance strain gages.

Procedures will be developed to make more reliable wire-filament strain gages, with particular emphasis on the development of better weld joints. More stable alloys will be used for gage materials, such as the Fe-Cr-Al alloys, which are not now being used in commercially available 1200°F wire strain gages. This special wire is already on hand as well as the fixtures for fabricating pilot gages for further evaluation. The state-of-the-art survey carried out several years ago will be updated. Specifications will be developed for procurement of strain gages with considerably improved performance and reliability at temperatures above 750°F, under steady-state and thermal shock conditions. With such gages, it should be possible to develop high-temperature nuclear components and accessory equipment having far greater structural reliability with respect to creep, fatigue, stress-rupture, etc, than is possible today.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Sodium Component Development

Project: Mechanisms in Sodium

Project Engineer: W.J. Hallett

Reporting Period: Fiscal Year 1965

General Order: 7593

AEC Category: 04-01-04-31

Subaccount: 1329

I. PROJECT OBJECTIVES

The objective of this project is to evaluate materials, configurations, and surface treatments for use in high-temperature liquid sodium or sodium vapor. Information generated in this project will be applicable to the designs of mechanisms such as control elements, handling equipment, and variable orifices which must operate in sodium cooled reactors.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

Testing of bellows at a stroke-to-compressed length ratio of 4:1 in 1200°F liquid sodium was reinitiated using test equipment and candidate bellows reclaimed from previous bellows work. A Belmetrics, 316 stainless steel bellows, which had previously been exercised in 1200°F sodium, was additionally tested and failed at 43,000 cycles (3-in. compression and extension stroke). The cycles attained represent 430% of design life. Preparations for testing an identical bellows in the test rig were nearly completed.

Bench testing of a motion-multiplying (4x) mechanism which consists of a 2-stage differential rack system was started. This mechanism contains no bearings as such, and shows great promise for applications where a long output stroke with a short input is desired, i. e., control elements and fuel-handling equipment. The test mechanism has been placed on continuous operation for cycling a 100-lb load over an 80-in. distance with a 20-in. input stroke. Cycling in this mode will continue until failure occurs, to provide basic information concerning problem areas associated with rack and pinion drives having motion ratios of 1:4. Approximately 4200 cycles have been logged by the test mechanism, with no sign of degradation.

Disassembly and inspection of a mechanism which utilized a sodium-vapor condensing and thermal barrier zone below a conventional rubber O-ring (rotary seal) were completed. This mechanism had been adapted to a vessel containing sodium, and the internals exposed to a pool of sodium at 1200°F for 5252 hr. The inspection revealed that the condensing plates (Figure 1) were very effective, in that no sodium vapor had reached the seal area, or the annulus between the rotary member and the housing. Internal to the rotary shaft were two actuator shafts (one within the other) which could be moved axially through a 4-in. stroke. The smaller shaft (3/8-in. diameter) was supported by the larger shaft (5/8-in. diameter) so that there was no relative motion between the two shafts when the larger was exercised. When the smaller shaft was raised and then lowered to its seat position, relative motion between shafts did occur. At disassembly, a narrow band of sodium compound was found to have been deposited within the shaft annuli. The locations corresponded to a level where the estimated temperature was 250°F.

7593-2503

7593-2504

Figure 1. Sodium Vapor Condensing Plates on Rotary Seal

Figure 2. Rotary Seal Actuator Shafts

Externally, the two shafts (Figure 2) exhibited distinct lines of demarcation in the area of the sodium deposit. Above the deposits, the shafts were clean and smooth. Below the deposits, there was light frosting for a short distance which diminished to discolored, but otherwise clean surfaces again. It was concluded, that in time, both shafts would have accumulated sufficient amounts of sodium compound to induce binding.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The excellent performance of the Belmetrics bellows alleviates to a great extent reservations concerning the use of bellows as seals in liquid sodium. Although this bellows did not duplicate the extraordinary performance of a similar bellows which had been tested during FY 1963, the 43,000 cycles reached is still a significant accomplishment. Test results from a third bellows of this type will provide additional data for the evaluation of bellows for use as seals in liquid sodium. Results from the inspection of the O-ring test mechanism indicate that moving parts in a sodium vapor environment will eventually become bound if a temperature gradient extending below the freezing point of sodium exists in the part-clearance spaces.

SECTION V
SODIUM-COOLED REACTOR ADVANCED DEVELOPMENT
CONTENTS

<u>Title</u>	<u>Identification</u>	<u>Page</u>
ADVANCED DEVELOPMENT OF SODIUM-COOLED REACTORS	Summary	V-1
CONTAMINANT REMOVAL TECHNIQUES	7641 - 1322	V-4
LARGE COMPONENT TEST LOOP	7641 - 1323	V-7
EVALUATION OF LONG TERM OPERATION ON SRE SODIUM SYSTEMS AND COMPONENTS	7641 - 1324	V-8
IMPROVEMENT OF METHODS FOR MAKING SODIUM-COOLED REACTOR-PHYSICS CALCULATIONS	7641 - 1331	V-16
SGR CRITICAL EXPERIMENT	7641 - 1332	V-18

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Advanced Development of Sodium-Cooled Reactors
Program Manager: D. T. Eggen
Reporting Period: Fiscal Year 1965

I. PROGRAM OBJECTIVES

The general objective of this work is the production of economically competitive power from sodium-cooled reactors. To accomplish this objective, the advanced development phase of the program investigates areas in sodium technology, fuels and materials, sodium reactor safety, and physics. Sodium heat transfer and reactor components are developed and tested to reduce costs, improve reliability, and increase the confidence level. Necessary research and development are provided to support advances in the field.

II. PROJECT MANAGER'S SUMMARY AND EVALUATION

The project to evaluate effects of long-term operation of components in the SRE primary system was completed. A significant contribution was made to sodium system technology by the detailed examination of a cold-trap, which demonstrated the removal of many coolant impurities in addition to oxygen and the reduction of fission-product concentrations to levels permitting unhindered maintenance. Inspection of the main intermediate heat exchanger showed that stainless steel components can withstand long exposure to high-temperature sodium without detrimental effects and provided valuable information about carbon mass transfer. In particular, it was demonstrated that carbon can be gleaned from a sodium system during normal reactor operation by hot-trapping and cold-trapping without the need for removal of components.

The project to investigate and develop techniques for monitoring and removing undesirable impurities in sodium (particularly carbon) was discontinued at the end of the first quarter. Useful information was obtained on sample preparation, analysis and in-vivo purification. The LCTL was shut down and placed on a standby basis at the end of the first quarter. The system was maintained as a service to other projects which utilized it during the year. At the beginning of the second quarter, the reactor physics projects were reoriented toward the fast reactor development program. The reactor safety projects were incorporated into the fast reactor program because of their direct application.

A. SODIUM TECHNOLOGY

The most valuable information obtained under this program resulted from the detailed examination of the SRE cold-trap. The results of cold-trap sample analyses established the cold-trap as an efficient process purification instrument capable of removal of many impurities besides oxygen. Detailed emission spectrometry results show evidence of cold-trapping 16 out of 22 traceable elements. Both emission spectrometry and carbon analysis reveal an axial impurity distribution through the trap similar to the Cs^{137} distribution found by gamma spectrometry. The results also indicate that Sr^{90} and Sb^{125} are collected by a different mechanism than cesium. Thus the trapping of

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

fission products is not solely a result of trapping carbon particles carrying the fission products. Any fission product released to the sodium by gross fuel failures, can be reduced by cold-trapping to concentrations sufficiently low for unhindered maintenance and system service activities. Efficient cold-trapping is obtained by establishing a thermal gradient through the trap. The reactor sodium, after seven years of service, remains acceptable for continued use.

The main intermediate heat exchanger was inspected during the disposal operation and was determined to be in good physical condition with no visible evidence of wear, buckling, or warpage. Welds on the tube sheet were dye-penetrant checked and found to be in excellent condition. Observations of the intermediate heat exchanger flow patterns have provided additional information, on the shortcomings of the existing baffle arrangement, which should be beneficial in future design considerations. Definite evidence was obtained showing the depletion of carbon sources in the hot-leg of the system and their deposition in the cold-leg. There are indications that the carbon solution rate and solubility levels in sodium are considerably lower than presently accepted values. The individual cylindrical boron-nickel rings on an SRE control rod were measured for swelling. Only the lower rings showed any diametral growth. This growth is a result of the (n, α) reaction. A maximum diameter growth of 0.5% was measured on the lowest poison ring. This ring had experienced an integrated thermal flux of 3.3×10^{20} nvt. The correlation between calculated thermal neutron dose and measured diameter growth was good.

Three pressure transducers from the primary system were found to be in good working condition. Valves removed from the SRE heat transfer system were cleaned and subjected to routine inspection. All were in good condition and were scheduled for reuse in the reactor. A swing checkvalve that did not operate was inspected. Larger clearances in the valve design would have improved the performance during normal operating conditions. More important, hot-leg installation would have promoted cleanup and restoration to service through the subsequent operation of purification equipment.

B. SODIUM REACTOR PHYSICS

Many of the methods that had been developed for sodium graphite reactor physics calculations were directly applicable to the study of fast reactors. Certain cross-section sets were already available and were used with multigroup diffusion codes developed in previous years. Improvements in the theory of resonance cross-sections led directly to development of the code TRIX which was used to calculate Doppler coefficients. Reactor physics calculation methods developed primarily for thermal sodium-cooled reactors became the base for early fast reactor calculation methods and indicated the requirements needed to make improvements in the data processing procedures.

The remaining area of work accomplished this fiscal year in the critical experimental program was the analysis and reporting of the intracell measurements which had been completed late the previous fiscal year. The analysis of these data completes the present experimental program for the study of sodium graphite reactors. This information, in conjunction with the previously obtained values of material bucklings and critical mass data, represents a considerable set of information on which to base a nuclear design effort. These data have

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

already been compared to analytical calculations wherever possible. The intra-cell parameters themselves indicate a need for improvements in the theoretical models used for reactor calculations. Any such changes made should consider the entire set of experimental data.

The program has also indicated where further improvements should be made in the experiments. These include the necessity for obtaining more accurate information on both the La^{140} fractional yields and the time-dependence of the gross fission product ratios. Further studies should be made of $1/v$ detectors and the effect of epithermal neutron attenuation in cadmium boxes.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program:	Advanced Development of Sodium-Cooled Reactors		
Project:	Contaminant Removal Techniques		
Project Engineer:	W. J. Hallett		
Reporting Period:	Fiscal Year 1965	AEC Category:	04-01-04-21
General Order:	7641	Subaccount:	1322

I. PROJECT OBJECTIVES

The general objective of this program is to investigate and develop techniques for monitoring and removing undesirable impurities which might enter the sodium coolant during reactor operation. Undesirable impurities are classified as those which adversely affect reactor construction materials and fission products that are either potentially hazardous to the public or make reactor maintenance difficult. A specific objective is to determine the process by which carbon (in sodium) is rendered capable of carburizing the structural materials.

Evaluation of the effect of the presence of carbon (and other impurities) and the performance of purification techniques depend not only upon the sensitivity with which concentrations can be measured but also on the ability to obtain representative samples from a system for analyses. Thus a second specific objective is the development of suitable sampling and handling techniques.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

Two aspects of the area of contaminant removal from sodium were studied: the effects of additives to sodium upon the carburizing capability of the carbon in sodium, and a review of the results of limited operation of a new packless cold-trap for oxide removal.

A set of light capsule tests were performed* in which it was found that gettering of oxides by zirconium did not affect the degree of carburization of stainless steel by graphite in the massive (not powdered) form. Further, it was found that carburization of stainless steel did not occur in the presence of excess silicon carbide in sodium gettered by zirconium. The sodium used for these tests contained approximately 150 ppm of calcium and 75 ppm of silicon.

A second set of capsule tests were performed using sodium previously treated to remove carbon capable of carburizing stainless steel. In this set of experiments, 50 gm of pretreated sodium and stainless steel test coupons were placed in four aluminized stainless steel capsules. A zirconium tab was added to getter the oxides. In these capsules, four gm of 270-mesh graphite, eight 1/4-in. cubes of graphite, one 1/2-in. cube of graphite, and a control sample containing no additives were subjected to a temperature of 1200°F for 100 hr. The analysis of the test coupons showed no significant carburization except on the one taken from the capsule containing the 270-mesh graphite.

*NAA-SR-10501, p III-4

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Initial continuous operation of a packless cold-trap, installed in the LCTL, took place during the third quarter of the fiscal year. This trap was essentially a pair of vertical concentric pipes with stilling baffles at the bottom to collect oxides. Provision existed at the bottom to drain the accumulated material. The trap was put in service to purify the LCTL sodium prior to its use in a series of instrumentation tests for another liquid metal program. During operation, the flow was maintained at 70 lb/min, and the trap contained approximately 500 lb of sodium.

The cold-trap was operated in two stages: purification of a 36,000-lb batch of 1000°F sodium, and the subsequent treatment of an additional 41,700 lb of 540°F sodium. Prior to initiating the first cold-trapping operation, the total sodium inventory in the LCTL was thoroughly mixed by circulating it through the system by pumping. A plugging meter reading indicated the oxide concentration to be 270 ppm, corresponding to 21.1 lb of oxide present. The sodium inventory was then divided, 41,700 lb being placed in the LCTL core tank and the balance put in the supply tank for cold-trapping.

7641-2501
Figure 1. Reduction of Oxide in
36,000 lb of Sodium

7641-2502
Figure 2. Reduction of Oxide in
77,600 lb of Sodium

During the first run, based on plugging meter readings, the initial oxide concentration of 270 ppm was reduced to 200 ppm after 75 hr of operation (see Figure 1). Thus the oxide initially present in the 36,500-lb batch (36,000 lb in the supply tank and 500 lb in the cold-trap) was reduced from 9.8 to 7.2 lb in the supply tank, with 2.7 lb in the cold-trap. At the end of an additional 73-hr operation at 70 lb/min, the oxide concentration (as indicated by plugging meter) was further reduced to 78 ppm, which corresponded to 2.8 lb of oxide in the sodium and 7.1 lb of oxide in the cold-trap.

At the end of the first run, the remaining 41,700 lb of sodium was admitted to the system. Since this sodium had 270 ppm oxide, it brought 11.3 lb of oxide with it. The combined inventory of 78,200 lb of sodium therefore contained 14.1 lb of oxide, which corresponded to a concentration of 180 ppm. After 20 hr of trapping at 70 lb/min (see Figure 2), the oxide concentration was reduced to 93 ppm or 7.2 lb of oxide in the sodium with 13.9 lb in the trap. After 27 hr additional trapping, the oxide level was 35 ppm (2.7 lb oxide in the sodium and 18.4 lb in the trap).

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

As this run continued, the plugging temperatures became so low as to be unreliable for estimating oxide levels.

III. EVALUATION OF PROJECT

The limited amount of work done on this project (resulting from its cancellation at the end of the first quarter) has shown that reasonably consistent data can be obtained when measurements of sodium impurities are made at the higher levels of concentration. By avoiding, for the present at least, those impurity levels where large measurement uncertainties exist and concentrating upon those where general agreement can be obtained between analyses, these two benefits accrue: (1) increased confidence in measurements made in increments toward lower levels of concentrations, and (2) better explanations of the behavior of the impurities in sodium toward materials of construction.

The data obtained from the capsule tests suggest a method for screening those impurities of concern and eventually pointing up a method for determining acceptable (in terms of materials of construction) concentration levels of impurities. The surface effects of these materials (graphite) upon carburization in the presence of other elements also indicate possible methods for controlling the reaction that may occur between graphite and sodium.

Although considerable information was accumulated by operation of a packless cold-trap, complete data analysis was not possible. The cold-trapping rate at a flow of 70 lb/hr appeared to be independent of oxide concentration over the range of 50 to 200 ppm; and the amount of oxide collected, up to 20 lb in the 500-lb trap. Even though a "tailing-off" in the oxide removal rate was observed (Figure 2), it cannot be concluded that the trap was filled, but possibly that an equilibrium was attained between the oxide concentration in the incoming sodium and the solubility of oxides present in the trap.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Advanced Development of Sodium-Cooled Reactors	
Project: Large Component Test Loop	
Project Engineer: W.J. Hallett	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-21
General Order: 7641	Subaccount: 1323

I. PROJECT OBJECTIVES

The project objective is to operate and maintain the Large Component Test Loop (LCTL) as a service to other projects requiring experiments in this system.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A 2- by 3-in. mechanical freeze-seal pump, surplus from the SRE, was re-built for use in the LCTL. The pump was installed in the transfer line in the drain tank pit, replacing a failed EM pump.

Maintenance work was conducted on the facility to maintain an operable test system.

A large air compressor was procured for the LSGR calandria core maintenance test (7519-5210) conducted at the LCTL. Piping was installed from this compressor to the tower deck where core maintenance work was performed.

Graphite handling tests were conducted at the LCTL facility as reported by 7519-5210.

The LCTL facility was utilized by the S8DS program (7568-3457) to test and calibrate two EM pumps and three magnetic flowmeters. These tests were conducted over a range of temperatures from 300 to 1300°F.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

As of October 1, 1964, the LCTL test facility was shut down and placed on a standby basis until certain experiments were performed. Activities were limited to maintenance necessary for protection of the system, and to periodic checking to ensure that the preheat and inert cover gas is maintained.

The LCTL remains available as a service to other projects having need to use this rather large and versatile experimental system. Modifications to the system made to date are expected to reduce maintenance requirements and to improve operating characteristics.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Advanced Development Sodium Cooled Reactors	
Project: Evaluation of Long-Term Operation on SRE Sodium Systems and Components	
Project Engineer: J. E. Owens	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-21
General Order: 7641	Subaccount: 1324

I. PROJECT OBJECTIVES

The objective of this project is to evaluate the condition of equipment removed from the SRE during the Power Expansion Program. Equipment is investigated to obtain information for improving the design and operation of future sodium systems. Specific components being studied are: (1) cold-traps for removal of sodium impurities and collection of fission products; (2) the main intermediate heat exchanger for warpage and wear from temperature and flow changes; (3) control rods for the effect of a high radiation field and high temperature on dimensions; and (4) other components such as valves and pressure transducers, for effects of long-term exposure to high-temperature sodium.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. COLD-TRAP

One cold-trap was successfully sampled. Its maximum surface radiation intensity was sufficiently low (≈ 7 r/hr) to permit sampling work to be performed outside the hot-cell. Sample borings were obtained by cutting through shrouds, fins and shell with a 5/8-in.-hole saw and hammering a sharpened 304 SS tube into the cold-trap deposit. A total of nine borings were obtained at the locations shown in Figure 1. Each boring was divided into approximately 10 samples for analysis.

The results of carbon analysis, gamma spectrometry, and emission spectrometry of samples removed from the cold-trap are listed in Table 1. Results obtained from sodium samples removed from the reactor system while the cold-trap was in operation are also listed for comparison. These are quite consistent with values obtained on the sample taken from the exit plenum from the cold-trap. Table 1 also shows a calculated "Collection Ratio" which is the ratio of the impurity concentration in the deposit in the cold-trap to that in the sodium external to the trap.

Emission spectrometry did not yield true collection ratios for the cold-trap. In several cases, the upper limit (~ 500 ppm) failed to give maximum impurity concentrations, and the lower limits of detection frequently did not allow determination of true concentrations in the coolant. However, it is significant that the detailed emission spectrometry results (Figure 2) show evidence of cold-trapping 16 out of 22 traceable elements.

As a first approximation, both emission spectrometry and carbon analysis reveal an axial impurity distribution through the trap similar to the Cs^{137} distribution found by gamma spectrometry. Radiation surveys of all eight

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

7641-2503

Figure 1. Cold-Trap Sample Locations

TABLE 1
ANALYSIS OF IMPURITIES IN SRE COLD-TRAP

Impurities	Location and Quantity		Collection Ratio*
	Cold-Trap	Coolant	
Carbon	144 to 1550 ppm	18 to 60 ppm	77
	γ and β spectrometry		
Cs ¹³⁷	$4.0 \times 10^2 \mu\text{c/g}$	$15 \times 10^{-2} \mu\text{c/g}$	2.7×10^4
Sb ¹²⁵	$4.3 \mu\text{c/g}$	$0.6^3 \times 10^{-2} \mu\text{c/g}$	6.8×10^2
	Emission spectrometry		
Fr	200 to >500 ppm	50 ppm	>10
Si	200 to >500 ppm	50 ppm	>10
Mn	50 to 500 ppm	>5 ppm	>100
Pb	5 to >500 ppm	10 ppm	>50
Cr	5 to >500 ppm	5 ppm	<100
Ni	10 to 300 ppm	5 ppm	60

*Comparison between impurity levels in cold-trap and in Na coolant

NAA-SR-11450

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Element	Sample No.				
	5N3	5N1	4N3	4N1	9N2
Ag	<5	<5	<5	<5	<5
Al	10	<10	25	<10	<10
B	25	75	<10	<10	<10
Ba	<25	<25	<25	<25	<25
Be	2	2	<1	<1	<1
Bi	<5	<5	<5	<5	<5
Ca	<25	<25	<25	<25	<25
Cd	<2	<2	<2	<2	<2
Co	25	<25	<25	<25	<25
Cr	>500	60	10	5	<5
Cu	200	75	50	50	10
Fe	>500	>500	200	>500	35
Mg	200	200	35	35	<10
Mn	300	500	50	60	<5
Mo	35	<10	<10	<10	<10
Ni	300	75	10	10	<10
Pb	>500	200	<5	100	<5
Si	>500	>500	200	200	10
Sn	10	<5	<5	<5	<5
Ti	50	50	<10	<10	<10
V	50	75	10	5	<10
Zr	<25	<25	<25	<25	<25

7641-2504

Figure 2. Emission Spectrography of SRE Cold-Trap Samples
(The upper limit for impurity determination by emission spectrometry with the existing laboratory equipment is 500 ppm)

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

cold-traps exposed in the SRE show an activity pattern of the same form, thus reflecting similar buildup of impurities in all the expended cold-traps. In contrast, the beta activity shows a different pattern. Although beta spectrometry was not performed on samples removed from the cold-trap, radiation surveys were performed on the samples at the time they were removed from the cold-trap, which gave both total beta and total gamma activity. The gamma activity agreed well with the results obtained later in the laboratory. The beta activity varied from approximately eight times the gamma activity at the bottom of the cold-trap to about the same as the gamma activity at the top. The principal beta emitter is probably Sr^{90} , which is one of the few beta emitter isotopes still active after the long cooling period. The varying ratio of beta activity to gamma activity resembles the varying ratio of antimony to cesium activity found along the cold-trap axis. Both results indicate that the beta emitter and antimony are collected by a different mechanism than cesium. The collection ratio for Cs^{137} was found to be higher than 10^4 compared with a collection ratio of ~100 for carbon. The different order of magnitude between the Cs^{137} and carbon collection ratios demonstrates that the trapping of fission products is not solely a result of trapping carbon particles carrying the fission products.

The large deposits found in the upper part of the first mesh section indicates that a filtering action takes place on the processed coolant. Available temperature data show that the temperature gradient across the cold-trap is split, with 2/3 of the temperature drop across the economizer section and 1/3 across the steel mesh trapping section. If the cold-trap is excessively cooled, the processed sodium will reach oxide saturation temperature (plugging temperature) in the economizer section (the upper plenum), and impurity separation will start. With no place to settle, the solids will be carried to the first section of the steel mesh, where it will lodge. This is the location where large gamma activity has been observed in all traps. Operational experience has proven that a new cold-trap will freeze off after a few hundred hours of operation if the coolant plugging temperature is high (~400°F), and the interval cold-trap temperature is kept low (250°F). Flow through the cold-trap may be restored by heating the trap until the oxide plug has been redistributed. The results lead to the conclusion that efficient cold-trapping is obtained by establishing a thermal gradient through the trap so that oxide saturation temperature is achieved only in the mesh section of the cold-trap. For the same cooling capacity this method of operation will allow processing of a much larger sodium flow.

B. HEAT EXCHANGER

The main intermediate heat exchanger (MIHX) was inspected during the disposal program. Apparent distortions were found adjacent to most of the major welds. However, pictures taken during initial installation showed the same deformations, proving the distortions developed during fabrication rather than during subsequent operation.

The entire shell was removed in an upper and a lower section. Internal inspections of the shell revealed three welding flaws, each consisting of a void in a weld seam penetrating ~50% through to the outer surface. There was no indication that additional deterioration had taken place around the holes during the seven years of operation.

7641-2505

Figure 3. Steamlines Visible in the Stain on the Upper Half of the Heat Exchanger Shell

distributed in the system, power operation would gradually exhaust carbon sources in the hot-leg and deposit them in the cold-leg.

The distinct stream lines evident in the stain indicate insufficient mixing of the sodium during normal operation. This results in agreement with conclusions reached from analysis of its performance during Core I operation. Measured temperature distributions at various flow rates formed the basis for that work. More overlap between baffles or baffles located with less spacing (including the curved section of the heat exchanger) would have improved the cooling mixing on the shell side of the heat exchanger. The hot-leg tube sheet was steam-cleaned and decontaminated. All welds were dye-penetrant checked and found to be in excellent condition. All tubes in the heat exchanger were checked. None was warped, bowed, or crushed. No plugging was found except in the final 18 in. of the cold-leg tubes adjacent to the tube sheet. Approximately 8% of the tubes were partially or completely plugged. The pressure of carbonaceous material in the tubes was traced by radiation surveys prior to removal of the heat exchanger from the system.

Radiation surveys made of the MIHX in 1960 and 1961 were correlated with the radiation survey made in 1964 after the upper shell of the heat exchanger was removed (see Figure 4). The larger intensity measured in October 1964 compared with December 1961 is a result of draining the heat exchanger and removing the upper shell prior to the latest survey. (The first five surveys were taken with sodium circulating in the primary system and with the secondary system drained.) The point of maximum activity shows a progressive shift

Oxide deposits which followed the stream lines of the coolant were found on the upper shell. Steam cleaning removed the oxides, but the stream lines remained visible as dark stains on the shell surface (Figure 3). The stain was heaviest in the cold-leg and became lighter in the hot-leg of the heat exchanger. Its consistency was like lamp-black. Slight rubbing removed the stain and disclosed a shiny metal surface below. This observation verified the increase of carbon solubility in sodium with temperature; and indicates that even if all carbon deposits initially were evenly

7641-2506

Figure 4. Radiation Intensity Along Cold Leg of MIHX

toward the cold end with increasing time of operation, indicating dissolution of the carbon deposits on the hot side. Thus, continued operation of the system resulted in the gradual removal of carbon deposits.

C. CONTROL ROD

Control rod No. 3 was examined in the hot-cell. Visual inspections and diametral measurements were made of all 18 boron-nickel rings. The upper rings all appeared like new. The bottom two rings appeared rusty brown. No blistering or other surface damage was observed. Only the lower rings and especially the bottom ring showed any diametral growth. This growth is a result of the (n, α) reaction which occurs in a boron control rod where an atom of helium and an atom of lithium is formed from a B^{10} atom for each neutron absorbed. Figure 5 shows the correlation between calculated thermal neutron dose and measured diameter growth. The listed nvt is only valid for the surface layer of the control ring. The large boron absorption cross-section causes a rapid attenuation of thermal flux below the surface. At a depth of 0.1 in., it has decreased to 10% of the surface intensity.

The relatively larger growth at the lower end of the control rod is related to the rod geometry. Additional thermal flux enters through the end surface and through the inner surface of the control rod. The expected growth rate for the lower poison ring neglecting end-effects is shown in Figure 3. It is estimated that the end-effects doubles the flux in the lower half of the first poison ring. The results show a reasonable proportionality between this estimate and diameter growth.

7641-2507

Figure 5. Correlation Between Neutron Dose and Diameter Growth of Control Rod

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

D. PRESSURE TRANSDUCERS

Three pressure transducers from the primary system were checked for reliability of performance and physical integrity after service in the reactor system since 1957. Two of the transducers apparently had never been in direct contact with the sodium. They were mounted in a vertical cavity, and gas trapped in the cavity transferred the sodium pressure to the pressure-sensitive membrane.

All three pressure transducers performed well on the test bench, showing linear response between applied pressure and output signal, with an accuracy of $\pm 2\%$ and with good reproducibility. The two transducers which had been operating in a gas atmosphere performed well from the first test. The one which had been in direct contact with sodium required the removal of sodium and sodium oxide from the pressure-sensitive membrane before its performance equaled the two others.

E. VALVES

Valves removed from the SRE heat transfer system were cleaned and subjected to routine inspection. All were in good condition and were scheduled for reuse in the reactor. The plug and the seat of five valves were carefully examined for corrosion or erosion, but neither condition was evident.

A system throttle valve (V-609) was leak-checked and the leak rate was found to be less than that for a new valve. This valve had operated at outlet temperature in throttled or partially throttled condition for extended periods.

A swing-check valve, which locked in the open position shortly after initial reactor operation, was subjected to an extensive examination. No corrosion or erosion of flapper or valve seat was found. A black carbonaceous deposit was found between the shaft surface and support bushings. In addition, it was found that the support bushings were not aligned. As a result, the shaft could not pivot freely in the bushings, but the flapper was turning around the shaft. Larger clearances in the valve design would have improved the performance during normal operation conditions. However, during the SRE exposure conditions the performance would have been only temporarily improved until a heavier deposit of carbonaceous material had built up. The check valve was located in the cold-leg of the system. If it had been located in the hot-leg of the system, hot-trapping and cold-trapping would gradually have exhausted the carbonaceous deposits in the bushing area and made the valve operable.

A topical report "The Effect of Long-Term Operation on SRE Sodium System Components," NAA-SR-11396, has been written giving a complete description of the work concerned.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The objectives of the program were fulfilled, and it is believed that the project has made a significant contribution to sodium system technology.

The most valuable information obtained under this program resulted from the detailed examination of the cold-trap. This work has established the effectiveness of the cold-trap for removal of many coolant impurities other than

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

sodium oxide. It has resulted in a better understanding of trap performance and capabilities which should permit even more effective utilization of this equipment in future sodium purification service. It has shown that fission products, released to the sodium by gross fuel failures, can be reduced, by cold-trapping, to concentrations sufficiently low for unhindered maintenance and system service activities. The same type of capability has been demonstrated for other large and varied contaminant inputs so that the reactor sodium, after seven years of service, remains acceptable for continued use.

This effort also has provided valuable information about carbon mass transfer in a sodium system operating with large temperature gradients. Definite evidence was obtained showing the depletion of carbon sources in the hot-leg of the system and their deposition in the cold-leg. The slow rate at which this process took place, and the relatively limited removal of carbon in the cold-trap may be indicative that the carbon solution rate and solubility levels in sodium are considerably lower than presently accepted values.

The component investigations have established the validity of component designs and of the fabrication techniques used in their manufacture. These investigations also have provided additional evidence of the compatibility of austenitic steel with liquid sodium for long-term service at temperatures to 1000°F. The components studied were shown to be free of major mechanical and operational deficiencies, but several areas were noted where improvements can be realized in future designs and installations. For example, the reliability of the check-valve operation could have been improved by increased tolerances on the moving member. More important, hot-leg installation would have lessened the likelihood of formation of the deposits which eventually interfered with its operation; and, in the event of deposit formation, would have promoted cleanup and restoration to service through the subsequent operation of purification equipment. Similarly, observations of the intermediate heat exchanger flow patterns have provided additional information on the shortcomings of the existing baffle arrangement, which should be beneficial in future design considerations.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program:	Advanced Development of Sodium-Cooled Reactors		
Project:	Improvement of Methods for Making Sodium-Cooled Reactor Physics Calculations		
Project Engineer:	H. A. Morowitz		
Reporting Period:	Fiscal Year 1965	AEC Category:	04-01-04-21
General Order:	7641	Subaccount:	1331

I. PROJECT OBJECTIVES

The general objective of this project is to develop reactor physics calculational techniques and codes for reactivity, neutron economy, and critical mass for sodium graphite reactors that are efficient, reliable, and accurate. Specific objectives include improvement in the methods for calculating lattice parameters, use of multigroup methods, and further development of the Monte Carlo method. Special emphasis is placed on resonance capture, epithermal fission, neutron spectrum, and flux distribution in lattice cells having a high degree of heterogeneity. Comparison will be made between calculated results and experimental measurements.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

At the start of the second quarter, the project objectives were reoriented toward the fast reactor program. The present report covers only the activities of the project as it was originally oriented.

Reports entitled "Thermal Neutron Spectrum Effects on Calculations for Graphite Lattices," NAA-SR-7892 and "Multigroup Calculation Methods with Application to the SGR Critical Experiment," NAA-SR-10531 (previously NAA-SR-Memo-7891) were issued. The effort to obtain from other laboratories the graphite scattering kernel of D. E. Parks for use with the THERMOS neutron thermalization code at elevated graphite temperatures was not successful.

Work was started on a FORTRAN Monte Carlo code for calculating the thermal flux distribution in lattice cells having complicated geometry. Specifications were given for the input data format and the output edit. A study was made of the stochastic procedures used in the free gas model for neutron thermalization so that thermalization routines available from other codes can be adequately evaluated for our use in the code being developed. The activity was terminated before the writing of the new code was started.

A review of the heterogeneous method (small-source theory) for making lattice calculations was started, in order to assess the utility of this method in connection with reactor calculations for the ASCR program. The central problem is determination of the fuel rod parameters, η^* , λ , and A^T . These quantities are analogous to the quantities η , f , and p , respectively, which appear in the 4-factor formula. The rod parameters are part of the input data for the code which calculates reactivities and power distributions. The KERNMAT code* was used for the reactivity calculations. Epithermal fission is not explicitly provided for in this code, so considerable effort was directed toward evaluation of approximate

*S. A. Raje, "KERNMAT. A Fortran code for the Heterogeneous Method of Reactor Calculations," CVNA-107 (September 1961)

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

Since the fast reactor program was initially directed toward study of coupled reactor systems, many methods that had been developed for sodium graphite reactors were directly applicable. Certain cross-section sets were already available and were used with multigroup diffusion codes developed in previous years. Improvements in the theory of resonance cross-sections led directly to development of the code TRIX which was used to calculate Doppler coefficients. Reactor physics calculation methods developed primarily for thermal sodium-cooled reactors became the base for early fast reactor calculation methods, and indicated the requirements needed to make improvements in the data processing procedures.

Studies performed during previous fiscal years have demonstrated the capabilities of the Monte Carlo method. The present effort was directed toward the construction of a FORTRAN code for making thermal flux calculations. This code would be easy to use, whereas the one presently available (RBU) is not. A satisfactory start was made toward development of the new code.

The heterogeneous method (small-source theory) studies with the KERNMAT code were not advanced to a point where reliable conclusions could be drawn. However, there were indications that some of the approximations that are made in KERNMAT would somewhat limit its usefulness for ASGR calculations. Nevertheless, it is clear that the heterogeneous method is a very powerful tool for the evaluation of certain effects in lattice type reactors. A code less restrictive in its theoretical basis than KERNMAT would be desirable.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Program: Advanced Development of Sodium-Cooled Reactors

Project: SGR Critical Experiment

Project Engineer: H. A. Morowitz

Reporting Period: Fiscal Year 1965

AEC Category: 04-01-04-21

General Order: 7641

Subaccount: 1332

I. PROJECT OBJECTIVES

The objective of the ASGR critical experimental program is to provide, at the earliest possible date, engineering physics information which is directly applicable to the latest ASGR concepts. This information will relate to both the optimization of proposed reactor designs and to suggested changes in future loadings of reactors currently in operation. Emphasis is placed on measurements which will implement the calculational techniques in use and provide additional information in areas where analytical means are presently inadequate. Theoretical support is included in this project to correlate the calculational techniques with the results obtained from the experimental measurements. Specific objectives include measurements of: (a) critical mass; (b) material buckling; (c) absorption cross-sections; (d) control rod investigation, including both reactivity worths and detailed flux mapping; (e) power distribution measurements within lattice cells; and (f) intracell measurements to provide information relating to resonance capture in U^{238} , fast fission factor, thermal utilization, and cell disadvantage factors.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

Lattices having the same moderator-to-fuel ratio as the proposed ASGR core have been studied, in addition to two other lattice spacings which bracket the design pitch. The center-to-center triangular fuel element spacings was 9.020, 11.015, and 12.056 in. The fuel elements were 18-rod, 1/2-in.-diameter, 3.02-wt% enriched uranium monocarbide fuel rods arranged in 2 concentric rings. Both wet and dry control rod concepts have been studied for each lattice pitch by placing sodium filled or empty 4-in.-diameter SS tubes in every fourth process channel where the other channels contain UC fuel elements. The following arrangements have been studied:

- 1) "Closed lattice," all channels filled with fuel elements;
- 2) "Open lattice with sodium," every fourth channel containing a sodium-filled, SS tube, others filled with UC fuel elements; and
- 3) "Open lattice with SS," every fourth channel containing an empty SS tube, others filled with UC fuel elements.

Intracell measurements, which were completed the previous fiscal year, consisted of determinations of the following quantities.

$$f = \frac{\text{thermal absorption rate in the fuel}}{\text{total thermal absorption rate}}$$

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

$$\delta^{28} = \frac{U^{238} \text{ fission rate}}{U^{235} \text{ fission rate}}$$

$$\delta^{25} = \frac{\text{epicadmium } U^{235} \text{ fission rate}}{\text{subcadmium } U^{235} \text{ fission rate}}$$

$$\rho^{28} = \frac{\text{epicadmium capture rate in } U^{238}}{\text{subcadmium capture rate in } U^{238}}$$

Data obtained from these measurements have been analyzed and the results are listed in the table.

The measurements of δ^{28} were made both by counting the La^{140} activity, a fission product common to both U^{235} and U^{238} , and by counting the gross fission products of each isotope. All measurements of δ^{28} were consistently higher than any corresponding calculation, regardless of the degree of sophistication of the calculation. The difference between measurement and 2-group calculations averaged $\approx 2.5\% \Delta k$, while the difference reduced to 0.6% or 0.7% Δk using 15 groups with fast flux weighting, in each instance the calculations giving the smaller reactivity. The difference in the two experimental techniques represents a difference of only $\approx 0.1\% \Delta k$. This measurement is discussed in full in NAA-SR-MEMO-11329, " δ^{28} Measurements in Uranium-Monocarbide and Graphite Lattices."

The thermal utilization measurements, f , were determined by the 2-foil technique using U^{235} foils to determine the U^{235} absorptions in the fuel, and a 1/v detector to determine absorptions in U^{238} and other 1/v materials. Good statistics were obtained for all values, the 9-in. closed lattice which had a small cadmium ratio giving rise, however, to considerable uncertainty in the thermal flux. Other values differed from calculation by $\approx 1\% \Delta k$. Careful measurements of this type should have their main limitation in the accuracy of the cross-sections used. The measurements of f and δ^{25} are reported in NAA-SR-MEMO-11313, "Measurements of Thermal Utilization (f) and δ^{25} in SGR Uranium-Monocarbide Lattices."

The values of δ^{25} were determined directly from data obtained for the f measurements. Measurement and theory are in agreement for the 11-in. closed lattice, but the measurements are $\approx 6\%$ smaller than calculated at both the 9- and 12-in. pitches, indicating that the general slope of the δ^{25} curve versus moderator-to-fuel ratio cannot yet be predicted.

Two techniques were used for the measurement of ρ^{28} . The cadmium ratio technique involves the use of bare and cadmium-covered U^{238} detector foils in the fuel. The 1/v subtraction technique eliminates the use of a cadmium-covered U^{238} detector foil in the fuel by using a cadmium-covered 1/v detector foil such as manganese, and normalizing with measurements made in a region having a Maxwellian neutron flux distribution. This reduces errors inherent in the cadmium ratio method of measurement. The measurements of ρ^{28} have been hampered by poor counting statistics obtained from the β - γ coincidence counting. Results using the 1/v technique are also dependent on the choice of 1/v detector, and may result because of slight variation from being a true 1/v detector, and/or

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

because of uncertainty in the epithermal neutron attenuation factors of cadmium for different reactor materials. These existing uncertainties will require further experimentation to eliminate them so that results are not being issued in a report at this time.

Two papers were given at the 1964 Winter Meeting of the American Nuclear Society. They were Part I-Experiment and Part II-Theory of "Material Bucklings of Uranium-Carbide Fueled Graphite Assemblies, "

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

Analysis of the intracell parameter data completes the present experimental program for the study of sodium graphite reactors. This information, in conjunction with the previously obtained values of material bucklings and critical mass data, represents a considerable set of information on which to base a nuclear design effort. These data have already been compared with analytical calculations wherever possible. The intracell parameters themselves indicate a need for improvements in the theoretical models used for reactor calculations, particularly in the case of δ^{28} . Any such changes made should consider the entire set of experimental data, since corrections for the different parameters may have compensating effects on the calculation of k_{eff} . Even though the clean core k_{eff} might be adequately calculated with compensating errors in the theory, the reactor properties over its lifetime cannot be predicted until all the intracell parameters are handled correctly.

The program has also indicated where further improvements should be made in the experiments. The measurements of δ^{28} have pointed out the necessity for obtaining more accurate information on both the La^{140} fractional yields and the time-dependence of the gross fission product ratios. These measurements would have been a logical extension of this program. The measurements of ρ^{28} indicate that further studies should be made of $1/v$ detectors and the effect of epithermal neutron attenuation in cadmium boxes used with these foils.

TABLE
SGR LATTICE PARAMETERS

Lattice	Thermal Utilization		ρ^{28}			δ^{28}			δ^{25}	
	Exp	Theory	1/v Method	Cadmium Ratio Method	Multi-group Theory	La^{140} Method	Gross Fission Product Count	Multi-group Theory	Exp	Theory
<u>9 in.</u>										
Closed	0.870 ± 0.015	0.842	7.3	3.9	5.15	0.077 ± 0.0016	0.072 ± 0.0038	0.0674	0.386	0.411
Open with SS	0.835 ± 0.005	-	3.8	3.0	3.80	0.073 ± 0.0015	0.067 ± 0.0034	-	0.303	-
<u>11 in.</u>										
Closed	0.832 ± 0.001	0.823	3.6	3.3	3.05	0.070 ± 0.0014	0.063 ± 0.0033	0.0559	0.246	0.246
Open with SS	0.786 ± 0.004	0.796	4.1	2.6	2.3	0.065 ± 0.0013	0.063 ± 0.0032	-	0.189	-
Open with Na	0.767 ± 0.002	0.762	3.2	2.7	-	0.069 ± 0.0014	0.064 ± 0.0034	-	0.205	-
<u>12 in.</u>										
Closed	0.824 ± 0.002	0.812	-	-	-	0.066 ± 0.0010	0.060 ± 0.0031	0.0538	0.189	0.200
Open with SS	0.755 ± 0.004	-	-	-	-	0.064 ± 0.0013	0.059 ± 0.0032	-	0.152	-

SECTION VI
HNPF-PNPF FUELS
CONTENTS

<u>Title</u>	<u>Identification</u>	<u>Page</u>
HNPF FUEL SURVEILLANCE	Summary	VI-1
HALLAM ALLOY FUEL ELEMENT SURVEILLANCE AND EXAMINATION	7617 - 1910	VI-2
HNPF CORE-I UC FUEL ELEMENT SURVEILLANCE AND EXAMINATION	7617 - 1912	VI-7
HNPF CORE-II UC FUEL ELEMENT SURVEILLANCE	7617 - 1914	VI-9
PNPF FUEL ELEMENT EXAMINATION	7615 - 1970	VI-11
PNPF CORE-II FUEL ELEMENT DEVELOPMENT	7629 - 1910	VI-18
PNPF CORE-II DESIGN	7629 - 1930	VI-22

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: HNPF Fuel Surveillance
Program Manager: B. R. Hayward
Reporting Period: Fiscal Year 1965

I. PROGRAM OBJECTIVES

The purpose of this program is to evaluate the performance of fuel elements in the Hallam reactor. Equipment, methods, and procedures will be provided to perform pre- and postirradiation examinations of fuel elements and fuel element components. The surveillance of the fuel operating parameters is to be incorporated into the performance evaluation.

II. PROJECT ENGINEER'S SUMMARY EVALUATION

As a result of the reactor shutdown early in the second quarter, the technical scope of this fuel surveillance program was reduced. The revised program included several visual examinations but no destructive ones. Based on the successful SU-9 irradiation of U - 10 Mo at HNPF conditions in SRE, the schedule for both NDT and DT examinations was extended to 3000 Mwd/MTU peak burnup for U - 10 Mo and 5000 Mwd/MTU for UC elements. The present peak burnup on U - 10 Mo is 2680 Mwd/MTU and 2035 Mwd/MTU on UC.

The washing procedures for U - 10 Mo fuel elements were demonstrated and one improvement added to the method to remove minor residues. The successful removal of either unwashed or washed process tubes was a major milestone. The dimensional analyses of the process tubes showed no change within the original specifications. The visually examined undamaged U - 10 Mo and UC fuel elements were all in excellent condition. These qualitative data verify the adequacy of the design. All Hallam Core I fuel elements have performed as planned.

The fuel surveillance program has assumed the responsibility for correlating, evaluating, and defining the current status of all Core I fuel elements. Various handling procedures on specific elements have resulted in some hardware failures. The storage atmosphere is an important factor. Most elements will be stored unwashed. Criteria for reinsertion are being developed.

The success of the SU-9 irradiation demonstrated the value of a large-scale lead experiment. This test substantially reduced the number of expensive destructive examinations on the Core I fuel. A similar test series on UC was initiated under the LSGR program. The two long-rod irradiations, NRX-86-1 and -2, have reached ~ 10,000 Mwd/MTU by the end of FY 1965. These two tests are being continued under this HNPF fuel surveillance program in FY 1966, due to the completion of all LSGR work at the end of FY 1965. These tests are operating well at approximately HNPF conditions and should provide valuable lead information on the Core I and Core II UC elements. These long-rod results coupled with the highly successful AI-3-11 hypo UC capsule results to 32,000 Mwd/MTU peak burnup show that the Core I and Core II hypo UC should easily meet or exceed the planned performance.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: HNPF Fuel Surveillance	
Project: Hallam Alloy Fuel Element Surveillance and Examination	
Project Engineer: B. R. Hayward	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-21
General Order: 7617	Subaccount: 1910

I. PROJECT OBJECTIVES

The primary objective of this project is to evaluate the in-pile performance of the HNPF Core I U - 10 wt % Mo fuel elements. This objective is to be accomplished by:

- 1) In-pile surveillance of the fuel elements, and
- 2) Performing nondestructive and destructive examinations of selected fuel elements at specific burnup levels.

Intercomparisons of these results will provide the basis for prediction of the burnup capability of the U - 10 Mo core elements in the HNPF.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. SURVEILLANCE

The highest exposure U - 10 Mo fuel element has an average accumulated burnup of 1322 Mwd/MTU and a peak burnup of 2680 Mwd/MTU. This peak is calculated by using a 1.14 peak-to-average flux radially across the element and 1.8 peak-to-average flux axially prior to August 1, 1964, and 1.7 peak-to-average axial flux subsequent to August 1, 1964. The U - 10 Mo element containing pre-irradiation measured fuel, and which has been tentatively selected for the first postirradiation examination (MF31 from core position 26), has achieved an average burnup of 1134 Mwd/MTU and a peak of 2297 Mwd/MTU. Fuel element performance appeared satisfactory up to the date of the reactor shutdown for moderator element maintenance.

After removal from the core, the temperature of the thermocoupled U - 10 Mo Core I fuel element was monitored in the storage cell. The maximum temperature measured was 247°F. This temperature compared closely with that predicted from temperature curves for an element with 2000 Mwd/MTU peak burnup, accumulated at a low burnup rate and with 2 mo of cooling time.

An investigation was conducted to determine and recommend the proper storage conditions for the HNPF Core I fuel elements, removed from the reactor, to assure minimum damage to the fuel rod cladding. The fuel elements were originally removed and placed in the storage cells unwashed for temporary storage. These cells were evacuated and backfilled with helium prior to insertion of the fuel elements. It was recommended that the temporary storage conditions be continued, since sodium by itself is not damaging to the cladding.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Moisture is required to react with the sodium to form sodium hydroxide. Whereas, with cleaned elements, moisture without sodium can be instrumental in intergranular corrosion with sensitized 304 SS. However, it was further recommended that each storage cell atmosphere be checked for the presence of moisture, just prior to reinsertion of the fuel element into the core, to assure that the elements had remained dry. The fuel element from any cell found to contain moisture would be examined. A detailed history of each fuel element handling operation has been recorded as part of the criteria to be reviewed prior to reinsertion.

B. POSTIRRADIATION EXAMINATION

The initial checkout of the nondestructive test equipment installed in the Hallam Maintenance Cell was conducted during the first quarter with the performance of a mock NDT examination. A full-size dummy fuel element with solid stainless steel fuel rods was used for this mock examination. The element was inserted into the maintenance cell, the process tube removed, and the dummy fuel bundle inserted into the fuel bundle cutting fixture. A fuel rod was then cut out of the bundle, transferred to the fuel rod stage, and fuel rod diameter measurements taken.

During FY 1965, several of the Hallam Core I fuel elements were visually examined in the HNPF maintenance cell, among them MF-107, MF-66, GF-4, MF-32, and MF-161.

MF-107, a U - 10 Mo fuel element, was examined to determine the cause of its stuck orifice drive. This element was first subjected to four wash cycles (steam only), after which the orifice drive operated freely. The fuel element was then examined in the maintenance cell to determine the cleanliness of the fuel bundle, and to measure the diameter of the Zr process tube. This tube was measured every foot at orientation of 0 and 90°. The maximum variation in diameter at any elevation was 0.005 in. The diameters measured varied from 4.414 to 4.431 in. which compares favorably with the drawing specification of 4.400 plus or minus 0.030 in.

The process tube was then removed from MF-107 and the fuel element was visually examined. The element appeared to be clean except for a few light salt deposits (Na_2O or NaOH) around some of the spacers, also a few crystalline deposits (NaOH) adjacent to or under the spacers. The original Zr process tube was reinstalled, and the element subjected to a 10-min water rinse followed by a 30-min steam cleaning cycle to remove the residue, after which it was returned to storage.

