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QUARTERLY PROGRESS REPORT
JULY, AUGUST, SEPTEMBER, 1969
REACTOR FUELS AND MATERIALS
DEVELOPMENT PROGRAMS
FOR FUELS AND MATERIALS BRANCH
OF
USAEC DIVISION
OF REACTOR DEVELOPMENT AND TECHNOLOGY
November 1969

AEC RESEARCH & DEVELOPMENT REPORT

ROUTE TO	DATE	INITIALS	REMARKS

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Edited

By

W. F. Sheely

NOVEMBER 1969

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<u>Quarter</u>	<u>Year</u>	<u>Report Number</u>
	<u>1966</u>	
First		BNWL-CC-694
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	<u>1967</u>	
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Second		BNWL-473
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	<u>1968</u>	
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Second		BNWL-842
Third		BNWL-919
Fourth		BNWL-971
	<u>1969</u>	
First		BNWL-1058
Second		BNWL-1144

CONTENTS

SUMMARY	1.1
FAST REACTOR CERAMIC FUELS RESEARCH .	1.1
CREEP OF UNIRRADIATED MIXED OXIDE FUELS.	1.1
IN-REACTOR CREEP OF MIXED OXIDE FUELS	1.1
THERMAL DIFFUSIVITY OF URANIUM OXIDE.	1.1
SCANNING ELECTRON MICROSCOPY OF NUCLEAR FUEL MATERIALS.	1.2
THERMAL EXPANSION OF MIXED OXIDE	1.2
PROPERTIES OF MOLTEN FAST REACTOR OXIDE FUELS.	1.2
PHASESTUDIES	1.2
IRRADIATION BEHAVIOR OF FAST REACTOR FUEL MATERIALS. .	1.3
IN-REACTOR THERMAL PERFORMANCE OF $\text{UO}_2\text{-PuO}_2$ FUEL . . .	1.3
THERMAL FLUX IRRADIATIONS OF HYPOSTOICHIOMETRIC $\text{UO}_2\text{-PuO}_2$ FUEL	1.3
FUEL SWELLING AND ANALYTICAL MODELING .	1.3
MIGRATION BEHAVIOR OF $\text{UO}_2\text{-PuO}_2$ FUELS.	1.3
IRRADIATION DAMAGE TO REACTOR METALS.	1.4
IRRADIATION BEHAVIOR OF STRUCTURAL MATERIALS	1.4
Irradiation Facilities Operation	1.4
Microstructural Modifications to Stainless Steels and Nickel-Base Alloys.	1.4
Production of Void in Nickel.	1.4
Irradiation Effects on the Fracture of Heavy Section Pressure Vessel Steels.	1.5
Irradiation Effects in Zircaloy-2	1.5
FUEL SUBASSEMBLY MATERIALS	1.6
EBR II Irradiations	1.6
Damage Analysis	1.6
Biaxial Stress-Rupture Studies	1.6
Mechanical Testing of Fuel Pin Cladding	1.7
RELATED FAST REACTOR SUPPORTING STUDIES.	1.7
Acceptance Testing and Clad Characterization .	1.7
Tensile Studies	1.7

Uniaxial Creep Rupture Studies	1.8
FAST REACTOR DOSIMETRY AND DAMAGE ANALYSIS	1.8
EBR-II DOSIMETRY TEST.	1.8
FAST REACTOR NEUTRON SPECTRA AND FOIL ACTIVATION CROSS SECTION EVALUATION	1.9
DAMAGE FUNCTIONS AND DATA CORRELATION	1.9
COMPUTER SIMULATION OF RADIATION DAMAGE	1.9
INTERACTIONS OF CARBON IMPURITY ATOMS WITH A TILT- GRAINBOUNDARY	1.10
RADIATION EFFECTS ON ABSORBING MATERIALS FOR CONTROL RODS	1.10
NUCLEARGRAPHITE	1.11
F-1 THERMAL EXPANSION	1.11
COMPUTER CALCULATIONS OF GRAPHITE OXIDATION IN HTGR. .	1.11
COBALT-60 ADDITION TO THE GAMMA IRRADIATION FACILITY .	1.11
NONDESTRUCTIVE TESTING	1.11
EDDY CURRENT METHODS	1.11
THERMAL METHODS	1.12
ULTRASONIC METHODS	1.12
NEW TECHNIQUES	1.12
ATR WATER LOOP	1.12
FAST REACTOR CERAMIC FUELS RESEARCH	2.1
CREEP OF UNIRRADIATED MIXED OXIDE FUELS - O. D. Slagle.	2.1
IN-REACTOR CREEP OF MIXED OXIDE FUELS - J. A. Basmajian.	2.5
THERMAL DIFFUSIVITY OF URANIUM DIOXIDE - J. L. Bates .	2.7
SCANNING ELECTRON MICROSCOPY OF NUCLEAR FUEL MATERIALS J. L. Daniel.	2.18
THERMAL EXPANSION OF MIXED OXIDES - P. G. Pallmer .	2.20
PROPERTIES OF MOLTEN FAST-REACTOR OXIDE FUELS - R. P. Nelson, J. J. Rasmussen, and O. D. Slagle . . .	2.21
Viscosity	2.21
Surface Tension	2.21
Compressibility	2.23

PHASE STUDIES - C. E. McNeilly and R. E. Woodley . . .	2.23
IRRADIATION BEHAVIOR OF FAST REACTOR FUEL MATERIALS	
J. A. Christensen	3.1
IN-REACTOR THERMAL PERFORMANCE OF UO_2 - PuO_2 FUEL	
J. A. Christensen	3.1
Melting Heat Rating for UO_2 -25 wt% PuO_2 Fuels	
L. A. Lawrence and J. A. Christensen	3.1
The Effects of Xenon on Fuel-to-Cladding Gap	
Conductivity - G. R. Horn	3.14
Fabrication of Fuel Capsule	
	3.18
THERMAL FLUX IRRADIATIONS OF HYPOSTOICHIOMETRIC UO_2 - PuO_2	
J. E. Hanson, R. D. Leggett [†] , J. W. Weber, L. A. Pember, D. Christensen, E. D. Jenson, and D. W. Dragnich . . .	3.36
Postirradiation Examination of BNW-1-1, 1-2, 1-3, and 1-5	
	3.36
BNW-1-7	3.44
BNW-1-8	3.48
FUEL SWELLING AND ANALYTICAL MODELING	
K. R. Merckx, J. A. Christensen.	3.56
Swelling Measurements	
F. E. Panisko	3.56
Stochastic Modeling	
K. R. Merckx	3.56
MIGRATION BEHAVIOR OF UO_2 - PuO_2 FUELS	
T. D. Chikalla	3.59
Shielded Electron Microprobe Examination of Irradiated Fuel (Transport of Components in a Temperature Gradient) E. D. Jenson.	
	3.59
Migration Behavior in Mixed Oxides	
C. A. Hinman	3.66
IRRADIATION DAMAGE TO REACTOR METALS	
	4.1
IRRADIATION BEHAVIOR OF STRUCTURAL MATERIALS	
L. D. Blackburn	4.1
Irradiation Facilities Operation	
A. L. Ward	4.1
Microstructural Modifications to Stainless Steels and Nickel-Base Alloys - I. S. Levy.	
	4.3
Production of Voids in Nickel	
G. L. Kulcinski and B. Mastel.	4.7

Irradiation Effects on the Fracture of Heavy Section Pressure Vessel Steels	
C. W. Hunter and J. A. Williams	4.14
Irradiation Effects in Zircaloy-2	4.33
FUEL SUBASSEMBLY MATERIALS - T. T. Claudson.	4.38
EBR-II Irradiations - P. K. McDaniel	4.38
Damage Analysis - J. L. Straalsund, H. R. Brager, and R. L. Fish.	4.39
Biaxial Stress-Rupture Studies - R. W. Barker	4.53
Mechanical Testing of Fuel Pin Cladding R. L. Fish	4.62
RELATED FAST REACTOR SUPPORTING STUDIES	4.68
Acceptance Testing and Clad Characterization M. M. Paxton	4.68
Tensile Studies - A. L. Ward	4.86
Uniaxial Creep-Rupture Studies - A. J. Lovell, J. F. Bates	4.89
FAST REACTOR DOSIMETRY AND DAMAGE ANALYSIS	
R. E. Nightingale	5.1
EBR-II DOSIMETRY TEST - W. N. McElroy, J. L. Jackson, J. A. Ulseth, L. S. Kellogg, and R. L. Simons	5.1
Reaction Rates	5.1
Multiple-Foil Derived Fluxes	5.3
Spectral Averaged Cross Sections	5.7
Single-Foil Derived Fluxes.	5.9
FAST-REACTOR NEUTRON SPECTRA AND FOIL-ACTIVATION CROSS SECTION EVALUATION - W. N. McElroy, R. J. Armani, and E. Tochilin	5.10
DAMAGE FUNCTIONS AND DATA CORRELATION - W. N. McElroy, R. L. Simons. and L. D. Blackburn	5.15
COMPUTER SIMULATION OF RADIATION DAMAGE	5.19
Cascade Formation and Annealing - D. G. Doran	5.19
Structure Damage in Imperfect or Strained Lattices G. E. Russcher	5.24
INTERACTIONS OF CARBON IMPURITY ATOMS WITH A TILT GRAIN BOUNDARY - R. E. Dahl, Jr.	5.25
Single Carbon Interstitials	5.26
Di- and Tri-Interstitials in a Grain Boundary Hole.	5.31

Carbon Migration in a 6-Degree Tilt Boundary	.5.32
RADIATION EFFECTS ON ABSORBING MATERIALS FOR CONTROL	
RODS - A. L. Pitner	.6.1
NUCLEAR GRAPHITE	.7.1
THERMAL EXPANSION - W. J. Gray	.7.1
COMPUTER CALCULATION OF GRAPHITE OXIDATION IN HTGR	
R. C. Giberson	.7.2
COBALT-60 ADDITION TO THE GAMMA IRRADIATION FACILITY	
D. E. Baker.	.7.2
NONDESTRUCTIVE TESTING - G. J. Dau	.8.1
EDDY CURRENT METHODS - THREE-DIMENSIONAL REPRESENTATION	
OF EDDY CURRENT TEST SIGNPLS - H. L. Libby	8.1
THERMAL METHODS - THERMAL TRANSDUCER METHODS	
D. R. Green	.8.4
ULTRASONIC METHODS - DETECTION OF COLD WORK AND GRAIN	
SIZE IN STAINLESS STEEL USING ULTRASONIC CRITICAL ANGLE	
REFLECTIVITY - F. L. Becker and R. L. Trantow	8.5
NEW TECHNIQUES - MOSSBAUER EFFECT BACKSCATTER STUDIES	
R. M. Ord	.8.11
ATR WATER LOOP OPERATION AND MAINTENANCE	
R. S. Hope	.9.1

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SUMMARY

FAST REACTOR CERAMIC FUELS RESEARCH

CREEP OF UNIRRADIATED MIXED OXIDE FUELS

The creep rate of polycrystalline UO_2 -20 wt% PuO_2 , 96% dense has been determined under various loads in four-point bending at the two temperatures of 1475 and 1560 °C. The activation energy was determined to be 110.0 kcal/mole. A study of the dependence of strain rate on stress indicated that the creep rate at a given stress depended on the previous stresses which had been applied to the sample.

IN-REACTOR CREEP OF MIXED OXIDE FUELS

The initial creep test capsule has been irradiated in the K-East Reactor specialty irradiation facility for 144 hr. The capsule was removed and examined. The heat transfer fluid (tin) had spilled from the containers and the specimens were broken. An improved design is being prepared.

THERMAL DIFFUSIVITY OF URANIUM OXIDE

The thermal diffusivity of "round robin" UO_2 specimens have been measured from room temperature to 2500 °C.

SCANNING ELECTRON MICROSCOPY OF NUCLEAR FUEL MATERIALS

Major improvements and additions to the scanning electron microscope have been completed to substantially increase the information content of micrographs produced on this instrument. An X-ray spectrometer to permit semi-quantitative analyses of observed microstructures and a rapid scan device have been added to the instrument. A hot stage, capable of about 1600 °C, is currently under design.

THERMAL EXPANSION OF MIXED OXIDES

An apparatus for measurement of thermal expansion of irradiated mixed oxide fuel is nearly complete.

PROPERTIES OF MOLTEN FAST REACTOR OXIDE FUELS

A preliminary measurement has been made of the viscosity of liquid UO_2 just above the melting point.

Stable pendant drops have not been formed on UO_2 rods 0.22 in. in diameter. This indicates that the surface tension of liquid UO_2 must be less than 600 dynes/cm if the liquid density is approximately 8.7 g/cm^3 .

Two attempts have been made to measure the compressibility of molten UO_2 . Both have resulted in furnace failures just above the melting point of UO_2 .

PHASE STUDIES

Measurements are being performed to determine the accuracy and precision of the present technique for measuring O/M ratios on nonstoichiometric mixed oxides. Another technique for determination of O/M ratios on milligram size samples is under development.

IRRADIATION BEHAVIOR OF FAST REACTOR FUEL MATERIALS

IN-REACTOR THERMAL PERFORMANCE OF $\text{UO}_2\text{-PuO}_2$ FUEL

The melting heat rating for $\text{UO}_2\text{-25 wt\% PuO}_2$ fuel was determined by precision methods to be 16.5 kW/ft for a fuel surface temperature of 950 °C. Fission rate distribution in fuels of fast compositions irradiated in thermal fluxes were measured and correlated with analytical predictions.

Measurements of the influence of Xe contamination on fuel-sheath heat transfer showed surprisingly slight effects. Addition of 40, 60, and 100% Xe to the HC fill gas in $\text{UO}_2\text{-25 wt\% PuO}_2$ fuel pins resulted in respective decreases in gap conductance of 15, 25, and 45%.

THERMAL FLUX IRRADIATIONS OF HYPOSTOICHIOMETRIC $\text{UO}_2\text{-PuO}_2$ FUEL

Detailed postirradiation evaluation of $\text{UO}_2\text{-25 wt\% PuO}_2$ fuel pins irradiated to as high as 160,000 MWd/T in MTR/ETR is in progress. Clad deformations and in some cases gross failures were observed. Significant overall and local inhomogeneities in Pu and fission fragment concentration were observed with the shielded electron microprobe.

FUEL SWELLING AND ANALYTICAL MODELING

Design of basic swelling experiments involving $\text{UO}_2\text{-25 wt\% PuO}_2$ specimens is near completion. Existing stochastic models were shown to provide suitable input for CYGRO and DEFORM.

MIGRATION BEHAVIOR OF $\text{UO}_2\text{-PuO}_2$ FUELS

Fuel-clad reactions and subsequent redistribution of clad constituents in the fuel were studied with the shielded microprobe and described in terms of the thermodynamics of fuel-clad-fission product systems. A basic model which describes fuel constituent migration in response to concentration, temperature, and thermoelectric potential gradients typical of oxide fast fuels is being formulated.

IRRADIATION DAMAGE TO REACTOR METALS

IRRADIATION BEHAVIOR OF STRUCTURAL MATERIALS

Irradiation Facilities Operation

A total of 36 corrosion specimens were discharged from the ETR at the conclusion of Cycles 101 and 102.

Core I testing in the ATR has been completed. Core II is scheduled to end and Core III to begin on September 15, 1969. Cycle 1 startup is scheduled for October 13, 1969.

Microstructural Modifications to Stainless Steels and Nickel-Base Alloys

Good improvements in the high-temperature tensile and creep-rupture properties of 348 stainless steel are observed after thermomechanical treatments that develop a carbide-precipitated stacking fault structure, resulting in significant increases over the properties available in the normal austenitic stainless steel such as from the annealed and cold-worked conditions of 304, 316, and 348 stainless steel.

Production of Void in Nickel

Irradiation of three sources of high purity nickel at 450 °C to 3×10^{21} n/cm² (>0.1 MeV) has resulted in the production of voids. The average void size ranged from 280 to 400 Å and the density of voids varied from 1.8 to 5×10^{14} /cm³. The resultant volume increase was 0.64 to 0.81%. The nickel samples also experienced hardness increases of 35 to 41%.