The U - 10 Mo fuel element with the highest accumulated burnup, MF-66, was inserted into the maintenance cell in the unwashed condition to determine if the process tube could be removed from an unwashed fuel element. Although the process tube retaining pins were full of sodium, they could be turned and depressed to disconnect the process tube. A force of 600 lb was required to separate the fuel element and process tube. When first separated, the surface between the process tube and the upper hanger casting had the bright luster of metallic sodium which quickly oxidized (turned white) in the 3% oxygen atmosphere of the cell. The fuel element was covered with a thin white coating of

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

sodium oxide from approximately the middle of the hanger rod on down. A thick layer of sodium was observed in the end hardware, and between the fuel rods and spacers near the ends of the element.

To check efficiency of the water rinse step in the cleaning process, the process tube was reinstalled on MF-66 and the element was subjected to four 30-min steam cycles, followed by a 10-min water rinse. Due to a malfunction in the wash cell equipment, a 30-min steam cycle, scheduled to follow the water rinse, was not given to this element. The element was returned to the maintenance cell and the process tube removed. Visual examination of the element did not reveal any defects in fuel rods or fuel element hardware. The element was very clean and no salt or crystalline deposits were found. The process tube was reinstalled, and the element returned to storage.

Figure 1 shows the appearance of an unwashed fuel element in the vicinity of a fuel rod spacer. Figure 2 shows the typical residue remaining on a fuel element which has been subjected to four 30-min steam cycles. Figure 3 shows the cleanliness of a fuel element given four 30-min steam cycles and a 10-min water rinse.

7617-2501

Figure 1. Unwashed
Fuel Element

7617-2502

Figure 2. Residue on
Steam-Cleaned Element

7617-2503

Figure 3. Steam-Cleaned and
Washed Element

The two Core I U - 10 Mo fuel elements, MF-32 and MF-161, removed from the reactor without their process tubes, were visually examined in the maintenance cell (unwashed) and appeared to be in good condition. These elements were then washed and a process tube was installed onto MF-32; however, the process tube on the MF-161 fuel bundle bound up and could not be installed. Elements MF-32, MF-66, and MF-107 are equipped with process tubes and at the present time appear to be sound and fit for reinsertion into the reactor core. Element MF-161 is now stored without its process tube and its reuse is questionable.

Sixteen fuel element hanger rods have been cut off to permit rotation of the HNPF loading face shield. A seventeenth fuel element, MF-117, is also still in the core. This element was dropped 13 in., and pulled free of the hanger rod.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

C. FUEL ROD SHIPPING CASK

The fuel rod shipping cask, for shipping irradiated fuel rods from the HNPF to the CDHC, was received and an operational checkout was performed. The cask checkout included installation of the cask support assembly on the maintenance cell plug, and drilling and tapping of the holes necessary to secure the support assembly in place. The valve assembly was then installed in the cask support assembly. A mock cask loading was performed.

D. POSTIRRADIATION EXAMINATION OF SU-9

The postirradiation of SU-9, the U - 10 Mo-fueled HNPF Prototype element irradiated in the SRE, was completed and a report was issued. Examination of the variable orifice control device revealed that the failure of the orifice assembly occurred in the connection between the cable from the orifice bulb and the control drive rod located near the top of the shield plug. The failure was apparently due to improper swaging of the rod to the cable, as the cable had pulled free.

Metallographic examination of the fuel shows a nearly fully transformed material in the top section of the element, which operated at higher average temperatures and lower fission rates. Near the center of the element, which underwent higher fission rates and higher centerline temperatures, a transition type of structure was observed with the center portion having a lamellar structure of the transformation product ($\alpha + \gamma'$), and the peripheral portions having a single-phase (γ) structure. The bottom portion of the element closely resembles the as-cast structure of unhomogenous gamma phase, indicative of lower operating temperatures.

Examination of the cladding reveals that it is in a highly sensitized state with carbides precipitated at the grain boundaries and along the slip lines. Evidence of some carburization is shown from chemical analysis which gave bulk carbon contents as follows: near top of element, 0.21%; near center of element, 0.13%; and near bottom of element, 0.17%. These results compare with an as-fabricated carbon content of less than 0.08%. The cladding showed no indication of a case-type carburization; however, sensitization and carburization were fairly uniform across the specimens at the grain boundaries and slip lines.

The metallography of the fuel and clad was the principal effort in FY 1965. Previous reports showed that U - 10 Mo expected performance was ~6000 Mwd/MTU peak burnup.

III. EVALUATION OF EFFORT FOR FISCAL YEAR 1965

The Core I fuel element performance up to the time of the reactor shutdown remained satisfactory. The fuel elements removed from the reactor are being stored in helium-filled storage cells as they come out of the core, unwashed. The exceptions are those elements washed for visual examination under this program, and the elements which remain in the reactor.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

The postirradiation examinations of Core I U - 10 Mo fuel elements scheduled for FY 1965 have been postponed, due to the reactor shutdown for moderator element maintenance. The initial examination was scheduled to be performed at a peak burnup of 3000 Mwd/MTU; and the peak burnup obtained was 2700 Mwd/MTU. The initial fuel element examination will probably be conducted prior to reactor startup following moderator can maintenance, to confirm that the fuel is still in satisfactory condition and fuel element hardware and fuel rod cladding have not been damaged by storage.

The visual examinations conducted in the HNPF maintenance cell have shown that (1) process tubes could be removed and replaced on both unwashed and washed irradiated fuel elements; (2) to remove all sodium residue from a fuel element, the steam cleaning should be followed by a water rinse; and (3) visually the fuel rods and fuel element hardware were sound.

The checkout of the NDT equipment was plagued with a series of equipment problems, both with the NDT equipment and with the basic maintenance cell equipment. The NDT equipment malfunctions were corrected; however, the nondestructive test equipment has since been removed to make room for the moderator element examination equipment. The final checkout of the NDT equipment will be performed when this equipment is reinstalled.

The fuel rod shipping cask and accessory equipment operated well and appeared to be well built. Final analysis of the shielding will be made when irradiated fuel rods are inserted into the cask.

It is necessary to carefully evaluate all fuel element operational procedures during the removal from the core, examination, and storage. This fuel surveillance program bears the responsibility for determining the acceptance for reinsertion criteria for all Core I fuel elements. Development of procedures and methods will be required.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: HNPF Fuel Surveillance	
Project: HNPF Core I UC Fuel Element Surveillance and Examination	
Project Engineer: B. R. Hayward	
Reporting Period: Fiscal Year 1965	AEC Category: 04-01-04-21
General Order: 7617	Subaccount: 1912

I. PROJECT OBJECTIVES

The primary objective of this project is to evaluate the in-pile performance of 10 UC fuel elements which were inserted in HNPF Core I. This objective will be accomplished by: (a) surveillance of the in-pile performance of the 10 elements, and (b) performing nondestructive and destructive postirradiation examinations at various burnup levels.

Comparison of these results will provide a basis for predicting the irradiation behavior of UC fuel elements in an operating sodium-graphite reactor.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. SURVEILLANCE

The operating limits for the UC fuel elements in the HNPF Core I were revised to permit operation at up to 1800°F. The highest burnup UC fuel element has an average burnup of 1150 Mwd/MTU and 2035 Mwd/MTU peak. The peak center temperature reached, at a reactor power level of 250 Mwt, was calculated to be 1690°F. Performance of the UC fuel elements continued to appear satisfactory right up to the shutdown for moderator element maintenance.

Temperatures of the thermocoupled UC fuel element in the storage cell were monitored to provide a check on our "afterglow" heat calculations. The resultant peak reading of 275°F central fuel temperature, after 60 days cooling for an element with 2,000 Mwd/MTU burnup accrued at a low burnup rate, compared favorably with the temperature taken from predicted temperature curves for this burnup.

B. POSTIRRADIATION EXAMINATION

During preparation for moderator can examination, one UC fuel element (CF-4) was pulled from the reactor core without its process tube when subjected to a force of 3800/lb during removal. This fuel element was visually examined in the maintenance cell, in the unwashed condition, to check for possible damage. The bottom three spacers were badly bent and appeared to be pressing into the fuel cladding, and two of the rods had been badly scraped near the bottom. The Na₂O residue had been scraped off to expose the cladding surface, of which did not appear to have been penetrated. Because of these damaged spacers no attempt was made to replace the process tube, which was lost when that portion

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

of it above the retaining pins was torn out. This element will not be returned to the core.

The UC fuel elements have been stored in the unwashed condition and in a dry helium atmosphere. The storage cells were evacuated and then backfilled with helium. The safe storage of the 0.010-in.-thick, 304 stainless steel cladding is dependent upon the dry and inert atmosphere of the storage cell being maintained. The atmosphere of all the storage cells will be sampled before the fuel elements are reinserted into the core.

The initial postirradiation examination of an HNPF UC fuel element is scheduled to be performed prior to insertion of the first Core II UC fuel elements, or at 5,000 Mwd/MTu peak burnup. This initial examination is tentatively scheduled for FY 1966. The specification for the destructive examination of these elements has been completed.

A similar surveillance effort will be made on all Core I UC elements prior to their reinsertion into the core. The procedures and plan for the potential washing of UC elements, if deemed necessary, require further effort.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: HNPF Fuel Surveillance

Project: HNPF Core II UC Fuel Element Surveillance

Project Engineer: B. R. Hayward

Reporting Period: Fiscal Year 1965

AEC Category: 04-01-04-21

General Order: 7617

Subaccount: 1914

I. PROJECT OBJECTIVES

The objective of this project is to analyze and evaluate the performance of the HNPF Core II UC fuel elements with regard to fuel and cladding material stability, mechanical design of the fuel rod, and the performance of fuel element hardware. This project parallels the surveillance programs that are being performed for the HNPF Core I Fuel elements (both U - 10 Mo and UC). The objective will be accomplished by: (a) performing preirradiation measurements on selected Core II UC fuel material and cladding material, (b) surveillance of in-pile performance of selected elements, and (c) performing destructive and nondestructive postirradiation examinations of the aforementioned elements at various burnup levels.

Data obtained from this program will be utilized to predict Hallam HNPF Core II fuel burnup and useful fuel element life.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

This project, initiated during the final quarter of FY 1964, is directed primarily at objective (a) listed above during FY 1965. This project is being carried out in conjunction with the parallel to the fabrication of Hallam Core II.

The fabrication and checkout of a profilometer, developed for fuel slug diameter measurements, was completed. The profilometer consists of a stage to hold and move the fuel slug through a pair of opposed transducers which measure any variations from a standard diameter, and a strip chart recorder which records these variations.

A total of 984 fuel slugs were collected under the statistical sampling plan initiated for this program. Each sample was then cut to the next lower whole-inch length and subjected to the following.

- 1) dimensional measurements (diameter and length)
- 2) photography
- 3) gammagraphy
- 4) vacuum outgassing
- 5) assembly into 24 groups, each 159 in. long, to make up the fuel rods

The remaining UC segment from each of the sample fuel slugs was used for density measurements, and saved as archive specimens.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

The archive specimens were individually wrapped in aluminum foil, with the identification number marked on the foil, and encapsulated in evacuated glass tubes which have been packaged in 30-gal. drums and are stored in the vault.

The preirradiation measured fuel slugs have been fabricated into 24 fuel rods specially numbered V-1 through V-24. Each of these fuel rods has been dimensionally analyzed. Rods V-1 through V-8 have been shipped to Hallam and incorporated two rods each into Core II fuel elements CF 453 through CF 456. The two rods in each element are diametrically opposed. The preirradiation measurement effort of this project has been completed.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

Due to the extended shutdown of the Hallam Nuclear Power Facility for major repairs, only objective (a), the preirradiation measurements, has been initiated. The in-pile surveillance and postirradiation examination will not be initiated until after the HNPF is operating and the Core II Fuel elements are inserted.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program:	Piqua Technical Assistance Program		
Project:	PNPF Fuel Element Examination		
Project Engineer:	S. O. Arneson		
Reporting Period:	Fiscal Year 1965	AEC Category:	04-02-02-01
General Order:	7615	Subaccount:	1970

I. PROJECT OBJECTIVES

The objectives of this project are to examine and evaluate the performance of irradiated Piqua fuel elements. Cylinders in ten fuel elements have been given extensive preirradiation measurements and inspection. Systematic data will be obtained from the postirradiation examination of these cylinders with regard to film formation on the heat transfer surfaces, fuel swelling due to irradiation, and interdiffusion of the aluminum, nickel, and uranium alloy. These data will be analyzed and evaluated as a primary basis for establishing the performance capabilities of the PNPF Core I fuel elements.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. EXAMINATION OF PIQUA FUEL ELEMENT P-1071

The examination of the first fuel evaluation element from Piqua Core I was completed. This element was removed from the reactor after it had attained peak and average burnups of 1400 and 920 Mwd/MTU respectively. The results obtained from this examination were highly encouraging with respect to film formation and fuel growth.

The fuel elements consist of a uranium alloy clad with aluminum. The alloy is composed of uranium enriched to 1.94 ± 0.03 wt % in U^{235} . The alloy contains 3.5 wt % molybdenum, and 0.1 wt % aluminum as a stabilizing element. The aluminum cladding, which has a finned surface to increase the heat transfer area, is metallurgically bonded to the uranium fuel using an intermediate diffusion barrier of nickel 1.0 mils thick. The fuel element is shown schematically in Figure 1.

The film on the heat transfer surfaces of the fuel element was very thin (~ 0.14 mils, maximum), uniform and tightly adherent. It was barely detectable at the upper and lower ends of the fuel column and there was no buildup of film deposit in any of the crevices of the heat transfer surfaces. This film was similar to the films observed on OMRE fuel elements in that the inorganic components of the coolant were concentrated by a factor of $\sim 10^4$ in the surface deposits relative to their concentration in the coolant. The observation that the film (~ 0.5 mil thick) on the outside of the outer process tube was approximately ten times thicker than typical of the film on the heat-transfer surfaces indicated that the film formation was a direct function of the flow velocity of the organic coolant passing by the surfaces exposed to the coolant.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

7617-2504

Figure 1. Piqua Core I Fuel Elements

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

The valve plunger at the bottom of the inner process tube was frozen in the up or closed position and all of material in the inner process tube did not have a chance to drain out. Waxy material removed from the top of the inner process tube was a mixture of HB-40 and p-terphenyl. Some black particles in this waxy material were probably the result of excessive radiation damage to the coolant in the low-flow regions of the valve at the bottom of the element. These black particles probably caused the valve plunger to stick in the closed position.

Clad fuel cylinder dimensional change data showed maximum outer diameter increases of 0.39% on the outer cylinders and 0.37% on the inner cylinders with length increases up to 0.29% on the outer cylinders and 0.25% on the inner cylinders. Metallographic examination of the nickel diffusion barrier between the fuel alloy and the aluminum cladding revealed that the metallurgical bond had maintained its integrity in all the specimens examined, and that the unreacted nickel layer was always in excess of 1.0 mil thick, the minimum acceptable thickness, as fabricated. The results of the postirradiation examination of this fuel element have been published in NAA-SR-Memo-11019.

B. EXAMINATION OF PIQUA FUEL ELEMENT P-1091

The second evaluation element was scheduled to be removed after an average burnup of approximately 1700 Mwd/MTU had been attained. However, as the time approached for the second examination, another element, P-1091 in core position F-13, began to experience an increase in channel coolant temperature rise. Although this element was not one of the ten premeasured elements, it was selected for removal and examination because of the interest in the temperature rise. Most of the other information sought on evaluation elements could be obtained as well from a nonmeasured as from a premeasured element, namely, (1) general condition, (2) film formation, (3) nickel diffusion bond condition, and (4) fuel growth measurements. The fuel growth information, while reduced in accuracy from that possible by examination of a premeasured element, could nevertheless be determined with reasonable accuracy by statistical means.

When removed from the reactor, this element had reached peak and average burnup levels of 2600 and 1750 Mwd/MTU respectively, as calculated from thermal performance. The fuel element as received at the Atomics International Hot Laboratory was found to be in good mechanical condition.

The outer surface of the outer process tube had a black, loosely adhering deposit near the top, with most of the rest of the process tube appearing shiny, apparently where the film had come off. Diameter measurements on the outer process tube showed no difference between the pre- and postirradiation values. Temperature measurements on the surface of the outer process during examination ranged from 170 to 205°F. A relatively continuous, thin black film was observed on the inside surface of the outer process tube, appearing to be thickest in the lower half, corresponding to the higher temperature region.

Examination of the fuel cylinders showed a very thin, continuous film on the cylinders which had operated at lower temperatures and had been exposed to lower flux levels. The cylinders nearer the center of the fuel column also had

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

a thin film but it had blistered or peeled away from the aluminum cladding to form hollow blisters, which in some areas extended out into the coolant channel approximately 0.030 in. None of the coolant channels was blocked, and no carbonaceous growths were observed. The film generally was very brittle and was fairly easily removed with a nylon brush.

The film thickness followed the flux profile of the fuel element and ranged from an average low of 0.0004 in. to an average high of 0.001 in. The density of the film increased from inlet to outlet end from 1.4 to 1.5 g/cm³, while the ash content ranged from 5 to 10%, following no general pattern. The carbon-to-hydrogen ratios (~2) of the surface films were similar to those of films found on OMRE fuel elements. The film formation rate on this element was an average of 0.58 mils per operating year, as compared with an average of 0.11 mils per operating year for Element P-1071.

A visual examination of the inner process tube showed a relatively thin, discontinuous film over most of the outer surface, some of which apparently was scraped off when the inner process tube was removed from the fuel column. The inner process tube was nearly full of solidified organic which had not completely drained. When this solidified organic was melted and poured out of the inner process tube, it was found that there were small black particles dispersed throughout the entire length. These black particles represented approximately 5 wt % of all the material in the inner process tube. Some particles were as large as 2.5 x 1 x 0.3 cm, while a large number were 1 x 1 x 0.5 cm, and smaller. Some of the pieces were flat on one side, indicating that they had formed next to the inner process tube wall, while others were equally rough on both sides, indicating that there had been no wall contact. This material was nonmagnetic, and did not melt at temperatures up to 410°C. The density was 1.46 g/cm³ and the ash content was 0.14 wt % (a factor of ~50 less than surface film ash).

A sample of the coolant taken from the top section of the inner process tube was analyzed for high boilers (HB) to see if afterglow irradiation of coolant caused any significant increase in the HB content. The HB content was 6.5%, which agreed (within the accuracy of the method) with the HB content of PNPf coolant (6.8%) when the fuel element was removed from the reactor. This indicated no detectable change in decomposition products in the coolant by post-operating irradiation. This also indicated that the coolant in the inner process tube was being replaced by fresh coolant while the reactor was operating.

The plunger in the valve body at the bottom of the inner process tube was found to be stuck in the closed position. The drain hole in the side of the valve body was only partially open showing a very slow drainage of organic. The valve body was cut in two and found to be filled with a very hard, but porous black material. Some coolant was also found in the disassembled valve which further supports the evidence that the tube was not completely plugged.

Chemical analyses of the material removed from the valve body showed that it was similar to the black particles dispersed in the coolant inside of the inner process tube. It is also possible that this same material was the source of the black particles found in the coolant at the PNPf since they have comparable ash content.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

The observations associated with the inner process tube of the fuel element clearly point to a mechanical design problem with the drain valve from this tube. During reactor operation, this drain valve is closed, and allows the stagnant coolant to decompose in the region immediately above the valve. The decomposed material accumulates until it plugs the small coolant orifice through which the organic normally flows out of the inner process tube during reactor operation. The situation can easily be corrected on all newly inserted fuel elements by modification of the inner process tube drain valve.

Dimensional measurements on all of the clad fuel cylinders were completed and a statistical analysis of the data was performed to determine the magnitude of fuel growth. Since this element was not one of the ten premeasured elements fabricated for the Piqua fuel surveillance program, it was necessary to infer what the preirradiation dimensions must have been based on average values taken for the ten premeasured elements. These average values differed slightly from the normal specification values. For example, the nominal cladding OD of the inner cylinder is 4.096 in., with a tolerance of ± 0.014 in., whereas the average of 114 measurements on 38 premeasured cylinders is 4.087 in., with a standard deviation of ± 0.003 in. The maximum outer diameter change on the clad outer cylinders was +0.72%, and it can be said that at the 95% confidence level the change was within the range of +0.60 to +0.84%. The maximum outer diameter change of the clad inner cylinders was +0.61%, and the 95% confidence range is +0.46 to +0.73%.

The aluminum cladding and the nickel diffusion barrier were stripped from the U-Mo-Al alloy cylinders 2 and 3, and bare fuel cylinder measurements were completed. The maximum outer diameter change on the bare fuel was +0.60%, with a 95% confidence range of 0.56% to +0.64%. The maximum inner diameter change was +0.95% with a 95% confidence range of +0.81 to +1.02%, while the maximum length change was +0.50% with a 95% confidence range of +0.46 to +0.53%. The results, in general, are consistent with the clad fuel data. It was noted, however, that the outer diameter changes were slightly lower, and the inner diameter changes were slightly greater on the bare fuel than on the clad fuel. In the case of the outer diameters, the slight discrepancies are thought to be the result of measurement technique. It was easier to align the pi-tape over the bare fuel cylinder than over the finned surface of the clad fuel cylinder. Any misalignment would result in larger values for the outer diameters. In the case of the inner diameters, the slight discrepancies are thought to be due to a partial unbonding of the aluminum, from the inner surface of the fuel cylinder. This separation would account for the smaller inner diameter increases on the clad fuel cylinders. The outer diameter and length changes in general follow the burnup profile. The fuel growth per unit burnup was approximately that observed on Element P-1071, 0.21% $\Delta D/D$ per 1000 Mwd/MTU. The fuel growth data for Elements P-1071 and P-1091 are shown graphically in Figure 2.

A total of 18 specimens were core-drilled from the fuel cylinders for metallographic, burnup, and density specimens. On five of these specimens the aluminum had separated from the inside surface of the fuel, and on one specimen the aluminum separated from the outside surface of the fuel. This type of phenomenon is not considered to be of any great concern, especially on the inside surface, since it is thought that the aluminum would rebond, both mechanically

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Figure 2. Dimensional Increase and Burnup for
Piqua Fuel Element

and metallurgically, if the element would have been brought back up to temperature because of the aluminum's very low strength properties and higher thermal coefficient of expansion. This separation probably did not occur in-pile since the U-Ni intermetallic phase width was not in excess of what was expected. If the aluminum had separated from the fuel in-pile, the surface of the fuel cylinder would have operated at a substantially higher temperature, resulting in a greater U-Ni interdiffusion.

The metallurgical examination of the fuel specimens showed that on all specimens examined, there was at least 0.88 mils of unreacted nickel remaining. In general, the nickel consumed was found to be consistent with what would have been expected for equivalent time at temperature for this system out-of-pile, with no excessive nickel depletion noted on any specimen examined.

In summary, neither the small fuel swelling nor film deposits were sufficient to reduce the flow area enough to account for even one-fourth of the observed increase in outlet coolant temperature. The reason for the observed coolant outlet temperature increase is believed to be due primarily to a substantial increase in surface roughness, associated with the film deposits. Such a surface roughness increase would result in a decreased fuel element flow rate, giving rise to the observed temperature increase.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

The results of the postirradiation examination of this fuel element have been published in NAA-SR-TDR-11342.

Fuel disposal containers have been designed for use in shipping the disassembled fuel cylinders to the disposal agency following storage in the reactor pool at Piqua. Two of these containers have been fabricated for the elements examined thus far.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The postirradiation examinations of both fuel elements have been highly encouraging from the standpoint of fuel growth and surface film formations. The fuel growth appears to be still a linear function of burnup, and there were no observed excessive film deposits found on the heat-transfer surfaces of the fuel cylinders. Metallographic examination of the nickel diffusion barrier revealed no excessive depletion of the nickel. The only disconcerting point found was the presence of the coked material in the inner process tube and the valve body. This buildup might cause some problem if it reached a condition that resulted in no coolant flow through the inner process tube, or interfered with proper operation of the control rods.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Piqua Nuclear Power Facility Development	
Project: PNPf Core II Fuel Element Development	
Project Engineer: S. O. Arneson	
Reporting Period: Fiscal Year 1965	AEC Category: 04-02-02-01
General Order: 7629	Subaccount: 1910

I. PROJECT OBJECTIVES

The objectives of this project are the development of processes, specifications, and procedures for fabricating SAP-clad UO_2 fuel elements for the PNPf Core II loading; fabrication of the fuel element assemblies simulating the PNPf Core II design for out-of-pile testing; and fabrication of one PNPf Core II prototype fuel element.

This project provides for the refining of existing processes with specific application to the fabrication of the PNPf Core II fuel elements. Specific areas for development include finned fuel tube fabrication and end-closure techniques. Materials testing will be conducted to further develop and evaluate mechanical and metallurgical properties of SAP alloys. The materials testing program will provide the basis for preparing material specifications for the PNPf Core II fuel elements. End-closure, fuel loading, and assembly procedures will be refined to establish procedures for fabricating the mockup and prototype fuel elements. These procedures will form the basis for preparing process specifications for fabricating the PNPf Core II fuel elements. Mockup fuel element assemblies are to be fabricated to establish fabrication procedures and to confirm mechanical and hydraulic characteristics of the design. A prototype fuel element will be built to develop production technology for fabricating the PNPf Core II fuel elements. The prototype is planned for insertion in Core I to provide performance data under irradiation and environmental conditions similar to those expected in Core II.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

The literature was reviewed for information on the wear characteristics of SAP in an organic environment. Corrosion tests and operational experience with test elements at the OMRE indicate there will be little or no erosion of the SAP under the PNPf environmental conditions. Experimental data on the wear characteristics of SAP and Type 321 stainless steel in contact with themselves and with each other have been generated by Euratom in connection with the ORGEL program. These tests indicate that the wear of SAP on SAP is between 600 to 700 times greater than the wear of steel on steel under the same conditions, and that the wear of SAP on steel is more than four times as great as that of SAP on SAP. The significance of these results is limited primarily to material comparisons, since the test conditions probably do not correspond to the PNPf Core II fuel element vibrational modes and loading. Data on the fretting characteristics of SAP clad fuel elements and of SAP coolant channels in flowing hot organic have been developed by AECL.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

7617-2506

Figure 1. Tensile Strength of Slowly Extruded XAP-001 Tubing

7617-2507

Figure 2. Tensile Strength of Impact Extruded XAP-005

Figure 3. Tensile Hoop Strength of XAP-005 and XAP-001 Tubing

7617-2508

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Available SAP creep-rupture, thermal conductivity, and specific heat data were evaluated and design recommendations were released for XAP-001 and XAP-005.

Short-term tensile and pressure tests were completed for 14-fin tubing specimens from XAP-001 and XAP-005. The uniaxial tensile test results for XAP-001, extruded by a slow extrusion process, are plotted in Figure 1; and those for XAP-005 tubing fabricated by impact extrusion are plotted in Figure 2. The tensile hoop strengths of the two tubing lots, as determined by pressure burst strengths, are plotted in Figure 3. These tests indicate that the strength of the XAP-005 tubing is comparable to that of XAP-001, the reference PNPf Core II cladding, at lower temperatures; and is significantly higher at the elevated temperatures up to 900°F. This difference in properties between the two alloys may be attributable, in part, to the effects of fabrication variables between a slow hydraulic extrusion used for XAP-001 and the fast-impact extrusion used for XAP-005. The biaxial pressure tests also showed that the burst (transverse) strength of SAP alloy tubing is about 75% of the longitudinal strength, as determined by uniaxial tests.

SAP Alloy XAP-001 tubing was extruded, twisted, and straightened for two PNPf Core II designs; a 14-fin tube for 0.394-diameter UO₂ fuel and a 10-fin tube for 0.268-diameter UO₂ fuel. The dimensional inspection of the 1200 ft of tubing fabricated has shown that the vendor is capable of meeting the dimensional tolerances for these designs. This tubing provided the material for end-closure development efforts and the fuel rod cladding for the UO₂ hydraulic mockup. Additional SAP billet material of two alloys, XAP-001 and XAP-005, was procured to provide cladding material for the prototype fuel elements.

Several series of Magneform-eutectic bonding trials were performed to establish the parameters for end-closure bonding of the Core II fuel rods. Early results showed that 1100 Al end-plugs, currently used in the slower "collect" bonding technique, did not bond well using magneforming. Substitute materials (SAP and 6061 Al) were tried unsuccessfully, although in both cases the initial bonds appeared to be promising. Subsequently, 1100 Al end-plugs, using an alternate bonding cycle process, produced acceptable bonds. This process is being developed as the reference Core II bonding process. A process specification has been initiated and will be completed early next fiscal year.

A 2-ft-long mockup of the PNPf Core II fuel element was fabricated to demonstrate the feasibility of assembling the fuel rod bundle with a cumulative 0.025-in. gap across the rods. Because of the ease with which the mockup was assembled, little difficulty is anticipated for the assembly of the full-scale fuel elements with a similar cumulative gap of 0.025 in.

A full-scale mockup of a Core II element was fabricated to establish the fabrication procedure to be followed during fabrication of the Core II prototype fuel element. The mockup has been submitted for hydraulic testing which is scheduled to be completed early in FY 1966.

A UO₂ pellet specification for the prototype and Piqua Core II fuel elements was issued. The enrichment (3.8% U²³⁵) and dimensional design requirements for the prototype fuel pellets were established and a purchase order was placed

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

for delivery in August 1965. Procurement action was initiated for the SAP fuel tubes and other hardware items for the prototype element.

A production-oriented nondestructive testing program was initiated during the last quarter of the fiscal year. Tooling for performing the standard nondestructive tests (helium leak checking, penetrant inspection, dimensional measurements pressure testing) is being designed. Investigation of more sophisticated NDT techniques has established the following preliminary results:

- 1) X-ray and ultrasonic techniques can detect scratches, inclusions, and other discontinuities in SAP finned tubing, and measure the wall thickness.
- 2) Ultrasonic techniques can detect gross alloy segregation in SAP billets.
- 3) Ultrasonic inspection can detect unbonded areas in the eutectic bonded end closures.

Quantitative results on the above techniques are being developed. Recommendations will be made early next fiscal year as to which NDT techniques should be used for Core II production. Process procedures and specifications for these techniques will be established during the first quarter.

III. EVALUATION OF EFFORT DURING FY 1965

All material and process development efforts required for PNPf Core II have been completed. SAP properties have been evaluated experimentally and a cladding specification has been written. SAP tubes were successfully extruded to the PNPf Core II cladding dimensions and tolerances. A magnetic pulse (Magneform) technique was developed for making eutectic bonded end closures in fuel tubes on a production scale. Nondestructive testing techniques have been evaluated and several appear to be adaptable to production processes. All material and process specifications will be issued by early FY 1966.

The hydraulic mockup was completed six weeks later than planned due to scheduling and inspection problems in obtaining the SAP cladding tubes and stainless steel process tubes. Therefore, hydraulic testing will not be completed during this reporting period as scheduled but will be completed early in the next fiscal year.

The fabrication of the Core II prototype fuel element has been rescheduled for completion in October 1965. Two major factors contributed to this delay. The first was the decision to use enriched fuel and insert the element in Core I for operating experience. This required additional nuclear and thermal analysis to establish a firm design (some latitude was possible in design configuration if the element were not to be irradiated). Consequently, the UO₂ pellets could not be ordered until June 1965, and will not be received until July or August of 1966.

The second factor contributing to the delay was the late delivery of the SAP billets which were not received until May 15. Since extrusion of the billets to finned tubing will take 4 to 8 weeks, the tubing will not be available until July or early August. It is expected that the element will be completed in October 1965. Although this is four months later than originally planned, it should not delay the Core II fabrication schedule.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: PNP Development

Project: PNP Core II Design

Project Engineer: H. J. Rubinstein

Reporting Period: Fiscal Year 1965

General Order: 7629

AEC Category: 04-02-02-01

Subaccount: 1930

I. PROJECT OBJECTIVES

The objectives of this project are (1) to design a SAP-Clad UO_2 Core for use in the Piqua Nuclear Power Facility; (2) to design a prototype for insertion in Core I; and (3) to prepare a safeguards summary report for operation of the PNP with this core.

II. MAJOR ACCOMPLISHMENTS DURING FISCAL YEAR 1965

This project was initiated in the second quarter of FY 1965. Design criteria were established and an 85-rod standard fuel element design was selected. It was determined that the control rod positions should be fueled. Conceptual designs of control rod position fuel elements were prepared for evaluation.

Gross thermal and nuclear analyses were made for core performance at thermal power levels of 45.5, 60, and 80 Mw. The 45.5 and 60 Mwt power levels can be attained with a 61-element core loading and within the control capabilities of the existing 13 control rods. The 80-Mwt power level was found to require the full 85-element core loading and 19 control rods. Figure 1 shows the standard 85-pin element. Figure 2 shows the control-rodded element. An increase in UO_2 pellet costs, coupled with the reductions in UC fabrication costs experienced in the Hallam Core II fuel element fabrication program, prompted a brief reevaluation of UC fuel for Piqua Core II. A conceptual 61-fuel rod UC element design was prepared and evaluated. The UC element was found to be competitive with UO_2 with respect to cost, and to offer significant performance advantages.

A test assembly was made to check assembly and operating clearances of the fuel rods within the element. Fabrication of a hydraulic mockup fuel element was initiated.

III. PROGRESS DURING REPORT PERIOD

The detailed core nuclear and thermal core performance analyses were completed. The hydraulic mockup fuel element was completed, but the hydraulic loop was unavailable due to priorities established for the U-305 test element.

The detailed fabrication drawings and product specifications were prepared for the Piqua Core II, SAP-Clad UO_2 fuel elements. The enrichment for the prototype SAP-Clad UO_2 fuel element was specified, and the fuel and cladding ordered.

7617-2509
Figure 1. Standard 85-Pin Fuel Element
NAA-SR-11450

Figure 2. Control-rodged Element

NAA-SR-11450
VI-25

OSTI ID: 0000 19406

7617-2510

TABLE 1
PNPF CORE II POWER DISTRIBUTION

Element (typical)	Number of Equivalent Elements (N)	Normalized Power Factor (F)	Elemental Power for 45.5 Mw (kw)	Elemental Peaking Factor (ϕ_E)	Axial Peaking Factor (ϕ_A)
F-11 (control rod IN)	1	0.0142	607	1.24	1.50
F-9	6	0.0245	1048	1.29	1.50
D-11 (control rod IN)	3	0.0124	530	1.32	1.50
E-8 (control rod OUT)	3	0.0214	915	1.17	1.50
F-7	6	0.219	937	1.35	1.50
E-6	12	0.0181	774	1.40	1.50
F-5 (control rod OUT)	6	0.0153	654	1.27	1.50
D-5	6	0.0128	547	1.49	1.50
E-4	12	0.0125	535	1.48	1.50
F-3	6	0.0117	500	1.30	1.50
—	$\Sigma N_i = 61$	$\Sigma N_i F_i = 1.000$	$\Sigma N_i P_i = 42.77 \text{ Mw}^*$	—	—

*It is assumed that 6% of the total power is generated in the moderator region.

TABLE 2
PNPF CORE II HOT CHANNEL FACTORS

Coolant bulk temp. rise ($T_{\Delta T}$)	= 1.18
Film temp. rise (F_θ)	= 1.37
Heat flux or cladding temp. rise (F_C)	= 1.14
Gas gap temp. rise (F_G)	= 1.58*
Fuel temp. rise (F_F)	= 1.11*

*Does not include uncertainties in thermal conductivity

TABLE 3
PNPF CORE II CHARACTERISTIC PARAMETERS

Design power (Mwt)	45.5
Standard fuel elements (No.)	48
Control rod fuel elements (No.)	13
Core flow rate (gpm)	14,700
Core pressure drop (psi)	20.5
Core ΔT ($^{\circ}F$)	45
Core inlet temperature ($^{\circ}F$)	527
Zone 1 flow velocity (ft/sec)	13.5
Zone 2 flow velocity (ft/sec)	7.05
Avg rod power (kw/ft)	2.29
Max rod power (kw/ft)	6.69
Avg heat flux (Btu/hr-ft ²)	32,100
Max heat flux (Btu/hr-ft ²)	107,300
Burnout heat flux (Btu/hr-ft ²)	959,000
Max clad temperature ($^{\circ}F$)	691

*This temperature is higher than was predicted in the third quarter progress report, because that prediction was based on a fully orificed flow pattern rather than the 2-zone pattern now being considered.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

A further investigation of UC fuel for Core II was carried out. Zirconium was evaluated as an alternate cladding material. A report documenting this investigation was written and issued. The adoption of SAP-Clad UC fuel elements for Piqua Core II was recommended.

The Piqua Core II, SAP-Clad UO_2 Hazards Report was written, internally reviewed, and issued.

A. HEAT TRANSFER

After the early selection of standard element design of 73 large rods and 12 small rods for the standard element (Figure 1), a combined nuclear, thermal, and hydraulic parameter study was made to determine the control rod fuel element design. A configuration consisting of 80 small rods with ten 0.045-in. fins each was selected for the control rod element. Based on neutron flux distribution information obtained from the Nuclear Engineering Unit, a predicted discrete power distribution was calculated for Core II (Table 1).

Following the selection of a flow pattern, hot-channel factors (Table 2) were calculated. Pertinent thermal and hydraulic parameters (Table 3) were then calculated for the PNPf UO_2 Core II. An investigation of maldistribution of flow in the control rod elements is currently under way.

An investigation was made to determine the effect on pressure drop in the core caused by the twisted longitudinal fins of the 85 aluminum fuel pins. Pressure-drops of fin heights of 45 and 55 mils and fin spacing based on 10, 12, and 14 fins per fuel pin were calculated.

On the basis of this information, the selected fin design of 14 fins with 45-mil height was retained for the fuel pins. In order to establish reactor grid plate loading, pressure drops across the grid plates were established for several operating conditions.

The pressure-drop effects of the in-core filters were evaluated to determine increased loading on the grid plate due to fouling of the filters and fuel elements.

A 2-zone arrangement has been developed in which 24 of the outer elements are orificed to maintain a coolant velocity of 52% of the velocity in the 37 un-orificed inner elements. With this arrangement, the core hot-spot temperature under design operating conditions is 691°F .

The partial orificing scheme recommended for Piqua Core II consists of orificing 24 outer elements uniformly so that the hottest of these elements will have the same maximum surface temperature as the hottest of the 37 un-orificed inner elements.

The coolant velocities are 13.5 ft/sec in the un-orificed region, and 7.0 ft/sec in the orificed elements. With this orificing arrangement, a total core pressure-drop of 20.5 psi is expected, with a core flowrate of 14,700 gpm.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

B. NUCLEAR

In order to achieve a mechanically stable fuel element having a conservative maximum fuel temperature, attention was restricted to elements having 85 fuel rods. Since the overall element diameter is fixed, the restriction of 85 rods fixes the diameter over fins of an individual pin. (In order to fit snugly into a round element, two pin sizes are used. Seventy-three pins are 0.534 in. in diameter over fins, and 12 are 0.408 in diameter over fins). The fin height thus becomes the primary parameter affecting the nuclear characteristics of the core. Consequently, a series of calculations was performed to determine the fuel cycle cost as a function of fin height. The results are shown in Figure 1 for both 45.5 and 80 Mwt. These results show that a fin height of 0.045 in. is near optimum for both power levels; therefore, this value was selected. Details of the resulting fuel element are shown in Reference 2 (Figure 1).

The fully packed 85-pin element cannot be used in the 13 control rod positions. An annular type fuel element, utilizing pins identical to the small pins in the standard element, was designed for these locations (Figure 2).

1. Control Requirements

The following is a tabulation of control requirements for a 61-element core at 45.5 and 60 Mwt.

Core Power (Mwt)	45.5	60
Average specific power (kw/kgU)	9.76	12.9
Control Requirements (% $\Delta k/k$)		
Equilibrium xenon	2.60	3.04
ΔT organic	2.01	2.01
ΔT fuel	2.17	2.67
Burnup	1.58	2.07
Central rod stuck out	1.40	1.40
	<u>9.76</u>	<u>11.19</u>

The integral control rod worth of 13 rods in a 61-element core has been calculated, using 4-group transport theory, to be 12.0% $\Delta k/k$. Based on this, the shutdown margin is 2.24% at 45.5 Mwt, and 0.81% at 60 Mwt.

2. Kinetics Parameters

	<u>Initial Core</u>	<u>Equilibrium Core</u>
β_{eff}	0.0071	0.0057
l^* (sec)	38	38
Temperature Coefficients (% $\Delta k/k$ -°F)		
Fuel	-0.002	-0.002
Coolant	-0.0059	-0.0054
Moderator	-0.0076	-0.0071

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

The equilibrium cycle fuel management scheme for a 61-element core operating at 45.5 Mwt and a plant factor of 0.8 is basically a 10-batch program. Refueling shutdowns occur at intervals of 6300 Mwd/ or about every 6 calendar months. Fuel is discharged from the central part of the core at an exposure of 15,000 Mwd/MTU, and fresh fuel is loaded near the core periphery. Some of the partially burned elements are rearranged at each shutdown to replace the elements removed from the center, and to make room for those loaded near the edge. In general, as the center of the core is approached, the fuel elements are more depleted.

This type of fuel management results in significant power flattening, permitting all of the control rods to be removed from the core at the end of the cycle without exceeding the design power peaking factors. The feed enrichment for the fresh elements necessary to maintain this cycle is 2.8 wt % U^{235} .

The initial core is designed to simulate the equilibrium core by having two enrichment zones. The central 31 elements are enriched to 2.1 wt % U^{235} , and the outer 30 elements are enriched to 2.8 wt % U^{235} .

C. FEASIBILITY STUDY

A study of the feasibility of a UC-fueled PNPf second core was conducted at the request of the AEC. Flow distributions, hot-channel factors, and power distributions were estimated from calculated values for the UO_2 core. Using these factors, a parameter study was conducted to determine a feasible UC fuel element design. Because of the higher density of UC, it was possible to design a fuel element in an annular configuration, which can be used in either a standard core position or a control rod position. Pertinent hydraulic and heat transfer parameters were calculated for this design. This work is presented in a letter to the AEC describing the results of the feasibility study.

The higher thermal conductivity and higher uranium density of UC results in an elemental peaking factor which is much reduced, leading to a better utilization of the fuel. An element was selected, having two concentric rings of fuel rods, each ring containing 24 rods. The element is shown in Figure 3.

The necessary enrichment for a fuel management scheme similar to that described above for the UO_2 core was calculated to be 2.6% U^{235} . Fuel cycle costs were estimated for both the UC and UO_2 cores and are presented below.

Item	Fuel Cycle Costs	
	(mills/kwh)	
	UC	UO_2
Fabrication	1.56	1.61
Inventory	0.30	0.37
Burnup	1.10	0.96
Uranium Recovery	0.58	0.58
Total	3.54	3.52

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

The difference between these two figures is not considered significant, since they are well within the uncertainties of the estimates. Furthermore, the UC design was not cost-optimized as was the UO₂ design. Notice that the burnup component of the costs is significantly higher for UC. This is directly attributable to the less efficient conversion of U²³⁸ to Pu²³⁹ in the UC core. The larger and denser fuel pins in the UC element produce a smaller resonance integral (15.5 barns for UC versus 19 barns for UO₂). This fact, together with the higher moderator-to-fuel ratio of the UC core, causes a larger U²³⁸ resonance escape probability, and therefore a lower conversion ratio.

The effect of increasing the thickness of the outer process tube on the UC element was investigated. It was estimated that increasing the thickness from 0.010 to 0.020 in. would result in a reactivity decrease of about 1.0% $\Delta k/k$. An increase in enrichment of about 0.1 wt % U²³⁵ would be required to regain this reactivity.

The following table presents various parameters of interest for both the UC and UO₂ cores.

	UC	UO ₂
Core composition		
Coolant moderator (vol %)	71	59
Fuel (vol %)	20	28
SAP (vol %)	7.9	2
Stainless steel (vol %)	0.84	0.56
H/U atom ratio in core	4.1	3.0
Power peaking factors		
Radial	1.5	1.5
Axial	1.5	1.5
Elemental	1.05	1.30
Flux uncertainty	1.15	1.15
Total (product)	2.7	3.4
U Inventory (kg)	4370	4220
Equilibrium core parameters		
Feed enrichment	2.6	2.8
Discharge enrichment	1.21	1.49
Fissile Pu concentration in a discharged fuel element (gPu/kgU)	4.17	4.35
Average discharge exposure (Mwd/MTU)	15,000	15,000

D. UO₂ PROTOTYPE

A hydraulic and heat transfer parameter study for the UO₂ prototype fuel element, based on the standard Core II element design, was completed and presented at a design review meeting. Based on this and predicted performance data, the prototype design was accepted, and a prototype performance level and fuel enrichment were selected which will allow maximum power generation when placed in Core I.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

E. SAFETY ANALYSIS

The first draft of the Safety Analysis Report was completed and was reviewed by Atomics International's Civilian Power Reactors Committee. The technical specifications for Core II operation and the final Core II Safety Analysis Report are being prepared. The report is also being modified to allow insertion of one UO_2 element in the initial PNPf core. This was not part of the original project objective.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

Most of the design effort on the SAP- UO_2 Core was completed during FY 1965. The mechanical design is firm except for any perturbation resulting from the hydraulic tests which were not yet run. The prototype design is complete. Areas requiring further analytical and design calculation are as follows.

- 1) Fuel management program and fuel cycle cost optimization
- 2) Control rod worth and shutdown margins
- 3) Heat-transfer analysis of fuel-handling system
- 4) Final issuance of safety analysis report

The feasibility study for a SAP-clad UC core showed the UC to have many advantages over the UO_2 core. These are primarily due to the higher density and conductivity of UC. This allows an element design more closely approaching that of Piqua Core I. At this writing, no decision has been made to redesign Piqua Core II using UC as fuel.

SECTION VII
REACTOR TECHNOLOGY SAFETY AND RESEARCH
CONTENTS

<u>Title</u>	<u>Identification</u>	<u>Page</u>
GENERAL REACTOR TECHNOLOGY		
FISSION EFFECTS IN METAL FUELS.....	7652 - 4537	VII-1
SYNTHESIS, STRUCTURE, AND BEHAVIOR OF METAL- HYDROGEN SYSTEMS	7652 - 4552	VII-8
AGE MEASUREMENTS	7604 - 1311	VII-14
EXTRAPOLATION LENGTH MEASUREMENTS	7604 - 1312	VII-18
CROSS-SECTION ANALYSIS AND APPLICATIONS PROJECT	7604 - 1313	VII-21
FAST-SPECTRUM DOPPLER MEASUREMENTS	7604 - 1330	VII-29
STRUCTURE, PROPERTIES, AND FISSION EFFECTS IN UC AND OTHER REFRACTORY, INTERSTITIAL FUELS	7551 - 4539	VII-36
STUDY OF BeO AS A REACTOR MATERIAL	7551 - 4549	VII-56
CONTINUOUS NEUTRON FLUX MONITOR	7616 - 4584	VII-72
MULTICYCLE REPROCESSING	7522 - 4612	VII-80
NUCLEAR SAFETY		
STUDY OF REACTOR HOUSING INSTALLATIONS	7603 - 1373	VII-85
FISSION PRODUCT RETENTION BY REACTOR COOLANTS	7608 - 1331	VII-86
MEASUREMENT OF DOPPLER COEFFICIENTS	7549 - 1351	VII-98

SECTION VII (Continued)

<u>Title</u>	<u>Identification</u>	<u>Page</u>
NUCLEAR SAFETY (Continued)		
ATMOSPHERIC DECONTAMINATION IN REACTOR ACCIDENTS	7640 - 1371	VII-110
PHYSICAL RESEARCH		
HIGH-TEMPERATURE CHEMISTRY ..	9305 - 5302	VII-125
RADIATION CHEMISTRY	9305 - 5305	VII-131
ELECTRONICS STRUCTURE OF METALS AND ALLOYS	9305 - 5401	VII-137
RADIATION DAMAGE IN CRYSTALLINE SOLIDS	9305 - 5404	VII-144

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Reactor Fuels and Materials Development — Other

Project: Fission Effects in Metal Fuels

Project Engineer: C. E. Weber

Reporting Period: Fiscal Year 1965

AEC Category: 04-40-02-09

General Order: 7652

Subaccount: 4537

I. PROJECT OBJECTIVES

The objectives of this project are: (a) to obtain a detailed understanding of the processes and mechanisms involved in fission damage, including those responsible for swelling of reactor fuel materials during irradiation and those arising from dynamic fission processes; and (b) as a result of such understanding, to propose and test procedures by which swelling can be minimized.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. NAA-118 CAPSULE

The swelling resistance of specially designed fine dispersion metal fuels that contain trace amounts of additives is being investigated at high fuel temperatures, 550 and 645°C (990 and 1152°F), and at burnup levels of 0.8 and 1.4 at. % burnup levels. Also incorporated into the experiment design are coarse dispersions, unalloyed uranium samples as comparison standards, and solution-quenched alloys.

A temperature-controlled capsule has been built, worked perfectly, and the samples were irradiated to design burnup without exceeding the temperature design limits. The experiment has been discharged from the reactor and shipped to the AI hot cells for postirradiation examination.

B. FISSION GAS RELEASE MEASUREMENTS

The mechanisms of rare gas atom migration and rare gas atom trapping are being evaluated by postirradiation annealing of lightly irradiated metal foils. Specific emphasis is placed on evaluating whether the gas release conforms to or deviates from the classical solution of the diffusion equation,

$$F^2 = KDt \quad \dots(1)$$

where

F = fractional gas release

K = geometric constant

D = diffusion coefficient, and

t = time.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Several types of deviations from diffusional behavior, Equation 1, have been observed. Rather abrupt changes in the slope of the gas release curve are often reported in the literature. This study has shown that successive anneals at progressively higher temperatures can cause further gas release. Also, only a few tenths percent of the contained gas is released from the sample.

The common practice of assuming the validity of Equation 1 and evaluating either F^2 vs t or F vs $\sqrt{t}$ has been a rather insensitive measure of the modes of gas release. Some calculational errors are inherent in evaluating either F^2 or $\sqrt{t}$, as has been pointed out. An alternative technique has been found to be more sensitive, namely to evaluate the derivative solution,

$$F \frac{dF}{dt} = \frac{KD}{2} \quad \dots (2)$$

7652-2501
Figure 1. Activity Change per Unit of Time vs Function of Total Activity Released

7652-2502
Figure 2. Fractional Release x Change in Fractional Release per Unit of Time as a Function of Time

Using this approach, results comparable to those presented in Figures 1 and 2 have been obtained. A change in release mechanism during the latter stages of the annealing treatment becomes apparent. This second mechanism is not observed when the burnup is reduced to 10^{14} to 10^{15} fissions/cc.

The gas release from solids which contain internal traps has been simulated on an analog computer. This technique provides a rapid method for evaluating what types and distributions of traps can provide the experimentally obtained release curves, and also a method for interpreting the in-pile response from postirradiation annealing data.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

The model assumes a random distribution of internal traps and a random diffusion of the gas atoms within the solid. Specifically, the following equations are solved simultaneously for 12 adjacent nodes

$$\frac{\partial c}{\partial t} = D\nabla^2 c + KF - k_1 c^2 - k_2 c [A_0 + \psi] \quad \dots(3a)$$

$$\frac{\partial \psi}{\partial t} = \frac{K_3}{2} c^2 + D_0 \nabla^2 \psi - \frac{D_0}{D} A_0 \psi \quad \dots(3b)$$

where

c = concentration of gas atoms in the lattice

ψ = concentration of clusters in the lattice $\int_0^t k_1 c^2 dt$

F = fission rate

A_0 = initial trap concentration

$\frac{D_0}{D}$ = ratio of atom and cluster diffusion rates

K_i = constants

t = time.

$D\nabla^2 c$ and $D\nabla^2 \psi$ define the diffusional loss of gas atoms and clusters of gas atoms from the solid. Any or all terms can be removed from the system or changed in magnitude. The printed output is the concentrations c or ψ , the total gas release, or the correlation described by Equation 2.

The computations have shown that homogeneous nucleation limits the applicability of Equation 1 to burnup levels that are less than 10^{16} fissions/cc, and that heterogeneous traps, such as dispersed particles, must have a strong geometric bias, i.e., a large surface area or geometric size to be effective in controlling the migration range of gas atoms. Calculated fractional release data are generally higher than the experimental values. The parameter ratio of fission to diffusion rate again becomes a characteristic value, although the previous solutions appear to have oversimplified the general relationships.

C. FISSION-INDUCED PLASTICITY

The nature of fission-induced creep and of fission spike damage mechanisms are being investigated by cyclotron bombardment of fuel materials of different crystallographic structure at a variety of strain rates and temperatures.

An apparatus that permits testing at temperatures of nearly -196 to 300°C, with stress levels of 10^2 to 10^5 psi with $\pm 1\%$ precision, and with strain resolution of 3×10^{-5} , has been constructed. All of the components were first proof-tested individually, the apparatus has been assembled and is being evaluated as an integrated system.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

This system permits simultaneous testing of four samples with continuous monitoring of strain load and temperature. The cyclotron beam intensity can be controlled and transient responses to the presence or absence of irradiation can be evaluated.

In a parallel effort, the data for creep of alpha uranium which have been published in the literature have been reviewed and analyzed in a consistent manner, using some of the more recent evaluation techniques. A pattern has begun to form wherein it may be possible to define regions of creep that have characteristic activation energies for prescribed levels of applied stress and temperature. Unfortunately, no similar correlation has been attempted previously, and there are regions of the general relationship that lack sufficient data to provide a firm correlation. The proof tests of the cyclotron device and evaluation of the test samples that are to be tested in this device will provide opportunities to explore these poorly defined load regions.

D. COMPUTER STUDIES

Past theoretical studies of rare gas atom mobility in solids have involved application of metallurgical theory concerning the nature of diffusional processes to systems for which no experience factors exist. Such evaluations have led to the conclusion that the energy of migration could conceivably be as low as one-half or as large as twice the activation energy for self-diffusion in the host lattice. Even these limits are not extreme values.

The newer theoretical approaches developed in recent years involve the simulation of a solid metal by means of a digital computer, and this technique permits the study of diffusional processes. Since the migration and the gas atom trapping concepts are important to the further development of swelling resistant alloys, computations of the most probable diffusion processes for rare gas atoms in metals are in progress.

Fortunately, the prodigious amount of computer programming that would normally be required to develop a computational code can be circumvented by applying current results being generated under Project 9305-5404, "Radiation Damage in Crystalline Solids." Under this project, a large amount of programming has already been done for the purpose of computer-simulation calculation of defects and damage states in metals. The small amount of reprogramming necessary to include impurity species has been reported under this project title. Further collaboration has involved the development of gas atom-metal interaction potentials from the known metal-metal and gas-gas atom potentials; and this development is also reported under the 9304-5404 heading for continuity.

The specific problem selected for the initial study is the diffusional jump of a rare gas atom from one substitutional site in a metal lattice to an adjacent, unoccupied site. This is the simplest useful computation and the knowledge gained from the study will provide direction for more complex problems, such as evaluation of the binding energy of two xenon atoms in such a lattice.

The initial problem is perhaps best discussed with reference to Figure 3 in which the potential energy is schematically plotted against distance. The abscissa represents the distance between two nearest neighbor atoms of an ideal

Figure 3. Potential Energy
vs Distance

solvent metal lattice. The zero of energy on the ordinate represents the total energy of a large but finite number of solvent metal atoms plus a single xenon atom situated at a substitutional site with nearby solvent metal atoms relaxed into an equilibrium condition, but with no vacancies nearby. This condition is called Configuration I. The upper curve of the figure represents the change in potential energy of the system as the xenon atom diffuses by a two-member exchange mechanism with an adjacent solvent metal atom. The height of the potential energy barrier in this case is drawn very high in order to represent what is in all probability a physically unrealizable phenomenon, i.e., a non-vacancy diffusion mechanism can be ignored.

The lower curve of Figure 3 represents the changes in potential energy accompanying vacancy mechanism of diffusion. The first change that occurs takes place prior to the actual diffu-

sional jump; if a xenon atom is originally at $x = 0$ and the vacancy at $x = 1$, a relaxation will occur and the xenon will move toward the vacancy by the amount of Δx , while the vacancy remains at $x = 1$, and the potential energy of the system will decrease its neighboring vacancy, plus the neighboring solvent metal lattice atoms, is the starting point of our diffusional study and is called Configuration II. The overall diffusional jump will involve an exchange of the xenon atom and the vacancy, with the xenon atom moving to $x - (1 - \Delta)x$ and the vacancy to $x = 0$; and this configuration will be called Configuration III. Obviously, Configuration II is related to Configuration III by a mirror plane of reflection at the point $x = 0.5$. The detailed computer study involves moving the xenon atom from Configuration II to Configuration III by a series of incremental steps.

At each intermediate step, the xenon atom is fixed in position and then two facts concerning the given intermediate configuration are determined: (1) the relaxed positions of as many neighboring solvent metal atoms as is consistent both with obtaining an equilibrium configuration, also with the storage capacity of the computer; and (2) the potential energy of the configuration will be determined. It is expected that the energy curve resulting from a number of intermediate steps will resemble the lower curve in Figure 3, where the energy of activation with respect to Position II is much lower than that for the nonvacancy diffusion mechanism. The ultimate test of the initial study will be a comparison of this computed energy of activation and the observed values. Deviations will of course be expected, due to approximations that must be made in the computer study; but a correlation with observed energies of activation should permit further refinement of the computer study.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

The computer program has been modified to the extent that it is usable for the present purposes (although in this type of work constant minor reprogramming is always necessary), and some tentative calculations have been carried out. These calculations had two objectives: (1) to proof-test the modified computer program; and (2) to evaluate the magnitude of the energy involved in the hypothetical Configuration I that has been discussed in connection with the figure. Although a main goal is to evaluate the diffusion of xenon in uranium, the initial studies will involve the xenon-copper system because good Cu-Cu potentials are available. It must be emphasized, however, that none of these potentials is derived from first principals, but are instead arrived at by fitting theoretical models to empirical data. However, Brinkman has developed an approximate potential for uranium that will be used in later evaluations; and hopefully an even more accurate uranium potential will be obtained.

An initial hand-calculation was first carried out for the energy of Configuration I with no relaxation of the copper atoms about a xenon atom on a substitutional site; it resulted in a large value of approximately 54 ev. The computer program verified this value on the "first iteration" which provides a measure of the code reliability. The computer program is so arranged that on "successive iterations" increasingly greater amounts of relaxation of the copper atoms are introduced. The iterations are repeated until a steady state has been reached indicating an equilibrium value. It is also necessary to demonstrate that other nearby configurations do not have lower equilibrium energies. On the first run some 20 iterations were made, and the energy steadily decreased to an approximate value of 26 ev. Further iterations will be run and alternate potentials will be used to improve on this first estimate. That this value is preliminary can be seen by comparing it with the estimated 12.7 ev obtained by Rimmer and Cottrell [(Phil. Mag. 2, 1345 (1957)) using a completely different potential and a somewhat different calculational procedure than has been used in the present study. One of the next steps will be to repeat the Rimmer-Cottrell calculation, using their exact potential and exact calculational procedure.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The in-pile irradiation of alloys containing dispersed phases of low-level additives at operating reactor temperatures has progressed to the hot-laboratory examination stage.

Considerable progress has been made in the area of measuring fission gas release and interpreting the mechanism of gas atom migration in metal fuels. The coupling of the analog computer codes with experimental data will permit a more detailed and more comprehensive evaluation of the effectiveness of various types of dispersed phases in trapping gas atoms and gas bubble clusters. It also provides a bridge between the significance of postirradiation annealing testing and in-pile performance within the framework of existing theories of fuel material instability. More important, the existing phase of this study includes evaluation of re-resolution of particles or gas atom clusters, and these aspects can be examined with some hope of major accomplishments.

The digital computer evaluations of the most probable rare gas atom migration processes and binding energies among colliding gas atoms utilized the more recent applications of computer studies of material structures. Though the size

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

of these impurity atoms is cumbersome, their potential lack of chemical attraction with lattice atoms makes definition of their motion important to other solid-solution processes. Ultimately, the binding energy with crystalline imperfections will be evaluated.

An in-cyclotron device has been designed and constructed. Detailed studies of the influence of temperature, burnup, and applied stress on in-pile properties of fuel materials can now proceed. In view of the correlations of out-of-pile creep data made possible by using data generated from other laboratories, and the importance of in-pile properties in defining the origin of "cavitation," such measurements obtain additional importance. In principle, the concepts of inter- and intragranular strength properties define the tendency for cavitation swelling. Yet the existing technology suggests both material weakness and also the existence of this form of swelling. Uranium supports no load during irradiation, and still the stacked Calder Hall fuel pellets do not undergo massive deformation. This anomalous behavior can now be explored with precise control of all experimental variables.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Program: Reactor Fuels and Materials Development — Other	
Project: Synthesis, Structure, and Behavior of Metal-Hydrogen Systems	
Project Engineer: C. E. Weber	
Reporting Period: July 1964 - June 1965	AEC Category: 04-40-02-09
General Order: 7652	Subaccount: 4552

I. PROJECT OBJECTIVES

The objectives of this project are to: (a) prepare research on new binary and ternary hydrides; (b) select correlated studies of physical, thermodynamic, and mechanical properties; (c) study the structure, solid-state physics, and nature of bonding in hydrides as relates, primarily, to thermal stability and movement to its defect structure; (d) apply advanced experimental and theoretical techniques to study fission gas mobility and radiation damage in various hydride structure so as to optimize the high temperature swelling resistance of this class of materials.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. SOLID-STATE EFFECTS

1. Self-Diffusion of Zr in ZrH_x

A study of the diffusion coefficient of Zr in epsilon-phase zirconium hydride has been undertaken with the end objective being to arrive at a better understanding of fission gas damage from a knowledge of the mobilities of the various ions and atoms in the hydrided fuel systems.

The method selected for measuring the self-diffusion of Zr in zirconium hydride is a tracer method, whereby Zr^{95} deposited on the surface of a specimen is allowed to diffuse through the lattice. After a suitable time at temperature, the γ activity is measured as a function of depth from the surface.

Two methods for coating the hydride specimens with Zr^{95} were tried. At first, attempts were made to vapor-deposit metallic Zr^{95} . A proper balance between film thickness required by activity considerations and thickness required from integrity of the deposit could not be achieved. This approach was abandoned when it was found possible to apply Zr^{95} oxalate solutions, which were subsequently fired to the oxide. The Zr^{95} oxalate was separated from a Zr^{95} -Nb⁹⁵ solution by passing through a solvent extraction column containing a quaternary ammonium resin.

Several methods for removing thin layers from annealed specimens for γ counting were tried. A method, described by Pawel and Lundy,* of anodizing and stripping was tried at first. This process has been used to remove exceedingly thin layers from some materials. With zirconium hydride, an excellent

*R. E. Pawel and T. S. Lundy, J. App. Phys. 35, No. 2, p 435 (1964)

anodized film was easily obtained, but the film simply could not be stripped off the surface. Finally, this approach was dropped in favor of a mechanical grinding method. After several attempts, an apparatus design was arrived at which allowed removal and collection of surface layers with an accuracy of 0.00003 in. The apparatus is essentially a precision lapping device.

The total activity of the removed surface layers and of the fresh surface layers are both measured after each grinding operation. The activity contribution from material at a depth below the undisturbed surface is corrected for by subtracting a background value measured with an 0.040-in. aluminum disc between specimen and counter. Using this correction, activity plots of surface versus depth agreed quite well with those determined from the removed surface layers.

All of the above experimental techniques were given a trial run, then actual measurements were started on specimens of both coarse and fine-grained $\text{ZrH}_{1.7}$. The experimental conditions and the results of the diffusion coefficient calculations are given in Table 1. The experimental data are plotted in Figures 1 and 2. Two diffusion coefficients are reported in Table 1, one for the data to a depth of about 0.001 to 0.003 in. into the specimen, and the other at greater depths (up to 0.10 in.). It is presumed that since the initial surface area is covered with tracer and the surface area is much greater than the grain boundary area, the diffusion coefficient measured near the surface of the specimen is characteristic of bulk volume diffusion. At greater distances, grain boundaries and other defects in the structure continue to act as rapid transfer sites, and predominate as the concentration gradient decreases. The diffusion coefficients measured well below the surface eventually become characteristic almost entirely of grain boundary diffusion, with a very small contribution from bulk diffusion.

Figure 1. Zr in ZrH Activity Penetration

Figure 2. Diffusion and Grain Size

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 1
DIFFUSION COEFFICIENTS IN $ZrH_{1.7}$

Treatment Time (hr)	Temperature (°C)	Sample Type	Diffusion Coefficient (cm ² /sec)	
			Volume	Volume + Grain Boundary
87	802 ± 1	Fine-Grained	2.5×10^{-12}	1.2×10^{-10}
		Coarse-Grained	5×10^{-12}	1.2×10^{-10}
113	832 ± 1	Coarse-Grained	4.3×10^{-12}	
66		Fine-Grained	2.1×10^{-12}	
46.25	908 ± 1	Coarse-Grained	1.0×10^{-11}	
41	935 ± 1	Fine-Grained	1.1×10^{-11}	6.6×10^{-10}

The diffusion coefficients for coarse and fine-grained material may be different. The activation energy for volume diffusion in both materials is of the order of 30,000 cal/mole, based on the limited data plotted in Figure 2.

2. Electron Microscopy of ZrH_x

Electron micrographs of thinned specimens of epsilon-phase zirconium hydride prepared from reactor grade zirconium sponge are shown in Figures 3 and 4.

The overall structure is believed to be characteristic of ordered domains within the interstitial lattice. The c-axis of the tetragonal structure is oriented in different directions within various domains of a given grain of material.

The two-fold symmetry defects observed as relating to the complex (ZrCH) structure based on the abnormally large unit cell probably arise as a consequence of local strain fields, and could provide some measure of the symmetry of the dispersed phase.

Multiple spot diffraction patterns continually occur, and indicate differing interplanar spacings (or "d-spacings"). They are indicative of a twinned structure with some existing substructure in the twinned crystal. This has been verified by dark-field studies where individual diffraction spots have been examined and different regions of the crystals have been preferentially scattered to specific diffraction spots. In brief, a twinned relationship seems to exist in the ϵ -phase material.

There is some indication that the ϵ -phase transformation is analogous to a martensitic transformation in steel, with composition rather than temperature being the driving force for the transformation. The variation in c/a ratio with increased hydrogen content makes such a transformation mode practical. In essence, we have an accommodation twin which relieves developed shear stress; and it occurs in a macroscopic basis so that a substructure within the twinned structure must result.

7652-2506

Figures 3 and 4. Micrographs of Zirconium Hydride

3. Fission Gas Mobility in ZrH_x

Various factors affecting the rate of release and the total amount of release of recoiled fission gas atoms from the zirconium hydride lattice under investigation include: (1) temperature of annealing initial concentration of gas atoms, (2) purity of the hydride with respect to interstitial elements, (3) surface condition, and (4) dehydriding at constant temperature across a phase boundary. In FY 1965, temperature and concentration dependence factors were stressed.

The temperature dependence over the dose range from 5×10^{14} to 2×10^{17} fissions/cc can be expressed by the equation:

$$\ln F = m \ln T ,$$

where

F = fractional release

T = temperature in K° , and

m = slope.

A plot of the slope of the above equation as a function of dose rate is shown in Figure 5. The dose dependence per indicated rate is small, but a distinct difference is observed between sponge and iodide materials. A plot of fission gas release at $850^\circ C$ versus dosage (Figure 6) again shows a difference in behavior between sponge and iodide materials, with sponge material releasing significantly

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Figure 5. Temperature Dependence of Fission Gas Release

Figure 6. δ -Phase Material Fission Gas Release

less fission gas, possibly because of heterogeneous nucleation on impurities. Fractional release at 850°C ranged from approximately 4 to 8% for sponge material, and from 6 to 21% for iodide material. Saturation in fission gas release at each successive temperature is indicative of nondiffusional behavior.

B. TERNARY HYDRIDES

The search for new ternary hydride compounds has continued, with the major emphasis this year having been on the Zr-B-H, Ti-N-H, and U-C-H systems. Table 2 summarizes all of the systems studied since the inception of the program, and indicates their tendencies to form ternary compounds.

TABLE 2
TERNARY HYDRIDE SYSTEMS STUDIED

System	Compounds Formed	Cell Constants (hexagonal unit cells)		
		a_o	c_o	c_a
Zr-C-H*	$ZrC \cdot ZrH_x (x \rightarrow 2)$	3.347	5.469	1.634
Zr-N-H	$ZrN \cdot ZrH_x (x \rightarrow 2)$	3.288	5.468	1.663
Zr-B-H	None			
Zr-O-H	None			
Zr-Si-H	None			
Ti-N-H	$TiN \cdot TiH_2 (x \rightarrow 2)$	3.041	5.023	1.651
Ti-C-H*	$TiC \cdot TiH_2 (x \rightarrow 2)$	5.042	1.635	
U-C-H	None			

*From work of D. T. Peterson, Ames National Laboratory

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

The mechanism for ternary compound formation is now fairly well established. A necessary step is formation of a substoichiometric refractory compound lattice, which then reacts with hydrogen. TiN , for example, is unreactive with hydrogen, but TiN_{1-x} absorbs hydrogen readily. The substoichiometric structure is created by reaction of excess metal with the refractory compound.

The reaction of hydrogen with UC containing excess uranium results in formation of the simple binary hydride UH_3 . The carbide phase appears not to absorb any hydrogen.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

Very significant progress has been made in the three major areas of self-diffusion, fission gas mobility, and ternary hydride preparation.

The experimental methods for measuring self-diffusion of Zr in zirconium hydride have been perfected, and preliminary measurements made. These indicate significant contributions from both bulk volume and grain boundary diffusion. A further separation of these effects will be the subject of next year's activities.

The fission gas mobility studies have definitely established that migration of xenon through the hydride lattice is by some process more complicated than simple diffusion. Relatively impure zirconium hydride provides sites for heterogeneous nucleation, and prevents simple diffusion of rare gas atoms.

The mechanism for formation of ternary hydride compounds has been discovered, and many new compounds in the series have been synthesized. Identification and characterization have, thus far, been primarily by x-ray diffraction. Other properties will be measured in the coming year.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program:	General Research, Reactor Physics Studies		
Project:	Age Measurements		
Project Engineer:	H. A. Morewitz		
Reporting Period:	Fiscal Year 1965	AEC Category:	04-40-01-01
General Order:	7604	Subaccount:	1311

I. PROJECT OBJECTIVES

The objective of this project is to investigate the slowing-down properties of fission neutrons in various metal and moderator mixtures. These measurements are compared with Monte Carlo calculations, to determine the accuracy of the microscopic, fast, neutron cross-section data used in obtaining group-averaged cross-sections for reactor design codes. In many cases, a direct measurement of these cross-sections is not feasible over the energy range needed. In such cases, the cross-section must be calculated; in other cases the measured cross-section must be evaluated. Age measurements represent an indirect method of checking out these cross-sections.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. AGE OF IRON-WATER MIXTURES

The age measurements in several iron-water moderator mixtures were completed. The theoretical analysis has been compared in considerable detail with corresponding experiments.⁽¹⁾

Comparison of the Monte Carlo calculations with experiments for the neutron age and higher moments is presented in Table 1.

TABLE 1
NEUTRON AGE AND HIGHER MOMENTS IN IRON-WATER MIXTURES

Fe/H ₂ O		Age (cm ²)	M ₄ (10 ⁴ cm ⁴)	M ₆ (10 ⁸ cm ⁶)
0.465	calc.	30.5 ± 0.2	10.2 ± 0.2	1.3 ± 0.1
	exp.	30.3 ± 0.5	10.2 ± 0.3	1.3 ± 0.1
0.908	calc.	36.3 ± 0.2	13.0 ± 0.2	1.6 ± 0.1
	exp.	37.4 ± 0.5	14.1 ± 0.2	1.9 ± 0.1
1.737	calc.	47.3 ± 0.3	20.2 ± 0.2	2.8 ± 0.1
	exp.	46.4 ± 0.5	20.0 ± 0.3	2.9 ± 0.1

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

The effect of the inelastic scattering in iron on the neutron age and higher moments is given in Table 2.

TABLE 2
EFFECT ON NEUTRON AGE AND HIGHER MOMENTS OF
INELASTIC SCATTERING IN IRON

Fe/H ₂ O		Age (cm ²)	M ₄ (10 ⁴ cm ⁴)	M ₆ (10 ⁸ cm ⁶)
0.465	(*)	30.5 ± 0.2	10.2 ± 0.2	1.3 ± 0.1
	(†)	36.7 ± 0.3	18.0 ± 0.5	4.1 ± 0.4
0.908	(*)	36.3 ± 0.2	13.0 ± 0.2	1.6 ± 0.1
	(†)	46.7 ± 0.3	27.3 ± 0.7	7.0 ± 0.6
1.737	(*)	47.3 ± 0.3	20.2 ± 0.2	2.8 ± 0.1
	(†)	66.3 ± 0.5	52.3 ± 1.4	18.0 ± 2.0

*Inelastic scattering in iron included.

†No inelastic scattering in iron.

Calculations for several additional Fe/H₂O mixtures ($0.05 \leq \text{Fe}/\text{H}_2\text{O} \leq 0.465$) support the theory that the neutron age and higher moments are nonlinear functions of the Fe/H₂O volume ratio.⁽¹⁾ This nonlinearity appears to arise from competition between several effects of the replacement of water by iron. The first effect is the decrease in slowing down efficiency due to the reduced hydrogen atom density; this effect tends to increase the values of the neutron age and higher moments. The second effect is inelastic scattering in iron, which tends to decrease strongly the values of the age and higher moments, as shown in Table 2. A third effect, moderation of the neutrons by elastic collision with the iron nuclei, is expected to be small because of the great moderating power of hydrogen.

B. AGE OF ALUMINUM WATER MIXTURES

Age measurements in mixtures of aluminum and water were completed in assemblies with metal-to-water volume ratios of 0.500, 1.000, and 2.000. A preliminary theoretical analyses of the experiments have been made using three sets of aluminum cross-sections^(3, 4, 5) with a single set of angular data to describe the elastic scattering anisotropy in aluminum. The results of these calculations are presented in Table 3.

The effects on the neutron age and higher moments of inelastic scattering and isotropic elastic scattering in aluminum are being analyzed. The data from the experiments on aluminum-water mixture are still being reduced.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 3
NEUTRON AGE AND HIGHER MOMENTS IN
ALUMINUM/WATER MIXTURES

Al/H ₂ O	Cross-Section	Age (cm ²)	M ₄ (10 ⁵ cm ⁴)	M ₆ (10 ⁸ cm ⁶)
0.500	Ref. (3)	40.8 ± 0.3	1.92 ± 0.04	3.85 ± 0.25
	Ref. (4)	40.8 ± 0.3	1.95 ± 0.04	4.13 ± 0.22
	Ref. (5)	41.3 ± 0.2	1.97 ± 0.03	3.99 ± 0.16
1.000	Ref. (3)	57.0 ± 0.3	3.27 ± 0.05	7.01 ± 0.21
	Ref. (4)	56.5 ± 0.3	3.26 ± 0.05	7.21 ± 0.22
	Ref. (5)	58.4 ± 0.3	3.50 ± 0.05	8.12 ± 0.30
2.000	Ref. (3)	90.3 ± 0.04	7.21 ± 0.09	19.86 ± 0.80
	Ref. (4)	89.0 ± 0.4	6.94 ± 0.08	18.24 ± 0.41
	Ref. (5)	92.6 ± 0.4	7.52 ± 0.08	20.40 ± 0.44

C. PUBLICATIONS DURING FISCAL YEAR 1965

- J. A. DeJuren and R. K. Paschall, "Thermal-Neutron Transmission Through Plane Cadmium Covers," Nuc. Sci. and Eng. 20, 314 (1964).
- R. K. Paschall, "The Age of Fission Neutrons to Indium-Resonance Energy in Water," Nuc. Sci. and Eng. 20, 436 (1964).
- H. Alter, "Age of Fission Neutrons to Indium-Resonance Energy in Water (Part II - Theory)," NAA-SR-Memo 8862 (1964).
- R. K. Paschall, "The Age of Fission Neutrons to Indium-Resonance Energy in Zirconium-Water Mixtures (Part I - Experiment)," NAA-SR-Memo 9503 (1964); a less detailed account has been accepted for publication by Nuclear Science and Engineering.
- H. Alter, "The Age of Fission Neutrons to Indium-Resonance Energy in Zirconium-Water Mixtures (Part II - Theory)," NAA-SR-Memo 9965 (1964); a less detailed account has been accepted for publication by Nuclear Science and Engineering.
- R. K. Paschall, "The Age of Fission Neutrons to Indium-Resonance Energy in Iron-Water Mixtures (Part I - Experiment)," NAA-SR-Memo 10219 (1965); a less detailed account has been accepted for publication in the Journal of Nuclear Energy.
- H. Alter, "The Age of Fission Neutrons to Indium-Resonance Energy in Iron-Water Mixtures (Part II - Theory)," NAA-SR-Memo 10918 (1965); a less detailed account has been accepted for publication in the Journal of Nuclear Energy.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

Theoretical calculations of the neutron age and higher moments of the slowing-down distribution for several iron-water and aluminum-water mixtures were compared with results of the experimental program of age measurements. The reasonable agreement between measured and calculated values for the neutron age and fourth moments indicates that the cross-section and associated angular data used in the calculations are satisfactory. The discrepancy between the measured and calculated sixth moments is believed due to uncertainty in the extrapolation of the measured distribution to infinity. While the contribution of the extrapolated portion of the curve is negligible for the case of the second and fourth moments, it represents a large fraction of the measured sixth moment. The value of the measured sixth moment is highly sensitive to the method of extrapolation.

The technique of measurement and the theoretical analysis have been developed to a high degree, permitting routine measurements of the age and higher moments with high precision. The total, the elastic scattering, and the differential elastic scattering cross-sections for oxygen, hydrogen, carbon, zirconium, and iron have been validated to a satisfactory degree from 1 to 10 Mev.

REFERENCES

1. H. Alter, "The Age of Fission Neutrons to Indium Resonance Energy in Iron-Water Mixtures (Part II - Theory)," NAA-SR-Memo 10918, (1965).
2. R.K. Paschall, "The Age of Fission Neutrons to Indium Resonance Energy in Iron-Water Mixtures (Part I - Experiment)," NAA-SR-Memo-10219, (1965).
3. H. Alter and H.J. Lorenzi, "Preliminary Graphical Display of the Neutron Cross-Section Master Tape," NAA-SR-Memo 8904, August, 1963.
4. L. Forsberg, "Neutron Cross-Sections for Aluminum," AE-117, August, 1963.
5. E.D. Joanou and C.A. Stevens, "Neutron Cross-Sections for Aluminum," NASA-CR-54260, November 1964.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: General Research, Reactor Physics Studies

Project: Extrapolation Length Measurements

Project Engineer: H. A. Morewitz

Reporting Period: Fiscal Year 1965

AEC Category: 04-40-01-01

General Order: 7604

Subaccount: 1312

I. PROJECT OBJECTIVES

The objective of this project is to investigate the diffusion properties of neutrons in different materials such as water, graphite, and beryllium, using both steady-state and pulsed techniques. As the neutron temperature from a steady-state source is different from that of a pulsed source, a comparison between the two results provides a check on the accuracy of the scattering kernel. The technique is a fairly sensitive test, as small changes in the geometric size of the medium cause correspondingly small changes of the asymptotic neutron spectrum. A good scattering theory should be able to predict the diffusion parameters which may be calculated from these experiments.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. EXTRAPOLATION LENGTH, PULSED

Extrapolation length measurements were performed in a 4- and 6-in. water cube. Tentative results are about 0.52 cm for both cases. This is in disagreement with theoretical calculations of about 0.35 cm, but in agreement with a recently reported⁽¹⁾ value of 0.556 cm. The measurements reported here were done with a D-D as well as D-T source to check on the effect of room-return background. In addition, the delay after the neutron pulse prior to opening the counting gate was varied to check on the effect of harmonics. Any systematic errors introduced by higher harmonics or room-return background were not detected by the above mentioned techniques.

The $\alpha - B^2$ measurements in diphenyl were recently completed at 85°C. Although the data analysis is not yet finished, the values of the decay constants are much higher (~5%) than those reported by Blackshaw and Waltner⁽²⁾ at 77°C. We have taken considerable pains to eliminate room-return background by using the D-D instead of the D-T reaction for small sizes, and by surrounding the moderator with massive amounts of paraffin and boron. It is questionable if valid data can be taken beyond a buckling of $\sim 0.22 \text{ cm}^{-2}$. Measurements at 105°C have started.

Measurements⁽³⁾ of the shape of the fundamental mode of a thermalized neutron pulse in graphite for rectangular stacks have yielded a value of the extrapolation length, $d = 1.825 \pm 0.025 \text{ cm}$. The pulsed-neutron decay constants in graphite have been measured over a buckling range from 1.946×10^{-3} to $1.230 \times 10^{-2} \text{ cm}^{-2}$, and fitted to 3- and 4-term power series in the buckling. To fit points at bucklings greater than $6.0 \times 10^{-3} \text{ cm}^{-2}$, a cubic fit is necessary. An iterative method of fitting the decay constants results in an extrapolation

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

length of 1.78 ± 0.01 cm. The diffusion constant was $(2.0896 \pm 0.0093) \times 10^5$ cm²/sec, and the diffusion cooling constant was $(3.77 \pm 0.35) \times 10^6$ cm⁴/sec at a density of 1.689 g/cm³. A technique of correcting the decay constants for the effect of spectral cooling has been developed.^(4,5) The decay constants corrected to room temperature can be fitted as a linear function of the buckling. This method iterates on the heat-transfer coefficient, γ , and gives a value of $\gamma = 1633 \pm 89$ sec⁻¹ for graphite. The heat-transfer coefficient relates the rate of energy transfer from a moderator to a cooled-neutron spectrum. With this approach, the entire buckling range can be fitted with three parameters.

The pulsed measurements in graphite were concluded during this fiscal year, and a paper was accepted for publication by Nuclear Science and Engineering.

B. THEORY

The analysis of pulsed decay experiments using the concept of the thermalization time constant was developed^(3,4) and applied to graphite, beryllium, light, and heavy water measurements performed at this and other laboratories. This technique is superior to the usual parametric analysis for the following reasons.

1. Because of the fewer constraints, D_0 can be obtained with a smaller error.
2. The analysis of the data does not depend upon the number of terms employed in a power series of the buckling.
3. The thermalization time-constant has a precise physical meaning (relaxation time for a thermal spectrum to approach its maxwellian equilibrium distribution). It is a value which can be calculated from basic concepts. For a finite medium the thermalization time constant, t , is given by

$$\frac{1}{t} = \frac{1}{t_\infty} + \frac{2w}{3} D_B B^2 = \frac{1}{t_\infty} \frac{T_0}{T_B},$$

where

- D_B = the diffusion constant,
- T_B = the temperature of the spectrum,
- T_0 = the temperature of the medium, and
- w = defined by $D_B = D_0 (T_B/T_0)^w$.

C. STEADY-STATE EXTRAPOLATION LENGTH

Measurements of extrapolation length and diffusion length have been completed for graphite stack sizes of 4 x 4 x 24 in., 12 x 12 x 50 in., 20 x 20 x 75 in., and 28 x 28 x 100 in. Neutron flux distributions were obtained by beta-counting Dy-Al foils which had been irradiated at various locations in the stacks. The vertical measurements were made about 1/3 of the way out from the center along a diagonal of the stack in order to minimize third harmonic effects near the bottom arising from mismatch between the reactor thermal column and the stack. Horizontal measurements made near the bottom of the stack were corrected for third harmonic effects. The horizontal distributions were fitted to a cosine curve and the vertical measurements to an exponential curve.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Because of the crystalline structure of graphite, neutrons having energies below the Bragg cut-off energy will have a negligible scattering cross-section and therefore a very long mean free path. In small graphite stacks, neutrons above the Bragg cut-off energy will be scattered out the side of the stack much faster than neutrons of lower energy. The smaller the stack size, the more significant the effect will be. The standard equation for determining the diffusion length, $1/L^2 = \gamma^2 - 2(\pi/a)^2$, is not expected to be valid in stack sizes where a significant number of cold neutrons are present. The diffusion length is imaginary for stack sizes smaller than about 20 x 20 in.

Measurements have also been made in a 36 x 36 x 74 in. stack and in a 44 x 44 x 74 in. stack, but the results have not been analyzed yet.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

Analysis of die-away experiments has been developed in terms of γ , the neutron-moderator heat transfer coefficient. This technique has been demonstrated to be superior to the standard parametric analysis methods. Codes have been developed to calculate γ and the error associated with it.

The pulsed neutron experiments in graphite were completed.

As a result of these efforts, the interpretation of pulse neutron experiments has been placed on a firm foundation and it is expected that comparisons with the steady-state experiments can now be meaningful.

REFERENCES

1. Gaerttner, et al., "January-March 1965 Progress Report of R. P. I. Linear Accelerator Project"
2. G. L. Blackshaw and A. W. Waltner, J. Nuc. Energy 17, 341 (1963)
3. S. Davis, J. A. DeJuren and M. Reier, "Pulsed-Decay and Extrapolation-Length Measurements in Graphite," Nuc. Sci. and Eng. (to be published) (1965)
4. J. A. DeJuren, "Spectral Decay Correction of Pulsed Neutron Data," Symposium on Pulsed Neutron Research, Karlsruhe, Germany (1965)
5. J. A. DeJuren, R. Paschall and E. Vaughn "SCAP, an IBM 7094 Code for the Spectral Analysis of Pulsed-Neutron Data," NAA-SR-Memo 10941 (1965)

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Program: General Research, Reactor Physics Studies

Project: Cross-Section Analysis and Applications Project

Project Engineer: H. A. Morewitz

Reporting Period: Fiscal Year 1965

AEC Category: 04-40-01-01

General Order: 7604

Subaccount: 1313

I. PROJECT OBJECTIVES

The objectives of this project are related to the production of a good, up-to-date, basic set of nuclear data. To accomplish this we intend to: (a) survey, analyze, and compile existing experimental and theoretical information on neutron cross-sections and angular data; (b) provide automated accessibility to users of cross-section data (microscopic data, group libraries); (c) provide automated reference to and retrieval of cross-section information; and (d) use nuclear model calculations to help fill gaps in cross-section data.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. LIBRARY DEVELOPMENT

Group constants consisting of the absorption, transport, fission, and production ($\nu\sigma$) cross-sections, as well as the total transfer matrix (elastic plus inelastic) were calculated by the processing programs GRAVE,⁽¹⁾ PRISM⁽²⁾ and CONTRACT⁽³⁾ for the following isotopes/elements.

Hydrogen	Potassium	Uranium ²³³
Deuterium	Chromium	Uranium ²³⁴
Beryllium	Iron	Uranium ²³⁵
Carbon	Nickel	Uranium ²³⁶
Oxygen	Zirconium	Uranium ²³⁸
Sodium	Molybdenum	Plutonium ²³⁹
Aluminum	Thorium ²³²	Plutonium ²⁴¹

The group constants, with the exception of the transport cross-section, were produced by averaging the microscopic data over a flux spectrum calculated by the ELMOE⁽⁴⁾ code for a given isotopic mixture. The transport cross-section was then averaged over the current spectrum computed by ELMOE for the same mixture. This latter weighting procedure was used since analysis shows that this method provides the best values of the diffusion coefficient.

The group-averaged absorption, transport, and fission cross-sections were obtained with the GRAVE program, while the total transfer matrices were computed with the PRISM code. Available inelastic level data were used in calculating the total transfer matrices, and the evaporation model was used when no level data were available. The relation between energy loss and angle of scatter for elastic scattering was rigorously taken into account by means of a 9th order Legendre expansion of the differential elastic scattering cross-section.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

7604-2501
Figure 1

7604-2502
Figure 2

7604-2503
Figure 3

7604-2504
Figure 4

7604-2505
Figure 5

7604-2506
Figure 6

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Sixteen- and 23-group cross-section libraries were generated for the transport theory code DTF⁽⁵⁾ (a Fortran version of DTK), as well as 16- and 18-group constants for the diffusion theory codes AIM,⁽⁶⁾ FAIM,⁽⁷⁾ and CAESAR.⁽⁸⁾ Four multigroup libraries in a 12-group structure were prepared for analysis of a plutonium-uranium core design study. An additional four multigroup libraries in 15-group structure were prepared to aid in an analysis related to the Argonne cross-section comparison. Eight libraries in 15-group structure were prepared to help analyze the sodium coupled core reactor design. The distinguishing feature of this multitude of group libraries was that each library used a unique neutron spectrum as the cross-section weighting function.

A 54-group library based on up-to-date nuclear data was prepared for FORM⁽⁹⁾ (a Fortran version of MUFT), and is currently available for the following isotopes/elements: hydrogen, deuterium, carbon, oxygen, aluminum, chromium, iron, nickel, and zirconium.

A special 35-group library designed for fast reactor studies was developed for use with the CAESAR and/or ULCER⁽¹⁰⁾ programs.

Additionally, in order to evaluate a series of ZPR-III critical experiments, four multigroup cross-section libraries in a 35-group structure have been prepared. The libraries, which include group constants for carbon, oxygen, aluminum, chromium, iron, nickel, U²³⁵, and U²³⁸, are used with the ULCER diffusion theory code.

B. OPTICAL MODEL DEVELOPMENT

Several nuclear model codes, ABACUS II,⁽¹¹⁾ OPTIC,⁽¹²⁾ NEARREX,⁽¹³⁾ and 2 PLUS⁽¹⁴⁾ were made operation in the Atomics International computing system. Data produced by these programs have been used to fill gaps in measured data as the need for knowledge of the information became acute. The computer code, 2 PLUS, was developed to treat a deformed even-even nucleus with a 0+ ground state and one excited level (2+). This code was used to analyze elastic and inelastic scattering data in the energy region from 500 kev to 3 Mev for Ti, Ci, Fe, Ni, Zr, Th²³², and U²³⁸ with promising results.

Additional modifications to the program, 2 PLUS, have been made which greatly improve its calculational capabilities. This was accomplished by treating additional levels as spherical model levels according to the standard Hauser-Feshbach compound nucleus model. This new version of 2 PLUS has been used to calculate angular distributions and strength functions for Th²³² and U²³⁸. The nuclear deformations were taken from electromagnetic measurements of Stelson and McGowan.⁽¹⁵⁾ During the analysis, the only nuclear well parameters which varied with energy or nuclide were the real well depth and the imaginary well depth. The imaginary well depth was determined chiefly from fitting measured values of the neutron S-wave strength functions for each nuclide. The experimental values of the strength functions were supplied from an analysis of measured resonance parameters.

Figures 1 through 8 represent final calculations at two-incident neutron energies for Th²³², and two energies for U²³⁸. There is only one measured inelastic angular distribution for each isotope (~500 kev). All other inelastic

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

7604-2507
Figure 7

7604-2508
Figure 8

Figures 1 through 8. Final Calculations at Two-Incident Neutron Energies

angular distributions are predictions based on fitting the experimental elastic angular distributions at each energy. At energies above 1 Mev, it was assumed that there were so many available final-state channels that the compound nucleus contribution to any one would be negligible.

This analysis has been compared with the spherical optical model analysis of Th^{232} and U^{238} by Auerback and Moore.⁽¹⁶⁾ The improvements in these calculations are twofold; first, a consistent set of optical model parameters for both isotopes, and second, better fits to the experimental inelastic angular distributions.

It has been noted that the deformation has a marked effect on the diffraction pattern of the elastic angular distribution. In addition, there appears to be an important forward scattering effect for inelastic scattering which cannot be accounted for by a spherical model Hauser-Feshback calculation.

There are several difficulties apparent in the accompanying figures. First, the diffraction patterns of the elastic angular distribution do not match the experiments exactly. Since the coupling of inelastic channels has a strong effect on these patterns, the extension of this model to include a second excited level is needed. There also appears to be insufficient compound nucleus elastic scattering at the lower energies, as evidenced by the discrepancies at large angles. An incorporation of Moldauer's modification to Hauser-Feshback's theory would tend to correct this discrepancy.⁽¹⁷⁾

C. PROCESSING PROGRAM DEVELOPMENT

A tape conversion program, TCP, which permits intercomparison between Aldermaston and Atomic International Evaluated Nuclear Data Files, was completed. Several examples are shown in Figures 9 through 14. This program is being extended in order to intercompare with the data files of Los Alamos, Livermore Radiation Laboratory, and Brookhaven National Laboratory.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

7604-2509

Figure 9

7604-2510

Figure 10

7604-2511

Figure 11

7604-2512

Figure 12

7604-2513

Figure 13

7604-2514

Figure 14

Figures 9 through 14. Examples of Comparisons by use of a Tape Conversion Program

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

The program CHAD which automates the handling of angular distribution data was completed. Also completed were CONTRACT, a program to reformat GRAVE/PRISM punched output, and MALINDA, a program to manipulate and list Nuclear Data.

D. AI NUCLEAR DATA FILE

Angular distribution data in the form of LEGENDRE expansion coefficients have been compiled for all isotopes contained on the Atomics International Evaluated Nuclear Data File (AIENDF). Inelastic scattering data for excitation of individual nuclear levels was compiled for the following element/isotopes: carbon, oxygen, sodium, aluminum, iron, uranium²³⁵, uranium²³⁸, and plutonium²³⁹.

An intensive literature review was completed as of 6/65 in which the latest available data on U²³⁵, U²³⁸, Pu²³⁹, Pu²⁴⁰, Pu²⁴¹, Pu²⁴², xenon, and samarium were collected. This information is being analyzed and evaluated preparatory to updating and extending the AIENDF.

Resolution of the discrepancy in the Be (n, 2n) reaction cross-section threshold was accomplished. Two experiments in which the reaction cross-section was folded into the fission spectrum gave similar results,

$$\sigma_{n,2n} = 107.5 \pm 4 \text{ mb and } 115.5 \pm 16 \text{ mb} .$$

When the reaction threshold is taken at 1.84 Mev., the fission spectrum integrated cross-section is 151 mb; when the threshold is taken at 2.7 Mev, the computed result is 114 mb. Since the Be (n, 2n) cross-section is known reasonably well above ~3.7 Mev, the line for the excitation function is constructed between 3.7 Mev and the threshold at 2.7 Mev from phase space momentum considerations. The Be (n, 2n) cross-section, in this interval, is obtained by choosing the cross-section value at 3.7 Mev and letting it fall to the threshold as $(E-2.7)^{1/2}$.

E. PUBLICATIONS DURING FISCAL YEAR 1965

NAA-SR MEMO-10626, "Production of Hydrogen and Helium in Reactor Fuel-Cladding Material," H. Alter (1964).

"Atoms International Automated Cross-Section Program," H. Alter and H. J. Lorenzi. Transactions of Am. Nuc. Society V, 243, (1964).

"Optical Model Analysis of Neutrons Scattered from Non-Spherical Nuclei," C. L. Dunford, J. T. Reynolds, and H. Fenech. To be presented at APS meeting, 6/65, at New York.

"Calculation of (n, p) Cross-Sections," N. C. Francis, C. L. Dunford, and C. R. Lubitz. Transactions of Am. Nuc. Society 8, 215, (1965).

NAA-SR-TDR-10884, "CONTRACT, A Program to Reformat GRAVE/PRISM Punched Output," R. A. Baline and B. Lemke, (January 1965).

NAA-SR-MEMO-10008, "MALINDA, Manipulation and Listing of Nuclear Data," D. W. Roeder (October 1964).

NAA-SR-MEMO-11231, "CHAD, Code to Handle Angular Data," R. F. Berland (June 1965).

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

A system of Fortran programs which generate multigroup constants has been written and extensively checked out for the IBM 7094. The system is highly automated in that, given the user's input specifications, the system extracts the microscopic cross section data from magnetic tapes, the Atomics International Evaluated Nuclear Data File (AIENDF), averages the desired data over either a specified or calculated spectrum, and then forms group constants in either a few-group or a man-group structure.

Doppler-broadened cross-sections over the resolved region are optionally calculated by the methods of Chernick and Vernon⁽¹⁸⁾ or by the intermediate resonance representation of Goldstein and Cohen,⁽¹⁹⁾ which treats interference between resonance and potential scattering. Unresolved resonances for $l = 0$ and $l = 1$ neutrons are calculated in the narrow resonance approximation, with scattering interference optional. A Doppler-broadening table for the interference-scattering component in Rothenstein's small-interference approximation is incorporated, as is a Doppler "J" table of improved accuracy.

The average number of neutrons emitted per fission is represented by polynomial fits to the measured data, and the coefficients for these polynomials are stored as the AIENDF.

Two complementary procedures are used to calculate matrices for inelastic scattering: the energy levels of the target nucleus are taken as discrete when both the levels and their probabilities of excitation as functions of the energy of the incident neutron are known or can be reasonably estimated; when such detailed information is lacking and above the region of resolved levels, the calculation is based on the Weisskopf evaporation model. Level excitation data are stored as the AIENDF, while data on the nuclear temperature are obtained from the Weisskopf level density formula.

Elastic-scattering matrices are generally computed by permitting the discrete level treatment for inelastic scattering to degenerate to the special use of scattering by a single level with zero threshold energy. The elastic scattering treatment of light elements has been adapted from the ELMOE program.

The final averaged group constants are punched on cards, binary or decimal, or placed directly on magnetic tape, in formats suitable for direct use with the transport theory, diffusion theory, slowing-down theory, and Monte Carlo neutronic codes in use at Atomics International.

The above mechanism by which multigroup libraries are rapidly constructed has been improved and refined. Comparison of such libraries with a series of critical experiments provides a means of evaluating the library group constants directly and the basic microscopic data indirectly. Continued progress in improvement of the processing programs, CHAD, PRISM, and TCP (Tape Comparison Program), is bringing the concept of a highly automated nuclear data system closer to fruition. A method of averaging the transport cross-section over a current spectrum has been shown to produce more acceptable diffusion coefficients than the previously accepted method of reciprocally averaging this cross-section with a flux spectrum. Modifications to existing nuclear model programs, graphical display of results, punchout options to provide card data for use with processing programs have enhanced the usefulness of these programs in view of the overall objectives of the cross-section program.

REFERENCES

1. R. A. Blaine et al., "GRAVE, A Group Averaging Program to Produce Multigroup Cross Section Libraries," NAA-SR-MEMO-9267 (1964)
2. B. D. O'Reilly et al., "PRISM, A Program to Calculate Total Transfer Matrices," NAA-SR-MEMO-9500 (1964)
3. R. A. Blaine and B. J. Lemke, "CONTRACT, A Program to Reformat GRAVE/PRISM Punched Output," NAA-SR-TDR-10884 (January 1965)
4. A. L. Rago et al., "ELMOE, an IBM 704 Program Treating Elastic Scattering Resonances in Fast Reactors," ANL 6805 (January 1964)
5. Internal NAA document - to be published
6. H. P. Flatt and D. C. Baller, "The AIM-6 Code" NAA Program Description (internal document) January 1961
7. D. C. Baller, "The FAIM Code, A Multigroup, One-Dimensional Diffusion Equation Code," NAA-SR-7137 (April 1962)
8. D. C. Baller, "CAESAR, A Multigroup, One-Dimensional Diffusion Equation Code," AMTD-200 (April, 1963)
9. D. J. McGoff, "FORM, A Fourier Transform Fast Spectrum Code for the IBM-709," NAA-SR-MEMO-5766 (September 1960)
10. D. S. Vargofcak et al., "ULCER, A One-Dimensional, Multigroup Diffusion Equation Code with Upscatter," NAA-SR-MEMO-9891 (May 1964)
11. E. H. Auerbach, "ABACUS-2," BNL 6562 (November 1962)
12. C. R. Lubitz, private communication
13. P. A. Moldauer et al., "NEARREX" - ANL 6978 (1965)
14. C. L. Dunford, PhD Thesis, Massachusetts Institute of Technology (July 1964)
15. P. M. Stelson and F. H. McGowan, Annual Review of Nuclear Science 13, 163 (1963)
16. S. O. Moore and E. H. Auerbach, BNL 818 (T-317)
17. P. A. Moldauer, Phys. Rev. 123, 968 (1961)
18. J. Chernick and A. R. Vernon, NSE, 13, 649, (1958)
19. R. L. Goldstein and E. R. Cohen, NSE, 13, 132, (1962)

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program:	General Research, Reactor Physics Studies		
Project:	Fast Spectrum Doppler Measurements		
Project Engineer:	H. A. Morewitz		
Reporting Period:	Fiscal Year 1965	AEC Category:	04-40-01-01
General Order:	7604	Subaccount:	1330

I. PROJECT OBJECTIVES

The objectives of this project are (a) to further refine the techniques for measuring Doppler reactivity coefficients, (b) to investigate the Doppler coefficients of various fuel and fertile materials in a variety of fast spectra, (c) to provide better understanding of temperature-induced reactivity changes by analytically and experimentally investigating these changes at cryogenic as well as very high temperatures, and (d) to investigate resonance-overlap and interference effects, and the effect of varying the surface-to-mass ratio.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

Measurements have been made in three different test regions designated as Cores 10B, 12, and 13. Core 10B simulated an unmoderated fast reactor and had a calculated median fission energy (mfe) of 195 kev at the center; Core 12 differed only in having a greater aluminum density which produced a mfe of 156 kev. Core 13 simulated a 30 v/o carbon-moderated reactor with a mfe of 62 kev. This range of spectra was further extended by surrounding the sample position at the center of the reactor with polyethylene blankets of various thicknesses.