Irradiation Effects on the Fracture of Heavy Section Pressure Vessel Steels

Studies are underway to determine irradiation and strain aging embrittlement in 12 in. A 533B base plate, irradiation embrittlement in 12 in. submerged arc weldment, and the irradiation preparation for 4 in. thick specimens.

Twelve 1 in. compact K_{IC} fracture mechanics specimens from center through 1/4T position have been irradiated in the M-3 loop of the ETR to approximately 1.5×10^{19} n/cm² (>1 MeV) at 535 °F, as well as 29 tensile specimens irradiated to the fluence range of 1.5 to 4.8×10^{19} n/cm². These irradiated specimens are currently being tested.

A feasibility study for irradiating 4 in. thick compact K_{IC} specimens has been completed, and an engineering design study is now under way to irradiate capsules in the ETR reflector.

The weldment study is using both 1 in. compact K_{IC} specimens oriented parallel to the weldment and 1/2 in. DCB specimens oriented perpendicular to the weld to determine the toughness gradient through the weld, and tests on the latter are nearing completion.

Irradiation Effects in Zircaloy-2

Tensile tests on Zircaloy-2 specimens, irradiated at 280 °C in the ETR to a fluence of 21.5×10^{22} n/cm² (E > 1 MeV), indicate no evidence of saturation of irradiation damage. Analysis of transmission electron microscopy data reveals a strong correlation between tensile properties and microstructure. However, though defect clusters and dislocation loops were in evidence, no voids were observed in either the as-irradiated or postirradiation-annealed material.

FUEL SUBASSEMBLY MATERIALS

EBR II Irradiations

There are five swelling and structural materials experiments in EBR-II. Ten BNW structural materials experiments have been discharged from EBR-II. Subassemblies X-019 and X-067 were discharged.

Damage Analysis

The initial results from immersion density measurements on a series of AISI 316 tensile specimens, which had been irradiated in the EBR-II to a maximum fluence of 3.0×10^{22} n/cm² ($E > 0.1$ MeV) at temperatures ranging from 700 °F to about 1400 °F, suggest a maximum in the temperature dependence of swelling near 1050 °F.

Analysis of a model for the effect of void-loop interactions on swelling indicate that the range of applicability is restricted to values of swelling below that where significant effects of void-loop interaction may be expected to occur.

An observation of voids in irradiated stainless steels under two different contrast conditions suggests that, for purposes of void diameter measurement, the dark border noted in the absorption contrast condition should be considered part of the void.

Helium analyses were obtained from seven 304 and 316 SS samples, which had been irradiated at a variety of positions in EBR-II, and evaluated in terms of alloy content, neutron spectra, and neutron energy.

Biaxial Stress-Rupture Studies

Biaxial stress-rupture tests on AISI Type 316 SS irradiated to $\sim 1 \times 10^{22}$ n/cm² ($E > 1.0$ MeV) in the EBR-II are compared to as-received control data at test temperatures

of 1000, 1200, 1400 °F. Stress-rupture test data at the various test temperatures correlate well with the Theta parameter.

Mechanical Testing of Fuel Pin Cladding

Burst tests have been conducted on 2-3/4 in. fueled specimens taken from PNL-1 series fuel pins. The specimens tested had been irradiated to a fluence of 4.0 to 5.8×10^{21} n/cm² ($E > 0.1$ MeV) and at cladding OD average temperatures of 830 to 910 °F. Sections from the upper end of the fueled column failed at greatly reduced strength and ductility as compared to specimens from the lower end. The specimens from the upper end failed by an intergranular process while the lower end failed by the expected transgranular mode.

RELATED FAST REACTOR SUPPORTING STUDIES

Acceptance Testing and Clad Characterization

Composition and metallography over-checks have been completed on "H" lot tube lengths. Tensile tests on tubing lots E, F, G, and H at RT, 800, 1000, 1200, 1400, and 1600 °F at strain rates of 0.01 and 1.0 min⁻¹ have been completed.

Tensile Studies

Tensile test results are reported for a series of AISI 304 SS specimens irradiated in EBR-II subassembly X-022. Some of the data show that much greater strengthening occurred in specimens irradiated to $\sim 6 \times 10^{21}$ n/cm² (total) at 770 °F than in those irradiated to $\sim 8 \times 10^{21}$ n/cm² at 870 °F, indicating a strong sensitivity of tensile properties to irradiation temperature.

The data suggest that irradiation to 4×10^{21} at 725 °F does not result in strengthening, but inconsistencies in some tests must be resolved before a final conclusion can be reached.

Uniaxial Creep Rupture Studies

The results of creep-rupture tests on aged 304 and 316 SS indicate in general, that aging reduces rupture-life except at low stress levels. Creep-rates of aged specimens generally fell within the scatter band of the control specimens.

Stress-rupture data for irradiated control 304 SS have been parameterized.

Creep-rupture tests on welded specimens indicate that the Stick-Electrode type weld produces better rupture life than do the TIG or MIG processes.

Analyses of transient strain in 316 SS specimens irradiated at 900 °F to a fluence of $\sim 1 \times 10^{22}$ n/cm² indicate it is reduced with increasing stress.

FAST REACTOR DOSIMETRY AND DAMAGE ANALYSIS

EBR-II DOSIMETRY TEST

As a result of the EBR-II Run 31F dosimetry test, reaction rates for ~ 20 fission and nonfission flux monitors are now available for r and z coordinates (in cm) from (0, 0) to (33, 60). A relatively high degree of accuracy has been established for these results by interlaboratory calibration studies. These reaction rates together with the SAND-I1 code are being used to generate energy-dependent integral fluxes (i.e., total flux, >0.1 , flux >1.0 , etc.) and corresponding measured values of spectral averaged cross sections, $\bar{\sigma}_m$. The availability of these $\bar{\sigma}_m$ values will permit the more accurate definition of flux levels and fluences for individual flux monitors [such as $^{54}\text{Fe}(n,\gamma)$ and $^{58}\text{Fe}(n,\gamma)$] that are being used in structural and fuel subassemblies irradiated in EBR-II.

FAST REACTOR NEUTRON SPECTRA AND FOIL ACTIVATION CROSS SECTION EVALUATION

Multiple foil and time-of-flight derived neutron flux-spectra and foil reaction spectral averaged cross sections have been compared for a core center and surface location for a bare ^{235}U assembly. The good agreement in the results found for the core center position establishes increased confidence in current microscopic cross section data and multiple foil results. On the other hand, the lack of such agreement for the core surface measurements suggests that previous photo-plate measurements appear to have underestimated the mean energy of the Godiva leakage and fission spectrum.

DAMAGE FUNCTIONS AND DATA CORRELATION

A preliminary form of a damage function for 304 SS has been obtained for the irradiation induced change in total elongation. A tentative conclusion is that low-energy neutrons (below ~ 0.1 to 1 MeV) contribute significantly to this damage.

COMPUTER SIMULATION OF RADIATION DAMAGE

A paper has been prepared describing a study of the sensitivity of certain point defect simulations to variations in the interatomic potential. The production of stereo-photographs from oscilloscope displays has been adapted to aid in the visualization of displacement spikes. Simulated short term annealing suggests that annealing enhances the production of mobile interstitial by (n,γ) recoils relative to that by high energy knock-on atoms.

The SLIDES computer code is being programmed and is now undergoing debugging.

INTERACTIONS OF CARBON IMPURITY ATOMS WITH A TILT- GRAIN BOUNDARY

The computer simulation of lattices shows ordering of a grain boundary by carbon atoms. In a cell containing 1275 iron atoms, one carbon atom added to the misfit region of the grain boundary essentially caused complete local ordering of the structure. It is apparent that a small concentration of carbon can drastically alter the strength of iron-based metals and alloys. Insight into precipitation mechanisms may be gained from the qualitative accommodation and migration data. Relatively large carbon clusters can be accommodated in misfit regions of a grain boundary without stressing the adjacent grain and thus their formation is energetically favorable. These clusters would function as pins impeding grain boundary motion or transmission of strains from one grain to the next and thus would harden or strengthen a metal. The region in which the formation and migration of interstitial impurities is influenced by the presence of a grain boundary, is quite small, no more than a unit cell in width.

RADIATION EFFECTS ON ABSORBING MATERIALS FOR CONTROL RODS

A subassembly containing boron carbide specimens was discharged from a Hanford reactor after an estimated 3% ^{10}B burnup. Sample temperatures ranged from 550 to 1130 °F. The boron-carbide was in five forms: 60 and 80% TD powder, and 65, 90, and 99% TD pellets. All samples showed apparent high gas release, and the pellets exhibited considerable degradation as a result of the irradiation. Most severe deterioration was in the highest density pellets (99% TD), where the surface became quite powdery.

NUCLEAR GRAPHITE

F-1 THERMAL EXPANSION

The c-axis lattice thermal expansion of POCO grade AXZ-5Q1 graphite was measured both before and after irradiation at 1000 to 1100 °C to 1.2×10^{22} n/cm². The average CTE between 25 and 425 °C for the unirradiated material was $27.1 \pm 1.0 \times 10^{-6}/^{\circ}\text{C}$; for the irradiated material it was $26.8 \pm 3.0 \times 10^{-6}/^{\circ}\text{C}$. This is well within the accepted value of $27.8 \times 10^{-6}/^{\circ}\text{C}$ for single crystals.

COMPUTER CALCULATIONS OF GRAPHITE OXIDATION IN HTGR

The GOP program is being modified to enable calculation of the 1100 MW(e) HTGR graphite chemistry system.

COBALT-60 ADDITION TO THE GAMMA IRRADIATION FACILITY

An additional 360,000 Ci of ⁶⁰Co was added to the gamma irradiation facility. The total amount of ⁶⁰Co in the source was 620,000 Ci. The maximum dose rate was 3.87×10^7 R/hr as determined by ceric sulfate dosimetry. An apparatus is being modified to study the radiolytic rate of reaction of CO₂ with H₂ in the 100 to 1000 vpm range in helium.

NONDESTRUCTIVE TESTING

EDDY CURRENT METHODS

Typical eddy current test signals from a two-frequency tube tester were studied as an aid in the development of a digital computer method for the separation of test variables. The results of this study will provide faster and more accurate means to analyze more test variables at an increased number of test frequencies.

THERMAL METHODS

A new thermal transducer system has been developed which has high speed capability. The transducer has been tested on several types of specimens; however, some modification will be necessary for real time processing. This system should enable better interpretation of the thermal image and lead to automated data acquisition.

ULTRASONIC METHODS

The critical angle ultrasonic reflectivity technique has been applied successfully to the measurement of cold work and grain size in FFTF fuel pin cladding. A resolution approaching +2% of cold work was achieved. The method shows considerable promise but will require more development.

NEW TECHNIQUES

A Mössbauer backscatter system has been developed capable of over 130% peak signal to nonresonant background measurements. This was achieved using special shielding techniques and the latest available equipment. Mössbauer spectroscopy can be performed in the order of minutes using "as is" specimens.

ATR WATER LOOP

The loop has operated intermittently at full power of 250 MW since 8/16/69. A total integrated power of 6,000 MWd has been achieved. Apparent radiolytic decomposition of the coolant water is causing higher O_2 pressures than anticipated.

FAST REACTOR CERAMIC FUELS RESEARCHCREEP OF UNIRRADIATED MIXED OXIDE FUELS - O. D. Slagle

The creep rate of polycrystalline UO_2 -20 wt% PuO_2 has been determined under various loads in four-point bending at 1475 and 1560 °C. The samples were prepared from coprecipitated powders which had been sintered in argon-8% hydrogen at 1700 °C to a density of 96% TD. The final average grain size was 20 microns. A representative microstructure is shown in Figure 2.1. Creep measurements were conducted in dry argon-8% hydrogen resulting in a final O/M ratio of 1.98.

FIGURE 2.1. *Representative Microstructure of UO_2 -20 wt% PuO_2 Creep Specimen Before Testing*

Table 2.1 compares the strain rates measured for a series of tests in which the temperature was varied between approximately 1475 and 1560 °C. The data did not exhibit any trend either increasing or decreasing. The activation energy was determined to be 110.0 kcal/mole by fitting a least squares line to a plot of the logarithm strain rate versus inverse absolute temperature. This activation energy is equal to the value for stoichiometric UO_2 reported by Bohaboy and Asamoto⁽¹⁾ for high compressive stresses (greater than 6,000 psi) and is larger than the value of 90 kcal/mole that these authors observed for the low stress region (less than 2,000 psi). Armstrong et al.⁽²⁾ have reported a value of 91 ± 8 kcal/mole for UO_2 in four-point bending at a maximum tensile stress of 10,000 psi. In comparison, this activation energy lies at the upper end of the range of values which have been reported for stoichiometric UO_2 .

TABLE 2.1. *Comparison of Strain Rates Taken at Two Different Temperatures with a Maximum Fiber Stress of 4131 psi*

<u>Maximum Strain Rate</u>	<u>Temperature</u>	<u>Maximum Strain Rate</u>
0.538	1563	
	1476	0.145
0.592	1560	
	1479	0.144
0.642	1563	
	1482	0.124
0.642	1563	
	1478	0.132
0.472	1561	

The data were taken in the order listed.

The strain rates are given in units of 10^{-6} hr^{-1} and temperatures are in °C.

Activation Energy = 110.0 kcal/mole.

The results of varying the stress at each of the temperatures is shown in Figure 2.2. The order in which the data was taken is listed by numbers beside each data point. For the data taken at 1475 °C, two different lines have been fit to the data. For the initial increasing stress data, a slope of 1.08 was found, while with decreasing stress, a slope of 1.75 was observed. These results suggest that the creep rate at a given stress depends upon the stress to which the sample has been previously subjected. These results also indicate that the application of stress to a sample changes the microstructure and/or dislocation arrangement in such a way as to lower the creep rate after returning to the lower applied stresses. Data at 1560 °C were taken in the order of the largest stress first, decreasing and then increasing. This data does not show any constant difference between data obtained during the decreases and increases in applied stress. A comparison in the slopes of the lines shows that the slope of the line at 1560 °C agrees most closely with the data at 1475 °C taken after the highest load was applied and the stress decreased. These results suggest strongly that the creep strain rate of mixed oxide fuel is dependent on the order in which the tests are made.

Previous stress dependence determinations made on UO_2 by Bohaboy and Asamoto⁽¹⁾ and by Poteat and Yust⁽³⁾ has shown that at low stresses (less than 2,000) a linear dependence of strain rate with stress exists. At higher stress (above 6,000 psi), the dependence becomes stress to the fourth or fifth power. These findings had been made using compressive creep. Because of the stress distribution in four-point bending, a change in dependence would not be as evident; however, no indication of any change in stress dependence can be seen. Nevertheless, its effect was much smaller than the scatter in the data and the variations introduced by the order the stresses were applied.

FIGURE 2.2. Strain Rate Versus Stress for $U_{0.2}Pu_{0.8}O_{4.0}$

After creep testing, a microstructural examination of the sample revealed that extensive cracking had occurred on the tensile face of the sample. Figure 2.3 shows a crack which had originated at the tensile surface. In addition to the cracks which developed under tension, there is an enlargement of porosity ahead of the crack. These observations suggest that pore growth may play an important role in the creep mechanism of this material.

100 micron

Neg 92256

250X

FIGURE 2.3. *Micrograph of $U_{0.8}Pu_{0.2}O_2$ Sample After Creep Testing Showing a Number of Cracks Which Have Developed*

IN-REACTOR CREEP OF MIXED OXIDE FUELS - J. A. Basmajian

The initial capsule to measure the in-reactor creep properties of mixed oxide fuels by four-point bending of beams has been irradiated. Two samples of UO_2 - 20 wt% PuO_2 fuel (1 in. $\times$ 0.06 in. $\times$ 0.25 in.) were irradiated for 144 hr in the K-East Reactor Specialty Irradiation facility at a nominal temperature of 900 °C.