Primarily, measurements were made of the reactivity and temperature coefficients of reactivity of uranium²³⁵ (metal and oxide), uranium²³⁸ (metal and oxide), and thorium (metal), and the variation of reactivity with surface-to-mass ratio of the metals.

The blankets used consist of polyethylene plates of various thicknesses which were substituted for core materials around the sample position at the center of the test region. Diffusion theory calculations are made for each blanket to obtain an estimate of the median fission energy at the center for use as a spectral indicator. Polyethylene blankets of thicknesses 0.125, 0.160, 0.213, 0.250, 0.375, and 0.500 in. were used in Core 13, producing spectra with median fission energies of 16.3, 10.4, 5.2, 3.2, 0.75, and 0.27 kev. In Core 10B, the 0.125-in. blanket produced a spectrum with a median fission energy of 29.3 kev. These values should be taken as only approximate, due to the poor simulation by the calculation of the geometrical arrangement of the polyethylene. A small gap exists in the blanket at the parting plane of the separable-half reactor and this produces a severe perturbation in the low-energy flux, as indicated by fission traverses with U²³⁵ and Pu²³⁹ fission counters.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Blankets composed of boron carbide have also been used, but the results are somewhat less easy to interpret due to the general flux depressions caused by absorption.

As a sample is heated in a spatially uniform neutron flux, its reactivity may change due to thermal expansion of the sample and thermal agitation of the constituent atoms. The first of these reactivity effects is termed the expansion effect; the second, the Doppler effect. Both of these effects vary considerably according to the neutron energy spectrum, and for fissile materials the expansion effect changes from positive at low energies to negative at higher energies. The Doppler effect, at least for U^{235} , may exhibit the reverse of this behavior, that is, negative at low energies and positive at higher energies.

The temperature coefficients of thorium and uranium²³⁸, which in most spectra are predominantly due to the Doppler effect, are shown in Figure 1 for different calculated median fission energies. In soft spectra, the Doppler effect in thorium is very strong, providing a good demonstration of the variation of the reactivity change with temperature. This variation is displayed in Figure 2, where the change in the reactivity worth of thorium reactivity with temperature is shown for several blankets. The curvature, which is predicted by theory, is readily apparent.

7604-2515

Figure 1. Temperature Coefficient of Thorium and Uranium²³⁸ for Different Calculated Median Fission Energies

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Figure 2. Variation of Reactivity of Thorium With
Temperature in 0.125-in. and 0.375-in.
Polyethylene Blanket in Core 13

7604-2516

The competition between the expansion effect and the Doppler effect is much more severe in U^{235} , and so considerable effort has been required to separate the two. Determination of the expansion effect has been attacked by several methods. Perhaps the most precise of these involves comparison of the reactivity of a full-density U^{235} sample with that of a reduced-density sample. The full-density sample weighs 184 gm, and has a density of 18.9 gm/cm^3 ; the reduced-density sample is composed of pellets produced by a powder metallurgical technique and weighs the same, but has a density of 15.2 gm/cm^3 . Thus it simulates the spatial distribution of a sample at a very high temperature (actually far above the melting point), but whose atoms are at room temperature. The results of measurements by this method for various calculated median fission energies are shown in Figure 3. A similar measurement in Core 12 with full- and reduced-density pellets weighing 19.4 gm agrees with the full-size sample results.

As a sample expands, its reactivity may change due to two effects, namely, the change in absorption of incoming neutrons because of the reduction in self-shielding, and the change in absorption of neutrons produced in the sample because of the increase in the escape probability. The self-shielding effect tends to be positive while the second effect, termed the self-multiplication effect, is negative. Theoretical calculations of the balance of these effects have been made for several core-blanket configurations and the results are shown in Figure 3. The discrepancy between the calculated and experimental values for soft spectra may be due to neglecting resonance self-shielding in the calculations.

7604-2517
Figure 3. Reactivity Change Due to Expansion of U^{235} for Different Calculated Median Fission Energies

7604-2518
Figure 4. Determination of Doppler Reactivity Change by Difference Between Measured Total Reactivity and Measured Expansion Reactivity in 0.500-in. polyethylene Blanket in Core 13

Measurements with a reduced-density sample of U^{238} and a thorium-zirconium alloy sample indicate a much smaller contribution by the expansion effect; in core 10B, approximately 2% of the U^{238} temperature coefficient was due to the expansion effect, while in Core 13, with the 0.375-in. polyethylene blanket, approximately 15% of the thorium temperature coefficient was due to the expansion effect.

By combining the measurements of the expansion effect and the temperature coefficient of U^{235} , a measure of the Doppler effect may be obtained. This is shown for Core 13 with the 0.500-in. polyethylene blanket in Figure 4. By applying this method to all the measurements with U^{235} , the variation of Doppler reactivity with spectrum may be obtained. Figure 5 shows the measured values for the expansion effect and the total temperature effect of U^{235} as a function of median fission energy. The Doppler reactivity obtained by subtracting the expansion effect from the temperature effect is also shown.

7604-2519
Figure 5. Variation of Reactivity of U^{235} Due to Temperature and Expansion and Derived Doppler Reactivity for Different Calculated Median Fission Energies

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

An additional method has been applied to the determination of the expansion effect in U^{235} . This method takes advantage of the density change associated with the transition from the α -phase to the β -phase at 665°C . Reactivity measurements are made with the sample below this temperature and just above. The results of several sets of measurements are shown in Figure 6. These sets are normalized in the 600 to 650°C temperature range. An abrupt reactivity change at the transition temperature is evident.

7604-2520
Figure 6. Variation of Reactivity of U^{235} With Temperature Showing Discontinuity at α - β Phase Transition Temperature

Quantitative interpretation of the α - β results has been hampered due to a discrepancy between the published values of expansion across the α - β transition and the values determined for this sample by radiography. The length of the sample at different temperatures has been measured by means of radiographs taken of the heated sample; for precise measurement of the change in length caused by the phase change, double exposure radiographs have been made. In this technique, the film is lightly shielded during the exposure at a temperature just below the transition temperature, the shield removed, and another exposure made at a temperature just above the transition temperatures. The results of these measurements are compared with published values for linear thermal expansion in Figure 7.

The variation of U^{235} reactivity with surface-to-mass ratio was investigated in detail for Core 13 and Core 13 with a 0.375-in. polyethylene blanket, by measuring the reactivity of several different samples. These results are shown in Figure 8. It is significant that while the Core 13 results indicate a decrease in specific reactivity with increasing surface-to-mass ratio, in the Core 13, 0.375-in. blanket the specific reactivity behaves conversely, increasing sharply as the surface-to-mass ratio increases. Similar measurements have been made using a stack of foils whose surface-to-mass ratio is varied by varying the length

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

7604-2521

Figure 7. Thermal Expansion of U^{235} Determined
by Radiographic Measurements of a Doppler-
Experiment Sample

7604-2522

Figure 8. Variation of Reactivity of U^{235} With Surface-to-Mass Ratio in
Core 13 and Core 13 With 0.375-in. Polyethylene Blanket

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

of the stack, and a rolled foil whose surface-to-mass ratio is varied by varying the diameter of the roll.

A cryogenic capsule was fabricated which used helium or nitrogen gas cooled by liquid nitrogen to cool the sample. Nitrogen gas proved unsatisfactory, due to liquefaction of the gas in the cooling coils. The lowest temperature attained was -165°C ; the reactivity change induced in a thorium sample was -0.005δ at -100°C . In order to achieve lower temperatures with greater control, the cryogenic capsule has been redesigned using a miniature Joule-Thomson refrigerator (Cryo-Tip, Air Products and Chemicals, Inc) as the heat sink. Temperatures as low as 20°K are anticipated.

Several measurements have been made to detect possible artifacts inherent in the techniques used. None of these has indicated the existence of artifacts which seriously affect the results of the current measurements. Briefly, these results are: comparison of the statistical distribution of a large number of individual identical measurements with the predicted distribution resulted in excellent agreement; oscillation of the temperature of a heater just outside the reactor indicated a negligible contribution; measurement of the noise reactivity of the reactor showed that the drift in reactor power produces a small negative reactivity signal constant in time, and so is cancelled in analysis of the data; measurement of the temperature coefficient of reactivity of the Lavite insulator material (the only significant material other than the sample that is heated) indicated a null effect. Effects due to the presence of U^{238} in the U^{235} sample, and U^{235} in the U^{238} sample are small and readily corrected.

The laboratory selected for fabrication of the plutonium dioxide samples experienced insurmountable difficulties and the order was cancelled. Another laboratory is being selected and will fabricate samples of Pu^{241} and high burnup plutonium also.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

Techniques for measuring the temperature coefficients of fissile and fertile materials in a wide range of neutron spectra have been well developed. Separation of the expansion effect from the total temperature effect has been demonstrated in the case of U^{235} . Design of an effective cryogenic capsule is proceeding, but has not reached the proof stage yet. Measurements with plutonium have been delayed due to fabrication difficulties.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Program:	Reactor Fuels and Materials -- High Temperature Materials Development		
Project:	Structure, Properties, and Fission Effects in UC and Other Refractory, Interstitial Fuels		
Project Engineer:	C. E. Weber		
Reporting Period:	Fiscal Year 1965	AEC Category:	04-40-02-09
General Order:	7551	Subaccount:	4539

I. PROJECT OBJECTIVES

The objectives of this project are as follows.

- 1) To use and develop, where necessary, the most advanced methods to prepare pure single and polycrystalline material of controlled structure and stoichiometry.
- 2) To precisely determine selected microstructural attributes and mechanical and physical properties required both to establish the potential for these materials as nuclear fuels and to provide a basis for interpreting and understanding irradiation behavior in terms of atomistic transport phenomena.
- 3) To apply current solid-state theory developed for metallic fuels to obtain an understanding of the processes and mechanisms involved in fission damage in order.
- 4) To apply knowledge gained therefrom, and also apply to these materials the general theory and experimental techniques developed to reduce and control swelling in metallic fuels and demonstrate such control in these refractory fuels.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. SECOND-PHASE DISPERSIONS IN UC

One type of improvement in the performance of carbide-type fuels being sought in this program is that of increased resistance to fission gas swelling at high temperature. Since it is expected that swelling can be at least partially controlled by incorporation of many small particles in the carbide, analogous to the dispersed phases successfully developed for metal fuels, a part of this program has been to produce uranium monocarbide containing dispersed particles of a second phase that are stable to temperatures well over 1200°C. At this stage of the development, composites consisting of UC with dispersed tungsten particles have been prepared which have desirable chemical, physical, and mechanical properties; and the second-phase particle concentration and size distribution fall in the acceptance range, although towards the minimum of that range. Preparations are being made for the next step, that of testing the swelling resistance during irradiation to high burnup of this quality material, to establish the effectiveness of 5×10^{12} particles/cc in fuel stabilization.

1. Preparation by Zone-Melting

The induction zone-melting procedure, by which a molten zone is passed along the length of a cast UC rod, has been adapted to the preparation of

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

tungsten-doped UC by winding a tungsten wire around the UC rod prior to zone melting. In this way, 0.5 to 2.5 wt % tungsten has been incorporated in the UC, the tungsten dissolving in the UC in the molten zone and precipitating out as particles on cooling. Similar experiments have also been made with molybdenum.

2. Characterization of W-Doped UC

Second-phase dispersed particles cannot be observed by optical metallography in UC containing tungsten because the tungsten particles are too small. The effect of tungsten addition can be observed optically in that the normally smooth grain boundaries in pure UC are changed to a convoluted or meandering shape as shown in Figure 1.

7551-2501

Figure 1a. Pure,
Slightly Hyper-
stoichiometric UC

7551-2502

Figure 1b. One wt % W
Stoichiometric UC

Particles were shown by electron microscope replicas. Pure UC is compared with W-doped UC at two different concentrations in Figure 2. Particles are present at about 0.2 microns in diameter and at a density of about 4×10^{12} particles per cc. Particles have also been seen in transmission electron microscopic examination of thin sections, but technique difficulties have thus far prevented us from obtaining an adequate photograph.

7551-2503

Figure 2a. Zone Melted UC Control
2b. 0.78 wt % W-Doped UC
2c. 1.2 wt % W-Doped UC

These electron microscope observations leave no doubt that fine particles are present, and that they are caused by the addition of tungsten. Further evidence was needed to identify the particles. Electron microprobe examination showed only that tungsten was present in uniform distribution, since the particles were too small to be resolved. The uniform distribution of tungsten throughout the doped samples, as determined by the electron microprobe, is shown by the data of Table 1. Further evidence was the finding of a definite x-ray pattern for tungsten. Together with the above information, the x-ray finding permits the inference that the tungsten-containing particles are indeed tungsten metal, or chiefly tungsten metal.

Preparation by zone-melting involves sweeping a molten zone along the length of the rod; and thus there is the question of a possible concentration gradient along the length. Chemical analyses to investigate this point confirmed that there was a gradient, but that it is a rather small one. In three different rods, concentrations of tungsten at points along the rod about an inch apart were 0.63 and 0.82% W, 0.60 and 1.22% W, respectively. In each case the tungsten concentration increased in the direction of motion of the molten zone. In an experiment using an uneven winding of tungsten on the UC rod, it was possible to actually reverse the tungsten gradient.

3. Phase Equilibria in the System UC-W

The occurrence of tungsten as free metal in UC is consistent with portions of the phase diagram for the system, as reported recently by Chubb and Keller of BMI. Although these authors found a ternary compound, UWC_2 , in the composition region of our experiments, free tungsten was a stable phase in contact with UC. The interesting region of the system at low tungsten concentration has not been investigated until now. A series of W-containing UC mixtures has been

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 1
ELECTRON MICROPROBE ANALYSIS FOR TUNGSTEN IN UC
(in counts/20 sec)

Sample Description	Location in Sample		
	Center	Intermediate	Edge
Control (no tungsten)	1255	-	-
	1289	-	-
	1277	-	-
	1343	-	-
	1329	-	-
	1299	-	-
"1%" tungsten 0.73% tungsten	1530	1684	1620
	1631	1641	1556
	1687	1585	1570
	1565	1652	1595
	1627	1655	1597
	1576	1641 1616	1604 1697
2.5 wt % W (nominal) 1.15% W	2001	1863	1867
	1920	1897	1903
	1912	1951	1914
	2004	1859	1879
	1904	1861	1893
	1957	1890 1910	1889 1858

prepared for annealing and quenching experiments to provide data on the extent of solid solubility of W in UC, which we expect to show a maximum at the temperature of the pseudo-binary eutectic in the system. The data reported in the preceding paragraph on tungsten gradients in zone-refined rods, together with analyses of the molten zone at the end of each run, give a measure of the distribution between liquid and solid phases at various tungsten levels. This yields a tentative indication of a portion of the diagram above the eutectic temperature, subject to possible deviations from equilibrium during zone refining. Ratios varying from 1.4 to 2.3 indicate the approximate shape of the liquidus and solidus curves just below the melting point of pure UC, but no temperature values are available. A solid solubility maximum of a few per cent tungsten seems probable, but experiments in progress are expected to yield precise values for a series of temperatures.

A knowledge of the phase diagram in the low-tungsten region is pertinent to planning and interpretation of heat treatment experiments as a means for controlling the particle size and number of particles of dispersed tungsten. The zone-refining procedure involves a moderately fast cooling rate, and fortuitously, this cooling schedule has produced a particle distribution believed to be suitable for swelling control. However, it is desirable and may be possible to produce

finer and more numerous particles by solution at high temperature followed by nucleation and growth by annealing at a somewhat lower temperature. Heat treatment experiments to date have shown that the particles are unchanged by heating to 1850°C for 2 hr; and in one experiment a change did occur, namely smoothing of the grain boundaries, on heating to 2350°C and quenching (Figure 3). However, this experiment was complicated by a simultaneous shift in stoichiometry to the "hyper" side, and it may have been this change which caused solution and retention of tungsten rather than heating and quenching. Further experiments with "hypo" material are in progress.

7551-2504

Figure 3. Zone-Melted 0.68 wt % W in UC Before and After Heating at 2350°C and Quenching (x500) Showing Grain Boundary Effect

4. Preparation by Arc-Melting

Arc-melting of buttons on a water-cooled hearth have also produced fine-particle dispersion of tungsten in stoichiometric UC, as illustrated by the surface replicas of Figures 4a and 4b. In this case, the cooling rate was probably much faster than in zone-refining and the particles are somewhat more numerous. A count of about 10^{13} particles per cc was calculated for the sample containing 2.5% W. The sample containing 1% W comprised fewer particles, suggesting that the quench was rapid enough to freeze some of the tungsten in solution. The arrangement of particles in lines in the 2.5% W sample is also indicative of a dendritic structure which may have been caused by rapid cooling.

Arc-casting of W-doped UC with a graphite hearth and molds is now in progress. This procedure may possibly produce fine dispersions directly, or may produce a mixture from which fine dispersions could be made by heat-treating.

The critical need in this development is for high burnup irradiation experiments to find out if improved swelling behavior is achieved. Improved hot hardness, as discussed in a later section, is an indication that W-doping changes mechanical properties in a favorable direction. It is hoped that reactor tests can be started within a few months.

A report, probably an open literature publication, covering this development is now in preparation.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Figure 4. Surface Replica Electron Micrographs of
Cold-Hearth, Arc-Melted, UC + 2.5 wt % W (top)
and UC + 1 wt % W (bottom)

B. PURE UC

1. Preparation of Single Crystals

Single crystals of UC were grown in the modified electron beam melter in each of the last five starts. Earlier difficulties were due to improper electron gun design during the equipment modernization. In two cases, seeding was used to predetermine orientation of the growth axis so as to make the (100) plane perpendicular to the rod axis. Traverse rates were 3 in./hr, and rotation of the bottom check was 55 to 60 rpm in all cases. The crystals were examined by metallographic and x-ray techniques; the data are summarized in Table 2. The crystals are strained and also contain small mosaic areas. Twist of the crystallographic lattice about the rod axis occurred in some cases. However,

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 2
ZONE-REFINED SINGLE CRYSTALS OF UC*

Sample Number	Starting Carbon Analysis (wt %)	Single-Crystal Length (in.)	Rod Axis Crystallographic Orientation	Reflected X-Ray Beam Divergence (0/mm ²)	Axial Twist of Crystal Lattice (0/in.)	Final Carbon Analysis (wt %)
260 181	4.65	3	7 to 14° from (100)	2	None	-
260 184	4.80	4-5/16	3° from (111)	4	< 2	5.1 to 5.2
260 185	4.75	3	0 to 2° from (100)	< 2	None	5.1 to 5.2
260 186	4.70	4-5/8	4° from (100)	3	< 2	5.1 to 5.2
260 187	4.75	4-7/8	3° from (110)	5	None	5.1 to 5.2

*All zone-refining runs were made in an electron beam melter in a vacuum ranging from 1 to 5×10^{-6} torr. Zone traverse rate was 3 in./hr for all runs. The freezing faces were rotated at approximately 60 rpm, creating a condition of shear in the molten zone.

this twist does not seem to be associated with the shear resulting from rotation of the bottom chuck. Thus, the twist observed in these samples is relatively small, ranging from 0 to about 4°/in., although bottom rotation was used during growth. In contrast, twists ranging from 6 to 15°/in. were observed in similar samples prepared without rotation.* Other factors that may have a bearing on the structure of these crystals include the longitudinal temperature gradient, time-temperature history, heating profile (or focusing of the beam current in the EBM), and impurities. These variables will be studied by varying the after-heater in the electron gun, traverse rates, and use of a long-time anneal after the crystal growth.

2. Control and Adjustment of Stoichiometry in UC

a. Zone-Melting Experiments

Zone-melting with electron beam heating requires a high vacuum atmosphere; and, because of rather rapid vaporization of uranium from molten UC in a vacuum, the product is always high in carbon. With induction heating, vaporization of uranium is effectively repressed and the effects of zone-melting itself on uranium-to-carbon stoichiometry may be observed. The fact that UC melts congruently means that the solid-phase composition should shift towards stoichiometric UC and the liquid phase should become enriched in either carbon or uranium, whichever element is in excess in the initial composition, assuming that equilibrium is approached during zone melting. Such effects have been

*From talk by A. K. Smalley on "Fine-Particle Metal Dispersions in UC," at Ninth Refractory Composites Working Group Meeting in Worcester, Mass. (August 18, 1964)

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

observed in a number of runs, showing up particularly in the enrichment of the molten zone in the excess element. However, a series of carefully controlled and documented experiments showed relatively little shift in composition due to zone refining, although in every case there was a shift in the expected direction towards the stoichiometric composition. This was true in regard to the proportion of second phase present, as shown by metallography; and was also true of the actual carbon content as shown by chemical analysis. Oxygen and nitrogen were also determined chemically and were low enough in all cases to be ignored as elements which could alter the stoichiometry by substituting for carbon. Chemical analyses are shown in Table 3.

TABLE 3
EFFECT OF ZONE-REFINING ON STOICHIOMETRY

Nominal Initial Carbon (wt %)	Zone Refining Traverse Rate (in./hr)	Section Analyzed Chemically	Chemical Analysis				
			Carbon (wt %)	ppm			
				Nitrogen	Oxygen	Iron	Silicon
4.6	0.5	Initial, arc-cast	4.48	210	70	200	75
		Zone-refined, bottom	4.56	375	55	100	<50
		Zone-refined, top	4.49	235	60	150	75
4.6	4.0	Initial, arc-cast	4.53	140	45	200	75
		Zone-refined, bottom	4.55	140	20	50	<50
		Zone-refined, top	4.53	150	40	50	<50
4.8	0.5	Initial, as-cast	4.78	255	960	100	75
		Zone-refined, bottom	4.795	230	65	75	<50
		Zone-refined, top	4.72	320	110	150	<50
4.8	4.0	Initial, as-cast	4.75	185	11	50	<50
		Zone-refined, bottom	4.75	260	16	<50	<50
		Zone-refined, top	4.73	225	12	<50	<50
5.2	0.5	Initial, as-cast	5.20	225	18	250	200
		Zone-refined, bottom	5.14	220	25	75	250
		Zone-refined, top	5.22	190	35	<50	75
5.2	4.0	Initial, as-cast	5.15	175	75	150	100
		Zone-refined, bottom	5.08	285	35	100	75
		Zone-refined, top	5.14	250	40	75	<50

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

The apparent interpretation of the results given in Table 3 is that there are only slight differences between the composition of liquid and solid at temperatures near the melting point of pure UC. Hence, many passes would be required to eliminate the second phase. Another point of interest is that of forming a carbon-deficient single phase UC, that is, UC_{1-x} , during zone-refining of hypostoichiometric material. Comparing the metallography of the first two samples of Table 3, it was found that the slow traverse rate, 0.5 in./hr, resulted in significantly less free uranium than the 4.0 in./hr sample, suggesting either that more of the free uranium dissolved in UC at the slower traverse rate or that the 0.5 in./hr rate was slow enough to sweep out uranium more effectively than was the 4.0 in./hr rate. X-ray lattice parameters are a measure of the amount of uranium dissolved in UC; that is, the defect lattice with uranium dissolved in it has a parameter lower than 4.960 Å, which is the value for stoichiometric UC. Patterns obtained for these two samples gave lattice parameters of 4.9672 and 4.9566, both lower than 4.960, and thus indicative of dissolved uranium. However, there is no indication that the first sample contains more dissolved uranium than the second; and it is concluded that free uranium was actually swept out more effectively at the slower traverse rate.

b. Decarburization of Hyperstoichiometric UC by Treatment in H_2

Single crystals, prepared in an electron beam melter under high vacuum, are almost always hyperstoichiometric with respect to carbon, as evidenced by the appearance of UC_2 platelets distributed through the UC matrix, the proportion of UC_2 being a semiquantitative measure of excess carbon. Excess carbon can be attacked and removed with hydrogen, as described by Nadler* for single crystals and by Smiley† for polycrystalline rods. It is desirable to obtain a direct comparison of decarburization kinetics for single and polycrystalline material in the same apparatus to gain further understanding of carbon diffusion processes in UC.

In a preliminary experiment, a small crystal, oriented with the 100 axis parallel to the rod axis, and initially containing 5.2 wt % of carbon, as measured metallographically, was heated at 1800°C in 16 mm hydrogen pressure for cumulative times of 4, 8, 16, 32, and 56 hr. At the end of each time increment, the crystal was cooled, removed from the furnace, and a small piece was cleaved from it. Then the remainder was reinserted in the furnace and heated for the succeeding time increment. The cleaved samples were analyzed semiquantitatively for contained UC_2 (i. e., excess carbon) by metallography. Surprisingly, it was found that the UC_2 platelets essentially disappeared during the first 4-hr heating period, indicating a reduction of the carbon content of the crystal from about 5.1 to very close to the stoichiometric 4.8 wt %. Also, the metallographic appearance of the samples did not change appreciably with further heat treatment.

In contrast, a polycrystalline sample, also initially 5.2 wt % carbon by metallography, required approximately 20 hr in a similar experiment to become stoichiometric in its metallographic appearance. Weight loss data are ambiguous

*H. Nadler, "Preparation of Uranium Monocarbide Single Crystals," NAA-SR-7465 NAA-SR-7465 (April 1964)

†W. G. Smiley and W. D. Turner, "Decarburization of Cost Uranium Carbide by Heating in Hydrogen," NAA-SR-7921 (August 15, 1963)

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

but are consistent with the metallography in that weight loss of the single crystal stopped after 8 hr but continued for 36 hr in the case of the polycrystalline sample.

The initial metallographic appearance of these samples is shown in Figure 5a for the single crystal and in Figure 5b for the polycrystalline sample. Comparison of these two photomicrographs suggests a mechanism for decarburization which is admittedly speculative, but interesting. The continuous platelets of UC_2 extending throughout the crystal could be channels for rapid carbon diffusion and permit more rapid decarburization of such a single-crystal structure than in the case of the interrupted UC_2 pattern of the polycrystalline sample. Eyre and Sole*, in an electron microscope study of UC_2 precipitates in UC, postulated that UC_2 crystallized from hyperstoichiometric UC in the body-centered-tetragonal form by a process of shear transformation plus short-range diffusion of carbon atoms. This is a relatively rapid process compared with diffusion of carbon atoms through a stoichiometric UC lattice. In order for this mechanism to be operative, the UC_2 crystals would have to persist at the temperature of decarburization, which was $1800^\circ C$. If such a conclusion could be established by further experiments, there would be important implications concerning the U-C phase diagram on the hyperstoichiometric side at this temperature level.

7551-2506

Figure 5. Hyperstoichiometric UC Single Crystal (left) and Hyperstoichiometric Polycrystalline UC (right), Prior to Decarburization

A later experiment showed a much slower rate of decarburization for a single crystal, with the 111 axis oriented parallel to the rod axis by comparison with the above result in which the 100 axis was parallel to the rod axis.

*B. L. Eyre and M. J. Sole, "Electron Microscope Studies of Uranium Dicarbide Precipitates in Uranium Carbide Single Crystals," Phil. Mag. 9, (100) p 545 (1964)

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Other explanations of the apparently rapid decarburization of the single crystal in this run are conceivable and should be investigated. One of these is the possibility of end-effects which would result in improper sampling by simply cleaving off one end of the rod.

c. Control of Carbon Content Using Alkali Metal as Carrier

The desirability of an alternative method to the hydrogen reduction technique for adjustment of carbon content of UC was pointed out (refer to the Annual Report for FY 1964), and a possible method was suggested. The proposed method was to employ a liquid metal as transport medium and a carbide-forming metal as a carbon sink. An experiment has been carried out to test the feasibility of the idea. Sodium was used as the carrier metal, and chromium in the form of a 0.003-in.-thick plating on the inside of a tantalum capsule as the carbon sink. A piece of zone-refined hyperstoichiometric UC was soaked for 48 hr at 1400°C. Examination of the UC showed decarburization had occurred only in a thin layer exposed to the sodium. Incomplete decarburization was expected because of the limited diffusion rate at 1400°C; but a more serious objection was the finding that transport of Cr and Ni to the UC had occurred, depositing a reaction layer.

Although the intent had been to follow this experiment with another using a higher boiling metal than sodium, in order to reach higher temperatures, this experiment points up some complications to be expected that would lead to a more extensive effort than is justified. Accordingly no further work on this method is planned for the present.

C. SELF-DIFFUSION STUDIES

Self-diffusion of both uranium and carbon in UC have been reported by W. Chubb, et al., at BMI, who found the diffusion coefficient for carbon to be three orders of magnitude greater than the diffusion coefficient for uranium at 1500°C; and a still greater contrast at lower temperatures because of the differing activation energies, 64 kcal/mol for uranium and 50 kcal/mol for carbon. These differences are expected in the metal-like structure of UC, and resemble the differences between diffusion rates of metal and interstitial element in metal-phase solutions of interstitial elements, such as oxygen in tantalum. One consequence of the diffusion rate of carbon being much faster than that of uranium is that processes which involve mass flow will depend on the slower moving uranium atoms. Another consequence is that rather rapid transport of carbon, other interstitial elements, and vacancies can be expected at moderate temperatures. Diffusion processes are of great importance in the interpretation of mechanical properties and fission damage behavior.

A rather high uncertainty was assigned by Chubb et al. to their diffusion measurements; furthermore, their sample was hyperstoichiometric. In an effort to achieve a more precise measure of these important constants, and to look for changes that may occur with variation in stoichiometry, preparations have been made and materials and equipment are now ready to measure self-diffusion in UC on this program. It is planned to vapor-deposit U²³³ and C¹⁴ on high-purity UC, and to measure U- and C-diffusion simultaneously on samples annealed in a furnace with close temperature control and high purity

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

atmosphere. The radioactive materials and analyzed UC samples are on hand, the furnace and the sectioning and counting equipment are on hand, and runs will soon be started.

D. MECHANICAL PROPERTIES

Measurement of certain mechanical properties of fuel materials and the interrelationships between these and other properties can provide insights into structures and dynamic behavior. Hot-hardness is an example of a property that is easily measured, shows trends expected to be similar to more pertinent mechanical properties and appears to be related to swelling behavior. The hot-hardness measurements are summarized in Figures 6a, b, and c. The distinct increase in hardness due to small tungsten additions to either hypo or hyperstoichiometric UC is noteworthy, and it is of further significance that a sample containing a similar small percentage of Si + Mo showed a similar increase in hardness. Since there is undoubtedly some tungsten dissolved in the UC, the effect could be one of lattice stiffening; or the increased hardness could be caused by the dispersed particles known to be present. The Si- and Mo-doped sample contains dispersed particles somewhat similar to the tungsten particles, as indicated by the similar grain boundary effect shown by optical microscopy in Figure 7a and b, and by the surface replicas of Figure 8a and b, which compare pure with doped material.

Other hot-hardness data indicate that hyperstoichiometric UC is harder than hypostoichiometric at temperatures above 600°C, which agrees with the expected hardening effect of UC₂ platelets as a second phase; but at low temperatures hypostoichiometric samples were harder, except at very low indenter load. Microscopic examination showed unhomogeneous deformation and cracking around indents made in hyperstoichiometric UC at room temperature compared with homogeneous deformation around indents in hypostoichiometric UC.

Figure 6. Effects of Impurities on Hot-Hardness at UC

7551-2508

Figure 7. Polished Sections (left) of UC (4.8 wt % C) and (right) of UC (4.6 wt % C) Containing 0.6 wt % Total (Si + Mo) Impurity

7551-2509

Figure 8. Electron Micrograph of Surface Replica of (top) Pure UC (4.8 wt % C) and (bottom) UC (4.6 wt % C) Containing 0.6 wt % Total (Si + Mo) Impurity x30,000

Other mechanical properties studied include internal friction, transient creep, and Young's modulus. All three of these measurements are made in the same furnace on a single sample during one heating cycle. Measurements have been made on nearly stoichiometric pure UC over a range of temperatures, and the results show sharp changes with temperature; but interpretation of the results is difficult. All of these properties are related to structures, and the internal friction, and transient creep depend in some way on diffusional processes. The data are presented in Table 4 and Figure 9. The sharpest changes occur in

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 4
YOUNG'S MODULUS AND INTERNAL FRICTION OF
NEAR STOICHIOMETRIC UC

Temperature (°C)	Young's Modulus (psi x 10 ⁶)	Internal Friction (Q ⁻¹ x 10 ⁻³)
25	31	2.8
200	32.0	2.8
400	32.5	2.0
600	31.5	2.8
800	29.0	8.0
1000	16.5	10.0
1200	16.5	10.0
1300	16.5	10.0

Figure 9. Transient
Creep of Stoichi-
ometric UC

the temperature range between 600 and 1000°C, namely, a decrease in stiffness as measured by Young's modulus, an increase in internal friction and an increase in the extent of deformation under stress. More extensive tests on samples of varying stoichiometry will be needed in order to draw conclusions relating to structural differences or diffusion processes.

E. DEFECT STRUCTURE OF UC AND REACTION OF CARBIDES WITH GASES

In addition to the nonstoichiometry in UC which shows up as a second phase of uranium metal or UC₂, the phase UC can be nonstoichiometric on the carbon-deficient side only, that is, UC_{1-x}. Although this is often described as solution of uranium in UC, it is structurally more significant to refer to vacancies in the interstitial or carbon lattice. The extent of this carbon deficiency has been studied by Buckley and by Magnier and Accary, and a maximum in the curve has been found at about UC_{0.96} at 1700 to 2000°C. Some quenching and annealing experiments on hypo-

stoichiometric UC samples performed here have partially verified these findings, but have also pointed up the deviation due to contamination that may result from annealing experiments performed in vacuum or inert gas atmospheres with partial pressures of oxygen or nitrogen above about 10⁻⁵ torr. There is some question as to how much effect gas contamination may have had on results reported in the literature.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

The substoichiometry range is appreciably more in the case of PuC than in the case of UC, and is much greater in the case of ZrC. It is expected that the concentration of lattice vacancies will play a significant part in some of the properties of fuels, particularly in some aspects of irradiation behavior.

The chief measurement made in the study of nonstoichiometry of UC has been the change in lattice parameter, with carbon vacancies causing a decrease in spacing from about 4.961 Å to perhaps as low as 4.9649 Å. Filling of vacancies with oxygen or nitrogen apparently will cause an increase in the parameter of UC_{1-x}, just as filling the vacancies with carbon would do, although when oxygen or nitrogen is substituted for carbon in a stoichiometric structure, the lattice parameter decreases.

One result of exposing substoichiometric carbides to gases of the interstitial elements, at high temperature, is that the vacancies are filled. However, at a normal atmospheric pressure of oxygen or air and a relatively low temperature, the principal result is formation of a scale of oxide. It is only at very low pressure and relatively high temperatures that reactions involving filling of vacancies and substitution of carbon for oxygen can be observed. Reaction with hydrogen is also an interesting way to fill vacancies in the carbon lattice, and it has been reported that two hydrogens enter the lattice for each carbon vacancy. This suggests that the concentration of vacancies could be measured by titrating with hydrogen.

In order to investigate the reaction of hydrogen to fill vacancies, a series of kinetic measurements at low pressures have been started, using ZrC as the initial carbide, in the expectation that under these conditions there will be a sharp change in kinetics at the point at which hydrogen has filled the vacancies and further hydrogen absorption involves formation of the ternary compound of Zr, C, and H₂ (see report on 7652-4552). The choice of ZrC to precede UC in the series was based on the wide stoichiometry range of this compound, from ZrC_{0.67} to ZrC_{1.0}, which provides a better opportunity to observe the expected kinetics than in the case of UC with a range of only UC_{0.96} to UC_{1.0}.

More extensive experiments have been performed on the reaction of UC with oxygen at low pressure. The first of these was an attempt to obtain high temperature x-ray data on a single crystal of UC, using a high-temperature x-ray camera with a vacuum atmosphere in which the specimen became oxidized sufficiently to completely obscure the UC x-ray pattern in about an hour at about 1000°C. Although during this run the ion gauge read 5×10^{-5} torr, later experiments indicate that the pressure at the sample may have been a little higher, probably 2×10^{-4} torr. The crystal dimensions were about 2 x 2 x 1 mm, with one of the large surfaces exposed to the x-ray beam. During the heating, the reflections due to UC disappeared and were replaced by reflections due to UO₂ and UC₂, both showing single-crystal-type reflections oriented with the original UC. On removal from the x-ray furnace, a thick surface coating was observed.

The coating was flaked off, and a powder pattern of it confirmed the presence of UC, UC₂, and UO₂. Rotating single-crystal photographs were also made of the flaked material, and these showed that the UO₂ and UC lattices were in nearly perfect crystalline register. The UO₂ diffraction maxima showed some broadening, but were still essentially single-crystal in character. Further, the

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

UC₂ lattice seemed to have its C-axis normal to the exposed sample surface, as evidenced by lack of reflections from the (100) planes on the diffractometer traces. The orientation of the A-axis of the UC₂ lattice with respect to the UC lattice could not be determined from the patterns now available.

In this experiment, the UC apparently reacted according to the equation

and the UO₂ and the UC₂ were oriented with the axes of the original UC so that single-crystal patterns were obtained for each.

The correspondence of axes that makes these orientations possible is as follows: UC, cubic, $a_0 = 4.96$ (100) corresponds to (100) of UO₂, cubic, $a_0 = 5.47$, a 10% difference; and (100) of UC also corresponds to (110) of UC₂, tetragonal, $a_0 = 3.517$, $c_0 = 5.987$ (110) = $[2 \times (3.517)^2]^{1/2} = 4.96$, an exact fit. It has been shown that this correspondence between axes of UC and UC₂ exists in UC-UC₂ two-phase crystals. Presumably the oxidation coating on the UC was an inter-growth of small oriented crystals of UO₂ and UC₂ which gave x-ray patterns similar to single crystals.

Some carefully controlled kinetic experiments were then run on portions of the same single crystal of UC, a crystal about 1/4 in.-in.-diameter oriented with the 100 axis parallel to the rod axis and close to stoichiometric but with a slight excess of carbon, enough to show UC₂ platelets metallographically but not enough to show UC₂ by x-ray. These experiments have been very simple and brief, but have yielded a great deal of information about the system U-C-O and UC-O₂ interactions, much of which is not yet organized for presentation. Data on one set of runs at 900°C in a vacuum microbalance are shown in Table 5.

TABLE 5
OXIDATION OF UC SINGLE CRYSTAL IN
FLOWING DRY OXYGEN AT 900°C

Pressure (torr)*	Duration of Run (hr)	Rate of wt-Gain ($\mu\text{gm}/\text{cm}^2 \text{ min}$)
5×10^{-6}	2	0.0 ± 0.1
1×10^{-5}	4	0.0 ± 0.1
2×10^{-5}	1.2	1.0
1×10^{-4}	1.25	3 - 5 [†]

*Pressure 5×10^{-6} is residual pressure at best vacuum. Other pressures maintained by pumping against leak.

†A definite acceleration in rate was noted during the 1-1/4 hr of the run.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

X-ray diffraction from the surface showed strong patterns of UO_2 and UC_2 , both oriented with respect to the UC. There was also a definite but weaker pattern of U_3O_8 . The U_3O_8 is presumed to be a result of a secondary reaction. The actual x-ray patterns of the original and oxidized UC single crystal surfaces are reproduced in Figure 10.

Figure 10. X-Ray Patterns of 100 Surface of
UC Single Crystal Oxidized at 10^{-4} torr

A later experiment produced a weight gain of about 10 mg/cm^2 , and a cross-section of the oxidation coating was observed to be 150 microns thick. This agrees within experimental error with the thickness calculated by assuming that the coating is $\text{UO}_2 + \text{UC}_2$ produced by reaction of UC with O_2 , thus providing more support for the x-ray evidence that this is the reaction occurring.

When another portion of the same single crystal was heated in dry oxygen at 10^{-4} torr at 1800 instead of at 900°C , no coating formed at all, and the sample lost weight due to vaporization of some compound of uranium. It is planned to explore some temperatures between 900 and 1800°C at this pressure and at lower pressures, because, from a practical point of view, it is important to

know the conditions under which oxygen is absorbed by UC as a coating or as solid solutions, and those under which uranium or uranium compounds vaporize.

F. FUNDAMENTALS OF FISSION DAMAGE

1. Fission Gas Release

A very limited number of experiments have been performed in which carbide samples were subjected to low-level irradiation in a nuclear reactor followed by measurement of the quantity of fission gas released during heating. The carbide materials behave qualitatively like metals and hydrides in that the quantity of gas release is temperature-dependent. Following a leveling off of activity after a period of time at a given temperature, additional gas release can be obtained by heating to a higher temperature; and several successive releases can be obtained in this manner. Further, if a sample which has leveled off at one temperature is heated to a higher temperature for a short time and then cooled to the lower temperature, additional gas is released. This behavior pattern makes it questionable to interpret gas release in terms of diffusional processes alone. Possible (partial) explanations may be found in terms of some surface reactions, such as oxidation, or desorption.

Samples of a hyperstoichiometric single crystal of UC, a UC - 20 mole % ZrC, and a UC - 1% W dispersion were irradiated and annealed. These showed some qualitative differences, but results are subject to question because of decrepitation during storage. The technique is regarded as potentially fruitful in assessing the merits of modified fuels, such as W-doped UC, and an increased activity in this area is planned, using an improved furnace which is now in operation.

2. X-Ray Examination of Irradiated UC

An exploratory experiment to investigate irradiation damage by means of x-ray techniques gave tentative indication that changes in structure can be observed in this way. A near-stoichiometric single crystal of UC was irradiated to a low level ($\sim 10^{14}$ nvt), and Lauegrams of the sample before and after irradiation showed an apparent broadening of spots; but interpretation was made difficult by the high degree of imperfection in the original single crystals.

It seems likely that the use of substantially more perfect UC single crystals would make the use of Lauegrams and other x-ray techniques such as Berg-Barrett and Lang camera fruitful procedures to study structural effects of fission. This need provides some impetus to a search for methods to produce improved single crystals, which incidentally could be much smaller for this use than those now produced by zone-refining. For example, suggestions have been made that growth from a liquid metal solution at temperatures much below the melting point might be successful in producing more perfect crystals. If a promising experimental setup can be devised, an exploratory run will be made. Lacking better crystals, only a very limited further study of x-ray techniques is planned.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The most notable achievement of the program this year has been development of uranium monocarbide containing dispersed particles of tungsten. Examination of the UC-W alloys prepared in the induction zone melter quickly showed a good possibility of preparing the desired very fine dispersions directly by this simple procedure. A substantial effort was therefore spent to investigate process parameters and to characterize the material. As a result, it is now possible to prepare samples of second-phase dispersions in partially enriched UC for in-pile tests; and such tests are being planned. Fine particles were also produced by cold-hearth arc-melting of UC-W mixtures, and tests are in progress on arc-melting and casting. The arc-melting procedures permit study of more varied starting materials, and are more readily adapted to commercial operation than is the zone-melting procedure.

The practical value of these materials as improved fuels is yet to be demonstrated, although improved hot-hardness has been found; and this is probably indicative of improved high-temperature mechanical properties in general. A much more significant measure will be the planned irradiation tests to measure swelling at high burnup.

Because the development of these dispersions is regarded as a significant step towards making improved fuels, further study of their basic behavior is justified. It is for this reason that experimental work now in progress includes heat-treating experiments with the objective of controlling particle size and population as well as a phase-diagram study of a part of the system UC-W.

By use of the electron beam zone refiner, 1/4-in.-diameter single crystals of UC can be grown on a more or less routine basis, and the excess carbon can then be removed by treatment with hydrogen. These near-stoichiometric crystals are useful for some research purposes; however, the crystals have subgrains and many other imperfections. Formation of highly perfect single crystals will probably require an entirely new method of growth. The hope of using the induction-zone refining equipment to grow stoichiometric single crystals in a single step had not materialized, in the first place because the refined rod has always been polycrystalline. Use of inert gas atmosphere instead of vacuum has been shown to inhibit uranium vaporization; and near-stoichiometric polycrystalline samples can be grown from feed material that is close to stoichiometric, but relatively little adjustment in stoichiometry due to zone-refining has been found. However, induction-zone refining has proved a very useful technique, especially for preparation of W-doped UC.

Progress made in measurement of mechanical properties favors continuation of these studies to develop interpretations relating to other properties. Preparations are nearly complete for self-diffusion measurements expected to be valuable in interpretations of mechanical properties and irradiation behavior. Not much effort has been put on study of fission damage resulting from low-level irradiation, and future effort in this area is expected to be on fission gas release kinetics, with studies on lattice structural changes partially limited by unavailability of highly perfect single crystals.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Careful attention has been given to the probable importance of the concentration of defects in the carbon lattice of UC for many of its properties which affect its use as a fuel. These defects include carbon vacancies produced by solution of uranium in UC, and substitution of oxygen or nitrogen for carbon or filling of vacancies by oxygen or nitrogen, which can occur readily by high-temperature exposure of UC to inert atmospheres or vacuum normally considered to be noncontaminating. An equipment improvement program has been under way as a result of which we will be able to analyze precisely for very small amounts of these contaminants in inert gases, and to provide the necessary purification. In the case of vacuum, ion-pumped systems capable of 10^{-8} torr or better will virtually eliminate significant O_2 or N_2 contamination. In the meantime, experiments in progress on high-temperature reactions with low-pressure gases, that is, of UC with O_2 and H_2 , and of ZrC with H_2 , are yielding a great deal of interesting information in an important area thus far largely neglected.

Talks and Publications during fiscal year 1965 were as follows: "Fine-Particle Metal Dispersions in UC," presented by A. K. Smalley at the Ninth Refractory Composites Working Group Meeting, Worcester, Mass., August 18, 1964; and "Reactions of UC with Oxygen at Low Pressures," by Guy Ervin, Jr. and K. T. Miller, to be submitted for publication June 1965.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Reactor Fuels and Materials: High Temperature Material Development	
Project: Study of BeO as a Reactor Material	
Project Engineer: C. E. Weber	
Reporting Period: Fiscal Year 1965	AEC Category: 04-40-02-09
General Order: 7551	Subaccount: 4549

I. PROJECT OBJECTIVES

The broad objectives of this program are as follows.

- 1) Preparative Research. To develop, understand, and utilize means of growing pure and doped BeO single crystals; to pursue to conclusion the study of special hot-pressing and sintering phenomena, and hence to exploit control of fabrication parameters in material preparation.
- 2) Characterization. To determine and understand the fundamental characteristics of the BeO crystal lattice and of polycrystalline BeO ceramics; lattice geometry, energy, defects, and transport; grain, grain-boundary, and pore structure, orientation, distribution, composition, and transport.
- 3) Mechanical Behavior. To study and understand the elastic, anelastic, and plastic deformation and fracture behavior of BeO in relation to the nature of the crystal, defects, slip, grain-boundary deformation processes, and the composition and bulk microstructural character.
- 4) Radiation Effects. To assist in furthering the understanding of the basic processes underlying fast neutron damage to BeO and the consequent effects on behavior.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

Progress during this last year in support of the project objectives was achieved in a number of individual and interrelated tasks, as follows.

A. CRYSTAL GROWTH

The BeO crystal growth effort this year has been devoted primarily to crystal production for experimental purposes; and the level of effort has been considerably reduced from the prior year. Improvements in the crystal growth technique and crystal-handling methods have been instituted as described in a later section on crystal defect evaluation.

The primary effort has been on the growth of large crystals from molybdate flux. Crystals up to 14 mm in length have been grown; some of the larger crystals are shown in Figure 1. This seems to be the practical upper limit of size obtainable with the present equipment. Significant scaleup in size of crucible, furnace, etc., and incorporation of known by unexploited process improvements probably would be necessary to produce significantly larger crystals.

One technique for producing larger crystals might be growth on seed crystals. An attempt to do this has been moderately successful. A group of crystals was

7551-2512

Figure 1. BeO Crystals

mounted on a Pt wire rig, as shown in Figure 2a. Each crystal was prepared in a specific fashion. In particular, selected crystals were shallowly ground away on (0001) , $(000\bar{1})$, and $(10\bar{1}0)$ faces. It was reasoned that dislocations would be introduced by the grinding operation, and that these dislocations could accelerate crystal growth. However, growth was neither accelerated nor inhibited by the grinding treatment. The seed rig after overgrowth is shown in Figure 2b. Several reasons have been advanced for this insensitivity to grinding pretreatment, including the hypothesis that the flux dissolved the damaged region during the initial period of saturating the flux. The experiment did confirm that growth on a seed crystal is feasible, with the overgrowth generally as good as the seed in quality. The habit and quality of the overgrowth reflects the flux environment, though, just as though it were self-nucleated rather than seeded.

7551-2513

a.

7551-2514

b.