The capsule was inserted into the reactor with continuous temperature monitoring. When the capsule was in a uniform neutron flux, the three thermocouples read between 875 °C and 900 °C. An auxiliary heater was used to control the temperature. A 100 °C drop in temperature occurred approximately 20 hr after

insertion. The reactor control revealed the drop in temperature to coincide with a control rod movement. In order to bring the temperature back to 900 °C, the capsule was inserted into a higher neutron flux and the heater power raised. For the balance of the test period, the heater operated at this power level. Two thermocouples failed 110 hr after the start of the experiment. The one operating thermocouple read 850 °C when the heater was shut off.

Examination of the capsule revealed that the capsule had tilted and that most of the tin used as a heat transfer medium between the fuel and the capsule environment had been spilled from the molybdenum "boat." Since the tin was found on one side and toward the leading end of the capsule, two modes of tilting must have been operative. The spill had to have taken place during initial insertion of the capsule or when the capsule was moved the following day. Since the capsule is at the end of a 35-foot long rod, the most likely cause of the rotation of the capsule was a slight angular rotation at the rod end being magnified to the approximately 30° rotation of the capsule. The vertical position indicator on the capsule was marked all the way to the end of the pushrod. The endwise tilt is probably due to a tilt in the experimental tube.

Postirradiation examination revealed that both specimens were broken, probably resulting from uneven thermal stresses due to loss of molten tin heat transfer medium. No apparent bending of the specimens was observed visually at low magnification in the hot cells. The SS-clad thermocouples which failed appeared to have reacted with the tin. Molybdenum cladding will be used to alleviate that problem in future capsules. Further modifications of capsule design and reactor irradiation procedures are being made to prevent capsule disorientation during insertion into the reactor.

THERMAL DIFFUSIVITY OF URANIUM DIOXIDE - J. L. Bates

The thermal diffusivity of "round robin" uranium dioxide was measured from near-room temperature to 2500 °C. Measurements were made as part of a program to compare the high temperature thermal conductivity/diffusivity of the same UO_2 measured at different laboratories using different techniques.^{*(4)} The "round robin" UO_2 for each participating laboratory was from a single source with a reported uniform composition and structure. The O/U ratio was 2.003 ± 0.003 with a density of 10.75 g/cm^3 at room temperature. The grain size of the sample was very large, easily detected by the naked eye. Samples for thermal diffusivity measurements at BNW were between 0.076 and 0.127 cm thick and were 0.625 cm in diameter. Three samples were measured, two approximately 0.076 cm thick and one 0.127 cm thick. Measurements were made on three samples in a purified, argon atmosphere at atmospheric pressure. The argon introduced into the furnace contained 1 ± 1 ppm oxygen and 5 ± 2 ppm water.

The thermal diffusivity was measured with a "flash" technique, using a laser to provide the thermal pulse. Measurements were conducted in a tungsten resistance furnace with tungsten and molybdenum heat shields. The sample was supported in the furnace in a UO_2 holder supported on a tungsten tube. Temperatures were measured with a tungsten-5%rhenium versus tungsten-26%rhenium bare thermocouple set in the UO_2 holder. The water content (dew point) of the furnace atmosphere was measured continuously. Oxygen content of inlet gas was also determined continuously using a solid electrolyte oxygen meter. Impurities in gas were determined spectrographically.

* This program was directed for the Division of Reactor Development and Technology, Fuels and Materials Branch by General Electric Company, Nuclear Materials and Propulsion Operation.

One sample, RR-3, was coated on both sides with a water suspension of graphite to determine if radiation penetration from the laser beam into the uncoated sample could result in possible experimental error. Measurements were made to 2500 °C before the coatings were applied, to 1000 °C with the coatings, and to 1500 °C after the coatings were mechanically removed from both surfaces.

Thermal diffusivity values were calculated using heat loss corrections using the method suggested by Cowan.^(5,6) No pulse time corrections were required. Corrections were made for density using the temperature - volume expansion relationship reported by Christensen.⁽⁷⁾

Thermal conductivity (λ) values were calculated using the heat capacity (C_p) data reported by Leitnaker et al.,⁽⁸⁾ the measured thermal diffusivity (a), and the measured sample density (ρ) corrected for temperature changes using data reported by Christensen:⁽⁷⁾

$$\frac{V_T}{V_0} = 1 + 9 \times 10^{-6} T + 6 \times 10^{-9} T^2 + 3 \times 10^{-12} T^3 \quad (1)$$

where V_T is the specific volume (cm^3/g) at temperature T in °C, and V_0 is the specific volume at °C. The heat capacity data of Leitnaker was modified slightly near 1400 °C to correct for the slight maximum and minimum in the C_p versus temperature curve resulting from the least squares fitting of the combined enthalpy experimental data. A line was drawn through the maximum and minimum curve in this temperature region, and a new equation for C_p as a function of temperature was determined.

$$\begin{aligned} C_p = & 5.4757 \times 10^{-2} + 8.4077 \times 10^{-5} T - 1.1425 \times 10^{-7} T^2 \\ & + 5.7510 \times 10^{-11} T^3 - 6.1208 \times 10^{-5} T^4 - 8.8911 \\ & \times 10^{-19} T^5 + 5.6447 \times 10^{-23} T^6 \end{aligned}$$

where C_p is in cal/g/°C.

The measurement conditions are summarized in Table 2.2. With the exception of RR-3, each sample was initially heated from 1000 to 1200 °C and cooled with measurements being made both on heating and cooling. Subsequent cycles were made to higher temperatures with measurements also being made both on heating and cooling. Generally, more data points were obtained on heating than on cooling. Samples were removed from the furnace after each cycle to examine the sample. These procedures were followed to determine if the sample had physically changed or if experimental errors could be introduced by altering the position of the sample in the UO_2 holder.

TABLE 2.2. Thermal Cycling Conditions for "Round Robin" UO_2

Sample Thickness, cm	Temperatures, °C			
	Cycle 1	Cycle 2	Cycle 3	Cycle 4 and 5
RR-1 (0.081)	16-1208 ^(a)	94-1216	266-2012	1326-1336 1087-2111
RR-2 (0.127)	17-1200 ^(b)	400-1010	88-1487	
RR-3 (0.081- 0.076)	399-2500	16-1012 ^(c)	25-1519	

-
- a. Because of instrument difficulties, precision of data was less than for other data.*
b. No thermal diffusivity data obtained.
c. Sample surfaces coated with graphite.

The thermal diffusivity are illustrated in Figures 2.4 through 2.9. A summary of the thermal diffusivity data is shown in Figure 2.10.

The data for all three specimens is the same only after the sample has been heated to approximately 1200 °C or above. The thermal diffusivity values between room temperature and 1200 °C during the first cycle and subsequent cycles before attaining a minimum temperature of 1200 °C are higher than after the sample had been heated to 1200 °C. The maximum

FIGURE 2.4. *Thermal Diffusivity of "Round Robin" UO₂ Sample RR-1, Cycles 1 and 2*

FIGURE 2.5. Thermal Diffusivity of "Round Robin" UO_2 ample RR- Cycles 3, 4, and

FIGURE 2.6. Thermal Diffusivity of "Round Robin!" UO_2 Sample RR-2, Cycles 1 Through 3

FIGURE 2.7. Thermal Diffusivity of "Round Robin" UO₂ Sample RR-3 Before Coating - Cycle 1

FIGURE 2.8. *Thermal Diffusivity of "Round Robin" UO₂ Sample RR-3, Graphite coated - Cycle 2.*

FIGURE 2.9. Thermal Diffusivity of "Round Robin" UO₂ Sample RR-3, Uncoated
Cycle 3

FIGURE 2.10 Summary of "Round Robin" UO_2 Thermal Diffusivity Data

decrease is approximately 6% at 200 °C and 8% at 800 °C, only slightly more than the estimated +5% precision of the data. However, the thermal diffusivity values of all three samples after heating to 1200 °C or higher were statistically the same over the entire temperature range to 2500 °C. This effect may result from a permanent change in sample composition or structure which is stable in the measurement environment only after heating above 1200 °C. An increase in stoichiometry could possibly reduce the conductivity at the lower temperatures.

The possible effects of the penetration of radiation into the sample from the laser pulse were investigated. Two methods were used to determine if the UO_2 was translucent to the 0.6943 μ radiation from the ruby laser crystal. Samples of two different thicknesses were used. If the sample were significantly translucent, the thermal diffusivity of the thinner specimen would be larger. The results show no difference in the thermal diffusivity in the thicker RR-2 specimen (0.127 cm thick) and the RR-1 and RR-3 specimens (0.076 cm thick).

The second procedure used was to coat both front and back surfaces of the sample (RR-3) with a graphite coating which was thick enough to absorb the radiation from the laser on the front surface and to prevent the optical detector from measuring temperatures into the sample through the back surface. After measurements to 1000 °C, the graphite coatings were mechanically removed from the sample and measurements made to 1500 °C. No differences in thermal diffusivity were observed during measurement, before coating, with coating, or after removal of coating (Figures 2.8 and 2.9). These two results demonstrate that radiation absorption beyond the sample surface from the laser does not significantly influence the accuracy of the thermal diffusivity results.

The thermal conductivity of the Round Robin UO_2 after heating to 1200 °C or above is shown in Figure 2.11. The conductivity reaches a minimum between 1600 and 1800 °C with an increase from 1800 to 2500 °C.

The evaluation of the data is continuing.

SCANNING ELECTRON MICROSCOPY OF NUCLEAR FUEL MATERIALS -

J. L. Daniel

Major improvements and additions to the scanning electron microscope have been completed to substantially increase the information content of micrographs produced on this instrument. The X-ray spectrometer accessory uses a nondispersive solid-state detector to record characteristic X-rays emitted from specimen components, and identification is achieved with a 512-channel energy analyzer. Specimen areas as small as about 0.1 μ in diameter can be analyzed simultaneously for all elements above atomic number 12. Test applications to a variety of specimens has revealed unsuspected compositions in several instances and valuable guidance in interpretation of micrographs is available for the first time. The analyses can be performed during examination of radioactive specimens, but background interference increases rapidly with radiation level. Methods for obtaining quantitative data and for correlating area analysis for individual elements with electron micrographs are being developed.

A hot stage for the SEM is being designed to permit in-microscope experiments to about 1600 °C. The stage will fit the standard microscope internal mounts and will include provisions to minimize interference from thermal electron emission and furnace incandescence. A cup-type of heating element is preferable to provide a small uniformly heated zone, but initial tests indicated that excessive heating currents would be required. Mesh and wire elements are being fabricated also for testing.

FIGURE 2.11 Summary of "Roun Robin" UO₂ Thermal Conductivity Data

Equipment for TV monitoring and video taping of dynamic experiments has been installed on the microscope. The TV monitor also allows rapid scanning of entire surface areas during initial examination of any specimen. Best achievable TV and video tape resolution is about 1000 Å.

THERMAL EXPANSION OF MIXED OXIDES - P. G. Pallmer

The lattice expansion of irradiated plutonium - uranium oxides can be determined with a high temperature stage on the X-ray spectrogoniometer by separating the diffracted X-ray pattern from the high intensity background. Pulse height discrimination of the proportional counter pulses is alone inadequate. A monochromator, which has been installed to selectively diffract the X-rays from the background radiation, allows enough lead shielding to be interspersed between the sample and counter to produce a satisfactory peak-to-background ratio. To minimize weight, a lead block 4 in. long was shaped to occupy the space directly between sample and detector tube. A sample of $(U_{0.75}Pu_{0.25})O_2$ irradiated to 65,000 MWd/ton was used to test the effectiveness of the lead shielding with the sample at the center of the goniometer. The effective linear absorption coefficient was 0.23 cm^{-1} , which will make possible a usable signal-to-background ratio.

Existing shields for scattered X-rays are backed by, or replaced by, at least 1/8 in. lead for operator shielding. Additional lead glass shielding can be provided if necessary. The installation and testing of glove box atmosphere controls and services is approximately 95% complete. The vacuum system is completed and tested except for the transfer of the furnace chamber and its flexible connecting bellows. High temperature experiments on unirradiated mixed oxide will begin next quarter.

PROPERTIES OF MOLTEN FAST-REACTOR OXIDE FUELS - R. P. Nelson,
J. J. Rasmussen, and O. D. Slagle

Viscosity

The viscosity of molten fast-reactor oxide fuels is being measured with an oscillating cup viscometer.⁽⁹⁾ The precision of the viscometer was improved by reducing the moment of inertia and the decrement of the empty system. The moment of inertia was reduced to 365 g/cm² by decreasing the mass of the scale disc and inertia bar. Substitution of the tungsten torsion wire with an annealed W-26 Re wire lowered the wire decrement to 0.0004.

A preliminary measurement of the viscosity of liquid UO₂ just above the melting point (2755 °C apparent melting temperature) was made. The system parameters were:

- Tungsten crucible radius 0.635 cm
- Torsion wire diameter 0.0127 cm
- Sample moment of inertia 14.3 g/cm²
- Sample decrement 0.00468
- Period of system 9.43 sec

A viscosity of 1.4 P was calculated, assuming a liquid oxide density of 8.7 g/cm³.^(10,11) Gamma radiographs of the solidified oxide indicated, however, that only partial melting may have occurred, so this value must be considered tentative. Further experiments will be made to refine this value and extend the measurements to 3000 °C.

Surface Tension

Attempts have been made to measure the surface tension of liquid UO₂ by the pendant drop technique. The 3-inch long tungsten capsule was machined to provide a temperature gradient of approximately 125 °C from the center of the capsule to the top where the rod was attached. Viewing the sample continuously

with an image orthicon camera, a drop was observed to form on the end of the sintered rod when the UO_2 melting point was reached. By the time an X-radiograph was taken, the drop had fallen from the rod. As the temperature was raised in an attempt to form new drops, whiskers began to form in the region just behind the end of the UO_2 rod. The rod tip grew larger and flatter as the temperature was raised until the tip eventually filled the capsule. In a second run, it was found that backfilling the capsule with 0.1 atmosphere of the gas mixture 75% He to 25% Ar reduced the amount of whisker growth. The initial pendant drop which formed fell immediately and no more drops could be formed. Calculations based on the estimated volume of the pendant drops, rod diameter, and a liquid UO_2 density of 8.7 g/cm^3 indicate that the surface tension of liquid UO_2 must be less than 600 dynes/cm.

A new pendant drop capsule will be constructed with a smaller temperature gradient from the center to the top of the capsule. The UO_2 rod will be reduced in size sufficiently for stable pendant drops to be formed. Two tungsten capsules have been prepared to obtain UO_2 surface tension data from the equilibrium meniscus shape. This technique will also allow the volume change on melting and liquid density to be determined. Preliminary results indicate that a gamma ray source will be required in order to see the meniscus. The X-ray source, even with a 300 kV potential, will not penetrate the lip of the UO_2 meniscus sufficiently to obtain the data necessary to calculate the surface tension. These meniscus data will be obtained in the new high temperature vacuum resistance furnace.

A second furnace for surface tension measurements of mixed oxides by the pendant drop method is being designed. The furnace will have a double shell of aluminum separated by 3/16 in. for coolant water. Tests have indicated that this will not cause excessive X-ray scattering. The furnace will

have a "D" shape to enable the film to be placed closer to the capsules than is presently possible. This will reduce the error resulting from geometrical uncertainties.

Compressibility

Two attempts have been made to measure the compressibility of molten UO_2 . Both have resulted in furnace failures just above the melting point of UO_2 . The first failure occurred when a leak developed in the feed-through seal around the single crystal tungsten transmission rod. The second failure was believed due to a crack which developed in the sealed crucible containing the molten UO_2 . This latter failure indicated that once the oxide has been melted and solidified in the crucible, further measurements on the same sample may result in crucible failure. Before attempting additional measurements, weld tests will be made in an attempt to fabricate more reliable crucibles.