Figure 2. Seed Crystal Assembly Before and After Growth

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Figure 3. Diffusion Coefficients for Be in Single-Crystal BeO

7551-2515

Figure 4. Be Diffusion in BeO

7551-2516

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

The crystal growth effort has been redirected toward an attempt to produce graphite crystals of high quality. We feel that problems encountered in prior attempts to grow graphite crystals were due to (1) uncontrolled crystal nucleation, and (2) excessively high growth rates. Two procedures are under consideration. The procedure that appears to have the greatest promise involves nucleation and growth of graphite crystals in a molten metal solvent. A number of metals can dissolve sizeable quantities of carbon; and in so doing the melting temperature of the solution is lowered by several hundred degrees, which is very convenient. It is planned to use the steady-state controlled thermal gradient technique with metal solvents. Furnaces for this program have been ordered, but are not expected to be delivered for several months.

The second procedure considered for growth of graphite crystals exploits the slow transport of carbon through a gaseous phase in a sealed container and in a temperature gradient. This is considered to be a more difficult experiment and less likely of success, and so is being given second priority in attention.

B. SELF-DIFFUSION

The studies of cation diffusion in BeO have been completed and reported. The measurement of cation diffusion in single crystals was extended to a greater number of samples in order to gain both improved data and statistical accuracy, also to investigate several minor points of concern. The single crystal data are shown in Figure 3; they are shown again in Figure 4 as a composite, with all other reported data on Be diffusion in BeO. The discussion presented in the previous annual report related the significant material characteristics to cation diffusion kinetics, as well as to the diffusion mechanism itself. The more recent data have not necessitated change in these interpretations, but rather have strengthened the previous arguments. The wide range of diffusion coefficients evident in Figure 4 illustrates the important role of impurities in controlling diffusion rates and consequently the kinetics of other diffusion-controlled processes such as creep.

Attempts to measure oxygen diffusion rates in single-crystal BeO have not been as satisfactory. Methods of applying O^{18} to the surface of crystals, in the form of BeO^{18} , have been satisfactorily developed. Upon analyzing the distribution of O^{18} as a function of depth into the sample following diffusion anneal, it was found the O^{18} concentration did not follow a smooth profile, but was distributed very unhomogeneously. The present interpretation is that microcracks in the surface of the crystal may be providing very efficient paths for O^{18} diffusion into the crystal interior. Such cracks have been proposed to account for some aspects of cation diffusion. The microcracks are generated even with the most gentle method of mechanical cutting and polishing. If this observation of unhomogeneous distribution of diffusant O^{18} is verified by other methods, the effort to measure oxygen diffusion in single crystals by sectioning methods will be terminated.

C. CRYSTAL ENERGY CALCULATIONS

The total internal energy, E_t , of a crystal can be approximated by a simple two-term Born potential involving the electrostatic energy, E_e , and the overlap repulsion energy, E_r , as shown in Equation 1.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

$$E_t = E_e + E_r = -Ae^2 z_1 \sum_i z_i r_i^{-1} + b \sum_i r_i^{-n}, \quad \dots (1)$$

where A is the Madelung constant, z_1 and z_i are the ionic charges, e is the electronic charge, b and n are constants for each material, r is the separation distance between a reference anion (chosen as Ion 1 in Equation 1) and its successive nearest neighbor ions, and the term $\sum_i r_i^{-n}$ is called the inverse n 'th power repulsion factor. For the present study of the wurtzite structure, the index i ranged over the first 25 shells of ions for the Madelung constant calculation, but only over the first 5 shells for the inverse power repulsion factor where, in the present case, n was taken equal to 12.

The Madelung constant, A , and the inverse 12th power repulsion factor, r^{-12} , have been calculated over wide ranges of the crystal parameters c_0/a_0 and u for the wurtzite crystal structure (where u is a measure of the MX bond length parallel to c_0). The results are plotted side-by-side for comparison in Figure 5 where the individual curves represent different values of c_0/a_0 and each is labeled by its c_0/a_0 value. The mirror symmetry about the value $u = 0.500$ reflects

7551-2517

Figure 5. Crystal Energy Data

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

the fact that in the ideal wurtzite structure the cations are located at either $u = 0.375$ or $u = 0.625$. The "neck" in the curves at $u = 0.500$ ranges from $c_0/a_0 = 1.633$ down to $c_0/a_0 = 1.35$ for the Madelung constant, and represents about a 3% change in A ; and for a similar range the inverse twelfth power repulsion factor changes by approximately 10%. Thus the r^{-12} plot is much more sensitive to variation in c_0/a_0 than is the A plot. Moreover, the increase in r^{-12} with a decrease in c_0/a_0 represents a real increase in the positive repulsion energy, E_r , whereas the increase in A with a decrease in c_0/a_0 represents a decrease in the negative electrostatic energy, E_e , as can be seen from Equation 1, where because of the minus sign, E_e decreases as A increases. Thus, E_r increases and E_e decreases as c_0/a_0 decreases.

The minima of the curves of different c_0/a_0 in Figure 5 are plotted in Figure 6 as heavy solid lines. The computer values of the Madelung minima are shown as open circles; the inverse 12th power repulsion factor points are shown as open triangles; and the experimental values for AlN, BeO, and hypothetical, ideal wurtzite ($c_0/a_0 = 1.600, 1.623$, and 1.633 , respectively) are shown as open squares. In the experimental region, the three curves are quite linear and lie close together although they do not overlap. From Equation 1 it follows (after a double differentiation with respect to r_1) that the extrema in E_e and E_r are maxima and minima, respectively; and from the physics it turns out that the E_e maxima are rather broad, whereas the E_r minima are rather steep. Thus, in the experimental region, it follows that the broad E_e maxima and the steep E_r minima lead to an "energy valley" in potential energy space in which the wurtzite compounds are constrained to lie. It seems that the reason why the wurtzite compounds are further limited to lie in such a narrow range of c_0/a_0 is that (as noted at the end of the first paragraph preceding) the A curve is falling in potential energy while the repulsive r^{-12} curve is rising; and this could well lead to a minimum that lies along the axis of the above energy valley and thus converts it into

7551-2518

Figure 6. The u -Values for Four Models

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

an elongated "energy bowl" in which the wurtzite compounds are constrained to lie. This point is still being investigated.

Four theoretical models of tetrahedral bond distortion have been considered, to find which model would most closely agree with the parameters c_0/a_0 and u as defined by the computed minimum energy valley over a wide range of distortion. The curves of u -values for these four models are illustrated in Figure 6 where they are labeled u_1 , u_2 , u_3 , and u_4 , corresponding to Models I, II, III, and IV, respectively. Models I and II were first proposed by Keffer and Portis (J. Chem. Phys. 27, 675 (1957) and correspond to the cases where (in the distorted tetrahedra that exist for $c_0/a_0 \neq 1.633$) Model I maintains the four tetrahedral bonds equal in length, and in Model II the four tetrahedral bond angles are kept equal. Models III and IV have been proposed in the present work and are based upon considerations of the close packing of spheres. These considerations lead to a fixed tetrahedral bond length (of certain selected bonds) of magnitude $r_{cp} + r_{tet} = 1.225r_{cp}$, where r_{cp} is the radius of a close-packed anion and r_{tet} is the radius of a cation that would just fit into an ideal tetrahedral interstice of closest packed anions with $c_0/a_0 = 1.633$. In Model III, the single bond parallel to c_0 is maintained fixed at $1.225r_{cp}$, while the other three bonds are allowed to vary with the distortion. In Model IV, the three bonds skewed to c_0 are held fixed while the single bond parallel to c_0 is allowed to lengthen as c_0/a_0 decreases; and finally, at $c_0/a_0 = 1.49$, the cation would be in the plane of the three lower cations as shown at the top of Figure 6.

Between Models I and II, the latter can be rejected, since curve u_1 in Figure 6 has the wrong slope. Between Models III and IV, the former can be rejected on two counts: (1) u_3 does not show the upturn towards the line $u = 0.500$ that either u_4 or the A or r^{-12} curves exhibit; and (2) contrary both to Model IV and to experimental observations, Model III predicts that the bond length parallel to c_0 should be shorter than the bonds skewed to c_0 . Thus we are left with Models I and IV, and here no choice can be made since their curves, u_1 and u_4 straddle the A and r^{-12} curves. Of course it is not to be expected that a single simple model will fit the facts, since the A and r^{-12} curves themselves diverge, as shown in Figure 6. However, one feature of Model IV should be mentioned; namely, that the A and r^{-12} curves do not intersect the line at $u = 0.500$ until they are permitted to do so by the principles of close packing, i.e., until a smaller value of c_0/a_0 than for u_4 .

This material has been published as an Atomics International report, NAA-SR-11150, June 1965, as well as having been presented to the American Physical Society at the April 1965 meeting in Washington, D. C. [Bull. Am. Phys. Soc. 10, 547 (1965)].

D. TPO CRYSTAL ALGEBRA

The first two papers of the Translation-Permutation Operator (TPO) crystal algebra have been published in Acta Crystallographica (December 1964 and March 1965). The first paper lays the foundations of the algebra in terms of: (1) the geometry of the atoms (A positions) and the plane, triangular interstices (B and C positions) of a closest packed $p6mm$ monolayer; and (2) the permutations induced among these A, B, and C positions by translations from one monolayer midplane to another. The algebra can be shown to form a mathematical

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

group under certain conditions, and combinations of unit TPO lead to formulae which represent unit cells of a large number of diverse crystals. The second paper derives from the first by noting that "stacking errors" in a physical crystal correspond to errors in the "ordering" of the TPO information. In the special case that the closest packed anions (but not the interstitial cations) are ideally ordered, there results a defect that has been termed an "Interstice Lattice Stacking Fault" which is abbreviated as ILSF. Theoretical examples were given for the polar ZnS structure, the nonpolar CdX₂ structure, and the complex spinal structure. Direct correlations were offered between the theoretical possibilities for the ZnS structure and the inversion twins found in BeO crystals.

These two papers are the first of a series that looks forward to the development of a new crystal algebra that is expected to be especially useful for the description of defective and deformed crystals. The algebra has several significant advantages. First, the information content of the algebra is highly descriptive and thus very useful as a study-aid for research into complicated crystals where simply describing the crystal is often a problem unto itself. Second, the algebra has a heuristic purpose of leading to new areas of research; for example, the second algebra paper concerning ILSF suggests that these defects might well be found in a wide range of structures where there might also be practical applications, e.g., the spinel-ferrite structure. Third, and perhaps most important, the algebra aims to provide a powerful tool for carrying out crystal physics calculations involving complex structures. To this end, the algebra is highly amenable to computer programming techniques. Using only a small amount of input information, the algebra can instruct the computer to generate its own atom coordinate system instead of having to laboriously write a long subprogram to do this; moreover, the algebra will permit ready changes of crystal parameters and each time will instruct the computer to generate the new set of atomic coordinates. Thus whole ranges of variations of crystal parameters can be studied instead of a few isolated cases which then require excessive interpolation to fit specific cases of interest.

E. CRYSTAL DEFECT EVALUATION

The defect structure studies started last year have been extended this year, principally by x-ray diffraction topography and related means of investigation. Near the end of this year a Lang-type camera for x-ray diffraction topography was obtained and put into use, thus considerably advancing the laboratory's capability in studying crystal defect structures.

These studies have had special significance to insight into crystal growth mechanisms and growth-related internal flaws. Following the observation that BeO crystals typically contained strain "cloud layers" (termed "strata") that were detectable by topographic methods, and identification of these as being related to periodic temperature variations, the temperature stability of the crystal growth furnaces was improved. This led to great reduction, but not total elimination, of the growth strata. The appearance of evidence, in the topographs, of intercrystal surface abrasion, led to changes in handling procedure for selected crystals, that minimize this type of surface damage.

As mentioned last year, evidence was found in the topographs for a screw dislocation located on the axis of a typical BeO prism or pyramid. An example

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

of this is shown in Figure 7. The existence of such a screw dislocation has been repeatedly confirmed in topographs, and manifested in still other ways. In Figure 8 is shown a very low-lying cone on the flat termination face of the inversion twin core at the growth end of the crystal. This cone is regarded as the buildup of a spiral growth-step immediately around the emergent screw dislocation. The size scale of growth-step height and separation is so small that the steps usually cannot be observed by electron microscopy, using conventional replication techniques. An unusual and rare example of an observable growth step pattern is shown in Figure 9. In this case, the growth steps are about 250 Å high and separated by $\sim 10\mu$. There is no question that the screw dislocation plays an important role in the growth of this type of crystal at low supersaturation ($\sim 2-3\%$).

7551-2519

Figure 7. Topograph of Etched Prismatic Crystal (002) Reflection

7551-2520

Figure 8. Reflection From Cone in Center of Core Face

7551-2521

Figure 9. Spiral Step Pattern Growth About Center of Core Face

A small amount of effort has been devoted to etching studies. One of the surprising findings was that prolonged etching in hot phosphoric acid can etch the matrix crystal away and leave the inversion twin core behind as a sharp spire. Evidently this is due to the selective attack of this acid on various crystal faces, i.e., pyramidal for the crystal matrix and near-prismatic for the core. Examples of this are shown in Figure 10. Also evident in this figure are hillocks, left by etching, on the originally first order pyramid faces. The cause of these hillocks is not known, but does not appear to be the presence of dislocations that can be seen in diffraction topographs.

Preliminary experiments have shown the usefulness of lithium molybdate (which also is the crystal growth medium) as an etchant. It is known to reveal screw dislocations emergent on (0001), for example. First observations show other features of etching that appear promising to study of dislocation structures in BeO, whether arising from growth or from subsequent treatment such as mechanical deformation.

7551-2522

Figure 10. Selective
Attack of Etching
Acid

F. RADIATION DAMAGE

The study of crystal structure effects resulting from neutron irradiation at low temperature ($<100^{\circ}\text{C}$) was continued along several lines of investigation.

It was previously reported that (hkl) and $(00l)$ reflections were duplex in nature, having a broad component with an off-center sharp peak component. The approximate shape of the broad component and the relative position of the sharp component of an (002) reflection from a crystal irradiated to 4.6×10^{20} nvt of fast neutrons (≥ 1 Mev), determined with a double-crystal spectrometer, are shown in Figure 11. Although the coordinates of this figure are laboratory coordinates for crystal orientation (δ) and diffraction angle (θ), the shape of the reflection in reciprocal space would differ only slightly. The breadth of the broad component was interpreted in terms of x-ray scattering from small strain regions surrounding interplanar clusters parallel to the basal plane, having diameter of roughly 75 Å and total height of 100 to 150 Å. The offset position of the sharp component is attributed to expansion of the bulk crystal lattice by random distribution of point defects. Both the defects and cluster-strain region are represented in Figure 12.

It was determined that although the intensity of the (002) broad component rose rapidly with irradiation above 10^{20} nvt, its shape did not change significantly. This suggests that the strain regions surrounding basal plane clusters do not change size with irradiation, but, instead, increase in number as irradiation proceeds. If this is a valid interpretation, it appears that the strain regions achieve an equilibrium size quickly after nucleation, and increased diffraction intensity simply reflects greater numbers of such regions.

The existence of β -BeO in irradiated and crushed BeO has not been confirmed. It was reported last year that very small particles of β -BeO were observed, by electron microscopy, in material derived from irradiated and crushed single crystal BeO. This led to an intensive examination of both single crystal BeO and polycrystalline BeO (from General Electric Co., Cincinnati) samples which were irradiated at low temperature ($<100^{\circ}\text{C}$) to doses up to 5×10^{20} nvt, and then crushed. Since no evidence was found for the β -BeO phase, the original observation is unconfirmed and unexplained. Although the existence of β -BeO in irradiated and crushed BeO cannot be said to be impossible, its observation in the electron microscope presently appears to be at least uncommon.

In spite of this, the potential significance of the α - β transition relative to radiation damage remains intriguing. It would be expected that isomorphic structures would show the same general radiation damage effects. This does not appear to be the case here, though, since α -SiC, having tetrahedral coordination in common with BeO but not exhibiting a phase change comparable with that in BeO, shows nearly isotropic expansion with increasing radiation damage, whereas BeO radiation-induced expansion is highly anisotropic. The nature of the expansion in BeO is quite suggestive of the α - β transition. It seems not

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Figure 11. Map of Projected Intensity Distribution Expressed in Contours of Equal Intensity, in (002) Reflection for Crystal Irradiated to 4.6×10^{20} nvt Fast Neutrons, at Low Temperature ($<100^\circ\text{C}$)

7551-2523

7551-2524

Figure 12. Diagram of Matrix Structure Expanded by Randomly Distributed Point Defects, and Strain Region Around Interplanar Cluster

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

unlikely that the atomic displacements and annealing adjustments may create a metastable disordered structure, having some characteristics of the β -BeO crystal structure and giving rise to the expansion anisotropy. Confirmation or refutation of this conjecture is considered quite important to the theory of radiation damage in BeO.

A group of crystals irradiated at high temperature was received about mid-year. These crystals were irradiated to $\sim 10^{21}$ nvt (neutron energy ≥ 1 Mev) at a temperature of about 1000°C (courtesy of Carl Collins, General Electric Company). Even casual examination of the crystals revealed that the annealing effects at this high temperature were not sufficient to prevent physical degradation. The degradation was evident from the following.

- 1) Extensive splitting on prismatic planes.
- 2) Extensive surface damage on large crystals, with many wandering cracks, and some spalling of flakes from the surface.
- 3) Broad bands not reflective to x-rays, evident in x-ray diffraction topographs. Figure 13 shows this feature on a first order pyramid plane with a $(10\bar{1}0)$ Berg-Barrett pattern. The bands follow optically-visible surface cracks.
- 4) Colorless but opaque state due to internal cloudiness.
- 5) Strain was evident in x-ray diffraction rocking curves, which were broadened to the order of $1/4$ degree. By way of contrast, rocking curves before irradiation are typically on the order of $1/4$ minute (excluding instrumental broadening).
- 6) Streaking between certain reflections in the reciprocal lattice C^* direction, indicating the presence of stacking faults.

7551-2525

Figure 13. X-ray Diffraction Topograph Showing Broad Bands Following Surface Cracks

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

This last item indicates the relative stability of stacking faults that are regarded as being the interplanar clusters that were found to be formed during low temperature. Following irradiation at low temperature, the point defects disappear and the clusters grow in size during annealing at intermediate temperature; only at higher temperature ($>1200^{\circ}\text{C}$) do the clusters anneal out. It is not surprising, therefore, to find clusters (judging from the reciprocal space streaking) formed and retained during the high temperature irradiation.

In examining the crystals irradiated at $\sim 1000^{\circ}\text{C}$, it was found that a crystal structure other than that of normal BeO was observed. The origin of this structure was a thin shell coating most or all of the surface of every crystal. The structure was crystallographically related to that of the BeO structure, and differed most obviously in that the a-axis was quadrupled over that of BeO. The significance of this structure is indeterminate at the present time. The diffraction pattern closely resembles that of the compound BeAl_2O_4 , and possibly may in fact be this compound. The crystals may have had some very small patches of alumina powder clinging to them. In order to account for the widely-spread thin shells, however, it would be necessary to have had a considerable amount of alumina present, it would have had to migrate uniformly over the entire surface of every crystal and then react with the crystal to form the epitaxial coating. The alternate explanation is that the coating is a radiation-induced product deposited at the crystal surface by diffusion from the interior. Neither explanation is convincing at present, and resolution must await crystals, now being irradiated at high temperature, which have no exposure to Al_2O_3 . One further structure was found in a single case, also epitaxially related to the normal BeO; the character of this structure was not investigated.

BeO crystals of various shapes and sizes for a variety of experimental studies have been sent to General Electric Co., Cincinnati (Carl Collins) and Oak Ridge National Laboratory (G. W. Kielholtz) for neutron irradiation. It is expected the crystals will be irradiated in a schedule consistent with other planned irradiations at each laboratory, for a range of flux densities and total neutron doses, and over a range of temperatures up to 1200°C .

G. MICROSCOPY HOT STAGE DEVELOPMENT

One of the reports finished this year, based upon work and studies of prior years, deals with the development and use of a vacuum hot stage attachment to a research microscope. A pictorial representation of the furnace is shown in Figure 14. The furnace was designed for microscopy of transparent thin section samples at temperatures up to 2000°C over long periods of time, allowing time-lapse movies to be made of observable phenomena, such as grain growth, as one example. The report was prepared in both textual and film form.

H. PAPERS OF PRESENTATIONS

Reports and papers presented at professional society meetings and arising from this program are listed below.

- 1) "The Application of Radionuclides to the Determination of Phosphorus in As-grown Beryllium Oxide Crystals," C. D. Bingham and S. B. Austerman. Accepted for publication in International Journal of Applied Radiation and Isotopes.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Figure 14. Furnace for Long-Time, Hot-Stage,
Microscopy of Thin-Section Samples at 2000°C

- 2) "Electron Microscopy of Beta-Beryllia," A. H. Willis, S. B. Austerman, and K. Dearborn. *Nature*, 202, 689 (1964).
- 3) "Determination and Interpretation of Cation Diffusion in Single Crystal BeO," S. B. Austerman and J. W. Wagner. Presented at American Ceramic Society, Pacific Coast Regional Meeting, October 29-30, 1964.
- 4) "Self-Diffusion and Mass Transport in BeO," S. B. Austerman. Presented at American Ceramic Society, Pacific Coast Regional Meeting, October 29-30, 1964.
- 5) "Brittle and Plastic Behavior of Hot-Pressed BeO," G. G. Bentle, and R. M. Kniefel. Submitted for publication to American Ceramic Society.
- 6) "Grain Boundary and Surface Influences on Mechanical Behavior of Refractory Oxides — Experimental and Deductive Evidence," S. C. Carniglia, North Carolina State University Conference on "Grain Boundary and Surface Effects," November 16-18, 1964.
- 7) "Translation — Permutation Operator Algebra for the Description of Crystal Structures. I. Ideal Closest Packing," W. G. Gehman, *Acta Cryst.* 17, 1561 (1964).
- 8) "Hot-Pressing High Density, Small Grain Size Beryllia," Ralph E. Johnson, *Bull. Am. Ceram. Soc.* 43, 886-8 (1964).
- 9) "Determination of Cation Diffusion in Single Crystal BeO and Interpretation of Cation Diffusion in BeO," S. B. Austerman and J. W. Wagner. Submitted for publication to American Ceramic Society.
- 10) "Defect Structure in Large BeO Crystals," S. B. Austerman, J. B. Newkirk, and D. K. Smith. Presented at the 94th AIME Annual Meeting, Chicago, Illinois, February 14-18, 1965.
- 11) "The Elastic Constants of BeO at Room Temperature," G. G. Bentle. Submitted for publication to American Ceramic Society.
- 12) "Diffusion and Creep in Beryllium Oxide," S. B. Austerman and R. Chang. *J. Nucl. Mat.* 12, 337-9 (1964).
- 13) "Study of Defect Structures in BeO Single Crystals by X-ray Diffraction Topography," S. B. Austerman, J. B. Newkirk, and D. K. Smith. Submitted for publication to Journal of Applied Physics.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

- 14) "Graphite as Grain Growth Inhibitor in Hot-Pressed BeO," Kasimir Langrod. J. Am. Ceramic Soc. 48, 110-1 (1965).
- 15) "Multiple Thin Section Grinding. A New Tool," A. C. Hott and V. C. Davis. Submitted for publication to American Ceramic Society.
- 16) "Calculation of the Madelung Constant and Inverse-Power Repulsion Factors for the Wurtzite Crystal Structure," W. G. Gehman. NAA-SR-11150.
- 17) "Degradation of Growth Perfection in BeO Crystals by Silica," S. B. Austerman. Submitted for publication to Applied Physics Letters.
- 18) "Crystal Structure of Beta-Beryllia," D. K. Smith, C. F. Cline, and S. B. Austerman. J. Nucl. Mat. 14, 237 (1964). (Condensed form.)
- 19) "Crystal Structure of Beta-Beryllia," D. K. Smith, C. F. Cline, and S. B. Austerman. Acta Cryst. 18, 393-7 (1964) (Expanded form).
- 20) "Self Diffusion in BeO," S. B. Austerman, J. Nucl. Mat. 14, 248 (1964).
- 21) "Growth and Properties of Beryllium Oxide Single Crystals," S. B. Austerman. J. Nucl. Mat. 14, 225 (1964).
- 22) "Hot-Pressing for Nuclear Applications of BeO: Process, Product, and Properties," S. C. Carniglia, R. E. Johnson, A. C. Hott, and G. G. Bentle. J. Nucl. Mat. 14, 378 (1964).
- 23) "Variation of the Electrostatic and Repulsion Energies of the Wurtzite Structure with Variation in the c_0/a_0 and u -Parameters," W. G. Gehman. Bull. Am. Phys. Soc. 10, 547 (1965).
- 24) "Translation - Permutation Operator Algebra for the Description of Crystal Structures. II. Interstice Lattice Stacking Faults," W. G. Gehman and S. B. Austerman. Acta Cryst. 18, 375 (1965).
- 25) "A Study of Grain Growth in BeO Using a New Transmitted Light and Hot Stage," A. C. Hott and R. L. Eichelberger. Submitted for presentation at the International Conference on Sintering and Related Phenomena, Notre Dame University, June 21, 1965.
- 26) "A study of Grain Growth in BeO, Using a New Transmitted Light Hot Stage," A. C. Hott and R. L. Eichelberger, NAA-SR-11171.
- 27) "Dimensional and X-ray Diffraction Changes in Irradiated Single Crystal BeO," S. B. Austerman and K. T. Miller. Submitted for publication to Physica Status Solidi.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

A number of tasks have been advanced to, or near, the point of successful completion and termination, such as self-diffusion and BeO crystal growth. Data from other studies during the previous year were organized and appear in report or paper form as listed in the previous section.

The stock of high quality BeO crystals was added to during the year, so that an adequate stock for experimental purposes is at hand. Attention to crystal growth is being redirected toward graphite.

The examination of BeO crystals and evaluation of defect structures has contributed considerable insight into the nature of typical crystals and the mechanisms by which they grow from solution. In part, this has led to improved growth and handling procedures; in part, the crystals were characterized as to their preirradiation characteristics so that postirradiation properties could be most significantly evaluated.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

With respect to self-diffusion studies, the data accumulated with single crystals and the final interpretive studies confirmed and strengthened earlier conclusions. Because of the suspected oxygen diffusion along surface microcracks in the surface of single crystals, it may not be feasible to analyze oxygen diffusion in large crystals by sectioning methods; this point is still under consideration.

Radiation damage studies in BeO have progressed with clarification and reporting of damage structure due to low-temperature ($<100^{\circ}\text{C}$) irradiation. The nature of the type of point defects causing anisotropic expansion has received serious attention but has not been resolved. It is evident from the obvious appearance of single crystals irradiated at the higher temperature of $\sim 1000^{\circ}\text{C}$ that self-annealing at this high temperature is not itself adequate to prevent physical degradation during irradiation. Although some increase of radiation tolerance may occur at high temperature, the damage to single crystals indicates that the tolerable radiation dose at 1000°C is less than double that at 100°C . Crystals presently being irradiated should allow verification of this conclusion and should make possible more extensive investigation and interpretation than heretofore possible with single crystals. Evaluation of preirradiation defect structure and development of procedures for studying defect structures, both pre- and post-irradiation, will considerably enhance the radiation damage studies.

The studies on crystal energy calculations and crystal algebra constitute significant advances in theoretical treatment of crystal physics.

Crystal defect studies have produced some exciting developments, but have only scratched the surface. The studies to date lay the groundwork for more intensive studies on defect structure, both before and after subjecting the crystals to various kinds of damaging conditions. It is planned to extend these studies into the following year with a broadened scope, having emphasis on radiation damage effects. In order to fully assess radiation damage, evaluation of appropriate preirradiation properties and structure defects necessarily also must be performed. The broadened scope is planned to embrace refractory moderator and related materials instead of only BeO.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Engineering Development

Project: Continuous Neutron Flux Monitor

Project Engineer: C. R. Smith

Reporting Period: Fiscal Year 1965

AEC Category: 04-40-03-01

General Order: 7616

Subaccount: 4584

I. PROGRAM OBJECTIVES

The general objective of this project is to develop, test, and evaluate new types of gamma-insensitive neutron flux monitors for continuous operation in high-temperature, high-flux regions. The concept of monitoring neutron flux by measuring its effect on a stream of neutron sensitive fluid was tested prior to this fiscal year and found to be practical. Three gaseous systems had been evaluated. The best of these depended upon the neutron activation of a stream of argon gas to form radioactive Ar^{41} . Incore tests had confirmed that in an isothermal system the activity of Ar^{41} in the effluent gas is directly related to the neutron flux incore. These tests had also demonstrated that in a nonisothermal system, the activity in the gas stream would respond to changes in gas temperature as well as to changes in neutron flux level. It was, of course, desirable to neutralize the effect of temperature changes on signal. To this end, an analytical investigation of the laws governing gas flowrate and activation was undertaken in fiscal year 1964 and continued into fiscal year 1965. Several equations were derived whereby neutron flux could be expressed in terms of physically measurable parameters while nullifying the effect of temperature. Analog computer circuits were designed to solve these equations.

The objectives of this year's efforts were to: (a) derive and perfect suitable equations for measuring neutron flux, (b) design, build, and test analog computers to solve the best of these equations, (c) integrate these computers into the gas handling system and evaluate the applicability of the entire system for neutron flux monitoring, (d) explore the range of neutron flux which can be conveniently monitored with these systems, and (e) investigate time constants of the system with respect to changes in neutron flux and other operational variables.

II. TECHNICAL ACCOMPLISHMENTS IN FISCAL YEAR 1965

In following the development of this project through FY 1965, it will be helpful to refer to a simplified flow diagram for the system as shown in Figure 1. Argon gas is supplied to the system from a commercial cylinder of compressed argon. The flow of gas through the system is controlled by pressure reducers and fine-control needle valves. Before irradiation, the gas passes through a mass flowmeter which generates an electrical signal, Q_m , proportional to mass flowrate. The pressure of the gas near the outlet of the flowmeter is measured by both a pressure gage and pressure transducer. This pressure, P_1 , is considered the inlet pressure of the system. The gas next travels via small bore tubing through an incore activation chamber, V_{ac} , which is at temperature, T_{ac} and pressure P_{ac} , and in which neutron activation of the gas principally takes place. The gas, now containing radioactive Ar^{41} , flows through small bore

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Figure 1. Simple Argon
Flux Monitor

7616-2501

tubing out of the reactor and through a NaI(Tl) scintillator crystal which acts as a counting chamber, V_{sc} . Disintegrations of Ar^{41} passing through the chamber are detected by a photomultiplier tube; light pulses generated in the crystal are amplified, their energies are discriminated to favor detection of the 1.29 Mev γ from the decay of Ar^{41} and their countrate, C_{sc} , is recorded. The pressure of the argon gas at the scintillator, P_{sc} , is measured by a gage and pressure transducer before it passes through the outlet needle valve to an evacuated holdup tank.

A. DERIVATION OF NEUTRON FLUX EQUATIONS

Under conditions of constant flowrate and constant neutron flux, the number of Ar^{41} atoms in the activation chamber is given by

$$n^{41} = n^{40} \sigma \phi \tau_{ac} = \frac{N_o \rho_{ac} V_{ac}}{M} \sigma \phi \tau_{ac} \quad , \quad \dots (1)$$

where:

n^{41} and n^{40} = atoms of Ar^{41} and Ar^{40}

σ = the microscopic cross section of Ar^{40}

ϕ = neutron flux

τ_{ac} = residence time of gas in activation chamber

N_o = Avogadro's number

M = molecular weight of argon

ρ_{ac} = gas density in activation chamber

The ratio of n^{41}/n^{40} as the gas leaves the activation chamber is $\sigma \phi \tau_{ac}$. Since the time required for the gas to flow to the scintillation chamber is very short relative to the half life of Ar^{41} (110 min), this ratio remains unchanged. Therefore, we can say that the decay rate at the scintillation chamber is

$$D = \frac{\lambda N_o \rho_{sc} V_{sc}}{M} (\sigma \phi \tau_{ac}) \quad , \quad \dots (2)$$

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

where ρ_{sc} is the gas density in the scintillation chamber; and, if we define G as the ratio of disintegration rate to countrate, D/C_{sc} , we can write

$$\phi = \frac{C_{sc} M G}{\sigma \tau_{ac} \lambda N_o \rho_{sc} V_{sc}} \quad \dots (3)$$

The average residence time of the gas passing through the activation chamber is

$$\tau_{ac} = \frac{\rho_{ac} V_{ac}}{Q_m} \quad \dots (4)$$

Assuming ideal gas behavior,

$$\rho_{ac} = \frac{P_{ac} M}{RT_{ac}} \quad ; \quad \rho_{sc} = \frac{P_{sc} M}{RT_{sc}} \quad \dots (5)$$

Substituting Equations 4 and 5 in 3 and grouping all constants under K_1 (T_{sc} being considered constant) we obtain

$$\phi = K_1 \frac{C_{sc} Q_m T_{ac}}{P_{ac} P_{sc}} \quad \dots (6)$$

This is a generalized equation for neutron flux, valid regardless of whether flow is laminar or turbulent.

Solutions to Equation 6 obtained by using transducers and an analog computer are not practical if P_{ac} has to be measured in a high temperature, high-flux region. Measurement of T_{ac} with a thermocouple is feasible but undesirable.

If flowrates of the gas are held within the limits of viscous laminar flow and if the activation chamber is located midway between P_i and P_{sc} , we can express P_{ac} in terms of P_i and P_{sc} and Equation 6 becomes

$$\phi = K_2 \frac{C_{sc} Q_m T_{ac}}{(P_i^2 + P_{sc}^2)^{1/2} P_{sc}} \quad \dots (7)$$

A gas handling system and an analog computer designed to solve this equation were tested at the AE-6 reactor and at the STIR (Shield Test Irradiation Reactor).

The need for an incore thermocouple to measure T_{ac} can also be obviated. This requires the design of a modified system called the "Pigtail Circuit" shown in Figure 2 where a length of small bore tubing wrapped around the activation chamber becomes the flow controlling segment of the system. Using this system

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Figure 2. Pigtail Circuit for Monitor

and holding P_i and P_{sc} constant (and for argon gas only), the following equation can be derived:

$$\phi = K_3 C_{sc} Q_m^{0.434} \quad \dots (8)$$

A gas handling system based on this equation was built and tested at the STIR. A companion analog computer has yet to be designed.

B. DESIGN OF AN ANALOG COMPUTER FOR EQUATION 7

Figure 3 is a highly simplified block diagram of an analog computer used in solving the equation

$$\phi = K(C_{sc} Q_m T_{ac})(P_i^2 + P_{sc}^2)^{-1/2}(P_{sc})^{-1}$$

Figure 3. Simplified Computer

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

A detailed description of this circuit can be found in NAA-SR-11355 which will soon be published.

Each squaring function requires three amplifiers, one squaring circuit or bistable, and two transistor switches. Each of the two XYZ-1 blocks requires two amplifiers, two switches, and one bistable. All amplifiers are Philbrick Researches Model P-65A with the exception that model SP 656 amplifiers are used to amplify transducer outputs to suitable magnitudes for input to the computer.

C. DESIGN AND TEST OF GAS HANDLING CIRCUITS

The following combinations of tubes and chambers were assembled for tests at the AE-6 and STIR.

Reactor	System	Tubing			Chamber Volume cm ³
		l_1	l_2	l_3	
AE-6	No. 1	2 lengths, 34 ft ea 65 mil ID Cu	2 lengths, 6 ft ea 16.2 mil ID stainless steel	None	2-1/2
STIR	No. 2	2 lengths, 41 ft ea 65 mil ID Cu	2 lengths, 6 ft ea 16.2 mil ID stainless steel	None	2-1/2
STIR	No. 3	2 lengths, 41 ft ea 65 mil ID Cu	2 lengths, 6 ft ea 16.2 mil ID stainless steel	None	9
STIR	Pigtail	2 lengths, 41 ft ea 65 mil ID Cu	2 lengths, 6 ft ea 26-1/2 mil ID stainless steel	1 length, 3 ft ea, 10 mil ID stainless steel	24

Flowrate measurements were made through these systems under varying conditions for P_1 , P_{sc} , Q_m , and T. These measurements confirmed that flow-rate obeyed Poiseuille's Equation.

It will be noted that several sizes of tubing were used in each assembly. These were arranged symmetrically from the midpoint of the circuit; that is, the lengths of 65-mil ID Cu tubing were at the inlet and scintillator ends of the circuit. Closer incore, were the 16.2-mil ID lengths of stainless steel tubing. The activation chamber (and the 10-mil ID tubing, when used) was located at approximately midpoint along the circuit. Lengths l_2 , l_3 , and the activation chambers were insulated so that they could be heated electrically. The temperatures of the tubing and chambers were monitored with chromel-alumel thermocouples.

D. IRRADIATIONS AT THE AE-6 REACTOR

The AE-6 is a solution type reactor capable of a maximum power level of 1000 watts. The thermal neutron flux in the AE-6 central exposure facility at this power level is 4×10^{10} neutrons-cm⁻² sec⁻¹.

$$1. \frac{C_{sc}}{\phi} = f(P_i, P_{sc})$$

The first test of importance was to confirm Equation 7 while holding all factors constant except P_i and P_{sc} . We used System No. 1. Reactor power was 100 watts throughout. Count rate per unit flux was measured as a function of inlet pressure for two values of P_{sc} . The behavior of C_{sc}/ϕ as a function of P_i gave excellent agreement with theoretical curves shown in Figure 4. The value for ϕ in these experiments was the estimated flux at the activation chamber, not the signal from the flux computer.

Figure 4. Comparison of Gas Inlet Data

2. Stability

The next test was undertaken to confirm the stability of the flux computer signal at constant reactor power. The reactor was operated without cooling water at constant power levels of 200 and 300 watts. Under these circumstances, the control rods had to be withdrawn slowly to hold power constant. Using System No. 1 at constant flowrate, we recorded the flux computer signal. Simultaneously, we irradiated sets of gold, tungsten, and dysprosium alloy wires in

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

the vicinity of the activation chamber by use of a pneumatic rabbit. The activity of the wires confirmed the flux computer signal within about 5%. Some drift in computer signal was observed. We believe this was due to photomultiplier tube drift and this has been subsequently corrected.

3. Flux Computer Versus Reactor Power

Finally, we tested our flux computer over a three-decade range of reactor power. System No. 1 was again used. Mass flowrate was approximately constant and T_{ac} varied about 10°C due to core heating. The reactor was operated from 1 to 1000 watts. The results of this test are shown in Figure 5. The internal consistency of these data appears to be excellent. However, we observed that a 50-fold increase in reactor power from 20 to 1000 watts generated only a 42-fold increase in the flux computer signal. The reason for this has not yet been established.

7616-2505
Figure 5. Flux Signal vs Power

E. IRRADIATIONS AT THE SHIELD TEST IRRADIATION REACTOR

The STIR is a pool type test reactor. It is capable of generating a power level of 1 Mwt and a maximum thermal neutron flux of 1.2×10^{13} neutrons $\text{cm}^{-2} \text{sec}^{-1}$ in its main central exposure facility. Tests of the Argon Flux Monitor in this reactor are now complete but data reduction was still in progress at this writing.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Temperature, dynamic range, and response measurements were made on Systems No. 2, 3, and 4. In one of these tests, the ability of the Pigtail Circuit to monitor more than six decades of reactor power was demonstrated. The STIR was operated from 1 watt to 1 megawatt. Variations in reactor power of 0.1 to 0.2 watts while at 1 watt were clearly discernable to the Argon Flux Monitor. At 1 Mw γ heating increased the temperature of the activation chamber from 80 to 275°C. In the course of this temperature excursion, the count rate varied less than 2%. This demonstrates temperature compensation over this range at least. Our data have not yet been studied sufficiently to provide other details from these tests but these will be fully reported in NAA-SR-11356, a topical report now in preparation.

F. TOPICAL REPORTS

The following reports were prepared during this fiscal year and will be published shortly:

K. H. Kline, "Analog Computer for the Argon Flux Monitor," NAA-SR-11355, 20 pp

C. R. F. Smith and P. Spiegler, "Development of the Argon Flux Monitor Through GFY 1965," 50 pp

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

All of the objectives set forth in Section I have been attained. Some reduction of data from the STIR tests is still in progress, and these results will be included in NAA-SR-11356. Nevertheless, the following tasks have been successfully accomplished.

A. Two equations have been derived, either of which can be used with Argon Flux Monitor systems to measure neutron flux.

B. Argon Flux Monitor systems compatible with these equations have been built and tested successfully incore.

C. An Analog computer to solve one of these equations was designed and successfully tested during incore tests over a three-decade range of reactor power.

D. The Pigtail Circuit of the Argon Flux Monitor was shown to be able to monitor a range of more than six decades of reactor power.

E. The Pigtail Circuit of the Argon Flux Monitor was tested from 80 to 275°C at 1 Mwt and its signal appeared to be temperature compensated over this temperature range.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Advanced Development

Project: Multicycle Reprocessing

Project Engineer: G. E. Brand

Reporting Period: Fiscal Year 1965

AEC Category: 04-40-04-03

General Order: 7522

Subaccount: 4612

I. PROJECT OBJECTIVES

The objectives of this project are to complete the demonstration of low-decontamination methods for reprocessing uranium dioxide fuels irradiated to levels anticipated in power reactors, and to phase out all reprocessing studies. Evaluation of 2nd-cycle irradiated uranium dioxide is to be completed, and applicable reports written.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

Nuclear reactivity was measured before and after the second-cycle of irradiation and is reported in Table 1.

TABLE 1
EFFECTIVE FISSION PRODUCT POISON IN IRRADIATED
URANIUM DIOXIDE

Capsule NAA 57-	Pre-Second Cycle			Post-Second Cycle		
	Burnup (Mwd/MTU)	Equiv. Boron (ppm UO ₂)		Cumulated Burnup (Mwd/MTU)	Equiv. Boron (ppm UO ₂)	
		Thermal	Fast		Thermal	Fast
-3	5,700	117	127	15,200	151	132
-4	18,000	125	111	27,300	177	171
-5	21,000	190	111	31,300	229	215

The fission product contribution to the fuel's capture cross-section was obtained by determining the difference between the calculated and measured reactivity. The calculated reactivity was based on the sum of the U²³⁵ content as determined by mass spectrographic analysis and plutonium content as determined by radiochemical analysis. Computer calculations indicated that, for the level of burnup these materials had experienced, the fission product neutron cross-section should be essentially constant. The results of the reactivity measurements experimentally confirm this conclusion. The inconsistent result for the -5 capsule in thermal flux has not been explained.

Second-cycle irradiated UO₂ from three reprocessed, reirradiated batches was oxidatively deacid. Particle size diminution required three or four oxidation-reduction cycles to produce a sinterable powder. These data are presented in Table 2.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 2
SUMMARY OF DATA FOR REPROCESSED SECOND-CYCLE
IRRADIATED URANIUM DIOXIDE

Experiment Number	Irradiation (Mwd/MTU)	Ox-Red Cycles	Ignition Temperature (°C)	Tap Density (gm/cc)
NAA 57-5	31,000	1	-	-
		2*	-	2.73
		3*	-	2.71
		4*	-	2.65
NAA 57-4	28,000	1	380	2.68
		2	160, 410	1.94
		3	140, 350	1.46
NAA 57-3	15,700	1	310, 455	3.17
		2	200, 425	2.57
		3	125	2.30
		4	145, 350	1.57

*Furnace malfunctioned, ineffective oxidation-reduction

Uranium dioxide containing the equivalent of 100,000 Mwd/MTU in stable fission products and irradiated to 5,000 Mwd/MTU was oxidatively degraded and processed through two oxidation-reduction cycles. Behavior was similar to the 2nd-cycle irradiated UO₂.

Pellets were prepared from the second-cycle irradiated UO₂ using Carbowax 4000 as binder and zinc stearate as die lubricant. A minor malfunction of the powder dispenser caused the pressing of several oversize pellets. These were crushed and repressed. Pellets were sintered for 12 hr at 1730°C in flowing 25% H₂ - 75% Ar. A Deoxo purification unit was used. Pellet data are included in Table 3.

The low geometric density of the NAA 57-5 pellets is due to insufficient particle size reduction during oxidation-reduction, i. e., an insufficient number of effective cycles.

For comparison, a summary of results of the first cycle examination is presented in Table 4.

Radiochemical analyses indicated that decontamination was similar to first-cycle reprocessing-refabricating (Table 5).

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 3
SUMMARY OF PELLET DATA FOR REPROCESSED SECOND-
CYCLE IRRADIATED URANIUM DIOXIDE

Source NAA 57-	Boat	Pellet* Ident.	Diameter (in.)	% Theoretical Density†		
				Geom.	Air	Water
-4	Top	I	0.3111	92.9	95.9	-
		A	0.3112	92.9	94.9	-
		B	0.3107	94.4	95.6	96.1
		C	0.3098	94.9	96.7	-
		Avg.	0.3107	93.8	95.8	-
-3	Middle	I	0.3139	92.9	92.5	-
		A	0.3142	92.3	94.0	97.2
		B	0.3157	92.6	94.6	-
		C	0.3151	92.2	94.0	-
		Avg.	0.3147	92.5	94.2	-
-5	Bottom	I	0.3395	63.5	-	-
		RP1	0.3389	65.9	-	-
		RP2	0.3375	66.8	-	-
		RP3	0.3392	66.8	-	-
		SP1	0.3438	61.7	-	-
		SP2	0.3344	66.4	-	100.0
		SP3	0.3427	61.4	-	-
		Avg.	0.3394	64.6	-	-

*RP = pellet crushed and repressed, SP = single pressed.

†NAA 57-4, and -5 densities based on 10.80 gm/cc, NAA 57-3 densities based on 10.90 gm/cc.

TABLE 4
SUMMARY OF PELLET DATA FOR REPROCESSED FIRST-CYCLE
IRRADIATED URANIUM DIOXIDE

2nd Cycle Irrad. Exp. Number	Boat	Avg. Pellet Diam. (in.)	% Theoretical Density*	
			Geometrical	Air
NAA 57-3	Top	0.3024	94.1	98.1
NAA 57-4	Middle	0.3030	91.1	94.0
NAA 57-5	Bottom	0.3106	90.3	94.3

*Based on a theoretical density of 10.97 gm/cc

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 5
DECONTAMINATION FACTORS, SECOND-CYCLE AIROX
REPROCESSED-REFABRICATED URANIUM DIOXIDE

Source NAA 57-	Burnup (Mwd/MTU)	Sr ⁹⁰	CePr ¹⁴⁴	Cs ¹³⁷	ZrNb ⁹⁵	RuRh ¹⁰⁶
-3	15,200	1.0	1.0	>5	0.4	0.6
-4	27,300	1.0	0.9	>6	1.1	1.2
-5	31,300	1.1	1.2	>25	0.7	0.4

Uranium isotopic content was determined on a refabricated pellet from NAA 57-5 with the following results: U²³⁴, 0.06%; U²³⁵, 4.63%; U²³⁶, 0.63%; U²³⁸, 94.68%.

Processing equipment has been disposed of and the hot cave has been decontaminated.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The postirradiation examination and evaluation were completed. It has been demonstrated that the AIROX process can successfully close the UO₂ fuel cycle and provide fuel with physical properties comparable to production grade UO₂. All applicable reports have been written and submitted for publication.

The hot cave has been decontaminated.

Reports issued are as follows.

NAA-SR-9161, "Reprocessing of Uranium Carbide by a Nitride-Carbide Cycle. II. Kinetics of Nitride Conversion to Carbide, L. A. Hanson (October 15, 1964)

NAA-SR-9278, "Reprocessing of Uranium Carbide by a Nitride-Carbide Cycle. III. Complete Cycle and Fission Product Study," L. A. Hanson (October 15, 1964)

NAA-SR-8213, "Refabrication and Encapsulation of Highly Irradiated Uranium Dioxide," J. Guon, J. E. Bodine, and R. J. Sullivan (December 1, 1964)

NAA-SR-9335, "Comparative Fuel Cycle Evaluations, Low Decontamination Pyroprocessing and Aqueous Reprocessing. Part II. UC Fuel in a Thermal Reactor," K. L. Mattern and L. J. Colby, Jr. (February 15, 1965)

NAA-SR-10738, "Oxidation-Reduction Reprocessing of Uranium Carbide Reactor Fuel. II. Behavior of Plutonium and Fission Products," W. G. Smiley (June 15, 1965)

Further reports written are as follows.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

NAA-SR-10485 "A Rotary Kiln for the Controlled Oxidation of UC,"
S. Strausberg
NAA-SR-10486 "Preparation of UC by Carbothermic-Reduction of U_3O_8 ,"
S. Strausberg
NAA-SR-10624 "The Use of Additives in the Preparation of Uranium Carbide,"
L. A. Hanson
NAA-SR-10737 "Electrolysis of Uranium Carbide in Fused Salt Using Molten
Metal Cathodes," M. L. Iverson and R. J. Sullivan
NAA-SR-11235 "The Chemical Reactivity of Uranium Carbide," E. W. Murbach
NAA-SR-11340 "Pyrochemical Reprocessing of Uranium Carbide Summary
Report," E. W. Murbach and G. E. Brand
NAA-SR-11375 "Second Cycle AIROX Reprocessing and Pellet Refabricating
of Highly Irradiated Uranium Dioxide," J. E. Bodine, J. Guon, and
R. J. Sullivan
NAA-SR-11389 "Pyrochemical Reprocessing of UO_2 by AIROX Summary
Report, G. E. Brand and E. W. Murbach
NAA-SR-11401 "Kinetics of the Reaction of Uranium Monocarbide with Water,"
L. J. Colby, Jr.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Reactor Safety

Project: Study of Reactor Housing Installations

Project Engineer: A. A. Jarrett

Reporting Period: Fiscal Year 1965

AEC Category: 04-60-20-01

General Order: 7603

Subaccount: 1373

I. PROJECT OBJECTIVES

The general objective of this program is to determine adequate design requirements for construction of low-leakage reactor housing installations using conventional building materials. Specific objectives are: (a) to measure the air leakage rates as a function of pressure differential through typical components and materials; (b) to establish methods and techniques for economically reducing leakage; (c) to establish the validity of utilizing component leak rates for predicting the leak rates of full-scale structures; (d) to provide guides for the design and construction of reduced leakage structures; (e) to study the economic potential of low-pressure, improved-leakage, conventional construction as a means of containment; and (f) to publish the above results as a comprehensive manual.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

The second edition of the manual "Conventional Buildings for Reactor Containment," NAA-SR-10100, has been edited, printed, and distributed as required by the AEC contract. The manual represents a 4-yr testing program, and outlines a recommended method for design and construction of buildings to a specified leakage rate and pressure.