PHASE STUDIES - C. E. McNeilly and R. E. Woodley

BNW has developed a technique to determine the degree of nonstoichiometry in mixed oxide fuels. The basic method involves determination of the O/M gravimetrically after heating a non-stoichiometric oxide to 800 °C in an Ar- H_2 - H_2O atmosphere with a controlled oxygen potential of -100 kcal/mole O_2 .⁽¹²⁾ During the past quarter, attempts have been made to determine the precision and accuracy of the technique. The precision was evaluated on a series of 25 (U,Pu) O_{2-x} pellets (5 from each of 5 sintering runs) with an O/M ratio determined separately for each pellet (Table 2.3). Assuming all five pellets in each run have the same actual O/M, it has been shown statistically that the present technique gives O/M values that are reproducible to ± 0.0012 O/M units at 1 σ (68% confidence level) or 0.0024 O/M units at 2 σ (95% confidence). The data further show an almost insignificant difference between batches (± 0.0009 O/M units).

TABLE 2.3. *Results of O/M Gravimetric Measurements
to Determine Precision of Technique*

	<u>Starting Weight</u>	<u>Weight After Oxidation</u>	<u>Weight Gain</u>	<u>O/M Ratio</u>
1A	1.63900	1.64315	+0.00415	1.9577
2A	1.56175	1.56582	+0.00407	1.9564
3A	1.61417	1.61835	0.00418	1.9567
4A	1.58500	1.58917	0.00417	1.9560
5A	1.63487	1.63925	0.00438	1.9552
1B	1.58415	1.58836	0.00421	1.9556
2B	1.65200	1.65660	0.00460	1.9534
3B	1.51360	1.51738	0.00378	1.9582
4B	1.58448	1.58880	0.00432	1.9543
5B	1.53980	1.54393	0.00413	1.9552
1C	1.67878	1.68339	0.00461	1.9541
2C	1.57315	1.57725	0.00410	1.9564
3C	1.63960	1.63969	0.00451	1.9541
4C	1.53256	1.53667	0.00411	1.9552
5C	1.66600	1.67044	0.00444	1.9554
1D	1.50365	1.50757	0.00392	1.9564
2D	1.57208	1.57623	0.00415	1.9559
3D	1.59294	1.59695	0.00401	1.9579
4D	1.59506	1.59923	0.00417	1.9560
5D	1.61573	1.61972	0.00399	1.9587
1E	1.55833	1.56253	0.00420	1.9550
2E	1.61175	1.61605	0.00430	1.9554
3E	1.58200	1.58642	0.00442	1.9533
4E	1.54985	1.55400	0.00415	1.9552
5E	1.54385	1.54810	0.00425	1.9540

We have attempted to verify the accuracy by using the technique to oxidize some high purity uranium and plutonium metal to an O/M of 2.0000. The results obtained to date are inconclusive and further measurements are underway. When a small piece of the metal (0.5 to 1.0 g) is oxidized under the above conditions, there is a large volume of very fine oxide powder generated. This powder has a large surface area and tends to adsorb moisture from the circulating atmosphere giving a false weight gain. If we assume we started with a pure metal (O/M = 0.0000), these samples typically calculate out to an oxide O/M of 2.03 - 2.05. In the most recent experiments we have dried the atmosphere, at temperature, after oxidation is complete to reduce the partial pressure of water from 4.58 mm to 5.2×10^{-4} mm. This results in an immediate weight loss (within 1 to 2 hr) with a corresponding decrease in calculated O/M to values from 2.0003 to 2.0150. If the sample is left in this atmosphere at 800 °C for longer periods (several hours) reduction to a substoichiometric oxide occurs. This work will be completed next quarter and a final report will be written.

Other experiments have been performed in an attempt to devise a method for determination of O/M ratios on milligram size samples such as might be extracted from several positions along the radius of an irradiated mixed oxide fuel pin. This technique involves carbon reduction of the oxide and chromatographic determination of the amount of CO₂ produced. Outgassing experiments have been performed on both TSX and Poco AXF graphite crucibles. The procedure normally employed was to heat the crucible to about 1200 °C for 30 min in a stream of pure helium before collecting any of the evolved CO₂. The crucible temperature was then increased to about 1950 °C and maintained at this value for an additional 30 min during which CO₂ was collected from the helium stream. In a slightly modified procedure, the preliminary heating to 1200 °C was

performed under vacuum, and the collection of CO_2 at a crucible temperature of about 1950 °C was done in the absence of helium. The amounts of CO_2 collected during several runs with the Poco crucible are compared in the following table with that obtained during similar runs with a TSX graphite crucible.

TABLE 2.4. *CO_2 from Poco Crucible Runs Compared with CO_2 from TSX Graphite Crucible Runs*

Graphite	Run Number and Type	CO_2 Evolved, mole/g $\times 10^7$	Crucible Condition
Poco	1 He stream	3.24	New
	2 He stream	3.44	Re-exposed to air after Run 1
	3 He stream	3.48	Re-exposed to air after Run 2
	4 He stream	4.00	Remained in He after Run 3
	5 Vacuum	25.58	Re-exposed to air after Run 4
TSX	1 He stream	9.66	New
	2 He stream	6.05	Re-exposed to air after Run 1
	3 Vacuum	20.21	Re-exposed to air after Run 2

With respect to CO_2 evolution in a helium stream, the table shows that a decided improvement was obtained with Poco as compared to TSX graphite. Nevertheless, in both cases the amount of CO_2 collected must be considered excessive. The amount of CO_2 collected under vacuum is obviously excessive; furthermore, neither crucible could be thoroughly outgassed in a reasonable length of time, even under high vacuum at 1950 °C.

Because of the outgassing difficulties with graphite crucibles, a crucible was machined from pure tungsten. In using a tungsten crucible, powdered graphite must be added to the fuel sample. However, with small amounts of finely-divided graphite, outgassing will not be a problem. Furthermore, the amount of chemisorbed oxygen should be insignificant compared to the chemically-combined oxygen of the fuel sample. Experiments will continue next quarter.

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IRRADIATION BEHAVIOR OF FAST REACTOR FUEL MATERIALS

J. A. Christensen

IN-REACTOR THERMAL PERFORMANCE OF UO_2 - PuO_2 FUEL

J. A. Christensen

Melting Heat Rating for UO_2 -25 wt% PuO_2 Fuels

L. A. Lawrence and J. A. Christensen

Restriction to rod powers which preclude fuel melting impose serious limitations on core design, power rating, and peak flux in the FTR. Melting heat ratings have been inferred and are used in establishing reference designs; however, available data include so much uncertainty that application of prudent safety factors probably unduly penalize reactor design power. Current efforts have been directed toward more rigorous measurement of melting heat rating for fast oxide fuels. Results to date show, with increased precision, values similar to those previously assumed. For UO_2 -25 wt% PuO_2 , the quantity $\int_{T_m}^T KdT$ was established at 43 W/cm. This is equivalent to a 950 °C linear rod power of 16.5 kW/ft.

Three capsules, each containing two fuel pins, were irradiated for 10 hours under conditions which assured thermal and structural steady-state. Specimens were then rapidly quenched to preserve structures representative of peak power operation. Each pin was surrounded by a continuously reading calorimeter consisting of a heavy-wall, aluminum sleeve with thermocouples at two radial positions. Heat generation rates measured in this fashion were supported by postirradiation burnup analysis for ^{140}Ba , ^{95}Zr , and ^{141}Ce . Postirradiation examination of the capsules consisted of removing the center 1/2-in. of fuel from each pin for metallography, beta-gamma autoradiography, and alpha autoradiography. A second sample adjacent to the first was removed from each pin for radio-chemical burnup determinations. Figures 3.1 through 3.3 show

Neg 0694109-1

FIGURE 3.1. Postirradiation Structures of
Experimental Pins 1 and 2

Neg 0694109-2

FIGURE 3.2. *Postirradiation Structures of
Experimental Pins 3 and 4*

Neg 0694109-3

FIGURE 3.3. *Postirradiation Structures of
Experimental Pins 5 and 6*

the macrographs and autoradiographs of the six pins. Figure 3.1 shows that Pin 1 operated well below melting. Pin 2 has large equiaxed grains near the center. It is postulated that this pin operated very close to center melting. Pins 3 through 6 exhibit the characteristic structures associated with fuel that has operated partially molten for a short period of time. The alpha autoradiographs of these four pins show segregation of the plutonium toward the fuel center. The subgrain structure in the six pins is evident on the photomicrographs of the polished and etched cross-sections (Figures 3.4 through 3.9). Measurements from these photographs of fuel melting radii are summarized in Table 3.1 along with fuel characteristics and test conditions.

Melt radius was correlated with heat rating and the correlation extrapolated to zero melting to yield the melting heat rating (Figure 3.10). A typical fuel cross-section contains these different indications of melting:

- The outer edge of the central porous region which correlates with the heat rating at shutdown.
- The outer edge of the fully dense region which correlates with the maximum heat rating.
- A weighted average location between these two indicators which correlates with average heat ratings determined by postirradiation burnup analyses.

Each of these three correlations extrapolated to the same melting heat rating, 30 kW/ft.

Flux depression corrections are required to apply these measurements, which were made in a thermal flux, to fast flux environments. These were obtained by measuring radial burnup profiles in several irradiated cross-sections and numerically integrating the results to yield the volume averaged flux in the fuel. The fuel pins used for these measurements were irradiated at low enough heat ratings to preclude migration of

Neg 0694108-1

0.10cm

FIGURE 3.4. *Postirradiation Microstructure of Pin 1*

Neg 0694108-6

0.10cm

FIGURE 3.5. Postirradiation Microstructure of Pin 2

Neg 0694108-4

0.10cm

FIGURE 3.6. *Postirradiation Microstructure of Pin 3*

Neg 0694108-5

0.10cm

FIGURE 3.7. Postirradiation Microstructure of Pin 4

Neg 0694108-3

0.10cm

FIGURE 3.8. Postirradiation Microstructure of Pin 5

Neg 0694108-2

0.10cm

FIGURE 3.9. Postirradiation Microstructure of Pin 6

- (q/L) MAXIMUM VERSUS R_m/R_0 , R_m = EXTENT OF POROSITY
- (q/L) END OF LIFE VERSUS R_m/R_0 , R_m = INNER BOUNDARY OF PORE FREE ZONE
- ▲ (q/L) BURNUP VERSUS R_m/R_0 , R_m = INNER BOUNDARY OF PORE FREE ZONE

Neg 0694034-6

FIGURE 3.10. Correlation of Heat Rating with Fractional Fuel Melting

TABLE 3.1. Fuel and Test Characteristics and Results

- Pre-Irradiation

Composition	UO ₂ -25 wt% PuO ₂
Oxygen/Metal	1.96 ± 0.01
Density	95% TD
²³⁹ Pu/Pu	87%
²³⁵ U/U	Natural
Diameter	.320 in.
Average Fuel-Clad Gap	.003 in.
Cladding	304L SS
- Irradiation Conditions

Rate of Power Increase During Charging	60% per hr
Rate of Power Decrease During Discharging	3% per second
Time at Steady-State Power	10 hr
Calculated Fuel Surface Temperature at Steady-State	950 °C

- Postirradiation

Pin No.	R _{melt} /R _o , %		Heat Rating, kW/ft		
	R ₁ /R _o	R ₂ /R _o	(Q/L)Max.	(Q/L)End Life	(Q/L) ^(a) Burnup
1	0	0	22	22	26
2	0	0	29	29	29
3	25	17	(b)	(b)	31
4	43	25	31	30	35
5	39	30	32	32	36
6	50	35	33	32	36

R₁ = Extent of PorosityR₂ = Inner boundary of Pore Free Zone

(a) Assuming 186 MeV/fission

(b) Thermocouple failed during irradiation

the isotopes of interest. The measured depression was found to be 0.55 for a $^{235}\text{UO}_2$ -25 wt% PuO_2 fuel with a 0.320-in. diameter, which is 10% less than that calculated using the THERMOS code. Figure 3.11 shows the measured flux profiles for the pins in the calibration capsule. The dashed curves correspond to the width of the samples (0.025 in.). In each case, THERMOS gives a lower flux profile and, therefore, a larger flux depression factor than actually measured.

The result $\int T_m K dT = 43 \text{ W/cm}$, plotted along with the current FTR 950 °C design values in Figure 3.12, shows a 5% increase in the integrated thermal conductivity over the design value.

Fabrication of capsules for studying fuel-sheath gap, heat transfer and porosity, restructuring, irradiation, and stoichiometry effects on fuel heat transfer is in progress (Table 3.2).

The Effects of Xenon on Fuel-to-Cladding Gap Conductivity

G. R. Horn

Mixed-oxide fuel pins are normally fabricated with helium in the fuel-to-cladding gap so that adequate heat transfer rates between fuel and sheath can be attained. This is particularly important in fast reactor fuel pins because they normally operate with very high surface heat fluxes and these high heat fluxes can give rise to very high temperature drops across the gap.

Recently, it has been suggested that some experimental fast reactor fuel pins be fabricated with a mixture of helium and xenon in the gap. In particular, it was proposed that unencapsulated mixed-oxide subassemblies to be irradiated in EBR-II be "tagged" with unique mixtures of xenon isotopes so that, in the event of a fuel element failure, the identity of the failed element could be determined quickly and easily by

**.320 DIA. STAINLESS STEEL
CLAD WITH Zr-II THERMAL DAM**

**.350 DIA. HEAVY WALL
STAINLESS STEEL CLAD**

**.350 DIA. STAINLESS STEEL
CLAD WITH Zr-II THERMAL DAM**

**.376 DIA. STAINLESS STEEL
CLAD WITH Zr-II THERMAL DAM**

Neg 0694034-5

FIGURE 3.11. *Relative Flux Profiles in the Calibration Pins*

Neg 0694034-2
 FIGURE 3.12. Integrated Thermal Conductivity for UO_2 -25 wt% PuO_2 Fuels

TABLE 3.2. Specimen Fabrication Schedule

Capsule Pin	Tests ^(a)				Schedule	
	Gap Mils	Fill Gas	Bulk Density, % TD	O/M	Pin Completion Date	Irradiation Completion Date
GAP CLOSURE KINETICS						
FP-33-1A	2	He	94%	1.96	Completed	10/3/69
1B	11	He	↓	↓	↓	
2A	2	He-33% Xe				
2B	11	He-33% Xe				
3A	2	He-60% Xe				
3B	11	He-60% Xe				
4A	2	Xe				
4B	11	Xe				
FP-37	(same as FP-33)					
DENSITY EFFECTS TEST						
FP-38-1A	2	He	83% Vipac	1.96	12/12/69	1/5/70
1B	↓	↓	83% Pellet	↓		
2A			87%			
2B			94%			
3A			90%			
3B			94%			
4A			90%			
4B			94%			
FP-39			(same as FP-38)			
STOICHIOMETRY EFFECTS TEST						
FP-310-1A	2	He	94%	1.94	10/20/69	11/3/69
1B	↓	↓	↓	2.00		
2A				1.94		
2B				2.00		
3A				1.98		
3B				2.00		
4A				2.00		
4B				2.00		
BURNUP EFFECTS TEST						
Capsule Pin	Variable Parameter					
	Burnup, MWd/MIM					
FP-311-A	0		(b)			
B	40,000		(b)			
FP-3/2 A	0		(b)			
B	80,000		(b)			

(a) Test variables represent target values only

(b) Depends on availability of high burnup fuel

analyzing a sample of the reactor cover gas. Tagging would be accomplished by adding approximately 1 ml of xenon to each fuel pin so that the fill gas composition would be about 10% Xe-90% He. While this technique has definite advantages to the operation of the reactor, there was some concern that the addition of 10% xenon to the fuel pin fill-gas would significantly affect heat transfer characteristics across the gap. Thus, the results of experiments which involved tagged fuel pins might not be typical and might not be directly comparable to the results of experiments which involved nontagged pins. Therefore, an irradiation test was conducted to directly compare in-reactor the heat transfer characteristics in xenon-tagged versus nontagged fuel pins.