All test specimens, equipment, and the metal-panel model were dismantled and disposed. The concrete model is being used as a containment structure for fission product release studies. The large test cell was shipped to Hanford.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

With publication of the second edition of the manual, the objectives of the program have been met. It is believed that the manual provides useful and reliable technical data in the design, construction, and inspection of low-leakage, conventional structures whose leakage characteristics are predictable and controllable. When typical practices are followed, which are clearly defined, success in the construction of low-leakage reactor building enclosures is now a reality. It is believed that the manual will serve as a guide to architects, designers, field inspectors, contractors, and regulatory authorities.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Reactor Safety

Project: Fission Product Retention by Reactor Coolants

Project Engineer: A. A. Jarrett

Reporting Period: Fiscal Year 1965

AEC Category: 04-60-10-01

General Order: 7608

Subaccount: 1331

I. PROJECT OBJECTIVES

The general objective of the project is to determine the ability of reactor coolants to retain specific fission products released to the coolant stream as a result of fuel cladding failure or fuel meltdown. Specific objectives of the project are: (a) determination of the ability of sodium to retain radioactive iodine as a function of such parameters as sodium temperature, elapsed time from incident, sodium-to-fuel and sodium-to-gas volume ratios and geometric relationships, condition of released iodine; and (b) determination of the ability of boiling sodium and sodium in the vapor phase to retain radioactive fission products.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. EXPERIMENTAL EFFORT

The majority of experiments were specifically designed to evaluate the extent of fission product retention in sodium under various fission product injection conditions. Experiments were also performed to evaluate the extent of fission product retention associated with the injection of fission products into a sodium bond from high-burnup uranium carbide fuel. Plating and deposition studies were performed in conjunction with removal of the Sodium Reactor Experiment (SRE) primary loop. These latter studies were supported by other projects, but are reported here because of their application to fission product retention.

1. Capsule Rupture Experiments

During the latter part of the fiscal year, several changes were made in the capsule rupture experiment design. One of these, necessitated by the large volume of sodium and the higher level of radioactivity to be used, was containment of the equipment in a large inert gas experimental enclosure. A suitable concrete structure with 6-in. shielding walls was located which had been used in previous AEC project work.* A sketch of the experimental arrangement and the blockhouse is shown in Figure 1. Provision has been included in the experimental arrangement to fill the system with clean sodium, transfer the sodium to the plenum simulator, inject fission products into the plenum simulator in various forms, sample the residual off-gas as well as the sodium in the plenum, and dump the sodium content into waste containers. The structure and experimental arrangements have been completed and are undergoing preliminary vacuum and flow tests. The test fuel specimen for the first test has been irradiated, and the first capsule rupture experiment is imminent.

*Reactor Containment Study

7608-2501

Figure 1. Test Enclosure

2. Condenser Discharge Experiments

A condenser discharge melting device was designed and tested during the fiscal year, and several experiments have been performed. A block diagram of this device is shown in Figure 2. This technique permits injection of fission products from a melted or vaporized preirradiated foil into sodium vapor or vacuum with subsequent transport through a sodium plenum. During calibration tests which were conducted this year, the device indicated a probable time duration of approximately $300 \mu\text{sec}$ during which electrical energy is dissipated in the uranium foil, and a maximum energy in the present model of approximately 910 joules. The particle size spectra resulting from the discharge into the foil appear to have two predominant peaks, one in the $1\text{-}10 \mu$ size and the other larger than 10 microns.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

7608-2502

Figure 2. Plenum Experiment Flow Diagram

In the first of several supplementary experiments conducted this year, the fuel and sodium melting capabilities (provision is included for enhancing the vapor density of sodium atoms in the reaction chamber by vaporizing a small sample of sodium) were verified by discharging the condenser through various uranium and uranium-sodium configurations. Following these tests, the fission products resulting from the discharge were swept through potassium iodide solution with no sodium present. Results were reported in the second quarterly progress report. Replacement of the sampling arrangement shown in Figure 2 with the Maypack diffusion tube concept shown in Figure 3 was then accomplished, and the primary experiments were begun.

7608-2503

Figure 3. Maypack Diffusion Tube Arrangement

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

The first experiment was designated as OP-2, and the system conditions were reported in the third quarterly progress report.

Following the condenser discharge, the gas stream and sodium plenum were sampled. Results indicated an insufficient quantity of radioactivity was transported out of the reaction chamber to provide a measurable quantity in either the sodium or the gas-sampling media. The design of the gas flow system through the reaction chamber was subsequently modified, and a series of test experiments were initiated before beginning work on the CD-1, CD-2, and CD-3 series.

The purpose of the first two test experiments performed this year was to characterize the penetration and diffusion characteristics of fission products resulting from the melting and vaporization of uranium with and without a sodium plenum simulator in the system. The system conditions of these experiments were thus similar, with the exception of the sodium plenum and, both are shown in Table 1.

TABLE 1
SYSTEM CONDITIONS FOR CONDENSER DISCHARGE EXPERIMENTS

Total sodium content in sodium experiment (gm)	13.9
Depth of sodium in sodium experiment (cm)	2.7
Temperature of sodium (°F)	800
Helium flow rate through sodium (cc/min)	200
Helium velocity through sodium (cm/sec)	10.4
Total helium flow (cm ³)	3×10^3
Fuel mass (gm)	7.0×10^{-2}
Helium flow through diffusion tubes (cm ³ /min)	50
Temperature of transfer tube to sodium plenum: (°F)	500

It was observed during these tests that the transfer tube between the reaction chamber and the sodium plenum simulator retained a considerable quantity of fission products, even though heated to 200°F. The relative deposition of these isotopes is shown in Table 2.

In Tables 3 and 4 a comparison is shown of the Maypack component retention for the two experiments. It will be noted from the earlier figure that the copper screens preceded the carbon-loaded paper.

Data were also obtained from the diffusion tubes located at the juncture of each Maypack component. Figure 4 presents these data which were obtained in the various diffusion tubes during the experiment in which a sodium plenum was employed, and Figure 5 presents a comparison of similar diffusion tube data with and without the sodium plenum in the system.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 2
RELATIVE DEPOSITION OF RADIOACTIVITY
IN TRANSFER TUBE

Isotope	Activity (Normalized to BaLa ¹⁴⁰)
Ru ¹⁰³	7.7
Cs ¹³⁷	2.1
ZrNb ⁹⁵	2.7
BaLa ¹⁴⁰	1.000
I ¹³¹	362

TABLE 3
DISTRIBUTION OF ISOTOPES IN MAYPACK COPPER SCREENS*

Condition	BaLa ¹⁴⁰	I ¹³¹	Ru ¹⁰⁶	Cs ¹³⁷	ZrNb ⁹⁵
With no sodium	1.0	20	6.7	1.9	2.1
After bubbling through so- dium plenum	1.0	6.6	6.5	2.4	2.3

*Arbitrarily normalized to BaLa¹⁴⁰ to compare the two experiments

TABLE 4
DISTRIBUTION OF ISOTOPES IN MAYPACK
CARBON FILTER PAPER*

Condition	BaLa ¹⁴⁰	I ¹³¹	Ru ¹⁰⁶	Cs ¹³⁷	ZrNb ⁹⁵
With no sodium	1.0	17	6.7	1.7	2.2
After bubbling through so- dium plenum	1.0	8.4	6.5	2.4	2.3

*Arbitrarily normalized to BaLa¹⁴⁰ to compare the two experiments

7608-2504

Figure 4. I^{131} Activity After Bubbling Through Sodium Plenum

7608-2505

Figure 5. I^{131} Activity After 2nd Tube

3. Uranium Carbide Fuel Rod Measurements

During the fiscal year, three high burnup uranium carbide fuel test rods were obtained from the fuel development project (AI3-12, 81-2, and 81-4), and samples of the cover gas, sodium bond, and cladding were obtained. The burnup levels ranged from approximately 10,000 Mwd/ton to approximately 30,000 Mwd/ton, and were sampled at various times after removal from the reactor. The most successful of these examinations was performed during the first quarter when the acquisition of cover gas, sodium bond, and cladding samples from a high-burnup UC capsule experiment (AI3-12) permitted an implied measurement of fission product retention under equilibrium conditions. This particular fuel rod capsule had been inserted into the GETR in May, 1963 and was removed June, 1964; estimated burnup was 20,000 Mwd/t. The rod was 18 in. long and 0.5 in. in diameter and contained 71.6 gm of U^{235} , 644 gm of U^{238} , and 34.1 gm of C. Surrounding the rod was a sodium bond of 14.6 gm in 10-mil stainless steel cladding.

The radioactive inventory of a cover gas sample obtained from this fuel rod is shown in Table 5. (The actual amounts measured have no significance, since some of the gas activity was apparently lost due to a leak in a thermocouple weld. Therefore, the results are normalized to Kr^{85} for comparison).

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 5

RADIOACTIVITY OF COVER GAS SAMPLE AT
REACTOR OUT-TIME

Isotope	Measured Activity (μc)	Relative Activity (Measured Activity Normalized to Kr^{85})
Kr^{85}	1.0×10^{-5}	1.0
$\text{Xe}^{131\text{m}}$	1.2×10^{-5}	1.2
I^{131}	3.6×10^{-7}	0.036
Xe^{133}	1.1×10^{-4}	11.0

In Table 6 are shown the normalized data of Table 5 and a similarly normalized estimate of the fuel rod inventory. (The fuel rod inventory is also normalized to Kr^{85}).

TABLE 6

COMPARISON OF RELATIVE ISOTOPIC CONTENT OF
THE ROD COVER GAS (MEASURED) AND THE ROD
INVENTORY (ESTIMATED)

	Normalized Cover Gas Activity, (μc)	Normalized Fuel Rod Inventory (Estimated)
Kr^{85}	1.0	1.0
$\text{Xe}^{131\text{m}}$	1.2	1.7
Xe^{133}	11.0	354
I^{131}	0.036	15.3

Rather than the I^{131} being 15.3 times more abundant than the Kr^{85} in the cover gas, as might be expected from the estimated results, there was actually only 0.036 times as much I^{131} in the gas space as Kr^{85} . Therefore, the iodine must have been depleted by transport through the sodium bond, or by plating or some other phenomena. Since no I^{131} was found on the cladding, these results indicate an apparent iodine retention of $15.3/0.036$, or 415; or, to express the results in terms comparable to previous release figures, $1/415$ or 0.2%.

The data represent the only measurement of I^{131} in the cover gas of any of the fuel rods studied (probably because of the short decay-time in this case), and are reported in spite of the lack of quantitative evaluation. Attempts to measure a similar ratio in the other experiments were unsuccessful. However,

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

data were obtained from the other fuel elements concerning the deposition of other radioactivity on the cladding, as reported in the third quarterly progress report.

Some conclusions have also been drawn concerning the ratio of Xe^{131m} to Kr^{85} , and Xe^{133} to Kr^{85} in the gas space. If it is assumed that the deviation from the anticipated ratio shown in the table may be attributed to radioactive decay during transport, a fuel transport-delay time of 5 to 26 days can be inferred from these data.

4. SRE Radioactivity Deposition and Plating Studies

As reported previously*, an opportunity was provided during this fiscal year to examine sodium-coolant and piping samples from several locations in the primary loop of the Sodium Reactor Experiment (SRE). These samples were considered to be of interest because the SRE experienced a partial fuel melting in 1959, due to coolant blockage in several fuel channels. As a result, fission products were introduced into the system, and subsequently plated on the piping. Since plating and deposition of fission products have been observed to a considerable extent throughout the project, it was felt that information on this subject would be valuable to reactor safety analyses and other areas in which the ultimate fate of fission product is of interest. Figure 6 shows a typical sector of the cold-leg SRE piping and the type of black deposition which was noted on the stainless steel surface. The deposition has been tentatively identified as a

7608-2506

Figure 6. Photo and Autoradiograph of SRE Spool L-103
after 26-hr HCl Etch

*Quarterly Technical Progress Report, AEC Unclassified Programs (July-September 1964) NAA-SR-10501

NAA-SR-11450

VII-93

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

carbonaceous compound probably resulting from introduction of tetralin into the SRE just prior to channel blockage. It is of interest to note the comparable autoradiograph and the extent to which the radioactivity deposition is associated with the visual stain. A repetitive etch technique was then employed, using dilute nitric acids to ascertain extent of penetration and degree of fixation of the radioactivity plated on the piping surface. The results of this etching are presented in Figure 7.

7608-2507

Figure 7. HND₃ Rinse and Isotope Removal

A radiochemical examination of a section of the SRE main sodium hot-leg spool piece (L-101) was also performed this fiscal year. Samples from the bottom and top of each end of the pipe were selected for analysis. Each sample was etched in 2.5 N nitric acid (Figure 7).

In practice, it was found that virtually all activity from the hot-leg samples was removed in the first two baths. Aliquots of each bath were drawn and counted on a 400-channel pulse height analyzer counting system. The results are presented in Table 7.

5. Experiment in the KEWB

Following the melting of the sodium immersed UC fuel disc in the KEWB which was performed late in fiscal year 1964, sodium and off-gas sampling measurements were completed this year. Results of this experiment (IP-5) (as noted in the progress report from the first quarter of FY 1965) showed primarily that the gas sampling streams were blocked with molten sodium during the time of the KEWB burst. It was noted however, that the UC foil was successfully melted. Considerable difficulty was experienced in recovering UC fragments from the sodium using the butyl cellosolve dissolution technique and a supplementary experiment which was performed to evaluate the extent of butyl cellosolve - UC reaction showed no appreciable effect. It may be speculated however, that either the high temperature sodium environment, or the radiation environment modified the situation, although these are merely conjectures.

Although a second UC melting experiment was planned this fiscal year, it was decided to conduct the experiment as a capsule rupture experiment. It is planned therefore, in the proposed capsule rupture series.

B. ANALYTICAL AND REPORTING ACTIVITIES

During the subject fiscal year, the results of the project data and information were compiled for utilization in a document describing safety aspects and problem areas of sodium-cooled reactor systems. Results of the project were also communicated, and discussions were held with the representatives from the French safety evaluation team. At the request of the FARET personnel, discussions were also held concerning appropriate values for iodine retention for use in the hazards analysis.

A "Fission Product Retention and Plating Behavior in Liquid Sodium Systems," was presented at the International Symposium on Fission Product Transport and

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 7

SRE HOT-LEG SPOOL RADIOLOGICAL EXAMINATION RESULTS
(Microcuries per 100 g of Metal)

Location and Rinse	Cs137	Ru106	Mn54	Co60
"Close End"				
Bottom				
1st rinse	0.36	-	-	0.072
2nd rinse	0.0054	0.0072	-	-
Top				
1st rinse	0.81	0.41	0.52	0.25*
2nd rinse	0.022	0.022	0.015	0.030*
"Far End"				
Bottom				
1st rinse	0.030	0.21	0.46*	0.16*
2nd rinse	0.0076	-	-	-
Top				
1st rinse	0.40	0.15	0.45*	0.17*
2nd rinse	0.13	-	-	-
Sodium sample	140	-	-	13

Accuracies: * = $\pm 20\%$, sodium sample = order of magnitude,
others = $\pm 10\%$

Release. A second has been submitted for presentation at the Fast Reactors Conference at Argonne National Laboratory. A topical report entitled, "High Temperature Experiments on Fission Product Retention in Sodium," NAA-SR-9287 was issued.

Work has begun and a rough draft is in preparation of another topical report describing the overall results of iodine and fission product release in sodium systems and under typical containment conditions.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

A. GENERAL PROJECT EFFORT

In general, the progress of the project during fiscal year has not been as rapid as had been anticipated. Part of this difficulty stems from the unavailability of the irradiation reactor (STIR) during the first six months of the fiscal year, but most of the problem arises from the technological developments required for research in this area.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Acquisition of the multichannel analyzer and gamma spectrometer this year has provided a great deal more potential in terms of the data-handling capability. This potential has been utilized during the latter part of the fiscal year in counting the numerous samples obtained from each experiment utilizing Maypack and diffusion tube data.

The experimental arrangement which has been assembled and tested in the inert-gas concrete blockhouse will provide great flexibility in future experiments. This remote, medium radioactivity-level, medium sodium-volume facility is now available with the flexibility to accommodate a variety of basic or simulation experiments at little cost for adaptation.

Peripheral information has been provided with the acquisition of data from the UC capsule experiments and the SRE piping samples. These results, which were not supported on project funds, are more in the nature of providing an indication of the problem areas in plating and deposition rather than solving any specific problems.

The condenser-discharge injection method has been demonstrated. It may be used to provide rapid change of variables and corroborative experiments at less cost than for complete reactor installations. The physical and chemical aspects of fission products released in this manner must be considered with those melted in a nuclear environment, and essential differences in retention or diffusion characteristics noted for sodium versus nonsodium systems.

B. EXPERIMENTAL EVALUATION

The contemplated experimental program using the condenser discharge technique has just begun. Table 8 presents a comparison of the AI diffusion tube data with that acquired by ORNL*.

TABLE 8
COMPARISON OF AI AND ORNL DIFFUSION TUBE DATA^(a)

Study and Tube	Apparent Diffusion Coefficient, cm ² /sec	
	S'1(b)	S'2
ORNL Iodine characterization studies:		
First diffusion tube	0.113	0.002
Second diffusion tube	0.081	0.011
AI sodium plenum studies:		
First diffusion tube	0.108	0.011
Second diffusion tube	0.108	0.002

(a) Data obtained from silver-plated diffusion tube results.

(b) S'1 and S'2 represent the two identified components of the diffusion tube radioactivity deposition.

*ORNL-3776, Nuclear Safety Program, Semi-Annual Progress Report

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Aside from minor deviations of slope in the second components (which, as ORNL points out, may not be diffusion-limited), the first diffusion tube coefficients in the AI sodium plenum agree very closely with those reported by ORNL. The S'l component has been tentatively identified in the ORNL studies as I₂ but in the AI case can probably best be characterized as a particle of ~7A°. A rather well-defined difference between the AI sodium plenum studies and the AI nonsodium experiment is pointed out in Tables 3 and 4. It may be noted that there are apparent differences in component retention apparently resulting from the sodium bubbling. These have in some manner changed the form and enhanced the retention characteristics of the I¹³¹ on both the copper filters and the carbon-loaded paper filters, while all the other isotopes reveal little change resulting from the sodium plenum transport.

While these results are of interest, it is in the application of these data to reactor systems that the real project objectives lie. Thus, deposition of the I¹³¹ on the transfer tubing may be of more ultimate significance in accurately assessing the fate of iodine in a sodium system than information about precise chemical and physical forms of the isotopes released from the sodium.

To date, the temperature gradient deposition tube built late last fiscal year has not been used in these experiments, as it was felt that the diffusion tube-Maypack arrangement might produce more information. This device may be employed in future condenser discharge experiments to supplement and corroborate the information obtained with present measurement techniques.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Reactor Safety

Project: Measurement of Doppler Coefficients

Project Engineer: A. A. Jarrett

Reporting Period: Fiscal Year 1965

AEC Category: 04-60-01-09

General Order: 7549

Subaccount: 1351

I. PROJECT OBJECTIVES

The general objective of this project is to provide a firm experimental basis for a rapid and inexpensive method of calculating the Doppler coefficient of reactivity that will be attractive to reactor designers. Specific objectives are: (1) to establish gold (with a simple resonance structure at low energies) and U^{238} metal (with an extended resonance structure) as resonance integral and Doppler effect standards to which other materials can be compared both in experiment and in theory; (2) to find and evaluate the magnitudes of systematic errors present in measurement techniques; (3) to test the range of applicability of present theory by making measurements over an extended range of surface/volume ratios and temperatures in simple $1/E$ spectra and spectra differing only slightly from $1/E$; (4) to test both theory and experiment by auxiliary measurements designed to emphasize differential effects; (5) to measure the Doppler coefficients of uranium metal and oxides, and compare to previous work; (6) to extend resulting experimental and theoretical techniques to other fertile materials, such as thorium metal and oxide, uranium alloys, and uranium carbide; and (7) to extend the studies to structural and control materials and those materials of interest to reactor design where insufficient basic resonance data are now available.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. GENERAL THEORY

The theoretical portion of this program has produced the $TRIX-I^{(1)}$ code for the analytical calculation of effective resonance integrals and the Doppler effect. $TRIX$ is the result of a complete reevaluation of the methods of Chernick-Vernon⁽²⁾ and Goldstein-Cohen,⁽³⁾ as exemplified by the $ARES^{(4)}$ codes. Many changes and refinements have been incorporated.

Among the most important of these is the improvement in the method of calculating escape probability approximations for lumped resonance absorbers.^(5,6) The new method now provides a rigorous method of calculating the mean optical chord length independently for each resonance. Refinements in treatment of the tails of the resonances have resulted in an improved calculational method which reproduces accurately both the 2200 m/sec cross-section and infinitely dilute resonance integrals for most isotopes.

An improved approximation to the scattering interference calculation was also incorporated.⁽⁷⁾ Comparisons with previous measurements in U^{238} indicate that this modification makes a considerable improvement, but that more work should be done in this area. (See also Section C, below).

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Even though TRIx is known to have minor shortcomings, the code represents an important advance for practical reactor calculations. The accuracy of TRIx, as assessed by comparison with available experimental results, is sufficiently greater than the older analytic methods for TRIx to have been adopted as the present standard at Atomics International for such calculations. Now in production status as an independent code, it is being incorporated as subroutines in CEASAR⁽⁸⁾ and other nuclear codes. Where comparison with the detailed numerical methods of ZUT⁽⁹⁾ has been possible, TRIx has been shown to produce equivalent results in nearly all cases, while requiring 10 to 50 times less computer time per case.

A determination of new parameters for the unresolved resonances of U²³⁸ and thorium has been made. The determination was made from new Columbia data.⁽¹⁰⁾ The main difference for U²³⁸ from previous data is in a larger level spacing. The new and previous values are compared in Table 1.

TABLE 1
COMPARISON OF PARAMETERS FOR UNRESOLVED
RESONANCES OF U²³⁸

Parameter	Γ_{N_o} (ev)	Γ_{γ} (ev)	Average Spacing (ev)
New value (Otter)	0.001875	0.0250	22.6
Previous value (ARES-II)	0.00183	0.024109	18.34

A careful statistical treatment for thorium was designed to determine the effect of higher-order resonances (higher than $\ell = 0$) and possibly missed resonances. The results are not appreciably different from the simple averaging of the Columbia data. Both results, however, are at variance with thorium parameters used previously.

B. EXPERIMENTAL

Previous Doppler measurements having appreciable precision have been limited to less than one decade variation in surface/mass ratio (S/M).^{*} Within this particular range, the so-called "practical" range, the two or three lowest energy U²³⁸ resonances account for more than 90% of the effective resonance integral, but for only about 10 to 20% of the Doppler effect. Accurate assessment of the validity of the theory are thus difficult within this limited range.

In the experimental portion of this program, the range of S/M has been extended to encompass three to five decades in Au¹⁹⁷⁽¹¹⁾ and U²³⁸, two materials

^{*}The equivalent parameter $s = 1/4N_{res} (S/V)$, the surface/volume ratio per atom of resonance absorber, is used instead of S/M in calculation and presentation of results.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

with relatively well known resonance parameters. Since the effect of the calculational treatment of any given parameter or approximation is a function of S/M , the wide range in the measurements can now provide a meaningful base for further improvements in theory.

Extension of the range of S/M towards zero also has the effect of putting greater emphasis on the high-energy portion of the resonance region, the range of interest in fast reactor Doppler calculations. It is the extreme sensitivity of the activation method, as developed in this program, which makes possible the large increase in range of S/M .

1. Evaluation of Experimental Errors

In order to provide the meaningful comparison with theory, both systematic and random experimental effects must be well understood. As a minimum, the direction and limiting magnitudes of all such errors must be known. In addition to its advantages in the theoretical aspects of this program, gold provides a nearly ideal experimental material. The large cross-section, convenient half-life, lack of interfering activities, and easy machinability lend themselves well to studies of experimental error sources.

A basis was provided from within the data itself, for evaluation of error sources: the ratio of the activity of two power-monitor foils always irradiated under identical conditions. The ratios of the activities of each of these foils are the measured quantities. The ratio of the measured quantities in any pair of counters must be 1.0 unless there is some abnormality in the counters. The measured standard deviation in these ratios can be divided into two parts. The first part is counting statistics, which were evaluated separately by the data reduction code. The second part can consist only of the combination of the mean short-term drift (for periods up to about 1 hr) of the counters and of the geometrical nonreproducibility of any given sample holder between counters. The difference between the measured standard deviation and the counting statistics (computed in quadratic form) is the magnitude of the standard deviation of the combination of short-term drift and sample holder reproducibility. It amounts to 0.1%. The same data were used in an autocorrelation technique to determine if there was any significant trend in counter reproducibility as a function of time. There is at least an 80% chance that such did not occur; there was almost certainly no significant drift in the ability of any of the three counters to measure properly the ratio of the activities of two foils.

After the above ratio was computed, the calibration factors as determined previously by a combination of theory and experiment were applied to the data. The calculation was then repeated with the normalized data. The final ratios had a mean of 1.0. A broadening of the standard deviation can be attributed solely to foil calibration factors. The quadratic difference gives the standard deviation of the monitor foil calibration factors. This amounts to 0.065%. The calibration factors for the experimental foils could not be checked in the same manner, since there was never a large enough sample population. These errors, however, must be smaller because most of the sets of sample foils were much more uniform, and nonuniformity introduced most of the uncertainties.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Every effort was made to place the power monitor foils in exactly the same location in a flat flux. The monitor foils were then placed in precision holders for counting. It was not possible to use the same holders in every case. The monitor foils were counted randomly among a set of about 50 holders. The combination of the error from the geometric differences between the holders and the position of the monitor foils during irradiation were determined by finding the mean and standard deviation of the corrected and normalized power monitor ratios for each counter separately. The quadratic difference between the new standard deviation and the previous one is the magnitude of the error introduced by the combination of sample exposure holder and counting holder nonreproducibility. It amounted to 0.13%.

Uncertainties in the measurement and application of counter paired-pulse resolution are a potentially serious source of error in these experiments. Through application of careful statistical techniques and the development of new methods, (12) this potential source of error has been reduced by a considerable amount and can be evaluated for each measurement. The final uncertainty from this source is expected to have a mean somewhat less than 0.2%.

In the present experimental situation, the thickness of the samples is one of the experimental variables. The variations in sample thickness can also introduce unwanted changes in counter efficiency by changing geometry and self-attenuation of the gamma rays. In auxiliary experiments, the effective mass absorption coefficient for our counters, the variation in efficiency with foil vertical position, and distribution of activity as a function of depth within the foil were all measured separately. In effect, the different foils were taken apart and measured layer by layer. With this information, independent and combined correction factors were calculated for each foil thickness, with a total uncertainty in these two parameters of about 0.1% (in a maximum correction of nearly 11%).

2. Neutron Spectrum

Only 10% of the infinitely dilute resonance integral of Au^{197} comes from resonances higher than the 4.9 ev giant resonance. The proportion becomes larger as the lump size increases (see Figure 1). In general, the Doppler effect is determined by the higher resonances in a proportion that changes rather drastically as the lump size grows (see Figure 2). It is thus very important that the actual neutron spectrum in which the experiment was performed be determined either by theory or by experiment.

For these experiments, the spectrum at the test location was computed in the 16-group FAIM⁽¹³⁾ diffusion theory approximation along with correction factors accounting for the difference between this and the reference (1/E) spectrum. It was not feasible to check the spectrum calculations directly by experiment, primarily because the resonance integrals of absorbers useful in the required energy range were not known with sufficient precision. Instead, the test region was mapped in three dimensions, using packets of five foil materials having primary absorptions distributed between cadmium cutoff and 132 ev. (The measurement of spatial dependence at fixed energies is equivalent in this case to measurement of spectrum at a fixed location.)

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Figure 1. Contribution to Resonance Integral by Energy Group for Au^{197} at 25°C (Values for groups 1 to 10 have been multiplied by 10)

7549-2501

Figure 2. Percent Contribution to β by Energy Group

7549-2502

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

The measured spatial distribution of absorptions agreed within measurement uncertainty with the FAIM calculations for the groups which include the higher energies 132 ev for Co⁵⁹ and 18 ev for W¹⁸⁶. The spatial distribution for the gold mapping foils (4.9 ev) and dysprosium (first epicadmium groups) did not agree with the shape of the calculated spatial distribution. The FAIM calculation on which spectrum was based clearly did not include as many neutrons in the lower energy groups as was indicated from the flux maps. The FAIM correction factors for departure from the reference spectrum are given in Table 2. It will be noted that these are functions of surface/mass ratio of the sample, and amount to a total difference of 0.7% for the range of interest.

TABLE 2
NON-1/E CORRECTION FACTORS FOR
10.6-in. LATTICE FOR Au¹⁹⁷ (550°C)

Surface/Mass (barns)	Effective Resonance Integral (barns)	Factor
150084	1010.2	1.0022
52551	906.56	1.0021
10818	309.10	1.0018
1802.5	623.82	1.0009
349.28	158.73	0.9991
127.35	108.50	0.9975
34.608	74.225	0.9952

To provide an estimate of the effect of the magnitude of errors resulting from reasonably inferrable uncertainties in neutron spectrum, calculations of new spectral weights were made, based on assumed spectra that differ from the FAIM spectrum in known ways. In the first case, the departures from the ideal 1/E spectrum were doubled. In the second case, the spectrum was assumed to increase linearly from Group 9 (which included the tungsten data already found to be in agreement with experiment) to cadmium cutoff, as inferred from the flux-mapping data. The effect is a function of surface/mass ratio, and is largest for the thickest foils. The results are shown in Table 3 for the thinnest and thickest foils. Even in the worst case, the total effect is quite small. The temperature-dependence of the spectral corrections, e.g., that part of the effect of most importance to the Doppler experiments, was negligible in all cases studied.

As further test of the accuracy of the spectral predictions, a second lattice was studied. This lattice was chosen to have a spectrum considerably softer than the standard lattice. The combination of AIM-6⁽¹⁴⁾ and TRIx successfully predicted the activation ratios actually observed.

ATOMIC INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 4
EFFECTIVE RESONANCE INTEGRAL OF Au¹⁹⁷ at 25°C
(barns)

Surface/Mass	Effective Resonance Integral From TRIX-I	Resonance Integral Experiment*
28.97	91.5	63.1 ± 0.6
124.77	146.8	125.8 ± 0.7
344.64	217.7	197.6 ± 0.6
1787.0	427.4	407.8 ± 0.9
11028.0	863.4	865.2 ± 3.2
65091.0	1315.6	1317.0 ± 15.2
147631.0	1431.2	1425.8 ± 2.9

*Arbitrarily Normalized by Calculation to Yield $I_{\infty} = 1583.4$ barns
(including epicadmium $1/v$).

Figure 3. Effective Resonance Integral for
Au¹⁹⁷ at 25°C

7549-2503

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 3
SPECTRAL EFFECT ERRORS

Spectrum	Resonance Integral (barns)	
	s = 124.7	s = 137600
In 1/E Spectrum	146.797	1423.547
In Actual Spectrum	146.228	
In Assumed Spectrum (twice above deviation)	145.751	1423.547
In Linearly Increasing Spectrum	146.140	143.547

The resonance integral of W^{186} , one of the isotopes used in the flux survey, is comparatively poorly known. Until recently, the best experimental value for the infinitely dilute resonance integral was 425 ± 100 barns. Calculations yield a value of 463 barns, but the uncertainties in neutron parameters make the calculated resonance integral uncertain to about 100 barns. Even though the mapping data compared gold to tungsten under poor conditions for resonance integral comparisons, it was possible to obtain an estimate for the resonance integral. (The calculation was made as a check on the calidity of the flux-mapping technique.) The experimental value is $480 +100 -200$ barns. Systematic uncertainties in relative counter efficiencies are the largest source of error and are responsible for the nonsymmetrical error statement. (It is possible in future measurements to cut the uncertainty to less than a third of this value.)

3. Results in Au^{197}

After all corrections were made, the final gold values were obtained by weighted means of three or more repeated measurements. In resonance integral measurements, the experiment yields a set of numbers that are proportional to the effective resonance integral at each surface/mass ratio. (The proportionality constant contains the product of the absolute flux and the absolute counting efficiency, neither of which can be determined with anything like the required precision.) Three considerations were used in determining the normalization constant: (a) The three thinnest foil types (largest values of S/M) have their effective resonance integrals determined principally by the giant resonance; (b) for these three values of S/M alone, the ratio of calculated to experimental value was a constant, indicating good agreement; and (c) the normalization should extrapolate to the excepted infinitely dilute value (1583.3 barns, including the epicadmium $1/v$ capture). The normalization was determined under the above considerations by an analytical weighted least squares fit and by an arbitrary ratio method. Both agreed.

The results are shown in Table 4 and Figure 3. The error statements in the table require some qualification. The quoted errors were arrived at by two independent means. The error analysis described above developed a

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

weighted variance, the expected or external error term. In terms of this expected error, and considering all sources of error except that due to neutron spectrum uncertainties and possible error in counter resolving time (both of which affect the experiment systematically, not randomly), the expected error has a confidence level slightly more than 80%. This can be interpreted as saying that either the expected error was too conservative or that it yielded errors that approximate 1-1/2 (standard deviation). The internal agreement of the data provides a second error estimator. In general, this is smaller than the external error estimator. The errors quoted in Table 4 are the larger of these two terms, and yield a confidence level slightly more than 90% (or two standard deviations) for inclusion of random errors alone. When the systematic error possibilities of known magnitude are considered, this confidence interval drops to about 70%, that is, one standard deviation.

The systematic errors from known sources would have the following effects. Errors in neutron spectrum are most likely to be in the direction which will raise the values of the effective resonance integral at low values of S/M, perhaps as much as a half to three-quarters of a barn. If there were errors in measurement of counter resolving time, they would have the opposite effect. The maximum effect would be about the same. Materials effects measurements reported previously indicate that if the materials of the capsule have an unexpected effect on the result, the effect would be in the direction of reducing the integral, again preferentially for low values of S/M. The magnitude of this effect would be negligibly small if present at all.

The corresponding results for measurement of the Doppler effect are shown in Table 5 and Figure 4. The same comments apply to the quoted errors. In general, the error sources and possible systematic errors are smaller for the Doppler measurements than for the measurements of the effective resonance integral, primarily because the Doppler effect is a derivative such that 1st-order errors in the integral either vanish completely or at least become 2nd-order errors in the Doppler effect.

TABLE 5
THE DOPPLER EFFECT IN Au¹⁹⁷ -
EXPERIMENTAL RESULTS

Surface/Mass (barns)	Doppler Coefficient ($10^4 \frac{\beta}{C} - 1/2$)
125	22.28 $\pm$ 2.01
345	4.73 $\pm$ 0.62
11028	17.84 $\pm$ 1.70
147600	15.84 $\pm$ 1.40

7549-2504

Figure 4. Temperature Derivative of Resonance Integral of Au¹⁹⁷
(averaged between 25 and 250°C)

C. COMPARISON OF THEORY AND EXPERIMENT

In general, the present measurements agree with calculations for large values of S/M (thin samples), and disagree for small S/M. The calculations overestimate the effective resonance integral by as much as twenty experimental standard deviations in some cases. The calculations apparently underestimate the Doppler effect, though not by as many experimental standard deviations.

The resonance parameters of gold were investigated as a possible source of the discrepancy. The spin assignments corresponding to some of the resonances are uncertain, and some are unknown. The choice of spin affects the resulting parameters greatly. In addition, the parameters themselves have their own attached uncertainty. A set of survey calculations investigated the overall effect of these uncertainties. In general, the choices of spin and experimental uncertainty in parameters were lumped together in such a way as to produce maximum and minimum limits. The overall effects are small. They are shown in Table 6. The largest effect, as expected, was obtained by eliminating entirely the scattering interference calculations.

At present, the working hypothesis is that the calculation does not properly truncate the integral at cadmium cutoff, thus yielding too large an integral. This is being investigated. If this is true, then it would affect only the comparison of the theory to cadmium-covered experiments and not materially affect actual reactor calculations.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

TABLE 6
RESONANCE INTEGRAL AT 25°C
(barns)

Surface/ Mass	Standard Case	Maximum Γ_y Group 9	Unknown J = 1 Group 9	Unknown J = 2 Group 9	No Scattering Interference	J(60.3) = 1
10.0	65.251	65.453	65.23	65.265	64.672	65.204
100.0	135.02	135.55	134.92	135.09	134.37	134.92
1000.0	335.97	336.87	335.6	336.24	335.42	-

Doppler Coefficient, β , at 287.5°C

	Standard	Maximum Γ_y	J = 1	J = 2	No Scattering Interference	J(60.3) = 1
10.0	0.001095	0.0010813	0.0010913	0.001090	0.0012243	0.0010895
100.0	0.0014472	0.0014286	0.0014377	0.0014540	0.0015144	0.0014359
1000.0	0.0015550	0.0015449	0.0015436	0.0015648	0.0015749	-

In addition to the above, the unresolved resonance parameters were cross-checked with the latest data by reconstructing the measured unresolved data of BNL-325 from our parameters. The agreement appears good.

The results of the comparisons so far indicate that additional work must be done on scattering interference calculations.

D. U²³⁸ MEASUREMENTS

Reduction of the primary data was continued. Preparations were made for experimental normalization of the sample planchettes. The coincidence spectrometer system was modified to improve temperature stability by incorporation of highly stable reference supplies and first-order-compensation of reference junctions in the discriminator systems.

Completion of the data reduction and data analysis for U²³⁸ is expected early in the next fiscal year.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

Data reduction and data analysis for the first phase of the experimental work, establishment of standards, has been completed for gold,⁽¹¹⁾ and is nearly completed for uranium-238. It appears that the goals of accuracy and completeness with respect to evaluation of the systematic errors in this experiment have been achieved. The wide range in surface/mass ratio introduced into the experimental method makes it possible to investigate the systematic effects (most of which

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

are themselves functions of surface/mass), and allow investigations of the accuracy of computational methods at this time. In so doing, improved techniques for both resonance integral and Doppler measurements have been developed.

The theoretical portion of the program has developed a calculational method exemplified by the TRIX code which has produced results equivalent, in most cases, to the much more expensive numerical methods. The fact that it requires nearly two orders of magnitude less computer time than previous methods adds to its usefulness, and eliminates the need to make simplifying assumptions or compilations and extrapolations of empirical data in design and safety calculations. It is this computational method that these experiments were designed to check.

The cross-check is in progress at this time. It has confirmed the previously suspected, but moderate, inadequacies in the theoretical treatment of scattering interference, and has uncovered a possible error in the theoretical treatment of cadmium-covered activities.

The primary goals of the project are being attained, although at a slower rate than had been hoped. The slower rate is attributed to the large amount of cross-checking and side investigation that has been needed as the project unfolded. The majority of such details have been completed, and the project is now making good progress.

REFERENCES

1. J. Otter, "TRIX-1, An Improved Resonance Integral Code," NAA-SR-MEMO. (To be published in 1965)
2. J. Chernick and R. Vernon, "Some Refinements in the Calculation of Resonance Integrals," Nucl. Sci. and Eng. 4, 649 (1959)
3. R. Goldstein and E. R. Cohen, "Theory of Resonance Absorption of Neutrons," Nuc. Sci. and Eng., 13, 132 (1962)
4. F. L. Fillmore and B. D. O'Reilly, "ARES-II," AMTD-130B, Atomics International (1963)
5. J. Otter, "Escape Probability Approximations in Lumped Resonance Absorbers," NAA-SR-MEMO-9744 (1964)
6. J. M. Otter, "Modified Rational Approximation to the Collision Probability in Lumped Resonance Absorbers," Trans. Am. Nuc. Soc. 7, 275 (1964)
7. W. Rothenstein, "Collision Probabilities and Resonance Integrals for Lattices," Nuc. Sci. and Eng., 7, 162 (1960)
8. D. C. Baller, "CAESAR," AMTD-200 (1963)
9. L. W. Nordheim, Nuc. Sci. and Eng. 12, 457 (1962)
10. J. B. Garg, et al., "Neutron Resonance Spectrometry III. Th^{232} and U^{238} ," Phys. Rev., 134, B985 (1964)
11. L. Beller and Harry Farrar IV, "A Wide Range Study of the Effective Resonance Integral and Doppler Effect in Au-197," Transactions ANS 8, 285 (1965)
12. L. S. Beller and Harry Farrar IV, "Experimental Studies in Paired-Pulse Resolution in Counting Systems," NAA-SR-MEMO-11132, (In press) (1965)
13. D. C. Baller, "The FAIM Code, A Multigroup. One-Dimensional Diffusion Code," AMTD-118 (1962)
14. H. P. Flatt and D. C. Baller, "The AIM-6 Code," NAA-SR-MEMO-9204 (1961)

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Reactor Safety	
Project: Atmospheric Decontamination in Reactor Accidents	
Project Engineer: A. A. Jarrett	
Reporting Period: Fiscal Year 1965	AEC Category: 04-60-40-04
General Order: 7640	Subaccount: 1371

I. PROJECT OBJECTIVES

The primary objective of this project is to study the production, distribution, and availability for dispersion to the environment, of sodium oxide particles and any concomitant fission products produced in large sodium fires. To most effectively carry out this primary objective, intermediate objectives are: to study particle production mechanisms, size distributions, and other removal phenomena, in order to determine the particulate suspension remaining for use as a source term for environmental dispersion and loading requirements for an air-cleaning system; and to determine possible artificial means of enhancing removal by increasing particle growth, fallout, and deposition.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. SUMMARY

1) Measurements were made in small diameter (4-in.) burnpots of the burning rate of liquid sodium as a function of oxygen concentration surface temperature, surface area, and atmospheric flow rate over the sodium surface. The burning rate was linear with oxygen concentration, flow rate, and burning area.

2) Measurements of the release and size distribution of the sodium vapor and its oxide were made as a function of the same parameters. The release was found to be proportional to flow and temperature of the surface.

3) It was demonstrated that sodium iodide tagged with I^{131} was released in concentrations less than the concentration in the sodium pool, and that the iodine was distributed in proportion to the surface or volume of the oxide particles.

4) The mass median diameter of particles measured near the surface of a burning sodium pool were found to vary from 1 to 2 microns, with geometric standard deviations of between 1.5 to 3.5 microns.

5) The development, calibration, and modification were conducted for impactors, impingers, settling chambers, and an electrical particle counter for measurement of particle size distributions.

6) The following papers and report were presented and published.

a) "Natural Mitigation of Radiological Hazards in Sodium Fires," at the Winter Meeting of the American Nuclear Society

b) "Sodium Fires and the Release Characteristics of Particulates and Fission Products," at the International Symposium on Fission Product Release and Transport under Accident Conditions, Oak Ridge, Tenn. (jointly with Project 7661-1342)

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

c) "Study of Sodium Fires and Particulate Release as Applied to the FRCTF," NAA-SR-MEMO-11403 (jointly with Project 7661-1342)

B. STUDIES IN LARGE FIRES WITH FORCED FLOW

Since efforts of this project and those of Project 7661-1342 (Fast Reactors Program) are conducted on a coordinated basis, most of the pertinent burning rate data obtained from large fires can be found in the reports of the latter project. The burning apparatus used to conduct the reduced oxygen experiments presented in this section is shown in Figure 1, and is referred to as the Large Fire 2 (LF 2) apparatus. It consists of a sodium preheat and transfer system, burn pot, burn chamber, oxide collection system (scrubber), and an exhaust system. Included are facilities for supplying and regulating the oxygen and nitrogen flow and concentration; measuring, recording, and controlling temperatures; measuring the oxygen outlet concentration; monitoring the humidity; sampling airborne concentration and particle distribution; and sampling scrubber water for sodium and tracer iodine activity. Provisions are made for sampling the activity level of the sodium in the preheat tank as well as the specific activity of the sodium while oxidation is occurring.

Figure 1. LF-2 Test Apparatus

7640-2501

After addition of sodium and iodine into the burning chamber, the bulk sodium temperature and activity levels were monitored at zero oxygen and zero flow. Upon reaching equilibrium conditions, the flow rate and oxygen concentration at

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

a given bulk sodium temperature were varied, and the following observations were made and reported in NAA-SR-MEMO-11403.

- 1) Bulk Na temperature profile in burn put
- 2) Gas temperature profile above burn pot and in adjoining carryover piping
- 3) Oxygen concentration entering and leaving burning region, and amount consumed
- 4) Rate at which Na is collected in scrubber (release rate R in lb Na/hr), and arrival rate $S = R/60Q$ lb/ft³ where $Q = \text{ft}^3/\text{min}$ flowing over sodium
- 5) Rate at which I^{131} activity is collected in scrubber (release rate ϕ in $\mu\text{c/hr}$), and arrival rate at $\theta = \phi/60Q$ $\mu\text{c/ft}^3$
- 6) Specific activity Σ of I^{131} in burn pot (in units of $\mu\text{c/lb Na}$)
- 7) Concentration of I^{131} activity and sodium in gas being transported to scrubber (in units of θ' ($\mu\text{c/ft}^3$) and S' (lb Na/ft³))
- 8) Particle size distributions, relative mass as function of aerodynamic diameter (mass median diameter and standard deviation)

To minimize duplication*, only the particle size data are discussed in this report. The table summarizes information obtained from Experiment LF-2 concerning particle size data and concentration data obtained with an 8-stage round jet impactor†. These devices measure the mass distribution versus the effective cutoff aerodynamic diameter, $D\sqrt{\rho C}$ in units of micron (gm/cm^3)^{1/2}. For a given jet and flow rate, particles having equal values of $D\sqrt{\rho C}$ will be collected with the same efficiency. Such particles will also have equal settling velocities, if laminar flow prevails. Column 3 of the table gives the size of the particle (in terms of the "aerodynamic" diameter, $D\sqrt{\rho C}$, which represents the 50 percentile of the mass collected; and column 4 is the computed geometric standard deviation for the distribution. D is the diameter in microns, ρ = density, and C is the Cunningham correction factor for slip. Column 5 states the sample position in the burning apparatus (see Figure 1). Column 6 is a computation of the mass median diameter, MMD-50 (50th percentile of mass). This is computed from Column 3 by dividing by $\sqrt{\rho}$, with $\rho = 2.27$, the density of Na_2O .

The count median diameter, CMD-50, is the geometric mean by number and is shown in column 7. The relation used for this computation is $\text{CMD} = (\text{MMD}) \exp(-3 \ln^2 \sigma)$.

The concentration of sodium collected at the sample position may be computed from the data obtained by the impactor, and is termed S_L (lb/ft³). The ratio of S_L to the concentration S computed from scrubber data is given in the 7661-1342 Progress Report.

The last row in the table was computed by taking an arithmetical average of the weight of sodium on each stage of each impactor used to obtain the data of column 3. Only samples taken at Position 1 were used. The other parameters in columns 6 and 7 are computed as described above. These averages and the geometric standard deviation σ are used to plot the curves in Figure 2, where the ordinate is the cumulative distribution of the mass of particles whose sizes

*The design of the impactor is by T. T. Mercer of the Lovelace Foundation.

(("A Cascade Impactor Operating at Low Volumetric Flow Rates," LF-5 (Dec. 1962)
†(See Annual Report of Project 7661-1342)

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

RESULTS OF AIR SAMPLING
(Particle sizes)

Test No.	Column No.							
	1	2	3	4	5	6	7	8
	α_I	Q	D ρC Aerodynamic Diameter	σ Geometrical S.D.	Sample Position	MMD-So (μ)	CMD-So (μ)	S_L/S
LF2-A	0	12	1.6	1.9	1	1.0	0.29	0.17
			2.6	2.5	1	1.7	0.14	0.27
			2.0	2.2	4	1.4	0.24	-
LF2-B	1.17	12.3	2.2	1.3	1	1.5	1.2	0.48
			2.8	2.7	3	1.9	0.99	0.43
			2.8	2.2	4	1.87	0.27	0.24
LF2-C	1.98	19.7	2.2	1.9	1	1.5	0.48	0.64
			2.0	1.6	4	1.4	0.65	0.36
			2.2	1.8	1	1.5	0.48	0.7
LF2-D	4.07	17.9	2.0	2.0	1	1.4	0.36	0.12
			3.0	1.8	4	2.0	0.70	0.53
LF2-E	0	200	2.0	1.8	4	1.3	0.41	0.7
			1.8	2.2	1	1.2	0.17	0.5
			Average		2.1	1.95	1	1.4

are less than a stated size as a function of the size parameters used. Since these graphs are on logarithmic probability paper, the cumulative mass distribution plots as a straight line. The geometric mean is the 50% size and bisects the symmetrical frequency curve.

The other curves in Figure 2 were drawn by using relationships given by Hatch* for comparing distribution of mass and number with distribution by volume and area. Rows 1 and 2 of the table give pertinent parameters of inlet oxygen α , and flow over the burning surface Q ft³/min.

Figure 3 gives the activity distribution of I¹³¹ in terms of the effective "aerodynamic" diameter (see Curve A), and also in terms of the diameter in microns (see Curve B). The ordinate is the measured percentage of x, the total activity associated with a given size plotted as abscissa.

These data were obtained by counting all samples used to compute the graphs shown in Figure 2-A. Stages 1 through 8 for all samples were grouped

*Industrial Dusts, Drinker & Hatch, McGraw-Hill, N. Y. (1936)

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Figure 2. Size Distributions

Figure 3. Distribution of I^{131} Activity on Sodium Oxide vs Particle Size

numerically (i. e., all Stage 1 samples were counted simultaneously), and counted as one sample. In this way, the I^{131} activity level was high enough to be measured.

The activity and mass distribution data from the LF-2 experiment represent the most consistent particle size data obtained to date for the aerosols released from vaporizing and/or oxidizing liquid sodium. One of the reasons for this is probably due to the somewhat more stringent control on the liquid surface temperature, the sampler used, and the sample analysis techniques. Even so, the mass distributions as measured in LF-2 do not differ significantly from those measured in the laboratory burn chamber, in tranquil settling chambers, and over the surface of small fires in open hoods. The ability to make repeated measurements of an equilibrium concentration and distribution allowed much greater confidence to be placed in the round jet cascade impactor and the data generated therefrom. By counting the activity on the individual stages of the impactor samples (even though low level counting techniques were used), it has been possible for the first time to characterize the radio iodine particulate sizes relative to those of sodium aerosols.

The curves of Figure 2 show the importance of more than one sampling technique. For example, the MMD-50 is 1.4 microns, a size for which 98% of the particles are smaller by number, while ~50% of the particles by number represent less than 2% of the mass. Thus, it is not too surprising that particle distribution obtained from impactor and counting methods (electron microscopy) differ. From Figures 2 and 3, it can also be seen that the I^{131} activity is equally

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

poorly distributed with the mass; i. e., the activity median diameter of ~0.7 microns is representative of only 13% of the mass and 96% of the activity is on particles representing 50% of the mass.

The LF-2 experimental data showed little variation of particle size distribution with oxygen concentration and, in fact, indicated a sodium release rate from the liquid sodium surface to the bubbler-scrubber which was independent of the oxygen concentration (burning rate was proportional to O_2 concentration). This implied that more of the oxidation occurred on the surface of the sodium than had previously been estimated, and that the release was proportional to the temperature (vapor pressure).

A similar relationship was observed for the release of I^{131} activity. In fact, the I^{131} release rate characteristics were observed to be essentially the same as those for the sodium aerosol. The relative specific activity of the released I^{131} expressed as $\mu c/lb$ was $\sim 1/4$ of the specific activity introduced into the burn pot, thus indicating a reduction by a factor of 4 between the activity level of the burn pot and the aerosol activity. As mentioned previously, 96% of the activity was on particle sizes representing 50% of the mass and 98% of the total number of particles.

C. SMALL FIRE MODELING

A number of "small" fires on a "laboratory" scale were conducted using approximately 3 lb of sodium in a 4-in.-diameter burn pot. The major purpose of the small fire experiment was to determine whether simple scaling laws can be developed which would relate the small fires to the large ones, so that a large number of parameters can be expeditiously studied. The two most important measured quantities to be compared were the burning rate of the sodium and the release rate, both expressed in units of $lb Na/hr-ft^2$ as β or R , respectively. A secondary purpose of the small experiment was to determine whether or not the burning rates and release rates were functions of the volume flow rate of the atmosphere supplied to the fire, since the large fires were terminated by experimental difficulties prior to obtaining sufficient data to define flow rate dependence.

A choice was made of liquid sodium temperature of $1000^\circ F$ and an oxygen percentage of 4% in the atmosphere, in order to enable a comparison to be made with LF-2 runs with these same parameter values. These values of sodium temperature and oxygen percentage were maintained throughout the entire experiment. A relatively small deviation occurred in one run which used 3.8% oxygen.

1. Description of Apparatus and Methods of Measurements

The burning chamber is a flanged, stainless steel pot 8 in. high by 4 in. in diameter (see Figure 4). A $3/4$ -in.-thick insulating ring separates the burning chamber from a hollow stainless steel cone bolted to the burn chamber. Attached to the cone is a curved pipe which carries the Na vapor and oxide to the bubbler. A separation was made in the pipe exhaust allowing outside air to come in contact with the sodium, thus oxidizing the sodium particles prior to their entering

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

the bubbler when zero oxygen release rate data was obtained. The bubbler had its own vacuum supply to aid in capturing the released material. The 2-stage bubbler system is composed of a 3600-cc chamber followed by a 75-cc impinger. Samples of the solution were taken each 15 min in order to measure release rates to the scrubber. The burning chamber was inserted in an insulated oven capable of achieving temperatures as high as 1800°F. The oven was placed in a large drip pan and the entire apparatus (excluding the bubblers) was placed in a fume hood.

6-2-65 7640-2504

Figure 4. Burning Chamber

The temperature was measured at four different locations in the burning region. Three of the thermocouples were connected to a multi-point recorder. The fourth thermocouple was connected to a temperature controller. Nitrogen and oxygen bottles were connected to flow meters and a mixing chamber, which in turn were connected to four inlet positions on the cone. An oxygen analyzer was used to determine the oxygen content of the inlet and outlet gas mixture, in a similar fashion to LF-2. The analyzer was also valved to the nitrogen supply and atmosphere so that the analyzer could be calibrated prior to reading. All computations in the small modeling experiments were identical with those of LF-2.

2. Experimental Results

Figure 5 contains 2 graphs of the burning rate, β/A , in lb sodium/hr-ft², plotted atmosphere flow rate in liter/min. The initial area of the fire A was 0.087 ft². Run 1 indicates a linear dependence between these quantities for the range of Q values used; and the relation can be expressed analytically as follows.

7640-2505

Figure 5. Burn Rate vs Flow

$$\frac{\beta}{A} = 0.098$$

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

If F is expressed in ft^3/min (to enable comparisons to be made with the large fires), then,

$$\frac{\beta}{A} = 0.098 \times 28.3 F = 2.77 F$$

$$\frac{\beta}{AF} = 2.77 \frac{\text{lb}}{\text{hr-ft}^2} \cdot \frac{1}{(\text{ft}^3/\text{min})} \quad \dots(1)$$

This result is now compared with a suitable large fire, LF2-D of Project 7661-1342. In that fire, 4.1% O_2 was used with a sodium surface temperature of 1000°F . One obtains

$$\frac{\beta}{A} = 1.6 \quad \dots(2)$$

where

$$A = 2.9 \text{ ft}^2$$

$$F = 18 \text{ ft}^3/\text{min} ;$$

thus,

$$\frac{\beta}{AF} = \frac{1.6}{1.8} = 0.089 \frac{\text{lb}}{\text{hr-ft}^2} \frac{1}{(\text{ft}^3/\text{min})}$$

The values given by Equations 1 and 2 should be compared on the basis of a "scale" factor. It has been suggested that such a factor be the ratio of the areas of the large and small fires; i. e., the ratio of the large to the small area should be compared with the ratio described in Equation 1 (small fire) to Equation 2 (large fire). This is tantamount to requiring an equivalent oxygen consumption region over the burning areas of the fire. Thus the comparison is made:

$$\frac{(\beta/AF)_{\text{small}}}{(\beta/AF)_{\text{large}}} = \frac{2.77}{.089} = 31 \text{ and } \frac{A_{\text{large}}}{A_{\text{small}}} = \frac{2.9 \text{ ft}^2}{0.087 \text{ ft}^2} = 33 \quad \dots(3)$$

Another way of looking at the mass release scaling is on the basis of an equivalent volume flow rate per unit area of the fire, i. e., a relation as follows: $\beta/A = K F/A$. By using the data from both small and large fires, we find:

$$K = 0.24 \text{ for small fires, Run 1}$$

$$K = 0.26 \text{ for large files, LF2-D.}$$

These data for 4% oxygen should be compared at other oxygen concentrations in order to further verify the validity of the scaling laws. Flow dependence should also be checked with large fires.

Run 2 in Figure 5 also shows a linear relation between burning rate and flow rate, but with an increase in the slope. In these experiments, complete consumption of the inlet oxygen occurred. Moreover, examination of the disassembled apparatus showed that the effective surface of the sodium had increased

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

markedly, due to presence of a slag of oxide and sodium mixture along the sides, this possibly providing a siphoning action drawing liquid sodium into the many cracks and crevices and thus presenting an increased burning area. This may be a problem in small-fire modeling which will require modifications in experimental technique. It may also be due to the fact that all of the zero oxygen release rate data versus flow was obtained before Run 2 began, thus encouraging vaporization and condensation.

Figure 6. Sodium Release Rate vs Flow

Figure 6 shows graphs of the measured sodium release rate for two cases, with and without oxygen for Run 2. A comparison may be made of the ratio of the release to burning rate, R/β , for the small and large fires. In the former, data were taken from Run 2, with 4% oxygen. The apparent nonlinearity of R/A may be real or just due to scatter in the data. For this reason, a range of R is taken from 0.18 to 0.22 lb/hr-ft² at a flow of 5 l/min. The value of β/A from Figure 5 for Run 2 is 1.13 lb/hr-ft² at 5 l/min and the ratio at the same flow rate is:

$$\frac{R}{\beta} = \frac{0.18}{1.13} \text{ to } \frac{0.22}{1.13}, \text{ or } 0.16 \text{ to } 0.20$$

This may be compared with a value for comparable large fire from LF2-D of the LF-2 experiment, where $R/\beta = 0.19$, and Q was 18 cfm for both R and β . Here the agreements between large and small fires again appear to be rather good when comparison of R/β are made for a constant flow rate.

One other comparison for similarity of results for the large and small fires can be made by studying the release rate of the sodium particulates to the theoretical release of sodium vapor at the temperature of the liquid sodium. Such a comparison is based on assuming that observed concentration at the scrubber is related to the vapor pressure P as $P = R/60Q$ KT/M , where K - gas constant, T - temperature, M - molecular wt, Q = flow rate ft³/min, R = lb/hr released to the scrubber.

A comparison of the experimental release rate of the Na to the computed release rate from the vapor pressure is shown in Figure 7. It is seen that the flow rate of 5.5 l/min, the experimental value becomes greater than 1.0. Since the concentration of Na vapor in the gas is directly related to the Na vapor pressure, a condition must exist in the experimental apparatus which enhances the condensation of vapor. It can be assumed that droplets are formed (which do not change the vapor pressure), due to a temperature gradient with resultant supersaturation. The droplets are then carried away with the gas mixture, thus encouraging more vaporization. Similar observations were made in LF-2 at high flow rates.

Figure 7. P_c/P_v vs Flow

3. Laboratory Burn Chamber Studies

The 6 ft x 46 ft³ laboratory burn chamber (Figure 2, Quarterly Project Progress Report for period Jan-Mar, NAA-SR-10850, p 57, VII) was used to measure the release and fate of particles in a closed system with an oxygen concentration of 1%. During the previous report period, some computational errors resulted in reporting incorrect data results. These data were presented in graphical form as representing the concentration and size distribution for particles settling in a tranquil environment. When the data were reevaluated, it was found that:

(a) the filter papers used for the last stage of the impactors were contaminated, thus giving too much weight to the smaller sizes; (b) the concentrations reported from impinger samples taken at a height of 3 ft below the top of the chamber were computed incorrectly. When these corrections were made, it was concluded by means of some theoretical calculations that: (a) some environment existed in the chamber which caused settling under conditions between that of tranquil and stirred; (b) the 8-stage round jet impactors were giving some results which were incorrect if settling was occurring.

Because of the theoretical attempt to analyze the data observed in the laboratory burn-chamber studies, a graphical method was developed for computing size and concentration of sodium oxide which remains airborne in any size container for either turbulent or tranquil environments. These data are presented in Figures 8 and 9. The data assume a given uniform concentration and distribution of sizes in a room at $t = 0$. They also assume that agglomeration occurs at a rate which is small compared to the settling rate. A particular case is selected for which the MMD = 1.4 and $\sigma = 1.95$ (see Curve B in Figure 2). From Curve B one may compute the fractional weight present initially for any given size and size increment in the manner here described.

Figure 8. Tranquil Settling

7640-2508

7640-2509

Figure 9. Stirred Settling

Let S_i be the mass concentration (in. lb/ft³) of the i^{th} size interval of particles at the end of the burning in the Laboratory Burn Chamber. These quantities are functions of time (due to settling), and their initial values S_{i0} are obtained from Curve B of Figure 2 by taking the appropriate difference for the i^{th} size interval. If $H(\text{ft})$ is the height of any room, and $v_i \text{ ft/hr}$ is the terminal settling velocity, then one may write for S_i :

$$S_i = S_{i0} e^{-v_i t/H}$$

It is clear that $\sum S_{i0} = S_0$, the initial concentration for all particle sizes that are airborne. Similarly, $\sum_i S_i = S$ is the airborne concentration at any time t . Figure 8 is a presentation of the curves of S_i plotted as functions of $t/h \text{ hr/ft}$. The values of S_{i0} were obtained from Figure 2, as stated previously; and values for v_i are based on Stokes Law. Figure 9 enables one to compute for any given room height H , and any time of interest, t , the distribution of particle masses as a function of size which remain airborne if turbulence exists. If a suitable sizing device adequately samples the distribution, then one may compute the existing distribution at any time later.

By way of illustration, assume the time interval of interest is the first 1/2 hr. Let $H = 22 \text{ ft}$; then $t/H = 1/44 \text{ hr/ft} = 0.02 \text{ hr/ft}$; for this value of t/H , only particles whose diameters exceed 7μ will have settled appreciably. For the purposes of this discussion, it is assumed no agglomeration had occurred. If agglomeration were important, the expression for S_i would involve a term depending on the square of the density of particles.

In contrast to stirred settling, particles may exist in a tranquil environment. If there is no turbulent mixing and no convection currents, then the particles are

considered to be settling in still air. The concentration of a given size of particle remains uniform below the level to which the uppermost particle of this size could have fallen at the moment of sampling in a room H ft from the top.

Figure 9 illustrates the distribution of various particle size groups as a function of time based on the same data in Figure 2. The nomenclature is the same as that used previously for S_0 , S_{i0} , and S . Initial values S_{i0} are obtained from Figure 2. If one considers a room of height H, then it will require a time $t_i = H/v_i$ for the i^{th} type of particle to settle completely. Thus, values of $t_i/H = 1/v_i$ will define the time interval required to completely settle the i^{th} type of particle. The lines of Figure 9 were drawn in this way. For any other value of H (some height in the room less than total height, measured from the top) and time to the parameter t/H will define the fraction of the room that is cleared for the particle type of interest.

It is also of interest to state the length of pipe L that a particle can travel before settling. The time the particle is airborne in the pipe is given by H/v_i , where H in this case represents the diameter of the pipe. It can be shown that the length L is given by $L = 4/3 H/v_i \times v_H$, where v_H is the horizontal velocity component. If one is interested in a pipe of given length L, then the equation can be used to define a critical particle size and velocity,

$$v_i = \frac{4}{3} \frac{H}{L} v_H ,$$

so that particles whose size is larger than that defined by v_i will fall out in the length L, whereas those particles of smaller size will not. In this way, if one knows the distribution of particles sizes and masses entering the pipe, one can compute the fraction surviving to impinge on a filter at the end of the pipe. One may also compute the concentration and distribution remaining airborne at any height H below the top of an initially homogeneously filled room or chamber.

4. Agglomeration Theory and Particle Detection

When agglomeration alone occurs in a particle environment, no change occurs in the concentration of mass which remains airborne; i. e., only the size distribution or number of particles changes with time. This is a standard relation and is well known. If one considers a closed environment like the laboratory test chamber described above, when one has a continuous source of particles one may wish to compute the maximum number of particles of a given size which may exist as an unagglomerated particle when it is produced at a constant rate. The fact that one size is never produced in a sodium fire limits the following analyses. Consider the rate of change of concentration (by number) of particles made at a constant production rate M particle/sec. Then

$$V \frac{dn}{dt} = -Vkn^2 + M , \quad \dots (4)$$

where

n = particles/cc

k = agglomeration constant $\text{cm}^3/\text{sec} \approx 4 \times 10^{-10} \text{ cm}^3/\text{sec}$, and

V = volume of container cm^3 .

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Upon integration, one obtains

$$n = \sqrt{\frac{M}{Vk}} \frac{(1 - e^{-t/\tau})}{(1 + e^{-t/\tau})} \quad \dots (5)$$

The significance of the equation is as follows.

- 1) at $t = \text{equilibrium}$, $n_{\alpha} = \sqrt{\frac{M}{kV}}$, and
- 2) the time constant, $\tau = \frac{V}{\sqrt{4MkV}}$.

Thus, the limiting concentration for particles of a given size (assuming a constant k) is dependent on the agglomeration and release rate. For example, if $M = 10^{10}$ particle/sec and $V = 10^5$ cc,

then

$$n = 1.6 \times 10^7 \text{ particles/cm}^3, \\ \tau = 80 \text{ sec.}$$

In 300 seconds, 3×10^{12} p/s would have been produced. The concentration of monosized particles would be 1.6×10^7 particles/cc, but 1.4×10^7 of these particles would have agglomerated.

If plating, settling, and flow terms are subtracted from the righthand side of Equation 4 and grouped as αn , the differential equation is

$$V \frac{dn}{dt} = M - Vkn^2 - \alpha n$$

The solution is then

$$n = \frac{2M(1 - e^{-t/\tau})}{\alpha(1 - e^{-t/\tau}) + \sqrt{4MkV + \alpha^2}(1 + e^{-t/\tau})},$$

where

$$\tau = \frac{V}{\sqrt{4MkV + \alpha^2}}$$

Once, again, the limiting concentration and time constant τ for a given size particle will depend entirely on agglomeration if $4MkV > (\alpha)^2$.

Because of the difficulty of computing rate of decrease in the concentration of a heterogeneous distribution of particles, this project attempted to develop the Royco multichannel particle counting system. Such a system would allow rapid monitoring of the number and size of particles as a function of time. The instrument has been calibrated and awaits testing.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

Evaluations of the release rate and particle size distribution of sodium and sodium oxide from small and large fires have been made. Several instruments have been designed, purchased, modified, and calibrated in order to determine particle size distributions. The activity distribution and release of simulated fission products from a sodium fire have been measured. ^{131}I tagged as NaI has been used exclusively. Other isotopes will be used as tracers in the future. Some evaluation of particle production and removal mechanisms has been made in controlled environments. Models for scaling fires have been initiated, and larger test facilities have been planned. Continuously monitoring the distribution and fate of released sodium and fission products in large controlled environments will enable the project to meet its ultimate objectives of evaluating the source term for filtration or environmental dispersion. Particle size distribution measurement and prediction has proven to be a formidable task. For this reason, more use will be made of automatic sampling and plating instruments which will allow a complete empirical characterization of the released particulates.

The initial results obtained with the small scaling experiments on sodium burning were encouraging with respect to apparently reproducing some of the experimental observations obtained in the large fire experiment (LF-2). If scaling experiments continue to provide this type of information it will enable a thorough study of the effect of important parameters and their variation on burning and aerosol release. However, it is anticipated that future plans will require continued use of large scale experiments in order to study other key variables pertinent to accident simulation which cannot be properly evaluated in small experiments.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Advanced Development Program

Project: High-Temperature Chemistry

Project Engineer: S. J. Yosim

Reporting Period: Fiscal Year 1965

AEC Category: 05-02-02-01

General Order: 9305

Subaccount: 5302

I. PROJECT OBJECTIVES

The project is divided into two general study areas, namely, fused salts and metal-metal salt solutions. The objective of the study of fused salts is to achieve an understanding of ionic melts (halides, oxides, oxy-salts, and glass systems) by: (1) establishing the nature of the species existing in ionic melts, (2) determining the physicochemical interactions between species, and (3) formulating systematic relationships between the physicochemical properties of fused salt systems and the molecular parameters of the species.

The objectives of the metal-metal salt study are: (1) to determine phase diagrams for, and the nature of interactions between metals and their salts at high temperatures, (2) to determine the species in these solutions, and finally (3) to predict the solubilities in metal-salt systems.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. PUBLICATIONS AND TALKS

The following papers have been published.

1. "Molar Volume and Surface Tension of Molten Metaphosphate Sulfate Systems," by B. B. Owens and W. W. Mayer, J. Am. Ceram. Soc., 47, 347 (1964)
2. "Calculation of Thermodynamic Properties of Fused Salts," by S. J. Yosim and B. B. Owens, J. Chem. Phys., 41, 2032 (1964)
3. "Melting Properties of KNO_3 Under Pressure," by S. E. Babb, Jr., P. E. Chaney, and B. B. Owens, J. Chem. Phys., 41, 2210 (1964)
4. "Thermoelectric Potentials in Molten Bi-BiI₃ Solutions," by D. O. Raleigh and L. E. Topol, J. Chem. Phys., 41, 3179 (1964)
5. "Thermodynamic Considerations in Metal-Metal Salt Interactions," by L. E. Topol, J. Phys. Chem., 69, 11 (1965)
6. "Melting Properties of the Alkali Nitrates to 10,000 Atmospheres," by B. B. Owens, J. Chem. Phys., 42, 2259 (1965)

The following papers are in press.

1. "The Excess Entropy of Mixing of Liquids," by S. J. Yosim, in J. Chem. Phys.
2. "The Electrical Conductivity of Molten Bi-BiCl₃ and Bi-BiBr₃ Solutions," by L. F. Grantham, in J. Chem. Phys.
3. "Electrochemical Studies of NO^+ and NO_2^+ in Concentrated Sulfuric Acid," by L. E. Topol, R. A. Osteryoung and J. H. Christie, in J. Electrochem. Soc.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

The following talks were given at the Southwest Regional Meeting of the American Chemical Society, December 5, 1964.

1. "The Electrical Conductivity of Molten Cadmium- Cadmium Halide Solutions," by L. F. Grantham
2. "High Pressure Phase Relationships and Compressibilities of the Alkali Nitrates," by B. B. Owens
3. "A Theory of Liquids," by S. J. Yosim.

The following talks were given at the American Physical Society.

4. "The Excess Entropy of Mixing of Liquids," by S. J. Yosim
5. "Corresponding States Melting Curve for the Alkali Halides at High Pressure," by B. B. Owens.

The following talks were given at the Fused Salt Symposium at the 127th Electrochemical Society Meeting.

6. "The Electrical Conductivity of Molten Cadmium-Cadmium Halide Solutions," by L. F. Grantham
7. "Effects of Temperature on Electrical Conductivities of Fused Salts," by S. J. Yosim and L. F. Grantham
8. "Properties of Molten Nitrates Under High Pressure," by B. B. Owens.

B. THEORETICAL STUDIES

1. Excess Entropy of Mixing of Liquids

An expression for the excess entropy of mixing two liquids isothermally was obtained from the entropy of mixing two fluids of rigid spheres, each at the volume of the corresponding liquid components, to form a solution of rigid spheres the volume of which was that of the liquid solution. The theoretical entropy of mixing of liquids was found to be ideal only if the diameters of the molecules are the same, the molar volumes of the components are equal, and the volume of mixing is zero. For small deviations from the above conditions, the excess entropy of mixing was proportional to the product of the mole fractions of the components. The equation was applied to nonpolar systems; considering the simplicity of the model, agreement between the calculated and observed values is considered to be satisfactory. This study has been completed and the results published.

2. Statistical Mechanics of Compressed Gases

The application of the scaled particle theory of liquids has proven very useful for calculating thermodynamic properties. Application of this theory to compressed gases has been initiated. One reason for this study is to test the theory over a much greater density range (from densities of low-pressure gases to those of gases compressed to the volume of liquids) than was possible in liquids. The first thermodynamic property to be studied is C_p , the heat capacity at constant pressure. Preliminary results for argon show good agreement even near the critical temperature.

3. Theory of Corresponding States Applied to High Pressure Melting Curve for the Alkali Halides

The effect of pressure upon the melting points of 14 of the alkali halides has been reported in the literature. However, the results have not been interpreted in terms of the theory of corresponding states which has been developed by Reiss, Mayer, and Katz (see Annual Report 1961). They found that a universal vapor pressure curve for fused salts may be given in the form

$$\pi = \pi(\tau),$$

where the reduced pressure $\pi = \kappa \lambda^4 P / Z^2$ and the reduced temperature $\tau = \kappa \lambda T / Z^2$. P is the pressure, T is the temperature, λ is the distance of closest approach, Z is the valence, and κ is a dielectric constant. We have now found that the high-pressure melting points of these alkali halides (in the fcc NaCl structure), when plotted in terms of the reduced parameters defined above, follow the single curve

$$\tau \times 10^{-3} = 3.22 \times 7.28 \times 10^{-4} \pi - 8.68 \times 10^{-8} \pi^2,$$

with an rms deviation of 2.5%. (As has been observed in other theoretical studies, LiCl did not conform to the other cation halides.) This universal melting curve should be very useful for predicting the melting points of the alkali halides at high pressure. This study has been completed and the results will be published.

C. TRANSPORT STUDIES

1. Conductivity Maxima in Fused Salts

Electrical conductivity maxima have previously been found by us in BiCl₃, BiBr₃, BiI₃, CdI₂, HgCl₂, HgBr₂, and CuCl when these salts were heated. The conductivities of additional salts have been measured to see how prevalent this phenomenon is. Maxima have been found in ZnI₂ (950°), SnCl₂ (875°), and InI₃ (480°), whereas in TiCl, TlBr, and TlI, which are regarded as more ionic salts, no maxima were observed. Thus far, there are 13 systems that have a temperature region where the temperature coefficient of conductivity is negative. If one uses the heat of formation per halide ion value at 298 as a measure of stability, then it appears that the less stable salts are more likely to have a maximum. For example, out of five salts which have heats of formation more negative than -40 kcal/X⁻, only one exhibits a negative temperature coefficient while out of the remaining 13 less stable salts, 12 possess this property.

2. Critical Temperature Studies in Fused Salts

Attempts are being made to measure the conductivity of fused salts above their critical temperatures. Thus far, the electrical conductivity of pure fused BiCl₃ was measured to 860°C, only 45° below the critical point. The specific conductivity increased from 0.36 at 230°C to a maximum of 0.58 at 425° then decreased to 0.16 at 860°C. In addition, the electrical conductivity of pure BiBr₃ was measured to 818°C (critical temperature is 950°C). The specific conductivity increased from 0.32 at 315° to a maximum of 0.35 at 425°, then

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

decreased to 0.14 at 818°. It is interesting to note that BiCl₃ from a conductivity standpoint appears to be quite ionic, even slightly below the critical temperature. The cells are being modified to permit higher temperature measurements.

3. Electrical Conductivities of Metal-Salt Solutions Near Critical Temperatures

The electrical conductivity of the liquid and vapor phases above a 30 mole % Bi-BiBr₃ melt was determined to 1025°C (75° above the critical temperature of the pure salt). The electrical conductivity of the solution continued to increase exponentially, even above the critical temperature of the BiBr₃ (14Ω⁻¹ cm⁻¹ at 1025°). However, the conductivity of the vapor phase was still only about 10⁻⁴ (Ω cm)⁻¹ at this temperature. This indicates that we are still below the critical temperature of the solution.

4. Electrical Conductivities of Cadmium-Cadmium Halide Solutions

Based on our electrical conductivity results of the Bi-BiX₃ systems, one would predict that in a system in which the subhalide polymer was extremely stable, no electron transfer should be observed; hence the electrical conductivity of such solutions should not increase appreciably as the concentration of subhalide is increased. The Cd-CdX₂ systems are ideally suited to check this hypothesis. Topol has found that the concentration of Cd⁺ ion is insignificant in Cd-CdCl₂ melts near the melting point; essentially all of the subhalide exists as the dimer. Presumably the same would be expected for the bromide and iodide systems. Therefore the electrical conductivity of 5, 10, and 20 mole % Cd-CdCl₂ and Cd-CdBr₂ as well as 5, 10, and 15% Cd-CdI₂ solutions were measured. The effect of Cd composition and temperature in the conductivity was not great, whereas in the bismuth systems the effect was in general large. In contrast to the Cd-CdCl₂ system at lower temperatures, where a decrease in conductivity was found as Cd was added, the conductivity of the Cd-CdBr₂ and Cd-CdI₂ systems increased at all temperatures as the Cd content increased. No evidence of conduction by an electron transfer mechanism was found. This completes the cadmium conductivity studies and the results will be published.

5. Electrical Conductivities of Mercury-Mercuric Halide Solutions

A study of the electrical conductivities of the Hg-HgX₂ systems has been initiated. The mercury halides have relatively low critical temperatures (~750°C), so that measurements in the supercritical region may be feasible. The first measurement was done on HgI₂ containing 15 mole % Hg. The conductivity (about an order of magnitude greater than the pure salt) increased from 0.335 (Ω cm)⁻¹ at 240° to a maximum of 0.375 (Ω cm)⁻¹ at 330°, then decreased to 0.120 (Ω cm)⁻¹ at 580°C. In addition, the electrical conductivity of a 30 mole % Hg-HgI₂ solution was measured. A maximum in the conductivity of this solution was also found, but at a higher temperature and concentration. The specific conductivity of the 30% solution increased from 0.47 (Ω cm)⁻¹ at 525°C to a maximum of 0.74 (Ω cm)⁻¹ at 420°, then decreased to 0.52 at 642°C. It is anticipated that with the modified cells, measurements can be carried out above the critical temperatures.

6. The Soret Effect in Metal-Salt Solutions

The Soret effect occurs when a mixture is placed in a temperature gradient; a separation takes place, resulting in an emf. At zero time, before separation of the components takes place, the emf is the Seebeck coefficient; at steady-state, the emf is related to the Soret coefficient. The steady-state thermoelectric powers of BiI_3 solutions containing 5, 10, 15, 20, 25, 30, 35, 50, and 80 mole % Bi have been measured at 500°C. The value at 500° was large at low metal concentrations ($-3100 \mu\text{v/deg}$ at 3% metal), and diminished exponentially to that of the initial thermoelectric power at 30% dissolved bismuth metal. From 30 to 80% bismuth, the initial and final thermoelectric powers continue to be identical, indicating no Soret effect at these compositions, even though the system contains two components.

D. HIGH-PRESSURE STUDIES

1. The Compression of the Solid Bismuth Trihalides

Compression measurements were made with a piston cylinder apparatus on the solid bismuth trihalides BiF_3 , BiCl_3 , BiBr_3 , and BiI_3 at 25°C at pressures up to 40 kilobars. The compressions $-\Delta V/V_0$ were calculated relative to P. W. Bridgman's compression data for iron. BiF_3 is a relatively incompressible material; $-\Delta V/V_0$ is only 0.054 at 40 kilobars. The compressions for BiCl_3 , BiBr_3 and BiI_3 are respectively 0.128, 0.176, and 0.152 at 40 kilobars. The anomalous order of the compression of the normal form of BiI_3 with respect to atomic number of the halogen led to the discovery of a phase transition in BiI_3 . This transition, $\text{BiI}_3(\text{I}) \rightarrow \text{BiI}_3(\text{II})$, is very sluggish at 25°C but occurs more rapidly at 100°C. The transition pressure at 100°C is estimated to be 24 kilobars. There is a volume decrease of 6.5% associated with the transition $\text{BiI}_3(\text{I}) \rightarrow \text{BiI}_3(\text{II})$ at 24 kilobars pressure. The fractional volume decrease, $-\Delta V/V_0$ [where V_0 is volume of $\text{BiI}_3(\text{I})$, 1 bar 25°C] for $\text{BiI}_3(\text{II})$ at 40 kilobars, is 0.206. The decrease in thermodynamic stability of $\text{BiI}_3(\text{I})$ with increasing pressure can be predicted from molar volume considerations.

2. Preparation of New Materials with High Pressure

Recently several new materials have been obtained in metastable forms after a high pressure-quench treatment (e.g. metallic indium antimonide and metallic indium telluride). An attempt was made to obtain metastable forms of Bi. (At 25°C Bi (I) forms Bi (II) at 25 kilobars, which subsequently is converted to Bi(III) at 26.5 kilobars.) However, the attempts were unsuccessful. The Bi(II) - Bi(I) reaction was found to be reversible even at liquid nitrogen temperature. In the case of Bi(III), this form was stable at liquid nitrogen temperature until the pressure was reduced to 5 kilobars. This experiment will be repeated with slight modifications.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

A. FUSED SALTS

The statistical mechanical approach for developing liquid theory so that the physico-chemical properties of molten salts can be calculated from equations

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

with no adjustable parameters continues to be very fruitful. The emphasis in the experimental approach continues on high-pressure and transport studies. Emphasis is being placed on the study of salts near and above the critical temperature. Such studies will prove informative in the transition from ionic gases to liquids.

B. METAL-METAL SALT SOLUTIONS

Emphasis is still being placed on experiments carried out above the temperature of complete miscibility as well as above the critical temperature of the salt. Such studies will shed considerable light on the gradual transition from salt to metallic behavior.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program:	Advanced Development		
Project:	Radiation Chemistry		
Project Engineer:	R. B. Ingalls		
Reporting Period:	Fiscal Year 1965	AEC Category:	05-02-02-01
General Order:	9305	Subaccount:	5305

Principal Scientists: R. B. Ingalls, R. A. Holroyd, G. T. Tiedeman

I. PROJECT OBJECTIVES

The objectives of this project are to delineate the fundamental physico-chemical processes occurring in the radiolysis of organic systems, and to study these processes in separate experiments designed to increase our understanding of their nature and their role in radiolysis.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. PAPERS AND PRESENTATIONS

The following papers have been published.

1. "Reactions of Deuterated Polystyrenes with Halogen and Deuterium Atoms," by L. A. Wall and R. B. Ingalls, J. Chem. Phys., 41, 1112 (1964)
2. "Thermal-Spike Model of Low LET Radiolysis: Temperature Dependence of Reactions Induced by Radiolysis in Liquid Toluene," by R. B. Ingalls, P. Speigler, and A. Norman, J. Chem. Phys., 41, 837 (1964)
3. "Methyl-C-14 Radical as a Scavenger in Studies of the Radiolysis of Saturated and Unsaturated Hydrocarbon," by R. A. Holroyd and G. W. Klein, J. App. Radiation and Isotopes, 15, 633 (1964)
4. "Radical Yields in the Radiolysis of Unsaturated Hydrocarbons," by R. A. Holroyd and G. W. Klein, J. Phys. Chem. 69, 194 (1965)
5. "Determination of Radiative Lifetime of Vibrationally Excited sec-Butyl Radicals using Methylethylamine as a Model," by G. T. Tiedeman, R. B. Ingalls, and Jack S. Margolis, J. Chem. Phys. 42, 2627 (1965)

The following papers have been accepted for publication.

1. "The ESR Spectra of Six Selectively Deuterated Biphenyl Negative Ions," by T. W. Lapp, J. G. Burr, and R. B. Ingalls, J. Chem. Phys.
2. "ESR Study of Excited Rare Gas Initiated Reactions of Polystyrene and Polymethylmethacrylate," by R. B. Ingalls and W. A. Young, in press, J. Chem. Phys.
3. "Radical Intermediates in the Mercury Photosensitized Decomposition of Cyclopropane," by R. A. Holroyd and G. W. Klein, J. Phys. Chem.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

The following paper has been submitted for publication:

1. "Radical Yields in the Radiolysis of Hydrocarbons - Part II - Neopentane, Isopentane, and Cyclopentane," by R. A. Holroyd and G. W. Klein, J. Am. Chem. Soc.

The following papers were presented.

1. "Mercury Photosensitized Decomposition of Propane, Isobutane, and n-Pentane at 1849Å," by R. A. Holroyd, presented at the Sixth Informal Photochemistry Conference at Davis, California, June 15-17, 1964
2. "Detection of Hydrogen Atoms in Irradiated Hydrocarbons," by R. A. Holroyd, presented at the Pacific Southwest Regional Meeting of A.C.S. held at Costa Mesa, California, on December 5, 1964
3. "Thermal Spike Model of Low LET Radiolysis," by R. B. Ingalls, presented at the Pacific Southwest Regional Meeting of A.C.S. held at Costa Mesa, California, on December 5, 1964
4. "Reactions of Hydrogen Atoms with Polystyrene - A Model for the Study of Biological Systems," by R. B. Ingalls, presented at the Laboratory of Nuclear Medicine and Radiation Biology in the Department of Biophysics at UCLA on February 16, 1965
5. "Primary Processes in the Radiolysis of Hydrocarbons," by R. A. Holroyd, presented at the Aerospace Corporation on March 22, 1965
6. "The Reaction of Hydrogen Atoms with Polystyrene" - A Model for the Study of Biological Polymers," by R. B. Ingalls, presented at the Pharmacy Research Conference at USC on April 5, 1965
7. "Evidence for Ionic Intermediates in the Radiolysis of Hydrocarbons," by R. A. Holroyd, presented at the Miller Conference on Radiation Chemistry in Port Marion, Wales, April 26-30.

B. RADIATION CHEMISTRY

1. Estimation of Activation Energies

The activation energy for the dissociation of the triphenyl cyclohexadienyl radical R,

was calculated to be 28 ± 3 kcal/mole at 500°C . This radical is likely to be an intermediate in both the pyrolysis and high-temperature radiolysis of o-terphenyl. Its dissociation will propagate a chain reaction and consequently may explain why the observed activation energy of pyrolysis of o-terphenyl, 71 kcal/mole, is lower than the energy required for initial bond cleavage.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

The activation energy was obtained by assuming that it was equal to the heat of dissociation of the radical, $D(R)$, which was calculated from the equation:

$$D(R) = \Delta H_f(\text{triphenylbenzene}) + \Delta H_f(H) - \Delta H_f(R). \quad \dots(2)$$

The heats of formation, ΔH_f , for the two structures involved were estimated from tables of bond energies. In the case of the radical R , $\Delta H_f(R)$ was obtained from the calculated heat of formation of triphenylcyclohexadiene and the known dissociation energy of an allylic carbon-hydrogen bond.

2. ESR Studies of Atomic Reactions

a. Polymer and Atom Survey

The reaction of nitrogen and oxygen atoms with "fluffed" samples of polymers was investigated by ESR techniques. For samples of polystyrene, poly-2,3,4,5,6-pentadeuterostyrene, poly- α, β, β - trideuterostyrene, and polyvinyl alcohol, featureless, very similar ESR spectra were observed. No fine structure was apparent in the spectra. There were minor differences between the oxygen and nitrogen treated series. Similar featureless spectra were observed when nitrogen atoms were reacted with fluffs of poly-m-methylstyrene, polyperdeutero-m-methylstyrene, and poly-p-isopropylstyrene.

In another series of experiments, deuterium and hydrogen atoms were reacted with samples of the latter set of polymers. For each polymer, different spectra were observed when deuterium was substituted for hydrogen. Therefore, it was concluded that the spin-unpaired species were not produced by abstraction reactions. Attempts will be made to relate the spectra of the species observed for these polymers to that of the cyclohexadienyl species previously proposed for polystyrene.

b. Mechanism of the Atom-Fluff Reaction

The temperature-dependence of the reaction of deuterium atoms with polystyrene fluff was investigated in detail. The results of this study showed that the activation energy for addition of deuterium atoms to the fluff is much smaller than the 10 kcal/mole activation energy expected for diffusion of deuterium atoms into solid polystyrene. Therefore, it is concluded that the addition reaction is probably confined to the polymer fiber surface.

3. Determination of the Radiative Lifetime of Vibrationally Excited sec-Butyl Radicals using Methylethylamine as a Model

A calculation was made to determine if radiation is likely to be an important mode of deactivation for vibrationally excited sec-butyl radicals. As a model of this radical the structurally similar compound, methylethylamine, was chosen. This differs from the radical only by the substitution of a nitrogen atom for a carbon atom, which should not perturb the vibrational behavior significantly. The radiative lifetime of the model compound was calculated to be 3.5×10^{-3} sec. Since, for vibrationally excited sec-butyl radicals, the time between collisions in typical gas-phase experiments is orders of magnitude smaller than 10^{-3} sec, radiation is not expected to be an important method of energy loss.

4. Ionic Intermediates in the Radiolysis of Organic Liquids

An experimental study has been initiated to determine the time required for geminate recombination of electrons with positive ions produced in the radiolysis of organic liquids. Irradiation of some compounds in which substituted phenethyl carbonium ions are expected to be intermediates has led to no rearrangements in the carbon skeletons of the molecules. These preliminary results suggest that geminate recombination occurs in less than 10^{-11} sec.

5. Radical Yields in the Radiolysis of Hydrocarbons

a. A Review of Techniques and Results

A review article has been prepared for publication which describes the existing chemical techniques for measuring radical yields such as iodine¹³¹, tritium iodide, and the C-14-labeled radical sampling techniques. Also, the results and conclusions obtained recently on radical yields are summarized. The review emphasizes the significant effect of structure on the yields of both fragment and parent radicals. A "track effect" has been demonstrated by a comparison of the scavengible radical yields with the product yields in neopentane, cyclohexane, and 2,2,4-trimethylpentane. This comparison shows that about 40% of the radicals produced react together in the radiation tracks or "spurs."

b. Neopentane, Isopentane, and Cyclopentane

Individual radical yields from neopentane, isopentane, and cyclopentane have been determined using the ¹⁴C₂H₅-radical sampling method. In neopentane only four alkyl radicals are present, whereas in isopentane nine are observed. In neopentane and isopentane, fragment alkyl radicals arising from the scission of carbon-carbon bonds, account for 79 and 41%, respectively, of the alkyl radicals formed in primary processes. The relative probability (per bond) of rupture of primary, secondary, and tertiary carbon-hydrogen bonds in isopentane is 1:3:9.

The observation that $G(\text{CH}_3)$ exceeds $G(\text{C}_4\text{H}_9)$ in both neopentane and isopentane and the fact that fragmentation is more probable for branched structures are attributed to the fact that decomposition of molecular ions is more probable for highly branched structures. In cyclopentane, 90% of the radicals are cyclopentyl though small amount of three additional C₅ radicals are observed.

In neopentane, a small yield of isobutyl radicals ($G = 0.16$) is observed which is believed to result from ionic isomerization. Evidence is cited which suggests that considerable combination of radicals occurs in tracks for low LET radiolysis. The fraction of methyl radicals which escape the track, $N(\text{CH}_3)/N(\text{CH}_3)_0$ is estimated to be ≤ 0.66 for neopentane.

c. 2,2-Dimethylbutane, 2,4-Dimethylpentane, and 2,2,4-Trimethylpentane

The carbon-14-labeled methyl radical "sampling" technique has been employed to measure the yield of radicals in several branched alkanes. The fragment alkyl radicals methyl, ethyl, and t-butyl account for 71% of the radicals formed in 2,2-dimethylbutane. Fragmentation is much less important (48%) in 2,4-dimethylpentane, and leads principally to isopropyl and isobutyl radicals. In 2,2,4-trimethylpentane, fragmentation accounts for 64% of the primary radical

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

yield. The yields of ethyl radicals from saturated hydrocarbons are found to fit a single empirical relationship,

$$G(C_2H_5) = 3.5(1.\underline{a} + 2.8\underline{b} + 7.9\underline{c})/(\underline{n} - 1)^2,$$

where $\underline{n}$ is the number of carbon atoms in the molecule and $\underline{a}$, $\underline{b}$, and $\underline{c}$ are the numbers of methyl groups attached to a carbon on which there are, respectively, 0, 1, or 2 other methyl groups. The primary yields of parent alkyl radicals have been measured, using ethylene to suppress hydrogen atom abstraction reactions. It is found that the probability of scission per bond of a secondary carbon-hydrogen bond is three to six times that of a primary bond for these alkanes. No such consistency is observed for tertiary bonds; the yield of tertiary octyl radicals from 2,2,4-trimethylpentane is nearly zero, and the tertiary heptyl radical yield from 2,4-dimethylpentane is only 0.3. It is thought that steric factors must be important in parent alkyl radical formation, and that radicals are formed by a bimolecular reaction; the primary process

is suggested. Evidence of a track effect has been obtained in the case of 2,2,4-trimethylpentane which indicates that only 57% of the methyl radicals, formed initially, escape from the track, the remainder undergoing bimolecular reactions in the track of the ionizing particle.

d. The Yield of Thermal Hydrogen Atoms from the Radiolysis of Saturated Hydrocarbons

The purpose of this study was to establish to what extent thermal hydrogen atoms are present in irradiated hydrocarbons. Ethylene, which is added in millimolar quantities to the hydrocarbon, was chosen specifically since it would only scavenge hydrogen atoms. The yield of ethyl radicals formed by the addition of hydrogen atoms to ethylene was measured using the $^{14}CH_3$ radical sampling technique. The results obtained for the yield of thermal hydrogen atoms per 100 e.v., $G(H)$, were for n-pentane, 1.8; for 2,4-dimethylpentane, 1.9; and for 2,2,4-trimethylpentane, 1.6. The ratio of $G(H)$ to the primary yield of cycloalkyl radicals was found to be 0.83 for both cyclohexane and cyclopentane. If the total cycloalkyl radical yield is 4.0, then these data indicate $G(H)$ is 1.8 for cycloalkanes. The yield of thermal hydrogen atoms apparently is not markedly affected by structural changes. The yield observed with ethylene is about one-half the yield reported in studies where solutes such as methymethacrylate and benzoquinone were used. The latter solutes apparently reduce the hydrogen yield not only by scavenging thermal hydrogen atoms but also by reacting with the precursors of molecular hydrogen which may be electrons.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

A significant contribution to radiation chemical theory has been made in the development of a thermal spike model of low LET radiolysis. This theory accounts successfully for the low-temperature dependence of yields of products, as is often observed in the radiolysis of hydrocarbons. The model also appears to allow one to predict beforehand yields of products during radiolysis at elevated temperatures from pyrolysis data. Further experimental tests of the theory are planned.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

The labeled radical sampling equipment has been set up and used to measure the individual radical yields from several pure liquid branched alkanes. Comparison of the results for several hydrocarbons has led to certain empirical correlations useful for the prediction of radical yields. This technique is also useful to monitor the effect on radical yields of various solutes, capable of interacting with intermediates such as ions, excited states, or hydrogen atoms. It has been established with the use of ethylene as a solute that thermal hydrogen atoms are formed in alkanes with a yield of 1.6 ± 0.2 atoms/100 e.v. independent of structure. Further work with other solutes is in progress.

Considerable emphasis has been placed in this period on the study of elementary reactions. The radiative lifetime of vibrationally excited secondary butyl radicals and the activation energy for the decomposition of phenylcyclohexadienyl radicals have been calculated, also the temperature-dependence of the rate of addition of hydrogen atoms to polystyrene has been measured.

A new experimental method for establishing the lifetime of ionic intermediates in organic radiolysis is being developed. This method is based on the determination of skeletal rearrangements known to occur in ionic intermediates in phenethyl compounds. These measurements may help establish the extent of ionic reactions in organic liquids.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Physical Research Program

Project: Electronics Structure of Metals and Alloys

Project Engineer: R. G. Breckenridge

Reporting Period: Fiscal Year 1965

AEC Category: 05-03-03-01

General Order: 9305

Subaccount: 5401

Principal Scientists: R. G. Breckenridge, L. J. Barnes, H. J. Fink,
W. J. Tomasch

I. PROJECT OBJECTIVES

This effort is devoted to the acquisition of knowledge regarding the electronic structure of metals and alloys (configuration of electronic energy states in momentum space) and the role of this structure in determining electrical, thermal, magnetic, vibrational, and alloying characteristics. Detailed information on the shape of the Fermi surface is provided by de Haas-van Alphen studies in magnetic fields up to 200 kilogauss, and the density of electronic states at the Fermi level is deduced from low-temperature specific heat measurements. The latter also yield information on lattice vibrational modes and on the interactions involved in superconductivity and magnet ordering. Further characterization of magnetic interactions is accomplished by means of conventional magnetic susceptibility techniques. Considerable effort is devoted to the exploration of high-field superconductivity with emphasis on thermodynamic and transport characteristics, as well as on the electron energy spectrum as deduced from thin-film electron-tunneling measurements. These experimental investigations are correlated with current theory, and attempts are made to characterize quantitatively the relationship between superconductivity and the normal-state electronic structure.

II. TECHNICAL PROGRESS DURING FISCAL YEAR 1965

A. PUBLICATIONS

1. W. J. Tomasch, "Influence of a Magnetic Field on Tunneling from a Type II Alloy Superconductor," to appear in June 1965 edition of Phys. Rev.
2. H. J. Fink, A. C. Thorsen, Earl Parker, Victor F. Zackay, and Lou Toth, "High Field Superconductivity of Carbides," Phys. Rev. 138, A1170 (1965)
3. R. R. Hake and L. J. Barnes, "Magnetocaloric Effects in a High-Field Superconductor," Proceedings of the IXth International Conference on Low Temperature Physics (Columbus, 1964), 513 (1965)
4. R. R. Hake, D. H. Leslie, and C. G. Rhodes, "Giant Anisotropy in the High-Field Critical Currents of Cold-Rolled Transition Metal Alloy Superconductors," Proceedings of the VIIth International Conference on Low Temperature Physics, ed. by R. O. Davies, Butterworths, p 342 (1964)
5. R. R. Hake and W. G. Brammer, "High-Magnetic-Field Specific Heat of a Low-Dislocation-Density Alloy Superconductor," Physical Review 133, A719 (1964)

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

6. R.R. Hake, "Thermodynamic Properties of a High-Field Superconductor," Rev. Mod. Phys. 36, 124 (1964)
7. R.R. Hake, "Some Thermodynamic Relationships for the Ideal Bulk Type II Superconductor," Type II Superconductivity Conference, Cleveland (1964) unpublished
8. R.R. Hake and J.A. Cape, "Calorimetric Investigations of Localized Manganese and Cobalt," Phys. Rev. 135, 5A, A1151 (1964)
9. J.A. Cape and R.R. Hake, "Localized Magnetic Impurity States in Ti, Zr, and Hf," to be published in Phys. Rev.
10. H.J. Fink, "Collective Modes of Vortex Lines in Type II Superconductors," North American Aviation Science Center Science Symposium, Ojai, October 25-27, 1964
11. L.J. Barnes and R.R. Hake, "Magnetocaloric Effects in a High-Field Superconductor," North American Aviation Science Center Science Symposium, Ojai, October 25-27, 1964
12. H.J. Fink, "Vortex Wave Spectrum of a Type II Superconductor," Phys. Rev. Letters 12, 578 (1964)
13. H.J. Fink, "Superconducting Surface Sheath of a Type II Superconductor Below the Upper Critical Field H_{C2} ," Phys. Rev. Letters 14, 309 (1965)
14. W.J. Tomasch and A.S. Joseph, "Experimental Evidence for a New Superconducting Phase Nucleation Field in Type-II Superconductors," Phys. Rev. Letters 12, 148 (1964)
15. W.J. Tomasch, "Persistence of Surface Superconductivity Beyond H_{C2} in a Type II Superconductor as Evidenced by Electron Tunneling," Phys. Letters 9, 2 (1964)
16. W.J. Tomasch, "Tunneling Between a Type II Superconductor and a Very Thin Superconducting Al Film in a Magnetic Field," to appear in the Proceedings of the IXth International Conference on Low Temperature Physics (1964)
17. A.S. Joseph and W.J. Tomasch, "Experimental Evidence for Delayed Entry of Flux into a Type-II Superconductor," Phys. Rev. Letters 12, 219 (1964)
18. A.S. Joseph and A.C. Thorsen, "Low-Field de Haas-van Alphen Effect in Copper," Phys. Rev. 134 4A, A979 (1964)
19. A.S. Joseph and A.C. Thorsen, "Low-Field de Haas-van Alphen Effect and Fermi Surface in Ag and Au," Phys. Rev. Letters 13, 9 (1964)
20. H.J. Fink, "Magnetoresistance of Silver," Physics Letters 13, p 105 (1964)
21. H.J. Fink, "Internal Currents and Magnetic Fields Close to the Surface for a Type II Superconductor for Applied Magnetic Fields Near the Upper Critical Field H_{C2} ," Phys. Rev. Letters 14, 853 (1965)

B. THEORETICAL STUDIES ON SUPERCONDUCTIVITY (H. J. Fink)

1. Theoretical Investigation of the Superconducting Sheath in Type II Superconductors

Considerable progress has been made in the understanding of the superconducting surface sheath. It was shown that the sheath does also exist in magnetic fields smaller than the upper critical field H_{C2} . The theoretical results were correlated to tunneling experiments [W.J. Tomasch, Phys. Letters 9, 104 (1964)] and good agreement was found. It was found that the energy gap near the surface for applied fields near H_{C2} is about 70% of the zero field gap, which is

considerable if one remembers that there is almost no superconductivity in the bulk of the metal at these fields. The internal magnetic field and the shielding current were calculated between H_{C2} and H_{C3} . It is remarkable that there are two supercurrents near the surface which flow in opposite directions and whose magnitudes are very large compared to the vortex currents just below H_{C2} . Above H_{C2} the magnitudes of these currents are exactly equal, thus providing no shielding to the sample (at least if the sample is in the lowest stable energy state). Below H_{C2} the currents are slightly different and give rise to a very small net surface current. The currents invert their direction within one coherence length from the surface. It is found that the variation of the internal fields from the external field and the magnitudes of the surface currents are unexpectedly large if the Ginzburg-Landau parameter κ is smaller than three.