Fabrication of Fuel Capsule

Four identical fuel pins were fabricated according to the specifications shown in Table 3.3 and the drawing shown in Figure 3.13. One-hundred percent inspection of the fuel pin components showed that all pellets and pins conformed to the specifications shown.

After assembly and before final closure, each pin was backfilled with a unique mixture of xenon and helium. The initial composition (as determined from postirradiation analysis) of the fill gas in each pin is shown in Table 3.4. The four pins were shrink-fitted into 0.2495-in. diameter cavities in an aluminum capsule which was 2.70 in. OD. The pins were positioned end-to-end in the capsule and were arranged in the following order: Pin 4, Pin 1, Pin 2, Pin 3. The entire capsule assembly was approximately 15 in. long excluding a 20-ft long aluminum tube used as a handle for charging and discharging operations.

FIGURE 3.13 Pins for Comparative Test of Helium-filled Fuel Pins

TABLE 3.3. *Fabrication Parameters for Experimental Pins*

Fuel Form	Sintered Pellets
Fuel Composition	25 wt% PuO ₂ —75 wt% UO ₂
²³⁵ U Enrichment	20 wt% ²³⁵ U in U
Pellet Density	93 ± 0.5% TD
Pellet Diameter	0.2135 ± 0.0005 inch
Fuel O/M	1.97 ± 0.02
Fuel Column Length	2.0 inches
Cladding Material	304 SS
Cladding ID	0.2195 ± 0.0005 inch
Cladding OD	0.250 ± 0.0003 inch

TABLE 3.4. *Initial Composition of Fill-Gas in Each Pin*

<u>Pin No.</u>	<u>Fill-Gas</u>
1	100% Xe
2	38% Xe - 62% He
3	60% Xe - 40% He
4	100% He

Irradiation

The capsule was irradiated in a horizontal position in a "rabbit" type irradiation test facility. It was inserted over a period of 1 to 2-min while the reactor was at full power and was irradiated 4 hr. During irradiation, the capsule was positioned in a region within the reactor where the flux is extremely constant over a considerable length (approximately 6 ft); therefore, all pins in the capsule operated at essentially the same heat rating. Postirradiation burnup analyses indicated that the pins operated at 32.5 ± 1.5 kW/ft. After irradiation, the capsule was withdrawn

from the reactor over a period of approximately 1 min. After 1 day of cooling, it was transferred to the Radiometallurgy Laboratory for destructive examination.

Results

The pins were removed from the aluminum capsule and the gas from each pin was collected and analyzed to determine the gas compositions that prevailed before irradiation. The results of these determinations were shown previously in Table 3.4.

Each pin was sectioned approximately 1 in. from the bottom of the fuel column (series a) and then again approximately 1/2 in. from the bottom of the fuel column (series b). Photographs and autoradiographs of these cross-sections are shown in Figures 3.14 and 3.15. Only three cross-sections are shown for series "a" because the ceramography sample from Pin No. 3 was inadvertently damaged and could not be reclaimed. The samples from series "b" were polished and etched and a 75X photomicrograph mosaic was obtained for each cross-section. Reproductions of these mosaics are shown in Figure 3.16.

Measurements were made on the mosaics and on enlargements of the photographs and autoradiographs to determine the radii to which various structural features extended in each of the cross-sections. The results of these measurements are summarized in Table 3.5 and are presented graphically in Figure 3.17. Both the melt radius and the grain growth radius represent isotherms in the fuel; it is therefore apparent from Figure 3.18 that temperatures within the fuel increased with increasing xenon content in the fill gas.

A burnup sample which included approximately 1/2 in. of fuel column was removed from each pin and analyzed. The results of these analyses, which are summarized in Table 3.6, indicated that the fuel pins operated at a heat rating between 31 and 34 kW/ft.

FIGURE 3.14. *Cross-Sections from Series A*

Pin #1
(100% Xe)

Pin #2
(38% Xe)

Pin #3
(60% Xe)

Pin #4
(100% He)

Photographs β - γ α
 Autoradiographs Autoradiographs

FIGURE 3.15. Cross-Sections from Series B

Pin No. 1

Pin No. 2

Pin No. 3

Pin No. 4

FIGURE 3.16. *Photomicrograph Mosaics of Cross-Sections from Series B*

TABLE 3.5 *Fraction of the Fuel Radius to Which Various Structural Features Extended(a)*

	Pin No. 4 (100% He)		Pin No. 2 (38% Xe-62% He)		Pin No. 3 (60% Xe-40% He)		Pin No. 1 (100% Xe)	
	Series A	Series B	Series A	Series B	Series A	Series B	Series A	Series B
Sub-Grain Structure (From beta-gamma Autoradiograph)	0.264	0.339	0.288	0.287	(b)	0.302	0.307	0.299
Fuel Melting (From Photomosaic)	(b)	0.506	(b)	0.517	(b)	0.535	(b)	0.626
Fuel Melting (From beta-gamma Autoradiograph)	0.498	0.520	0.540	0.535	(b)	0.567	0.634	0.642
Grain Growth (From Macro)	0.888	0.909	0.911	0.919	(b)	0.932	0.946	0.955

(a) Each point is an average of measurements made along at least four diameters for each photograph.
(b) Not available

FIGURE 3.17. *Radii to Which Various Structural Features Extended*

FIGURE 3.18. *Calculated Melt Radius as a Function of Gap Conductivity*

TABLE 3.6. *Burnup Analyses of Xenon Tagging
Experiment Fuel Pins*

Pin No.	Burnup (fission/ gram) as determined from ^{140}Ba - ^{140}La Concentrations After Short Cooling Time	Burnup (fission/gram) as determined from ^{141}Ce Concentrations After Long Cooling Time
1	2.16×10^{17}	2.10×10^{17}
2	2.30×10^{17}	2.06×10^{17}
3	2.25×10^{17}	2.14×10^{17}
4	2.34×10^{17}	2.07×10^{17}

Temperature profiles within the capsule were calculated for various fuel-to-cladding gap conductivities assuming a range of linear heat ratings from 31 to 34 kW/ft. From these profiles, Figure 3.18 which shows calculated melt radius as a function of fuel-to-cladding gap conductivity for a range of heat ratings, was constructed. By comparing the calculated melt radii to the observed values, the gap conductivity in each pin was determined as a function of heat rating. These values are summarized in Table 3.7 and in Figure 3.19. It is apparent from Figure 3.19 that this experimental technique does not yield precise absolute values for gap conductivities. Even though heat generation rates were known to $\pm 5\%$ and the melt radii were measured to $\pm 2\text{-}1/2\%$, derived gap conductivities varied over wide ranges.

A more meaningful treatment of these data was achieved by determining relative values of fuel-to-sheath heat transfer coefficients and in this manner canceling out systematic or consistent uncertainties that exist in the data. The post-irradiation burnup analyses showed that the heat generation rate for these pins was between 31 and 34 kW/ft. Previous measurements of fluxes within the irradiation facility have shown that heat generation rates should be essentially constant over the entire length of the facility. Therefore, although the absolute value for the heat generation rate was

FIGURE 3.19. Gap Conductivity as a Function of Gas Composition

TABLE 3.7. *Gap Conductivity in Each Pin as a Function of Heat Rating*

Assumed Heat Rating, (kW/ft)	Pin 4 (0% Xe— 100% He)	Pin 2 (38% Xe— 62% He)	Pin 3 (60% Xe— 40% He)	Pin 1 (100% Xe— 0% He)
31	2370-2540	2230-2400	2070-2300	1610-1710
32	2760-2980	2590-2800	2380-2670	1820-1930
33	3230-2560	3010-3290	2750-3120	2020-2170
34	3870-4190	3560-3950	3200-3710	2310-2470

not precisely known, it was known that it was essentially the same for every pin. On this basis, Table 3.8 and Figure 3.20 were constructed to show the relative amount by which gap conductivity decreased as xenon content of the fill gas increased. As Figure 3.20 shows, the addition of 10% xenon to the helium fill gas would decrease gap conductivity by less than 4%, while complete substitution of xenon for helium would decrease gap conductivity at most by 45%.

The results of this experiment demonstrated that the addition of xenon to the helium fill-gas in fast reactor fuel pins will cause a decrease in the fuel-to-cladding gap conductivity. However, the decrease is sufficiently small to be insignificant with respect to operating fuel elements. Experimental fuel pins can be tagged with 10 to 15% xenon without degrading the heat transfer characteristics to any significant extent.

The data indicate that gap conductivity could be decreased by 4% by xenon tagging experimental fuel pins in the EBR-II. Figure 3.21 shows the calculated temperature profile in a fast reactor pin operating at 15 kW/ft $\pm$ 5%. If the gap conductivity for that fuel pin was decreased by 4%, center temperature would be decreased by about 15 °C.

FIGURE 3.20. *Decrease in Gap Conductivity as a Function of Xenon Content of the Fill Gas*

FIGURE 3.21. *Calculated Temperature Profile in Fast Reactor Fuel Pin Operating @ 15 kW/ft $\pm$ 5%*

TABLE 3.8. *Decrease in Gap Conductivity as a Function of Xenon Content*

Assumed Linear Heat Rating kW/ft	100% He— 0% Xe			62% tie— 38% Xe			40% He— 60% Xe			0% He— 100% Xe		
	h_g	%	Dec.	h_g	%	Dec.	h_g	%	Dec.	h_g	%	Dec.
31	2370-	0		2230	0-		2070	3.0-		1610-	27.8-	
	2540			2400	12.2		2300	18.5		1710	36.6	
32	2760-	0		2590-	0-		2380-	3.3-		1820-	30.1-	
	2980			2800	13.1		2670	20.1		1930	39.0	
33	3230-	0		3010-	0-		2750-	3.4-		2020-	32.8-	
	3560			3290	15.4		3120	22.8		2170	43.3	
34	3870-	0		3560-	0-		3200-	4.1-		2310-	36.2-	
	4190			3950	15.1		3710	23.7		2470	45.0	

h_g = Gap Conductivity (Btu/hr ft² °F)

% Dec. = % Decrease Compared to Pure Helium

If the calculated temperature profile curve for the lower gap conductivity case was superimposed on the curve shown, the two would be nearly indistinguishable. It is apparent from this that uncertainties in heat generation rates are of far more significance in interpreting test results than the slight decrease in gap conductivity caused by adding xenon.

A further illustration of the insignificance of the xenon tag is shown in Figure 3.22 which shows the calculated xenon content in the fill-gas of a typical fast reactor fuel pin as a function of irradiation time assuming a 20% gas release rate. It is apparent from this figure that in a nontagged fuel pin the xenon content of the fill-gas will reach 10% within the first 3-1/2 days of irradiation.

The absolute values for gap conductivity derived from this experiment may be higher than one would expect in a

FIGURE 3.22. *Xenon Content of Helium Fill Gas as a Function of Irradiation Time Assuming Gas Release Rate is 20%*

typical fast reactor fuel pin operating under typical conditions. Since it was necessary to operate these pins with some center melting, thermal expansion of the fuel was such that a slight interference fit between pellets and cladding may have existed during operation. Therefore, instead of the hot gap that would normally be expected in typical fast reactor fuel pins, these pins apparently operated with fuel

in contact with the cladding. This effect does not adversely alter the basic conclusion drawn from this experiment. Studies at other sites^(1,2,3) indicate that if a difference in gap conductivity exists when the two surfaces are in contact, then a proportionate difference will exist when there is no contact. In particular, Ross and Stoute⁽³⁾ showed through a semi-empirical approach that gap conductivity is dependent on fill gas thermal conductivity even when the two surfaces are under pressure. Their derived relationship did show, however, that the dependency on fill-gas thermal conductivity decreases as interfacial pressure increases.

There are large uncertainties involved in determining absolute values of gap conductivity by the technique used in this experiment. Small uncertainties in heat generation rates, heat transfer properties of the capsule, and/or thermal properties of the fuel, lead to large uncertainties in derived values for gap conductivity. Furthermore, parameters that are difficult to control to a high degree such as manufacturing tolerances, and gas content of the fuel can mask the effects of other parameters being studied. Therefore, absolute values of gap conductivity determined by this technique are inherently quite imprecise. Conversely, the comparative values shown in Figure 3.20 represent a high degree of precision because the effects of all consistent or systematic uncertainties in the system were cancelled out. The same fuel and cladding was used for all pins, heat generation rates were constant over the length of the capsule, and if errors were made in determining heat transfer properties of the capsule, the same errors were made for all pins.

THERMAL FLUX IRRADIATIONS OF HYPOSTOICHIOPETRIC $\text{UO}_2\text{-PuO}_2$

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D. Christensen, E. D. Jensen, and D. W. Dragnich

The general status of the fuel pins being irradiated and/or examined are summarized in Table 3.9. The reported burnup values were derived from ^{148}Nd analyses of sections removed from the center of the fuel pins. The burnup values for those pins not yet examined were estimated from the reported fluence, assuming a linear relationship between fluence and burnup. Two of these capsules, BNW-1-10 and -12, are still undergoing irradiation to a goal exposure of 100,000 MWd/MTM. The other ten capsules have either completed their postirradiation examination or are in the process of being examined.

A complete fabrication history is being assembled for each of the BNW-1 experiments. This is to include analyses of the fuel from the powder stages through to the fuel pin, the processes involved, certification of capsule components, complete assembly details and all preirradiation measurements.

Irradiation histories are also being compiled for each experiment. Plots of temperature versus time are being studied to correlate the thermocouple readings to temperatures obtained from the Tiger V computer program analyses.

Postirradiation Examination of BNW-1-1, 1-2, 1-3, and 1-5Fuel Pin Recovery

Pins BNW-1-1, 1-2, 1-3, and 1-5 were recovered from their respective capsules in the BNW hot cells, cleaned in Dowanol and water, visually examined and photographed. The four pins were in excellent condition with no indication of a breach in the cladding. Sampling the inner capsules for fission gas was negative which supported the indication that all pins were intact. All pins were tightly bound inside this inner capsule.

TABLE 3.9. Summary Status of Irradiation Experiments Conducted Under Task FP-6

Test Designation	Description ⁽¹⁾ Fuel Form O/M		Facility	Status			
				Goal Exposure, MWd/MTM	Actual Exposure, MWd/MTM	Discharge Date	
BNW 1-1	Solid	1.93	MTR	100,000	(Est) 120,000	2-69	Being Exam. at ENW
BNW 1-2	Solid	1.93	ETR	50,000	(Est) 70,000	2-69	Being Exam. at BNW
BNW 1-3	Solid	1.96	MTR	100,000	(Est) 150,000	2-69	Being Exam. at BNW
BNW 1-4	Solid	1.96	MTR	50,000	71,000	6-68	Exam. completed at GNW
BNW 1-5	Solid	2.00	MTR	100,000	(Est) 160,000	2-69	Being Exam. at BNW
KNW 1-6	Solid	2.00	MTR	50,000	75,200	10-68	Exam. completed at BNW
BNW 1-7	Annular	1.93	MTR	50,000	<1,000		Lead tube failed. Exam. at BNW
BNW 1-8	Annular	1.93	MTR	100,000	29,000	1-68	Failed. Exam. at BNW
BNW 1-9	Annular	1.96	MTR	50,000	81,800	6-68	Exam. completed at LASL
BNW 1-10	Annular	1.96	CTR	100,000	(Est) 80,000	(Est) 2-70	Discharged 2-69 for interim exam. Recharged 5/69
BNW 1-11	Annular	2.00	MTR	50,000	87,400	8-68	Exam. completed at LASL
BNW 1-12	Annular	2.00	ETR	100,000	(Est) 84,000	(Est) 2-70	Discharged 2-69 for interim exam. Recharged 5/69

(1)

Fuel

25% PuO₂-75% UO₂

Pellets 0.212 in. OD (0.052 in. ID for annular pellets

Smeared density - 87.5% TD)

Pellet density - solid pellet 93% TD, annular pellet 95% TD

Fuel column length 13.5 in.

Cladding

OD - 0.250 in.

Wall thickness - 0.016 in.

304 SS

Thus, it was necessary to slit the inner capsules longitudinally in order to remove the pins without damaging them.