2. Magnetoresistance of Silver

Magnetoresistance experiments disclosed a sharp drop of the transverse magnetoresistance when the magnetic field is applied perpendicular to the (112) direction. This region is not surrounded by high peaks which is usually the case for two intersecting open orbit directions which are surrounded by aperiodic open orbits. This new anomaly in the magnetoresistance could be explained with the help of the topology of the Fermi Surface suggested by Pippard. In this model the open orbits along the (111) and (110) are primary and secondary open orbits which, as it turns out, do not intersect and therefore do not give rise to the high peaks which usually arise from the aperiodic open orbits surrounding the singularity.

3. High Field Superconductivity of Carbides

The high magnetic field properties of some refractory carbides were investigated at low temperatures. The upper critical fields for NbC, a solid solution of 40% NbC-60% TaC, and TaC are between 4.2 and 21.3 kG; MoC, Mo₅₆C₄₄, and Mo₆₀C₄₀ + 2% VC have upper critical fields between 80 and 120 kG; and Mo₃Al₂C has an upper critical field of 156 kG at 1.2°K. Mo₃Al₂C is the only known superconductor with the β -Mn crystal structure. From the available data the electronic specific heat coefficient γ , the thermodynamic critical field H_C , the Ginzburg-Landau parameter κ , and the lower critical field H_{C1} were estimated at the experimental temperatures.

4. Collective Modes of Vortex Lines in Type II Superconductors

The possibility of collective vibrations of Abrikosov vortex lines in Type II superconductors has been investigated theoretically. One finds two frequency branches; one "acoustic" branch derived earlier by Friedel, de Gennes and Matricon; and one "optical" branch whose energy gap at the wave number $K = 0$ corresponds to the cyclotron energy. The contribution of the vibrations of the vortex lines to the specific heat is also investigated.

C. ELECTRONIC TUNNELING STUDIES (W. J. Tomasch)

1. Type II Alloy Superconductors in a Magnetic Field

Current-voltage (I-V) characteristics of Alloy-AlO_x-Al diodes have been used to study tunneling between thick Pb-Tl films (2.5 - 7.5 μ , 4.3 - 13.5 at.% Tl) and

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

very thin Al films ($\approx 200 \text{ \AA}$) in fields H applied parallel to the diode plane. The observation of very high junction resistance for applied voltages greater than half the Al energy gap suggests that essentially all of the alloy surface retains a nonzero energy gap to at least twice the bulk thermodynamic critical field H_C . A form of sharp I-V structure persists to $H_C < H < 2H_C$, and has been tentatively interpreted in terms of a field-induced distribution of energy gaps in the alloy. The minimum energy gap, which may correspond to the energy gap at the alloy surface, has been evaluated numerically as a function of H for several compositions. Thick alloy films were employed since they exhibit bulk values of H_{C2} , the Ginzburg-Landau upper critical field. A spatially constant Al energy gap is believed to have been preserved through the use of very thin films. It is believed that the present results are typical of very smooth bulk specimens in which flux entry is delayed to beyond H_{C1} , the Ginzburg-Landau (G-L) lower critical field. Furthermore, these observations appear in harmony with a simple picture based on the Ginzburg-Landau-Abrikosov theory, taking the coherence length ξ as the spatial extent of an electron pair. Tunneling electrons presumably reflect conditions within roughly ξ of the surface. For $0 < H \leq H_{C1}$, the local energy gap ϵ should vary from its surface value ϵ_s to nearly the zero field value ϵ_1 ($H = 0$) over a distance ξ . This leads to a distribution of energy gaps $\epsilon_s \leq \epsilon_j \leq \epsilon_1$ ($H = 0$) which is consistent with the present observations if $\epsilon_s = \epsilon_m$. For $H > H_{C1}$, the presence of flux vortices should produce changes due to the spatial modulation of the energy gap. Absence of an abrupt change in ϵ_m at H_{C1} is probably attributable to delayed flux-entry phenomena which are known to exist in such films. The qualitative change in ϵ_m for $H_{C1} < H < H_C$ may signal vortex entry. Alternatively, it may reflect metastability characteristic of the internal flux density being less than the equilibrium value. Whether the high field limit for observing ϵ_m is determined by the Al films used or by more fundamental considerations (perhaps vortex modulation of the energy gap) remains an open issue pending further investigation. Eventually, as tunneling from vortex cores becomes more important, ϵ_m should tend to zero.

As reported earlier, tunneling data obtained in the presence of an applied transverse magnetic field allow determination of the G-L upper critical field, while zero field tunneling data yield a knowledge of the thermodynamic critical field. From these, one can calculate the G-L coupling constant κ which plays a central role in the theory. The composition and temperature dependence of κ has been studied over the ranges 4.3-13.5 atom % Tl and 1.1-4.2°K. After making allowances for the somewhat higher resistivities of the thick film specimens utilized, good agreement between the tunneling and bulk magnetization results was obtained as a function of composition. Furthermore, the temperature dependence of κ was found to be in good accord with the predictions of Gor'kov.

Although pure Pb does not have a κ value sufficiently large to be classified as a Type II superconductor, its value is large enough so that the sheath phase is stable for fields somewhat greater than the thermodynamic critical field. Tunneling data obtained in a longitudinal applied magnetic field have been used to obtain the effective κ of Pb utilizing the theory of Saint-James and de Gennes. The value obtained at 4.2°K agrees well with the value obtained by extrapolating the alloy data to zero concentration. This result also agrees with recent microwave impedance studies.

2. Pairing Interaction in Pb-Tl Alloy Superconductors

Recently developed first and second derivative capabilities have been employed to study the effect of alloying on the tunneling density of states (first derivative data), utilizing diodes quite similar to those described above. With the Al superconducting (1.1°K) such diodes yield current-voltage (I-V) characteristics which are indicative of a single, sharply defined alloy energy gap. With the Al normal ($1.4\text{--}1.5^{\circ}\text{K}$), the tunneling density of states data are fitted quite well by the Bardeen-Cooper-Schrieffer (BCS) density of states function (with thermal smearing) for voltages at least as great as twice the energy gap. At higher voltages ($\sim 4\text{--}6\text{ meV}$), departures from the BCS function are encountered similar to those seen in pure Pb, and are believed to be associated with structure in the phonon spectrum. To investigate this point, alloy second derivative data were compared with those for pure Pb, all observations having been made with the Al superconducting (1.1°K). The comparison is quite good, suggesting that alloying has very little effect on the phonon spectrum. In addition to tunneling measurements on films, electrical resistivity measurements have also been performed on foils (0 to 20 atom % Tl; 4.2, 77, and 300°K). The general form of these data suggest that the phonon and impurity contributions to the resistivity may be separated in a simple way, and that the lattice contribution at a given temperature is essentially independent of Tl concentration. In view of previously obtained evidence indicating a weak alloying effect on the electron density of states (at the Fermi level), it is believed that the present results are consistent with an electron-phonon interaction which is virtually independent of Tl concentration over the range investigated.

3. New Tunneling Effects in Thick Pb Films

It has been known for some time that Pb films thicker than a few thousand angstroms appear to exhibit two distinct energy gaps. Recent second derivative measurements have disclosed additional structure (not previously reported) near the energy gap region of the I-V characteristic. Some of the observed effects appear to be sensitive to specimen history. An exploratory investigation of these general phenomena has been recently initiated.

D. CALORIMETRIC AND MAGNETIC STUDIES OF TYPE II SUPERCONDUCTORS (L. J. Barnes)

Calorimetric studies which have revealed near ideal Type II superconducting behavior in a V-5 atom % Ta alloy ($T_c = 4.3^{\circ}\text{K}$, $\chi = 7$) have been continued. Additional specific heat data has been obtained at magnetic fields up to 8 kG (earlier work extended to 4 kG) and also at a low field of 120 G. The additional high field results along with the earlier work exhibit bulk second order transitions at the upper critical fields (H_{c2}) and in general are in fair agreement with the GLAG theory of Type II superconductivity. In a field of 120 G an anomalous increase in the specific heat was observed at a temperature corresponding closely to the expected lower critical field (H_{c1}) transition in addition to the usual well defined second order transition at H_{c2} . Specimen shape precluded determination of the transition order.

The calorimetric investigation has been broadened by studying the magnetocaloric effects in the same specimen mentioned above. Essentially the

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

temperature variations which occur on varying the magnetic field applied to the thermally isolated specimen were measured. Magnetic field variations produce near reversible isentropic temperature variations when the sample is in the mixed state at temperatures $T > 3^{\circ}\text{K}$. The measurements were in good agreement with the entropy-temperature-magnetic field diagram determined calorimetrically. However, at $T < 1.7^{\circ}\text{K}$ the magnetic field induced temperature changes are irreversible and become progressively more so at lower temperatures. At these lower temperatures continuous magnetic field increase produces discrete and rapid temperature increases which are presumably associated with discontinuous changes of the flux within the sample (so called flux jumping). At all temperatures the magnetocaloric effects, as expected, are confined to the mixed state ($H_{C1} < H < H_{C2}$). This work further confirms the general thermodynamic characterization of Type II superconductors by the GLAG theory. This investigation is being continued but a preliminary report has been presented. (R. R. Hake and L. J. Barnes, "Magnetocaloric Effects in a High-Field Superconductor," Proceedings of the IX International Conference on Low Temperature Physics, 513 (1965). (Paper presented at conference, August 31 to September 4, 1964).

In conjunction with the calorimetric studies measurements of the magnetic moment of various well-annealed and as-cast V-5 atom % Ta alloy specimens have been made. The well-annealed specimens exhibit near ideal Type II magnetization characteristics with a relatively small amount of irreversibility which is in accord with the calorimetric studies. The unannealed specimens reveal irreversible magnetic behavior as expected.

Detailed magnetization measurements have also been carried out on homogeneous well-annealed Pb-Tl alloys (up to 3% Tl). These dilute alloys were chosen in order to study the interesting region about the transition between Type I and Type II superconducting behavior. The transition can be swept through by varying the alloy composition or by varying the temperature of a particular alloy. K. Maki (Physics 1, 21 (1964) has proposed that in this transition region superconductors will exhibit characteristics of both Type I and II; namely, there will be two critical fields (H_{C1} and H_{C2}) and at both critical fields the superconductor will undergo first order transitions. The measurements so far reveal good agreement with the GLAG theory but no evidence has been obtained as yet of the peculiar behavior predicted by Maki. This work is still in progress.

In order to study the dynamic magnetization characteristics of various superconductors a sensitive technique of measuring the derivative of the magnetic moment with respect to the external magnetic field has been used. This technique yields a continuous recording of the derivative vs magnetic field. The pertinent features of the magnetization such as initial flux penetration at H_{C1} and the super-normal transition at H_{C2} are considerably enhanced. In addition, such dynamic phenomena such as "flux jumping" are clearly exhibited. This method of studying the magnetization characteristics can be closely correlated with the magnetocaloric and other measurements that involve continuous magnetic field changes. Such work is in progress. An additional bonus of the sensitivity of the technique is that the existence of the superconducting sheath state is demonstrated quite clearly and various of its properties including the critical field H_{C3} can be measured. This new line of investigation will be perused further.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

The derivative technique is being used to measure the lower critical fields (H_{c1}) of a series of Ti + V alloys. It is hoped that this study will shed some light upon the paramagnetic effect in high field superconductors. There is good evidence that the small paramagnetism of the electronic state in the alloys causes quenching of superconductivity at magnetic fields lower than the theoretically predicted ones. The study of such an effect is of importance to the practical utilization of high field superconductors.

III. EVALUATION OF EFFORT DURING FISCAL YEAR 1965

This year's activities have been unusually productive both in theoretical and experimental results. The Ginzburg-Landau-Abrikosov-Gorkov theory continues to demonstrate its capability of describing quantitative new facets of the behavior of Type II superconductors. The successful use of this theory to describe details of the sheath phase and to describe the vibration modes of vortex lines is an indication of the general validity of the concept. The electronic tunneling studies in superconducting diodes have proven to be an unexpectedly productive approach to several aspects of superconductivity. Many details of the influence of magnetic fields on the energy gap are clearly demonstrated as are the effects of alloy additions. Improved measurement techniques are also showing the influence of phonon-electron interactions and hitherto unrevealed details of the electronic structure. This subject will be actively pursued. Continued refinements of the more traditional studies of electron specific heats of superconductors are enhancing the sensitivity of the method and producing significant new information. The value of this work in developing high field superconductors continues to be very great, but now the method may be used to study other aspects of superconductivity.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Program: Advanced Development Program	
Project: Radiation Damage in Crystalline Solids	
Project Engineer: A. Sosin	
Reporting Period: Fiscal Year 1965	AEC Category: 05-03-03-01
General Order: 9305	Subaccount: 5404

Principal Scientists: A. Anderman, W. Bauer, F. H. Eisen, K. Garr,
D. W. Keefer, H. Neely, K. Thommen, A. Sosin.

I. PROJECT OBJECTIVES

The materials requirements of modern technology demand an increased understanding of the effects of lattice defects on the physical properties of crystalline solids and on their behavior when subjected to various kinds of radiation. Studies of radiation damage in solids not only directly yield information on the effects of radiation but also provide a means of investigating defects in solids. It is the objective of this project to study the production of defects in crystalline solids by radiation and to attain an understanding of their structure, their interactions with each other and with impurities and dislocations, their effect on the physical properties of solids, and their recovery kinetics. An understanding of these points should assist in the understanding of lattice defects produced in solids by other means and help make possible the development of materials of superior properties as the ability to better control their defect structure is developed. The materials chosen for study are metals, metal alloys, and semiconductors.

II. MAJOR ACCOMPLISHMENTS IN FISCAL YEAR 1965

A. TECHNICAL ACCOMPLISHMENTS

1. Recovery of Electron-Irradiated Aluminum and Aluminum Alloys (K. R. Garr and A. Sosin)

The experimental work on the pure aluminum and the aluminum alloys: Al - 0.060 atom % Mg, Al - 0.085 atom % Ga, and Al - 0.095 atom % Ag has been completed during the past year, and a paper is being written.

Analysis of the data for the pure aluminum indicates that there are two stages in the Stage III region (170 to 300°K). The first is a small substage centered at 190°K; the second, the main stage, is centered between 235 and 245°K depending on the purity and the dose. The first substage is sensitive to the residual impurity content (as determined by the value of the residual resistivity ρ_0) becoming smaller in magnitude as the purity is increased (lower value of ρ_0).

The residual impurities in the nominally pure aluminum also produce a large variation in the measured activation energy of the main stage of recovery in the Stage III region. The higher the purity of the material, the lower the measured activation energy, the values being 0.45 ev to 0.59 ev for the materials used in the study.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

The recovery in the Stage II region (65 to 170°K) for the pure Al shows substructure in the regions 68 to 88°K, and 120 to 140°K. The first of these regions (68 to 88°K) has two small stages centered at about 74 and 82°K. The latter stages appear to be sensitive to the residual impurities, decreasing in magnitude as the purity is increased. The first substage does not, however, show this same tendency and is believed to be an intrinsic recovery stage, that is, a stage due to some basic recovery process.

All of the alloys investigated in this study show a suppression of the recovery in the Stage I region (0 to 65°K) normally observed in pure aluminum, the amount of the suppression being 20 to 27% of the recovery observed in the pure aluminum specimen.

In the Stage II region (65 to 170°K), the recovery of the alloy depends on the particular solute added. For the Al - 0.060 atom % Mg alloy, the following results were obtained. Two recovery stages were observed in the Stage II region. The first is centered at 81°K, the second, at 127°K. Both stages are governed by a complex kinetic law; however, both appear to be uniquely activated stages.

The Al - 0.085 at % Ga sample exhibited a doublet (that is, two partially overlapping stages) stage, centered at about 111°K. This stage is governed by a complex kinetic law and does not seem to be uniquely activated.

The Al - 0.095 atom % Ag sample did not show any distinct recovery stages in the Stage II region. The addition of the Ag to the Al appears to have also suppressed the small amount of recovery noted in pure Al between 65 and 85°K. The recovery between 120 and 140°K observed in the pure Al was, however, also present in the Al-Ag alloy.

Stage III (170 to 300°K) recovery in the alloys is complex. The small stage centered at 190°K in the pure Al is more pronounced in the alloys and appears to cover a larger temperature range. There is more substructure in the Stage III recovery of the alloys compared to the pure material. All samples, except Al-Ag, show essentially complete recovery by 300°K)

The recovery of the Al-Ag sample was considerably different from that of the other alloys. During recovery of Stage III in the earlier irradiations, Al-Ag showed a decrease in resistivity followed by an increase and another decrease. This behavior persisted for several investigations of the recovery and then ceased. What followed was what appeared to be normal, but not complete recovery. This behavior can be explained on the basis of a clustering of Ag atoms which, after several irradiations and anneals, saturates and has little effect on the recovery.

The effective frequency factor, ν , for Stages II and III in the alloys (Al-Mg and Al-Ga) is about 10^{16} to 10^{17} sec⁻¹. These high values can be explained by a ratio of the atomic vibrational frequency (about 10^{12} to 10^{13} sec⁻¹) to the trapping (impurity) concentration. The effective frequency factor for the pure Al samples in Stage III varied concomitantly with the activation energy; the lower the activation energy (purer the material) the lower the effective frequency factor.

The results of the analysis are summed up in the following table. It is believed that the present data, particularly for the pure Al, give strong support to the two interstitial model.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Sample	Temp. of Stage (°K)	Iso-thermal Temp. (°K)	Reaction Constant	Activation Energy (E_a) (ev)	Temp. Range of E_a (°K)	Effective Frequency Factor ν
Al-0.06 _o atom % Mg	81	79	1.6 ₀	0.22 ± 0.03	76 to 86.5	3 × 10 ⁷
		80	1.6 ₀			
	127	126	1.6 ₀	0.33 ± 0.03	115.5 to 135	1 × 10 ¹⁶
		127.5	1.9 ₀			
Al-0.08 ₅ atom % Ga	235	230	2.3 ₀	0.63 ± 0.03	220 to 260	3 × 10 ¹⁷
	111	107.5	1.6 ₀	-	-	-
		109	1.4 ₅			
Al-0.09 ₅ atom % Ag	210	230	2.3 ₀	0.61 ± 0.03	230 to 260	4 × 10 ¹⁶
	-	230	-	-	-	-
Al pure	190	-	-	0.57 ± 0.03	180 to 200	-
$\rho_o = 1.42 \times 10^{-8} \Omega \text{cm}$	240	230	2.07	0.59 ± 0.03	225 to 260	6 × 10 ¹⁵
Al pure						
$\rho_o = 6.80 \times 10^{-9} \Omega \text{cm}$	234	-	-	0.53 ± 0.03	220 to 255	2 × 10 ¹⁴
$\rho_o = 6.58 \times 10^{-9} \Omega \text{cm}$	233	-	-	0.55 ± 0.03	220 to 255	2 × 10 ¹⁵
$\rho_o = 1.89 \times 10^{-9} \Omega \text{cm}$	248	-	-	0.46 ± 0.03	220 to 255	2 × 10 ¹²

2. Modulus Changes at Low Temperature — Dislocation Pinning in Pure Cu, Al, and Au (D. W. Keefer, A. Sosin, and J. C. Robinson)

A paper on the studies of dislocation pinning at low temperatures in electron-irradiated Cu has been accepted for publication by Acta Metallurgica. The conclusions reached in this paper are as follows.

1) The observed modulus stages, numbered I, III, and IV, result from the migration to dislocations of (possibly) crowdions, split interstitials, and vacancies, respectively. Although the presence of crowdions in Cu is not definitely established, a 2-interstitial model is clearly favored in explaining the results.

2) All three pinning stages appear to result from stress-induced diffusion of point defects to dislocations. The expected (time)^{2/3} dependence for this process was observed in both Stages I and III. The same time dependence has been observed for Stage IV by other investigators.

3) The measured activation energy for Stage III was 0.6 ev; that for Stage IV was about 1 ev. These energies agree with those found by other workers and are generally felt to be the energies of migration of interstitials and vacancies, respectively, in Cu.

4) Defects arriving at dislocations during irradiation and during subsequence annealing above 20°K appear to be tightly bound to the dislocations. No evidence of significant defect clustering, via defect migration down the dislocations, was observed upon annealing up to temperatures of around 400°K.

5) No evidence of vacancy-interstitial interactions on dislocations was observed, i. e., apparently vacancies arriving at dislocations in Stage IV do not often annihilate interstitials already there. We feel this and (4) above

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

are important in the general consideration of point defect - dislocation interactions and are presently exploring the possible implications of these results.

Several detailed isochronal measurements following electron irradiation at 20°K and following quenching from 450°C have been carried out on Al. In the case of irradiation, some difficulties have been encountered. It has been found that if an as-machined modulus sample is annealed in the cryostat and subsequently irradiated, little or no change in modulus is observed. Further, isochronal annealing results have been inconsistent. These difficulties have in part been traced to the dislocation structure of Al. A tentative explanation is as follows: From electron microscope work done elsewhere, the dislocations in annealed Al appear almost exclusively in subgrain boundaries. These dislocations may not be highly mobile, and therefore might not contribute significantly to the modulus defect. It has been found that if a sample is lightly cold-worked at room temperature prior to irradiation, definite pinning effects are observed upon subsequent low temperature irradiation. Possibly, cold-working increases the dislocation density within subgrains sufficiently so that modulus effects are observable.

Isochronal measurements carried out subsequent to irradiation of a cold-worked aluminum sample yield results which are not completely consistent with those obtained previously. Modulus Stage I, between 35 and 50°K, is still observed, followed by the usual region of depinning up to about 150°K. Modulus pinning Stage III, observed in previous work between about 150 and 240°K, is not now present, however. Instead, depinning continues up to about 240°K, at which point pinning Stage IV begins and continues to 360°K, and is complete by around 470°K, although the final modulus value is not steady. Dislocations are apparently annealing out at these temperatures. The reason for the absence of pinning Stage II is not yet clear, and detailed work on this point is intended.

It is interesting to point out that isochronal annealing results in the modulus measurements bear little relation to the results obtained by tensile testing methods. This appears to fortify our interpretation of irradiation-induced yield stress increases as being due to defects dispersed throughout the aluminum lattice.

Isochronal measurements following quenching of Al have yielded a well-defined pinning stage between 240 and 360°K, followed by depinning up to about 500°K. We therefore attribute modulus Stage IV in Al to vacancies; this assignment agrees with those made previously for Cu and Au.

Damage-rate modulus measurements of Au at a number of electron energies have been made. Although this work is not yet complete, the preliminary results indicate that near the threshold energy the damage rate in Au is somewhat less sensitive to electron energy than is the rate in Cu. This suggests that pinning which occurs during low-energy electron irradiation of Au is the result of long-range focusing events; these events are less sensitive to electron energy than are short range displacement processes. This conclusion is in agreement with that reached previously in this laboratory on the basis of resistivity measurements.

A paper on the modulus work in gold is presently being prepared for submission to a journal.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

3. Point-Defect Relaxations in Irradiated Metals (D. W. Keefer and R. S. Vitt)

A number of experiments have now been performed on one Cu internal friction specimen. The results of these experiments can be best described in two categories, those following very long irradiations, and those following more modest integrated fluxes. Referring to the former, two such irradiations, to defect concentrations of about 7×10^{15} , have been performed. Subsequent internal friction measurements at about 1 cycle/sec have been made between about 80 and 380°K. These data show a sharp rise in the internal friction (Q^{-1}) near 120°K (about 5×10^{-4} above background), after which Q^{-1} is nearly constant to 300°K. Above 300°K, the friction follows the normal background Q^{-1} , rising steeply with temperature to 380°K. The internal friction losses observed between 120 and 300°K are not clearly defined, appearing to be the result of several relaxation processes. Clearly, no peak is observed which may be attributed to the rotation or migration of the dumbbell, or split, interstitial. We presently feel that the relaxation strength of the dumbbell (if it exists at all) is smaller than has been predicted theoretically, and that there is no chance of observing its relaxation with the defect concentrations which are obtainable by our techniques.

The results obtained following low-integrated electron fluxes (defect concentration about 3×10^{-6}) appear to hold more promise. In these experiments, a definite internal friction peak is observed near 230°K. This peak appears to represent a true relaxation process, although this has not been definitely established by variation in the measuring frequency. The peak can be made to grow by annealing near 250°K, and is subsequently reduced by annealing at 300°K. We suggest that the peak is due to a point defect-dislocation interaction of some type and that the growth of the peak indicates defect migration to dislocations. This study will be continued in some detail since it appears that such measurements can yield valuable information concerning the exact nature of point defect-dislocation interactions.

4. Electron Irradiation Hardening of Metals (O. Buck, D. W. Keefer, and J. C. Robinson)

The equipment for this experiment is virtually complete, and very little in the way of testing remains to be done. Preliminary irradiations of aluminum will be performed shortly.

5. The Resistivity Recovery of Gold with Varying Residual Impurity Concentrations (W. Bauer)

Nominally, 99.99% (3×10^{-8} ohm-cm, source unknown), 99.99% (4.33×10^{-9} ohm-cm, Sigmund Cohn), 99.9999% (3.2×10^{-9} ohm-cm, Cominco) pure gold was irradiated and the isochronal recovery spectrum up to room temperature was investigated. Below Stage III, the recovery of the 5-9's and 6-9's gold is approximately the same, and the recovery of the 4-9's gold is suppressed by approximately a factor of 2 between 30 and 120°K. During the first part of Stage III the impure gold recovers faster, even to the extent that the recovery of 5-9's and 6-9's gold differ by a few percent. This indicates the importance of doing a definitive study on the effects of doping and further indicates that a thorough investigation of recovery in nominally pure 99.9999% gold is still

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

required. Studies of doped copper, currently undertaken by A. Sosin and H. H. Neely indeed emphasize this point. Since it is felt that no free migration of defects occurs below Stage III in gold, the suppression of recovery by impurities of low concentration indicates either the existence of long-range interaction between impurities and defects, or that a relatively large concentration of pairs of vacancies and interstitial-type impurities are produced dynamically during the damage process. The latter should reflect itself in an increased damage production as compared to the pure samples.

6. Irradiation of Oriented Gold Single Crystals (W. Bauer and A. Sosin)

Preliminary irradiations of gold single crystals with orientations such that the $\langle 100 \rangle$ or $\langle 110 \rangle$ directions were parallel to the electron beam have been completed. The thickness of the samples used ranged from 0.3 to 0.8×10^{-3} in. It is still too early to draw any definite conclusions about the directional dependence of the damage production.

However, the importance of the thickness of the samples, as has been pointed out by A. Sosin and K. Garr, is clearly indicated by our results. That is to say, the damage production in the thicker samples is within 30% of the damage production of polycrystalline gold. The method used for reducing the thickness of the samples has been refined to the extent that a reasonably reliable (20 to 30%) determination of the geometrical factor, used for the calculation of the resistivity increase, can be made. A full-scale effort will be launched to complete this work in the next year.

7. Evaluation and Interpretation of Threshold Displacement Energy Measurements (W. Bauer and A. Sosin)

This work has been completed during the report period and has been submitted for publication. A short summary is given here.

In this article we have attempted to analyze the difficulties which beset a determination of threshold energies, and to evaluate the various values of T_0 currently proposed by various investigators. The difficulties — lattice imperfections, thickness, and experimental sensitivity — are sufficiently severe to give rise to a discrepancy in proposed values of T_0 from 9 to 30 ev. We conclude that the threshold energy measurements of Bauer and Sosin are still the most complete available, and that the value of $T_0 = 16 - 19$ ev deduced from these measurements are the best current values. This conclusion is based on the following.

- 1) Experimental data from the purest and thinnest samples of copper as well as data from selectively doped copper.
- 2) Order of magnitude calculations supporting the existence of indirect displacements (focussing-impurity interaction) in the subthreshold range.

The assignment of $T_0 = 16$ to 19 ev is not presented as completely established. Further experiments are in order. In this regard we would suggest that a final assignment for the value of T_0 will require investigations which not only attempt to establish this quantity directly, but also quantitatively account for the observed displacement cross-section over a reasonable range in energies about T_0 .

8. Continuous Resistivity Recovery Spectrum of Irradiated Metals from 10 to 65°K (A. Sosin, W. Bauer, and H. H. Neely)

Experimenta have been performed to explore the fine details of the recovery spectrum of copper in Stage I using a continuous annealing technique. The mode of operation is one in which the difference of the resistivity of an irradiated and an unirradiated sample are measured simultaneously as a function of annealing temperature. Although our experimental conditions are close to ideal, we have been unable to achieve the required sensitivity in copper. This is due to the limitations imposed by resistivity measurements at temperature — deviation from Mathiessen's rule. It is intended in the near future to investigate other metals such as aluminum or platinum which may be more favorable from this viewpoint.

9. Damage Production in Platinum near 7°K by Electron Irradiation (W. Bauer)

The experimental part of this work has been completed during this report period and an abstract has been submitted for the APS meeting in Honolulu. The contents of the abstract are given here.

The increase in electrical resistivity per unit flux, $d\rho/d\phi$, of 99.000% pure 0.002-in.-thick platinum foils was measured as a function of incident electron energy from 1.3 to 2.1 Mev. Reasonable agreement with the theoretical displacement cross section, using a unit probability of displacement, is achieved with an effective threshold of 36 ev and a value of $\rho_f = 6.0 \times 10^{-4}$ ohm-cm/unit fractional concentration of Frenkel pairs. In gold the best agreement between the theoretical and experimental cross section was only achieved for an unusually low value of ρ_f . The ratio of $d\rho/d\phi$ for platinum to $d\rho/d\phi$ for gold is a constant (≈ 6) for energies above approximately 1.7 Mev, as expected for elements of similar atomic number, but decreases to zero near the threshold. These results are interpreted to indicate that (1) directional effects in the displacement process are more important in gold than in platinum, and (2) indirect displacement processes due to focussons are more important in gold near threshold than in platinum.

10. Impurity-Delayed Diffusion (A. Sosin and W. Bauer)

The problem of diffusion to inexhaustible sinks of an initially uniform concentration of defects with concurrent trapping and release of defects by impurities has been solved. The solution is applied to the case of spherical sinks representing grain-boundaries in a real metal. Numerical solutions have been worked out on an analog computer for a range of values of the different parameters — concentration of defects, concentration of impurities, diffusion lifetime, and defect impurity binding energy. The qualitative features of the calculations are in agreement with earlier work on "impurity-delayed chemical rate theory" (Damask and Dienes).

11. Stage IV Resistivity Recovery of Platinum after Electron Irradiation (W. Bauer and A. Sosin)

The experimental part of this work has been completed during this report period. A publication of this work in conjunction with the "impurity delayed diffusion" work, above, is being prepared. The salient features of this work are as follows.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

1) The Stage IV recovery in platinum is due to the diffusion of single vacancies.

2) Most of the recovery is adequately described by impurity-delayed diffusion.

3) This same mechanism describes the Stage IV recovery of deformed nickel.

4) Vacancy - Impurity-bind energies of the order of 0.7 are required for numerical agreement between theory and experiment. This implies that there may exist a strong interaction between selected impurities and vacancies in platinum and nickel. Preliminary experiments confirm this.

12. Directional Effects of Displacements in Iron (A. Sosin and W. Bauer)

A preliminary irradiation of a 0.0008-in.-diameter iron whisker has been performed. In large part, the experiment has proved the feasibility of this type of irradiation, especially the difficult crystallographic orientation. From multiple electron scattering considerations, the sample was somewhat too large for unambiguous interpretation of the data. Much of this report time has been spent in evaluating the importance of multiple electron scattering in order to establish a meaningful criteria for the whisker irradiations, and for that matter, other metals. It is hoped that a full-scale effort can be launched to complete this work in the next year.

13. Indium Antimonide Irradiations (F. Eisen)

The dependence of damage production rate for Stages III to V on sample orientation has been examined in a manner similar to that previously reported for Stages I and II, using a pair of samples having opposite (111) faces exposed to the electron beam. For Stage IV, the results are similar to those for Stage II in that the damage rate is higher with the (111) face exposed to the beam than with the (III) face exposed, and the relative difference increases as the electron energy is decreased. For Stage III, the damage rate was also higher for the (111) face, but the relative rates for the two faces were almost independent of the energy of the bombarding electrons. For Stage V, the damage rate at 500 kev is higher with the (III) face exposed to the electron beam. At 550 kev, the rates for the two faces are almost the same, and at higher energies the damage rate with the (111) face exposed to the beam becomes slightly higher than with the (III) face exposed. If these data are interpreted in the same manner as for Stages I and II, one would conclude that Stages III and IV are both due to the displacement of indium atoms. The reversal for Stage V may indicate that both indium and antimony displacements occur in this stage. This would not be surprising since, as mentioned later, the kinetics data indicate that several different defects with possible different activation energies for recovery are present in this stage. If III and IV are both due to indium displacements, one wonders why analogous antimony stages have not been observed. It could be that antimony displacements are for some reason unstable or that they are present and account for some of the donor type defects which are observed in p-type samples.

The observed variation of recovery temperature, T_c , with initial electron concentration, n_0 , for Stages III and IV in electron irradiated n-type InSb has been fitted assuming the same type of charge state dependence of the recovery as has been found to apply for Stage II. An appreciable change in T_c is only

found when n_0 is large compared with the intrinsic carrier concentration in the sample at T_C . For Stage V, this intrinsic concentration is larger than the highest n_0 for which recovery measurements have been made so that no shift in T_C with electron concentration would have been expected. In going to p-type samples with relatively low values of initial hole concentration, ρ_0 , one would expect little if any change in T_C for Stages III and IV, and no change for Stage V. In this way, it has been possible to identify these stages in p-type samples. T_C for Stage III has been observed to decrease from 145°K to 122°K as ρ_0 changed from $1.6 \times 10^{14} \text{ cm}^{-3}$ to $1.7 \times 10^{16} \text{ cm}^{-3}$ and T_C for Stage IV is observed over this range of ρ_0 , as expected.

The recovery kinetics for Stages III and IV have been observed to be independent of the energy of the bombarding electron between 1.0 and 1.95 Mev. The recovery data for Stage III can generally be divided into two exponentials in both n-type and p-type samples but in p-type samples it is necessary to use three exponentials to fit the data. These stages then may be due to the annihilation of two or three distinct close pair configurations with interstitial-vacancy separations greater than for Stage II.

The recovery kinetics for Stage V is a function of energy. If an attempt is made to fit the kinetics to a sum of exponentials, then the number of exponentials required increases as the bombarding electron increases. The activation energy for recovery, as determined from a Meechan-Brinkman analysis of the isochronal and isothermal recovery data also increases slightly as the electron energy increases. There is some dependence of the kinetics, after bombardment at a particular energy, on the temperature at which the isothermal annealing is carried out. No concentration dependence of the recovery temperature is observed at any point of the isochronal recovery curves. These data suggest that the recovery observed in Stage V is again due to the annihilation of interstitial-vacancy greater than for the pairs which annihilate in Stages III and IV. The changes in kinetics with bombarding energy are then due to the formation of new-pair configurations as the bombarding energy is increased. The changes of kinetics with annealing temperature can be interpreted as due to some interchange of the mobile defect between different positions before the annihilation of the interstitial-vacancy pair, which would be expected to depend on the annealing temperature. It is surprising that no indication of long-range migration of defects is found. This holds true for recovery observed in a still higher temperature stage (Stage VI centered at about 410°K). Here, as in the case of Stage V, there seems to be no dependence of the recovery temperature on defect concentration.

14. Recovery of Electron-Irradiated Copper Alloys (H. H. Neely and A. Sosin)

Cu + 0.03, 0.1, 0.3 atom % Au samples were irradiated at two different electron energies (0.7 and 1.1 Mev) to the same resistivity increase, to explore any effect on the sizes of the postirradiation recovery stages. The recovery was performed isochronally in each case. The results of the two investigations were closely identical with each other, but not with previous experiments at 1.1 Mev in which the integrated electron flux was greater by a factor of five. The major difference was that Stage IIc was enhanced in the present experiments by about a factor of two (or a total of 5, 9, and 28% respectively). Correspondingly, the resistivity was observed to recovery close to 100% in each sample following Stage III annealing here, whereas full recovery was not effected previously.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

The recovery in these alloys can be understood within the framework of simple impurity-trapping models with the possible exception of Stage IIc. The data indicates that the magnitude of this stage is essentially linearly dependent on trap density. However, the number of traps that appear to contribute to this stage is significantly lower than the impurity content of the samples.

15. Theoretical (A. Anderman)

The effort relating to the theoretical study of radiation effects in metals has been focused in these three major areas:

- 1) theoretical investigation of interatomic potentials
- 2) development and refinement of computer programs
- 3) computer calculation of selected events.

Several interatomic potentials have been developed. One is a composite potential formulated to study events in Cu. For $r < 2.0 \text{ \AA}$, the potential consists of a Born-Mayer function of the form:

$$\phi(r) = 0.0515 \exp [-14.03(r-2.547)/2.547] .$$

For $r < 2.0 \text{ \AA}$, the potential is represented by a Morse function whose range is assumed to extend to fourth nearest neighbor interaction. The Morse potential used is written as:

$$\phi(r) = 16.382 [45.56 \exp (-2.727r) - 2 \exp (-1.364r)]$$

The values of the Born-Mayer constants were chosen so as to cause the Born-Mayer function to merge smoothly with the Morse potential at $r = 2.0 \text{ \AA}$. The values obtained are close to those most used by the Brookhaven group in their corresponding study of Cu using only the Born-Mayer potential.

Potential constants have also been evaluated for a pure Morse potential in Cu and Al. Writing the Morse potential as

$$\phi(r) \text{ eV} = D (\beta e^{-2ar} - 2e^{-ar})$$

the results are:

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

Cutoff Distance	Morse Constants for Cu			Morse Constants for Al		
	α	β	D(ev)	α	β	D(ev)
1st Nearest Neighbor	1.43649	38.8204	22.5300	1.21012	31.4070	17.8378
2nd Nearest Neighbor	1.40465	40.0840	19.2627	1.17984	32.4580	15.0513
3rd Nearest Neighbor	1.36752	44.1413	16.5965	1.14008	35.9395	12.5610
4th Nearest Neighbor	1.36361	45.5600	16.3819	1.13481	37.2190	12.3115

As one check on the validity of the Morse potentials listed above, elastic constants were calculated from them using the Born equations. The results are summarized as follows.

Cutoff Distance	Elastic Constants Derived from Morse Potential for Cu (units: 10^{12} dynes/cm ²)		Elastic Constants Derived from Morse Potential for Al (units: 10^{12} dynes/cm ²)	
	C_{11}	C_{12}	C_{11}	C_{12}
1st Nearest Neighbor	2.1304	1.0652	1.323	0.661
2nd Nearest Neighbor	1.8564	1.2022	1.153	0.746
3rd Nearest Neighbor	1.9311	1.1648	1.215	0.715
4th Nearest Neighbor	1.9451	1.1579	1.228	0.709
Experimental Values*	1.7620	1.2494	1.230	0.708

*G. A. Alers and J. R. Neighbors, Revs. Mod. phys. 31, 675 (1959)

It is interesting to note that, in Cu, best agreement with experiment occurs for the 2nd nearest neighbor potential, while in Al the 4th nearest neighbor potential gives best agreement.

As a result of the investigation of the Morse potential, studies are now being conducted with the intent of developing an interatomic potential for metals from relatively first principles. Although this effort is still in its beginning stages, the work appears promising.

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

In addition to pure metal interactomic potentials, work has also been pursued towards developing a potential for the interaction between Cu and an impurity atom. The first impurity chosen for study is Xe. This selection was made for two reasons. The first is that firm experimental data exists concerning the nature of the Xe-Xe potential. Secondly, the behavior of Xe in a metal is of interest to a correlative effort being performed under Project 7652-4537 (Fission Effects in Metal Fuels) concerning a theoretical study of bubble formation as related to fuel swelling.

Several possible potentials for the Xe-Cu interaction were considered. The first approach consisted of obtaining modified Buckingham potentials for both the Xe-Xe and Cu-Cu interactions. This procedure was followed since Buckingham potentials are already available for the Xe-Xe interaction, [Intermolecular Potentials of A, Kr, and Xe; E. Whalley and W. G. Schneider, J. Chem. Phys., 23, 1644 (1955)] and, moreover, techniques have been developed for combining Buckingham potentials of unlike molecules [Forces between Unlike Molecules and the Properties of Gaseous Mixtures, E. A. Mason, H. Chem.. Phys. 23, 49 (1955)]

The constants for the Cu-Cu Buckingham potential were evaluated using the lattice spacing, sublimation energy and bulk modulus of Cu. Writing the modified Buckingham in the usual form

$$\alpha(r) = \frac{\epsilon}{(1-6/\alpha)} \left[\frac{6}{\alpha} \exp \left[\alpha \left(1 \frac{r}{r_0} \right) \right] - \left(\frac{r_0}{r} \right)^6 \right],$$

the constants for Cu, as a function of various assumed ranges of the potential in the lattice, were found to be as follows.

MODIFIED BUCKINGHAM POTENTIAL CONSTANTS FOR CU

Nearest Neighbor Beyond Which Interaction Assumed to be Negligible	α	$\epsilon(\text{ev})$	$r_0(\text{\AA})$
1st Nearest Neighbor	8.195	0.580	2.547
2nd Nearest Neighbor	8.343	0.485	2.624
3rd Nearest Neighbor	8.617	0.388	2.734
4th Nearest Neighbor	8.680	0.371	2.756

ATOMICS INTERNATIONAL
A Division of North American Aviation, Inc.

It turns out that all of the modified Buckingham Cu-Cu potentials derived are unsatisfactory in at least one respect, namely, that at small separations the $1/r^6$ term dominates the exponential term, thus giving rise to a quite unrealistic repulsive portion of the potential. However, the potentials do seem reasonable for values of $4 > 2.0 \text{ \AA}$. Within the bounds of this limitation, an initial study has been started of the energy of a substitutional Xe atom in a Cu lattice using a combined Xe-Cu Buckingham potential formed according to the combination rules suggested by Mason.

During the past year, some significant improvements have been made in the computer programs. Both speed and calculation and flexibility in application of the programs have been greatly enhanced. In addition, the program now provides for the introduction into the lattice of vacancies and impurity atoms as well as interstitial defects. All of these various defects may now be studied, either singly or in any combination desired.

Using the potentials and programs developed thus far, a number of calculations have been initiated. One interesting calculation concerns the defect lattice energy of a $\langle 100 \rangle$ split interstitial under the influence of a Born-Mayer potential. Approximately, the same energy was obtained in our calculation as was derived by the Brookhaven group. What is of interest is that our calculations were performed in a large crystallite without surface forces but whose surface atoms were held fixed. It turned out that those atoms which relaxed due to the presence of the defect were contained within a relatively small interior volume, and, hence, surface atoms did not affect the outcome of the calculation. The fact that our crystal model leads to the same result as the Brookhaven model employing rather elaborate surface forces suggests that such surface forces are not needed for this type of calculation.

B. PUBLICATIONS

1. D. Keefer and A. Sosin, "Electron Irradiation Hardening of Aluminum," *Acta Metallurgica* 12, 969 (1964)
2. D. Keefer and A. Sosin, "Thermal Depinning of Dislocations in Electron-Irradiated Copper," *Acta Metallurgica* 12, 1041 (1964)
3. W. Bauer and A. Sosin, "Threshold Displacement Energies and Sub-Threshold Displacements in Copper and Gold Near 10°K ," *Journal of Applied Physics* 35, 703 (1964)
4. W. Bauer and A. Sosin, "Point Defect Studies in Gold by Electron Irradiation at Low Temperatures. I - Threshold Displacement Energy and Displacement Cross Section," *Phys. Rev.* 135A, 521 (1964)
5. W. Bauer and A. Sosin, "Point Defect Studies in Gold by Electron Irradiation at Low Temperatures. II - The Resistivity Recovery Spectrum From 15°K to 240°K ," *Physical Rev.* 136 A255 (1964)
6. W. Bauer and A. Sosin, "Point Defect Studies in Gold by Electron Irradiation at Low Temperatures. III - The Resistivity Recovery from 240 to 340°K (Stage III)," *Physical Rev.* 136, A474 (1964)
7. J. C. Robinson, "Cryostat Design Permitting Localized Temperature Control from 20 to 870°K ," *Review of Scientific Instruments* 35, No. 8, 1017 (1964)
8. F. H. Eisen, "Orientation Dependence of Electron Radiation Damage in InSb," *Physical Rev.* 135, A1394 (1964)

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

9. F. H. Eisen, "Radiation Damage in Semiconducting III-V Compounds," Proceedings of the Symposium on Radiation Damage in Semiconductors, Royaumount, France
10. D. Deefer, J. C. Robinson, and A. Sosin, "Modulus Effects in Metals After Low Temperature Electron Irradiation," Part I. Cu," Acta Metallurgica, accepted for publication
11. A. Sosin and K. Garr, "Directional Effects in Electron Irradiated Cu Single Crystals," Physica Status Solidi 8, 481 (1965)
12. W. Bauer and A. Sosin, "Stage IV Recovery in Platinum," to be submitted
13. W. Bauer and A. Sosin, "Evaluation and Interpretation of Threshold Displacement Energy," to be submitted
14. A. Sosin and W. Bauer, "Impurity-Delayed Diffusion," to be submitted
15. W. Bauer, "Threshold Displacement Energy and Displacement Cross-Section of Platinum," in manuscript
16. A. Sosin and H. H. Neely, "Post-Irradiation Recovery in Irradiated Copper-Based Alloys - The Effect of Varying Gold Concentration," in manuscript
17. F. H. Eisen, "Recovery in Electron-Irradiated InSb I; Stage II," in manuscript
18. F. H. Eisen, "Recovery in Electron-Irradiated InSb II; Stages III, IV, and V," in manuscript
19. D. W. Keefer and J. C. Robinson, "Modulus Effects in Metals After Low Temperature Electron Irradiation, Part II - Gold," in manuscript
20. K. R. Garr and A. Sosin, "Recovery of Electron-Irradiated Aluminum and Aluminum Alloys," in manuscript
21. A. Sosin "On Kinetic Methods for the Determination of Activation Energies," to be submitted

III, EVALUATION OF EFFORT DURING FISCAL YEAR 1965

The work during fiscal year 1965 has seen some extensive probing into a number of areas investigated less thoroughly previously. We continue to give emphasis to dynamic irradiation effects as influenced experimentally by directional effects, to interatomic potential evaluation as playing the dominant role for defect theory, and to the role of impurities in the analysis of point defect characteristics. With respect to directional effects, a considerable effort has been made to define the experimental limitations inherent in experiments designed to explore displacement anisotropy. Only electron irradiation of highly thin, oriented single crystals can be expected to yield unambiguous data. The importance of establishing a reliable interatomic potential to be used as a basis for theoretical calculations has been made in previous reports. Our more recent successes in arriving at some physical basis for a choice is encouraging, even if tenuous. The importance of attaining a reasonable understanding of the role of impurities has also been made before. The extent of our effort here indicated our concern for this problem area.

The deepening exploration in the above areas has limited our activities in the study of radiation effects in bcc metals (and hcp metals, as well). We intend to mount a major effort here immediately. Until now, such an effort has been delayed by manpower consideration but more by facility considerations.

ATOMICS INTERNATIONAL

A Division of North American Aviation, Inc.

Mechanical effects, as altered by irradiation, have been examined on a broader front. The complexities in the effects, as demonstrated by the differing effects in the series of metals, copper, gold, and aluminum, must be attributed to the nature of dislocation characteristics. From one point of view, our observations are among the most important made with respect to dislocations in metals. By comparison, electron microscopy is unable to achieve the atomic resolution implicit in the modulus measurements. Tensile testing, similarly, involves stresses in a range above that required for the detailed atomic probing involved in modulus and internal friction measurements. Furthermore, our combination of modulus measurements and tensile testing does effect an examination of dislocation behavior at high and low stress levels.

The data describing recovery in electron irradiated InSb are now fairly complete, and a reasonable model can be constructed on the basis of these data. The most important missing element in a complete understanding of radiation damage in InSb, is a knowledge of the energy level structure of the defects. Some of this can be inferred from the damage production data obtained in doing the recovery work. These results indicate that the most important problem here is to understand how defects which are thought to be close interstitial-vacancy pairs can have only acceptor levels associated with them.