Profilometry Measurements

After being photographed at 0°, 120°, and 240° along the entire length, pins BNW-1-1, 1-2, 1-3, and 1-5 were measured with the profilometer. Diameters were measured continuously along the longitudinal axis at 0°, 45°, 90°, and 135° around the circumference. Figures 3.23, 3.24, 3.25, and 3.26 show the diameter changes at the fuel pin cladding plotted as deviations from the nominal preirradiation diameter of 0.250 in. The diameter at 180° was plotted from the original traces to verify the 0° curve shape.

Summary results from analysis of the profilometry data are shown in Table 3.10.

TABLE 3.10. BNW-1 Cladding Diameters

Pin No.	Maximum Cladding OD, (a) in.	Maximum Diameter Increase Compared to the Plenum	
		In.	$\frac{\Delta D}{D}$ %
GNW-1-1	0.2518	0.0015	0.6
BNW-1-2	0.2578	0.0079	3.1
BNW-1-3	0.2505	0.0	0.0
BNW-1-5	0.2564	0.0064	2.6

(a) Maximum from the average of four diameter measurements around circumference at 0 degree, 45 degrees, 90 degrees, and 135 degrees made at 1 in. increments along the longitudinal axis.

The curves in the figures show that the diameter increase occurred on BNW-1-1, 1-2, and 1-5 near the bottom of the fuel. The curves for BNW-1-3 show diameter increases and decreases at different axial locations for each of the

FIGURE 3.23. Plot of Diameter Measurements BNW-1-1

FIGURE 3.24. Plot of Diameter Measurements BNW-1-2

Lot of Diameter Measurements BNW-1-3

FIGURE 3.26. Plot of Diameter Measurements BNW-1-5

circumferential angles. Comparison of curves 90 degrees apart on BNW-1-3 indicates the pin cladding has assumed an ovality with the major and minor axis changing angular position at several locations along the longitudinal axis. There is no significant increase in the diameter at any axial location calculated by averaging the diameters at the four circumferential positions. In the comparison of maximum diameter increases on a basis of the estimated fuel exposure, Table 3.9 does not show a consistent behavior with exposure. However, much more data on the thermal and power history as well as the postirradiation examination results (including burnup analyses) of these pins must be analyzed before valid comparisons can be made.

Close examination of the fine structure on the original recorder chart traces of the diameters on BNW-1-5 showed a consistent pattern of small diameter increases spaced approximately equal to the preirradiation fuel pellet length. An attempt is being made to correlate these diameter changes with the surface structure on a positive replica made of this pin and with the pellet-to-pellet interfaces and pellet crack structure observed in postirradiation neutron radiographs.

Gamma Scanning

Axial gamma scans for gross activity and for $^{95}\text{Zr}/^{95}\text{Nb}$ have shown several activity peaks on BNW-1-1, 1-2, 1-3, and 1-5. Spectra data collected along the length of the fuel pins including several directly on the activity peaks are being analyzed with a computer program GSSLRN (Guassian-Learn) to determine the major gamma emitting isotopes present. The activity peaks on the gross gamma scans corresponds to axial positions on the fuel pin neutron radiographs where high density material, presumably fission product ingots, can be seen in the central voids of the fuel. Preliminary analysis

from the BNW-1-1 spectra data on the computer program indicates that the fission product ruthenium is the major contributor to the activity peaks along the fuel except for one approximately one mil from the top of the fuel column. The spectra analysis indicates that cesium is a major activity contributor at this position. There is an indication that cesium has migrated more generally to the top of the fuel pin including into the plenum region. Analysis of the gamma scan data and spectra is continuing.

Fission Gas Sampling

Three of the four pins (BNW-1-1, 1-2, and 1-3) have been sampled for fission gas; volumes of gas collected were 184, 138, and 166 cm³ (STP), respectively. These are fairly consistent with the burnups estimated in Table 3.9 if 70 to 80 percent release is assumed. The gas is being analyzed isotopically. The fourth pin, BNW-1-5, will be sampled next quarter.

Shielded Electron Microprobe Examination

Transverse sections of irradiated specimens BNW-1-7 and BNW-1-8 have been examined in the shielded electron microprobe. Composition and irradiation data for these samples are summarized in Table 3.11.

BNW-1-7. A specimen from pin BNW-1-7 was examined for distribution of Plutonium and Uranium across the pin. Four radial scans of X-ray intensity of plutonium and uranium, 90° apart, showed plutonium-to-uranium ratios of 0.23 to 0.26 across the pin (neglecting absorption and fluorescence corrections). No

TABLE 3.11. *Composition and Irradiation Data for Samples Studied in the Shielded Electron Microprobe*

	<u>BNW-1-7</u>	<u>BNW-1-8</u>
Percent PuO ₂	25	25
Percent of U as ²³⁵ U	0.7	0.7
Exposure (MWd/T)	300	29,000
Linear Power Rating, kW/ft	14.6	21.5
Cooling Time (Months)	21	18
Reactor	MTR	MTR
Fuel Surface Temp, °C	850-900	1100-1200
Centerline Temp, °C	~2100	~2700
Fuel O/M Before Irradiation	1.93-1.94	1.93-1.94
Pellet Density, % TD	93	93
Radiation Level at 1 ft R/hr	1.8	80

consistent trend was apparent among the four scans. Many voids and cracks were present, resulting in output records similar to Figure 5.13C of BNWL-1058.*

The relative distribution of uranium and plutonium over an 80 × 80 micron area was determined and is illustrated in Figure 3.27. The electron probe is scanned over the sample at such a rate that individual X-ray pulses can be displayed on the oscilloscope screen. A time exposure of the screen during one complete scan shows a varying density of "dots" or pulses. The density of pulses in any area is proportional to the concentration of the element being excited. Figure 3.27 shows three areas of lower plutonium concentration while the corresponding area for the uranium concentration shows normal or possibly slightly higher uranium concentration.

* *Quarterly Progress Report, Jan., Feb., Mar. 1969, "Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology," by A. L. Bement, G. J. Dau, E. A. Evans, and R. E. Nightingale.*

FIGURE 3.27. *Distribution of Plutonium and Uranium Over 80×80 Micron Region of BNW-1-7. Concentration of Element is Proportional to Density of Dots. Both Photos are of the Same Area.*

The distribution of uranium, plutonium, and iron in an area near the outer fuel surface, superimposed on the specimen current, is shown in Figure 3.28. At the magnification used (300X), uranium and plutonium concentrations appear quite uniform. Isolated dots in the crack are due to the background from the radioactive sample. The background is worse for plutonium than uranium or iron. The background radiation is a serious problem at diffraction angles less than that for the plutonium M_a lines and also for much hotter samples. Iron was observed in cracks and voids, but not in the matrix of the sample. This could possibly be polishing debris arising from sample preparation. No fission product elements were observed.

*Uranium and Specimen
Current*

*Plutonium and Specimen
Current*

*Iron and Specimen
Current*

FIGURE 3.28. *Distribution of Uranium, Plutonium, and Iron over a 265 x 265 Micron Area of BNW 1-7. X-Ray Output (Pulses) is Superimposed on Specimen Current Photograph. All Three Photos are the Same Area.*

BNW-1-8. Figure 3.29 shows a mosaic of a sample BNW-1-8 used for microprobe analysis. The sample was polished with nonaqueous reagents to get full retention of fuel-NaK and fuel-cladding reaction layers but was not etched. The gross staining observed in the interior of the fuel is artifact and should be ignored. The inclusions in the central void are aluminum particles from a conductive resin used to mount the sample. This sample failed during irradiation and liquid sodium-potassium (NaK) alloy entered the pin. The gray annular region just inside the cladding is the fuel-NaK reaction layer. Between the fuel-NaK reaction layer and the cladding is a region of fuel-cladding reaction layer which appears as a strip of cladding with some structure to it. Between the fuel-NaK reaction layer and the fuel is a crack in which inclusions occur in some areas. A few small inclusions are scattered primarily among the columnar grains. A semicircular track half-way between the center and the cladding marks the boundary between the once-molten fuel zone and the columnar grains.

Plutonium was analyzed continuously from the cladding to the central void across eight separate radii. All eight of these scans were characterized by similar plutonium distributions. The fuel-NaK reaction layer shows increasing plutonium concentration toward the pellet center, with one or more sharp spikes of much higher plutonium content than adjacent areas. From the fuel edge of the transition region between columnar grains and unchanged fuel, the plutonium concentration increases slightly and then decreases slightly. A decrease of plutonium concentration by nearly a factor of two, then a subsequent rise of the same amount, both occurring over about 100 microns characterizes a transition region before the columnar grains. On entering the columnar grain region, a uniform rate of increase of plutonium is seen. The region from a point half-way through

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1000 Microns

FIGURE 3.29. *Mosaic of Sample of BNW-1-8 Used for Microprobe Analysis. Arrow Indicates Path of Scan of Figure 4. Specimen was polished to retain fuel-NaK and fuel-cladding reaction layers. This technique produced staining in the interior regions of fuel which should be ignored.*

the columnar grain region to the inner edge of the columnar grains is characterized by constant X-ray counting rate, but due to increasing radiation background, the plutonium concentration decreases slightly. The innermost region, the once-molten fuel, shows a sudden increase of about 10% in plutonium concentration. In some cases, a plutonium spike of about 10% higher concentration occurs at the edge of this region and more spikes of about a 10% increase occur near the central void. Figure 3.30 is the plutonium concentration as a function of the radial fraction r_x/R , defined as the ratio of the distance from the center to the inside radius of the cladding. The ordinate shows the relative distribution of plutonium.

Considering the fuel-NaK reaction layer and fuel interface, the ratio of plutonium-to-uranium appears very nearly the same in both regions. However, both the uranium and the plutonium appear about 10% more concentrated in the fuel. This would be expected if the reaction layer had a lower density than the fuel. This also indicates that whatever process caused the fuel-NaK reaction layer, affected uranium and plutonium to the same extent. X-ray photographs of uranium and plutonium distribution in the fuel-NaK reaction layer are given in Figure 3.31 superimposed on the specimen current. The uranium appears to be fairly uniformly distributed, while plutonium shows a distribution over the same area as uranium but also shows a series of bands of higher plutonium content.

Examination of the wavelengths of X-rays from the fuel-NaK reaction layer showed only lines caused by uranium and plutonium. No sodium or potassium was detected. This could be due to instrumental problems or to preparation techniques. Further efforts will be made to detect these elements.

Neg 0692855-1

FIGURE 3.30. Plutonium Concentration cross one Radius of BNW 1-8

Uranium

Plutonium

Plutonium

FIGURE 3.31. *Uranium and Plutonium Distribution in the Fuel-Nak Reaction Layer of BNW-1-8.*

Some inclusions were found between the fuel-NaK reaction layer and the fuel. One such area is shown in Figure 3.32 with specimen current, iron, nickel, and silver X-ray distributions shown. Ruthenium was also found in these inclusions in small amounts. Silver was found to be present to the extent of about 20% in one inclusion. The occurrence of silver in these particles was verified by the $L\alpha_1$, $L\beta_1$, $L\beta_2$, and $L\gamma_1$ lines at the proper wavelengths.

One inclusion in the matrix was examined for the identity of elements in it. A wavelength scan was made using a LiF crystal with a sealed proportional counter (xenon) and a PET crystal with a flow proportional counter (P10). The scan detected uranium and plutonium, but it could not be determined whether the elements were in the matrix or the ingot. The major element present was ruthenium, with some silicon. No molybdenum or technetium was observed.

The fuel-cladding reaction layer was examined for component translocation and presence of cesium. Figure 3.33 shows traces of iron, nickel, and chromium redrawn on the same chart. The abscissa of each component was adjusted so the low point of the major trough was in the identical position for each element. This trough appears to be the boundary between unchanged cladding and the fuel-cladding reaction layer; therefore, its use as a reference point is justifiable. To the right of this line is unchanged cladding, and no variations in any component are apparent. To the left of this trough the chromium and nickel have the same composition as the cladding for about 15 microns, then drop off rapidly. Iron concentration remains the same as the cladding up to the crack, with a high concentration of iron on the far side from the cladding, followed by a sudden decrease. No cesium was detected in the cladding or near it.

Specimen Current

Iron

Nickel

Silver

FIGURE 3.32. *Specimen Current, Iron, Nickel, and Silver Distribution Between Fuel and Fuel-Nak Reaction Layer of BNW-1-8.*

FIGURE 3.33. Counting Rate of Iron, Nickel and Chromium Across Clad and into Fuel-Clad Reaction Layer. Sample BNW-1-8

More refined preparation techniques are being developed in order to better analyze these reaction layers. Additional sections are being prepared from BNW-1-8, BNW-1-6, and BNW-1-4.

FUEL SWELLING AND ANALYTICAL MODELING

K. R. Merckx, J. A. Christensen

Swelling Measurements

F. E. Panisko

An irradiation experiment is being designed to determine effects of fuel density, temperature, temperature gradient, and burnup on irradiation swelling of UO_2 -25 wt% PuO_2 .

The irradiation capsule (Figure 3.34) consists of 36 duplicate fuel wafers with fuel of 93 and $\sim 100\%$ theoretical densities. These specimens will be cut from 0.250 in. diameter sintered and hot-pressed pellets. Bulk fuel temperatures will range from 700 to 1200 in half of the specimens and from 1200 to 1700 °C in the remainder. Temperature gradients of 0, 1000, and 4000 °C/cm grid burnups of 20,000, 50,000, and 80,000 MWd/T are being considered. A nominal external pressure of ~ 100 psi will be applied to the capsule.

Metallographic measurements of pore size and distribution will constitute the principal method of fission gas swelling characterization. Dimensional and bulk density measurements will also be made if specimens remain intact.

Stochastic Modeling

K. R. Merckx

The near-term objective of this task is to develop the quantitative procedure for estimating the parameters in a fuel swelling model using observations made on fuel microstructures. Work has been done with the general stochastic models and their interpretations to show that the predictions of bubble migration and growth can be directly interpreted in terms of the existing

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FIGURE 3.34 Fuel Swelling Capsule Schematic

computer techniques used to estimate fuel performance. These interpretations were carried out for the CYGRO and the DEFORM programs. The displacement rate due to a migration velocity, β , is

$$\dot{u}_m = \frac{4\pi}{3} \int_0^S \max s^3 \beta(s) n(s) ds$$

and the dilation due to the growth rate, A , is

$$\dot{\epsilon}_g = \frac{4\pi}{3} \int_0^S \max \lambda s^2 n(s) ds$$

If the bubble migration causes only radial displacements and the growth causes an isotropic dilation, the resultant displacement rates in the CYGRO finite element program are for migration are

$$\dot{D}_E^M = \begin{pmatrix} \dot{u}_m (R_K = 1) \\ \dot{u}_m (R_K) \\ 0 \end{pmatrix}$$

and for growth are

$$\dot{D}_S^G = \begin{pmatrix} \dot{\epsilon}_g \\ \dot{\epsilon}_g \\ \dot{\epsilon}_g \\ \dot{\epsilon}_g \\ \dot{\epsilon}_g \end{pmatrix}$$

where

$$\dot{D}_S = \begin{pmatrix} \dot{\epsilon}_r \\ \dot{\epsilon}_r \\ \dot{\epsilon}_\theta \\ \dot{\epsilon}_\theta \\ \dot{\epsilon}_z \end{pmatrix} \quad \text{and} \quad \dot{D}_E = \begin{pmatrix} \dot{r} (R_K = 1) \\ \dot{r} (R_K) \\ \dot{\lambda} (L) \end{pmatrix}$$

The corresponding interpretation of the steady-state model used in the DEFORM program is that the radial displacement is

$$\begin{aligned} \dot{u} = & \left(\frac{b^2}{r} \right) \left[\frac{\dot{b}}{b} + \left(\frac{\dot{\epsilon}_z}{2} \right) + 1 - \left(\frac{r}{b} \right)^2 - \int_{r/b}^1 \dot{\epsilon}_g \rho d\rho \right] \\ & + \dot{u}_m - \frac{b}{r} \dot{b}_m . \end{aligned}$$

In this approach the migration strain rates are given by equations

$$(\dot{\epsilon}_\theta)_m = \frac{\dot{u}_m}{r} ; (\dot{\epsilon}_r)_m = \frac{d\dot{u}_m}{dr} ; (\dot{\epsilon}_z)_m = 0$$

and the viscous strain rate is given by the equation

$$(\dot{\epsilon}_i)_v = \dot{\epsilon}_i - (\dot{\epsilon}_i)_m - \dot{\epsilon}_g = \frac{S_i}{2\eta} .$$

This development shows that the results of this type of modeling can be directly used in present fuel design procedures.

MIGRATION BEHAVIOR OF UO_2 - PuO_2 FUELS

T. D. Chikalla

Shielded Electron Microprobe Examination of Irradiated Fuel

(Transport of Components in a Temperature Gradient)

E. D. Jenson

The occurrence of metallic deposits containing iron, nickel, and chromium in the fuel of irradiated mixed oxide pins has been observed by shielded electron microprobe examination and has prompted a search into the source of these metals. Since the composition of the deposits is quite different than the stainless steel cladding and is variable, a selective mechanism of transport seems to be operative as opposed to particles of stainless steel from the clad or process equipment. A plot of iron, nickel, and chromium concentration

versus distance from the clad along a single large crack is given in Figure 3.35a. Since the electron beam penetrates thin particles, the observed compositions may be low, but the ratio of two elements analyzed simultaneously will not be affected. A plot of iron-to-nickel and another of chromium-to-nickel ratio versus distance is given Figure 3.35b. In these figures, the metallic particles beyond 650 microns were large and probably were not completely penetrated by the electron beam. The remainder of the particles were small and the beam probably penetrated to the oxide matrix and caused the discontinuity observed at 650 microns. The linearity of the iron-to-nickel ratio with distance from the clad is surprising and, together with Figure 3.35a indicates a smooth change in the rate at which these elements were deposited. It is apparent that the maximum rate of deposition of iron and nickel occur at different areas along the crack. Nickel deposited most rapidly at or beyond 800 microns. The area of maximum iron deposition rate cannot be defined, but is under 650 microns and may be near the clad.

Table 3.12 gives the iron, nickel, and chromium composition of other metallic deposits observed and their distance from the clad. None of these were in radial cracks visible on the surface examined but may have been in cracks running longitudinally at that point.

TABLE 3.12. Iron, Nickel, and Chromium Concentration of Metallic Deposits Not in Definable Radial Cracks

Particle Number	Distance from Clad (microns)	Percent Composition		
		Fe	Cr	Ni
1	660	29.4	0.7	2.2
2	300	63.5	0.8	20.8
	475	43.4	1.1	22.8
	650	42.1	1.1	24.8
	800	38.3	1.0	22.9
3	875	32.7	0.4	25.8
	Clad	67.0	17.6	10.4

Particle 2 was a single elongated deposit while the other two were small circular deposits.

FIGURE 3.35. a. Iron, Nickel and Chromium Composition of Deposits in Fuel Crack Versus Distance from Clad
 b. Iron-to-Nickel and Chromium-to-Nickel Ratios in Deposits in Fuel Crack Versus Distance from Clad

It is apparent that the composition is quite variable, with a chromium content of about 1% or less, while the nickel content varies from as little as 2% to two and a half times as much as in the clad itself. The nickel content was still increasing at the particle furthestmost from the clad. A general trend of lower iron content with greater distance from the clad is evident.

Workers at Argonne National Laboratory⁽⁴⁾ have proposed that fission product iodine is the transporting agent. Iodine formed in the fuel is transported to the cooler clad, reacts with the clad components to form gaseous metal iodides, and the iodides migrate to a region of lower chemical potential containing less metal iodide. Somewhere along the path up the temperature gradient, temperature and concentration effects combine to produce conditions such that decomposition of the iodides occurs, resulting in a deposit of the metal and releasing the iodine to participate in further metal transport.

The transport mechanism is unknown, but is assumed to be diffusion down the chemical potential gradient. Assuming a diffusion mechanism, the rate of transport should be proportional to the saturation vapor pressures. The vapor pressures of iron and chromium iodides at 1000 °K are 700 mm⁽⁵⁾ and 0.2 mm,⁽⁶⁾ respectively. Since chromium iodide has a much lower vapor pressure than iron iodide, its rate of transport should be much less and this may be responsible for the very small amount of chromium deposited far from the clad compared to iron and nickel.

A competing reaction which would tend to minimize the iodine available for such a process is reaction of fission product cesium with fission product iodine. Depending on the relative concentrations of Cs and I, as well as the stability of CsI, it is possible that Cs could completely tie up the iodine.

Calculations⁽⁷⁾ of the Cs and I concentrations in UO_2 -25 wt% PuO_2 after 50 days irradiation at a power level of 8.3 kW/ft and a burnup of 7850 MWd/T give results shown in Table 3.13.

TABLE 3.13. Total Atom Concentrations of Cs and I Isotopes Having Half-Lives Greater Than the Times Given After 50 Days Irradiation of UO_2 -25 wt% to 7850 MWd/T.

Element	Atom Concentration ($\times 10^{-18}$) atoms/cm ³		
	Half-Life Greater Than		
	1 hour	1 day	10 days
I	141.0	116.0	3.3
Cs	53.1	53.1	53.1

It is apparent that the Cs concentration is practically constant, but that iodine is decaying quite rapidly. Also, the iodine isotope concentration exceeds the cesium concentration by about a factor of 2 to 3 until only isotopes of half-lives greater than 10 days are considered. Considering iodine isotopes with a half-life greater than 10 days eliminates ^{131}I , which has a half-life of 8.05 days and a concentration of 113×10^{18} atoms/cm³ after 50 days irradiation. It appears therefore that the iodine concentration exceeds the cesium concentration considerably on a time scale reasonable with respect to the processes likely to be involved in transporting clad components into the fuel.

Thermodynamic data on the reaction

indicate the reaction should proceed toward formation of $\text{CsI}_{(g)}$ and therefore remove some of the iodine from circulation.

Free energy data for the above reaction are given in Table 3.14. (Calculations indicate that the majority of iodine should be monatomic at the temperatures of interest.)

SABLE 3.14. Free Energy of Formation of $\text{CsI}_{(g)}$ from Cs Vapor and Atomic I.

Temperature, °K	ΔG° Formation, kcal/mole
500	-72.8
1000	-62.8
1500	-52.4
2000	-42.0

However, other reactions may remove Cs before it can remove I, such as formation of cesium uranate, which appears to be thermodynamically reasonable⁽⁸⁾ in this temperature range.

Assuming that iodine is the transporting agent, the thermodynamics of formation and decomposition of metal iodides give some information on the conditions necessary for deposition. For the reaction

the equilibrium conditions is

$$K = \frac{P_{\text{MI}_{n(g)}}}{a_{\text{M}_{(s)}} P_{\text{I}_{(g)}}^n} \quad (3)$$

where a is the activity and the fugacities have been approximated by the partial pressure, p_i . The stainless steel clad is a solution and the activity of its components must be included in the calculation. The driving force to form a metal iodide can be seen to be the lack of $\text{MI}_{n(g)}$ at the surface initially, or its removal due to the transport process, allowing more to form and approach the equilibrium condition. The activity of the metal component decreases the MI_n concentration from that expected from the pure metal, as does the partial pressure of the gaseous I. During the transport process, metal can be deposited initially all along the path since the partial pressure of the transported

MI_n will exceed zero. Once metal is deposited along the path, more metal can deposit if the pressure of the transported MI_n exceeds that defined in Equation 4 at that point.

$$p_{MI_n(g)} = K \left[a_{M(s)} p_{I(g)}^n \right] \quad (4)$$

Requiring that the transport process of $MI_{n(g)}$ is operative to equate the chemical potential, μ_{MI_n} , of the metal iodides at two points leads to the expression

$$\mu_2 - \mu_1 = \mu_2^* - \mu_1^* + RT_1 \ln \left(\frac{p_2}{p_1} \right)^{T_2/T_1} \quad (5)$$

where the subscripts 1 and 2 refer to temperatures T_1 and T_2 and μ and p refer to that for MI_n .

Putting the expression for p from Equation 3 into Equation 5 leads to

$$\begin{aligned} \Delta\mu = \Delta\mu^* + RT_1 \ln \frac{(a_2)^{T_2/T_1}}{a_1} + RT_1 \ln \frac{(p_2')^{T_2/T_1}}{p_1} \\ + RT_1 \ln (K_1)^{T_2/T_1 - 1} + \frac{\Delta H (T_2 - T_1)}{T_1} \end{aligned} \quad (6)$$

where p' refers to the partial pressure of $I(g)$. If $\Delta\mu$ is less than zero, metal deposition may occur. Five factors are seen to influence $\Delta\mu$, namely, the activity of M in the source and deposit, the partial pressure of iodine at the source and deposit, the heat of formation (ΔH) of metal iodide from pure metal(s) and gaseous I , the difference in the chemical potential for the standard state at the two temperatures, and the value of the equilibrium constant. The various parameters in Equations (5) and (6) are under evaluation to determine their importance in the transport mechanism.

Migration Behavior in Mixed Oxides

C. A. Hinman

A computer modeling program called MTRAN (for mass transport) is being developed to calculate the combined effects of (1) a concentration gradient, (2) an electrical potential gradient (due to thermoelectric effects), and (3) a thermal gradient upon the kinetics of solid-state diffusion in ionic solids. At the present time, the computer model is being used to analyze the driving forces for diffusion in oxide fuel materials under nuclear reactor environmental conditions. The general approach of the modeling is to calculate the atomistic flux due to the thermodynamic "forces" developed within the system which result from the extreme thermal gradients encountered in the system (up to 4000 °C/cm). The flux equation for the present model is as follows: ⁽⁹⁾

$$J = - D(C,T) \left[\frac{\partial C}{\partial x} + \frac{qC}{kT} \frac{\partial \phi}{\partial x} + \frac{Q^*C}{kT^2} \frac{\partial T}{\partial x} \right] \quad (7)$$

where $D(C,T)$ is the temperature and concentration dependent diffusion coefficient, $\frac{\partial C}{\partial x}$ is the composition gradient, q is the electrical charge associated with the diffusing specie, C is the concentration, k is Boltzmann's constant and T is absolute temperature, $\frac{\partial \phi}{\partial x}$ is the thermoelectric potential gradient, Q^* is the heat of transport, and $\frac{\partial T}{\partial x}$ is the thermal gradient. Once a flux is calculated, a flux gradient can be determined which leads to the use of a relationship commonly called Fick's second law:

$$\frac{dC}{dt} = - \frac{dJ}{dx} . \quad (8)$$

A concentration change is then calculated from the flux gradient during the time interval dt , which gives a new concentration profile.

The general mechanics of the program seem to be debugged fairly well at this point in time. The input parameters being

used for the diffusion of oxygen in uranium oxide have been obtained from various sources. The concentration dependent diffusion coefficient for oxygen is that derived by Thorn and Winslow⁽¹⁰⁾ for hyperstoichiometric UO_2 . No such coefficient is available for hypostoichiometric UO_2 so that present calculations for UO_{2-x} use the diffusion coefficient for either stoichiometric UO_2 or allow the stoichiometry parameter in Thorn and Winslow's relationship to go substoichiometric. The invalidity of this practice has been noted. Thorn and Winslow's diffusion coefficient for oxygen in UO_{2+x} is given by:

$$D(\text{O/M}, T) = 0.0216 \left\{ (\text{O/M} - 2.0) + \left[(\text{O/M} - 2.0)^2 + 495.0 e^{-\frac{1.62}{kT}} \right]^{1/2} \right\} e^{-\frac{1.52}{kT}} \quad (9)$$

where O/M is the oxygen-to-metal ratio.

Thermoelectric emf data by Bates and Dau⁽¹¹⁾ have been curve fitted to give the following equation:

$$\theta = 2.021 e^{-4.79 \times 10^{-3} T} - 2.55 \times 10^{-3} \quad (10)$$

where θ is the thermoelectric power in volts/°K. The compositional dependence of this parameter is not known, and this dependency may be extremely important.

Computations for a parabolic temperature profile have been made with the maximum temperature at 2000 °C and the lowest temperature at 1200 °C, varying along a distance of 0.5 cm. Results of these calculations suggest that the experimental data which were used as input are inadequate to describe the kinetics of oxygen migration under these conditions. The mass transport rates under these conditions are so fast that a maximum value for the oxygen-to-metal ratio of greater than 2.5

and a minimum value of 1.7 are obtained in only eight minutes of diffusion time. The next step is to start modifying the input data and the method of calculation in an attempt to bring the model in better agreement with experimental results. It is intended ultimately to extend the model mass transport mechanisms in mixed oxides.

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IRRADIATION DAMAGE TO REACTOR METALS

IRRADIATION BEHAVIOR OF STRUCTURAL MATERIALS - L. D. Blackburn

Irradiation Facilities Operation - A. L. Ward

The purpose of this phase of the program is to provide for the timely accomplishment of irradiation, testing, and subsequent processing of the data obtained from a variety of reactor structural materials. Irradiations are performed in aqueous, gaseous, and molten metal environments. Elevated temperature irradiations and tests are emphasized. Special testing techniques, applicable to either irradiated or unirradiated specimens, will be developed.

Facilities Operation

The experiments discharged from the various Engineering Test Reactor (ETR) core positions for Cycles 101 and 102 are given in Table 4.1, along with the estimated neutron fluxes and fluences.

TABLE 4.1. *Summary of Experiments Discharged at the Conclusion of ETR Cycles 101 and 102*

<u>Experiment Designation</u>	<u>No. of Specimens</u>	<u>Type of Specimens</u>	<u>Estimated Flux, n/cm²/sec(a)</u>	<u>Estimated Fluence, n/cm²(a)</u>
GEH-20-313	36	Zr Alloys	1.06×10^{14}	9.1×10^{20}

a. Neutrons with energies greater than 1 MeV.

NOTE: Total specimens discharged, ETR G-7 Loop Cycles 101 and 102 = 36

Total specimens discharged, ETR G-7 Loop to date = 3998

The operating history of the ETR and the G-7 hot water loop for Cycles 101 and 102 appears in Table 4.2. The history for the out-of-reactor loop is shown in Table 4.3.

TABLE 4.2. *Summary of Operating History for ETR and G-7 Hot Water Loop - ETR Cycles 101 and 102*

<u>Operating History</u>	<u>ETR Cycle Number</u>	
	<u>Cycle 101</u>	<u>Cycle 102</u>
1. <u>Reactor History</u>		
Start up ^(a)	3-13-69	5-14-69
End of Cycle	4-26-69	6-21-69
Megawatt Days	5919	5212
Effective Days at 175 MW	33.82	29.78
Number of Scrams ^(b)	9	11
Number of Shutdowns	3	2
2. <u>G-7 Hot Water Loop History</u>		
Maximum Temperature	539 °F	530 °F
Effective Days Above 200 °F	35.7	31.1
Effective Days at Oper. Temp.	30.45	27.4
Operating Efficiency ^(c)	100%	100%
Temperature Efficiency ^(d)	85.4%	88.2%

a. Includes flux runs at low power.

b. Power drops below half of full power (175 MW) with immediate recovery.

c. Effective days of loop operation above 200 °F (95 °C) with respect to effective days of reactor operation at 175 MW.

d. Effective days of loop operation between 500 and 550 °F (260 and 290 °C) with respect to days of operation above 200 °F (95 °C).

TABLE 4.3. *Summary of Operating History and Discharge Schedule for Out-of-Reactor Loop, ETR Cycles 99, 200, and 102*

	Time, hr		
	<u>Cycle 99</u>	<u>Cycle 100</u>	<u>Cycle 101</u>
1. TIME-TEMPERATURE SUMMARY			
200 °C (392 °F)	78.8	16.5	44.8
218 °C (424 °F)	17.5	4.8	40.7
246 °C (475 °F)	19.9	33.6	40.9
282 °C (540 °F)	697.6	758.0	730.4

2. SPECIMEN DISCHARGES: Material, Specimen Type, and Number.

AISI Type 410 Tensile - 18

Total specimens discharged to date = 1613

The ATR is expected to become available for irradiation experiments in October 1969. ETR G-7 water loop use is expected to be discontinued in favor of the ATR facility at that time. Test Core I has been completed. Core II is scheduled to end and Core III to begin on September 15, 1969. Cycle 1 startup is scheduled for October 13, 1969.

Microstructural Modifications to Stainless Steels and Nickel-Base Alloys - I. S. Levy

The purpose of this program is to optimize the stability of austenitic stainless steels and nickel-base alloys to fast reactor environments through microstructural modifications. These modifications are produced by selected variations in chemical composition and/or by use of thermomechanical treatments.

In an earlier report,⁽¹⁾ we proposed the concept of using a microstructure consisting of carbide-precipitated stacking faults to produce resistance to fast reactor radiation damage in cladding and structural alloys. It was also noted that the

development of the stacking fault structure was characterized by a hardening response. During the subsequent two quarters,(2,3) we examined the effects of various processing parameters on the development of stacking fault hardening at 700 °C in 348 stainless steel.

This past quarter, we have been evaluating the as-treated tensile properties, the thermal stability, and initial creep-rupture properties of 348 stainless steel after several of these treatments. Table 4.4 indicates that a range of significant (approximately 200-250%) increases in strength, relative to the annealed (1950 °F/1 hr, RAC) strength, can be achieved at 700 °C through the use of thermomechanical treatments (BNW-TMT) to develop the stacking-fault structure.

TABLE 4.4. As-Treated Tensile Properties of AISI 348 at 700 °C

Treatment	0.2% YS	UTS	% Elongation	
			Uniform	Total
Normal Annealed Condition (1950 °F/1 hr)	16,200	40,100	25.4	39.1
BNW-TMT-(3)	31,100	41,100	18.4	33.4
BNW-TMT-(5-368)	38,400	44,700	6.5	9.1
BNW-TMT-(5- 519)	46,000	49,900	6.3	10.1

Table 4.5 shows that the thermal stability of such a structure is excellent, while the strengthening produced by cold working is rapidly lost.

Table 4.6 shows that room-temperature yield strength can also be increased by 50% with negligible loss in ductility, an important characteristic for possible cold-forming operations.

Figure 4.1 and Table 4.7 show that at high temperatures and high stresses, the creep-rupture properties of the thermomechanically-produced stacking fault structure are

markedly superior to those of the annealed conditions (1950 °F/1 hr, RAC) of the other austenitic steels and even to that of the cold-worked condition of 316 stainless steel. The excellent resistance of the tensile properties to high-temperature degradation (Table 4.5) makes suspect the abrupt rupture of the creep test on the TMT-treated 348. Therefore, as noted in Figure 4.1, this test will be repeated. However, even with this rupture life, the important second stage of creep is about 5 times as long as that in the annealed condition

TABLE 4.5. *Effects of Thermal Exposure at 700 °C on Tensile Properties of AISI 348 at 700 °C*

Treatment	Exposure Time, hr	0.2% YS	UTS	% Elongation	
				Uniform	Total
BNW-TMT - (5-368)	0	38,400	44,700	6.5	9.1
	1000	37,400	47,200	9.1	14.8
	2650	34,500	43,100	9.3	12.9
Annealed plus 27% Cold-Worked Condition (by Swaging)	0	57,200	66,000	1.9	18.8
	50	22,200	37,800	20.1	50.1
	1000	21,700	37,500	19.0	46.6

TABLE 4.6. *As-Treated Tensile Properties of AISI 348 at Room Temperature*

Treatment	0.2% YS	UTS	% Elongation	
			Uniform	Total
Normal Annealed	40,700	95,200	46.7	54.8
BNW-TMT- (5-368)	60,700	96,200	41.1	45.6

TABLE 4.7. *Comparison of Creep Strengths at 1200 °F and 30,000 psi*

Alloy	Minimum Creep Rate, %/hr
Annealed 304	2.0
Annealed 316	0.3
Annealed 348	0.003
20% CW 316	0.005
348:BNW-TMT - (5-368)	\$0.0001

COMPARISON OF CREEP-RUPTURE CURVES AT 1200°F AND 30,000 psi

Neg 0693909-5

FIGURE 4.1. Comparison of Creep-Rupture Curves at 1200 °F and 30,000 psi

These results are significant for they indicate that, besides the theorized radiation-resistance of this structure, its stable high-temperature strength leads to creep properties that are extremely attractive in terms of fuel-swelling restraint, reduction of plenum size, deformation-resistant ducts, etc, all of which lead to important economies, through higher burnup and core compaction, in construction and operation of LMFBR reactors.

Production of Voids in Nickel - G. L. Kulcinski and B. Mastel

An experiment has been initiated to study the effects of stress on the dissolution of voids in irradiated metals. Initially, the studies will be concerned with the annealing kinetics of voids in high purity nickel and future efforts will be directed toward the stainless steel system.

Three different sources of high purity nickel (hereafter referred to as JM, MRC, and Ni-270) were irradiated to a fast neutron fluence of 3×10^{21} n/cm² (>0.1 MeV) at 450 °C in the EBR-II reactor. The metallic impurity content of the nickel samples was approximately the same although there were slight variations in the interstitial atom concentration (see Table 4.8). The major difference was in the relatively larger oxygen content of the Ni-270.

Table 4.9 summarizes the preirradiation properties of the nickel samples after a 2 hr anneal at 700 °C in a vacuum of 2×10^{-7} Torr. The resistivity ratios were determined from resistances at 273 and 4.2 °K on a high sensitivity (0.01 μV) voltage measuring apparatus. The hardness values represent the average of 10 readings on each sample taken with a Vickers microhardness tester; the standard deviations are also given. Precision lattice parameter values were obtained using a Nelson-Riley⁽⁴⁾ extrapolation using copper radiation.

TABLE 4.8. *Impurity Concentrations of the Three Starting Stocks of Ni*

Metallic Impurities	ppm, wt		
	JM	MRC	270
Mg	2	<0.1	<10
Al	4	0.5	
Si	5	<1	<10
S			<10
K		3.0	
Ca	2	1	
Ti			<10
Cr		0.5	<10
Mn	1		<10
Fe	7	10	<10
Co		1.0	<10
Cu	5	<0.5	<10
Zn		<4	
As		<0.4	
Zr		3	
Ag	1		
Sn		<10	
Purity (Metallic only) wt%	99.997	>99.997	>99.99
<u>Interstitials</u>			
Carbon	50	90	100
Oxygen	57	50	265
Nitrogen	<5	<2	12
Hydrogen	6	1.5	5
Purity (wt%)	99.985	99.982	>99.95
Total Gas Content (At ppm)	580	280	1320

TABLE 4.9. *Pre-Irradiation Properties of Ni Samples^(a)*

	<u>JM</u>	<u>MRC</u>	<u>270</u>
Grain Size - Microns	30	45	45
Resistivity Ratio $R_{273}/R_{4.2}$	117	111	103
Hardness - dph	82 $\pm$ 3	80 $\pm$ 2	85 $\pm$ 4
Lattice Parameter $\text{\AA}$	3.5241 ± 0.0002	3.5239 ± 0.0002	3.5240 50.0002

a. After a 2 hr - 700 °C anneal at 2×10^{-7} Torr.

The Ni was irradiated in the form of foils 0.0075 cm thick and 11 mm in diameter. The foils were contained in a 304 SS can filled with high purity sodium. The temperature of irradiation was calculated from gamma heating rates along with a knowledge of the ambient reactor sodium temperature.

After irradiation, the samples were electrolytically thinned in a 70-30 mixture of methanol and nitric acid at -40 °C. A dual jet arrangement was used to wash away the viscous layer of nickel oxide formed during thinning.

Voids were found in all three types of nickel after irradiation (Figure 4.2). The general morphology of the voids is the same as that previously determined,^(5,6) namely octahedrons bounded by {111} planes with various degrees of truncation on {100} planes.

The denuding of voids at grain boundaries is not as simple as that reported previously for high purity nickel samples. Mastel and Brimhall⁽⁶⁾ and Bloom and Stiegler⁽⁷⁾ report that in nickel irradiated to $\sim 1 \times 10^{20}$ n/cm² (>0.1 MeV) at 380 °C, there is approximately a 1000 $\text{\AA}$ wide denuded zone around grain boundaries and twin boundaries. In the present study, this type of denuding was found to occur at some boundaries

JM

MRC

270

 0.1 μ

Neg 0693782-2
FIGURE 4.2. Typical Microstructure of Nickel After Irradiation to 3×10^{21} n/cm²
 (>0.1 MeV) at 450 °C. All foils are in (110) Orientation

(Figure 4.3a), while essentially no denuding was found at other boundaries (Figure 4.3b). In fact, the voids found near, and in some cases, on the grain boundaries were present in a higher density and smaller size than the voids in the center of the grains. There is some evidence that this phenomenon is connected to the misfit of adjacent grains and further work is being conducted to explore this possibility. The phenomenon was found in all three types of nickel samples.

The present samples are also lacking the dislocation loop structure found in nickel irradiated at lower fluences. (6,7) This may be attributed to the slightly higher temperature of irradiation (450 °C compared to ~380 to 410 °C) or the higher fast flux.

Void densities have been determined for the JM and MRC samples, and the Ni-270 results will be reported next quarter. The interference fringe technique was used to determine the foil thickness using the following values for the extinction distance:

<u>g</u>	<u>Extinction Distance Δ^o</u>
<200>	268
<111>	302
<220>	468

The average length of the (110) octahedral edge was usually determined from micrographs taken during a (100) foil orientation so that truncation effects could be ignored. The average (110) edge was determined by $\frac{\sum \ell N_{\ell}}{\sum N_{\ell}}$, where ℓ is the measured edge length and N_{ℓ} is the number of voids/cm³ with that length. The volume increase/cm³ for the octahedrons was calculated by the formula $0.47 \sum \ell^3 N_{\ell}$.

Table 4.10 summarizes the preliminary void size and density data for the JM and MRC samples. These are results obtained from one sample of each material in the as-irradiated

a) Typical Denuded Zone-occurs Preferentially
at Large Angle Boundaries

b) Formation of a Layer of Smaller
Voids Near Small Angle Boundaries

0.1μ

Neg 0693782-1

FIGURE 4.3. Grain Boundary Effects on Void Formation in Neutron
Irradiated MRC Nickel

TABLE 4.10. *Summary of Post-Irradiation Properties
of Irradiated Ni*

	<u>JM</u>	<u>MRC</u>	<u>270</u>
Mean edge of octahedron Å	270 ± 20	400 ± 30	(a)
Void Density x 10 ⁻¹⁴	5 ± 1	1.8 ± 0.4	(a)
Volume Increase %	0.81 ± 0.18	0.64 ± 0.14	(a)
Hardness Increase %	41 ± 8	40 ± 5	35 ± 11
Lattice Parameter Change - Å	+0.0005 ± 0.0003	+0.0005 ± 0.0003	0 ± 0.0003

a. To be reported next quarter.

state. In general, the voids tended to be smaller and more numerous in the JM Ni. The average void size was 270 ± 20 Å for the JM material while the octahedrons in the MRC samples were an average size of 400 ± 30 Å. The differences in void density are such that the amount of swelling to be expected from each sample is approximately the same: $0.81 \pm 0.18\%$ for the JM Ni compared to $0.64 \pm 0.14\%$ for the MRC Ni. These values compare fairly well to $0.72\% \Delta V/V_0$ determined from immersion density tests on bulk Ni-270 samples irradiated in the same capsule to the same fluence conditions. (8)

All three samples showed increases in hardness after irradiation. The changes ranged from 35-41% of the well-annealed hardness values and no doubt reflect the void-dislocation interactions.

Very little lattice parameter increase above experimental error was detected. Since the lattice parameter is mainly a measure of the free or trapped interstitial concentration, these results indicate that isolated interstitial atoms were not present after irradiation at 450 °C. This information,

coupled with the observation that there are no observable interstitial loops or clusters, means that the interstitials must have annealed rapidly during irradiation. It is most likely that they precipitated into rafts which rapidly coalesced into very large loops. Such loops would not be visible in transmission and would only show up as single dislocations after thinning.

Irradiation Effects on the Fracture of Heavy Section Pressure Vessel Steels - C. W. Hunter and J. A. Williams

Specimen Irradiations

Compact tension fracture mechanics specimens (CTS) and tensile specimens of ASIM A 533-B pressure vessel steel were rotated and exchanged in the irradiation assembly during the April outage of the ETR. The initial reactor charging of this assembly into the M-3 loop of the ETR was during February 1969, Cycle 101, and has been previously described.⁽⁹⁾ Four CTS and 15 tensile specimens were removed and replaced with unirradiated specimens. The end specimens in the assembly were rotated during cycle 102 to compensate for the flux gradients. The fast fluence of the specimens removed was 1.5 to 2.5×10^{19} nvt (>1 MeV), based on the fission spectrum of the reactor. Additional flux monitors positioned in the grip holes of the CTS are being analyzed. A plot of the fast flux distribution over the length of the irradiation assembly is shown in Figure 4.4.

Figure 4.5 presents the temperature distribution along the assembly length during full reactor power; these temperatures were measured at the center of the root of the machined notch of the CTS. The tensile specimen temperature during full-power operation was 503°F , as measured in the button head at a bulk water temperature of 500°F , and varied with the bulk water temperature. The typical water temperature was 498 to 503°F at full reactor power. There was a negligible temperature rise

Neg 0693069-2
FIGURE 4.4.

Flux Distribution Over the Length of the HSST Specimen Irradiation Assembly in the M-3 Loop of the ETR

Neg 0693069-6

FIGURE 4.5. *Temperature Distribution Over Length of ETR, M-3 Loop Irradiation Assembly. Temperatures measured at the center of the notch root of IT compact tension specimens. Temperature distribution measured at full power.*

of the water during its flow through the irradiation facility. The flow rate was 100 gpm and pressure 2000 psi. The temperature gradients, indicated in Figure 4.5 for full-power operation, decreased proportionately as the reactor power and water temperatures decreased, so that the gradients were compressed and the specimen temperatures converged at the water temperature during reactor shutdown. The reactor power and M-3 loop water temperature for cycle 101 are recorded in Figure 4.6.

Cycle 102 outage occurred in July at which time 8 CTS and 14 tensile specimens of ASTM A 533-B base material were removed. These were replaced with 8 CTS from the submerged arc weldment and 14 tensile specimens of base material which had been plastically strained for the strain aging embrittlement study. The 4 CTS that have occupied the center position of the assembly since charging during the cycle 101 outage will remain in position to accumulate a total fluence of 8×10^{19} nvt.

Initial heat transfer studies for the design of an irradiation capsule for 4-in. compact tension specimens (4T CTS) have been completed. The calculations were based on scattered gamma data from possible "equivalent" positions throughout the ETR; however, the results did establish the limits of feasibility of capsule irradiation. In addition, the results suggested some modes of capsule operation to gain a variety of irradiation conditions. The predicted temperature distributions are shown in Figure 4.7 for three planes normal to the vertical axis of the reactor. These planes correspond to the fracture planes of the three specimens in the capsule. The temperature distribution was calculated for the heat transfer gaps and gamma heat generation gradients shown, using heat transfer coefficients for a mixture of 50% He to 50% Ar in the gap. The results indicate that multiple irradiation temperatures could be obtained in the same irradiation capsule. For single irradiation temperatures, a varying gas gap over the length of the capsule would

Neg 0693069-9

FIGURE 4.6. Bulk Outlet Water Temperature of the M-3 Loop and Reactor Power for the ETR Cycle 101

(CALCULATED TEMPERATURE DISTRIBUTION
ETR 4T IRRADIATION CAPSULE)

Neg 0693069-8

FIGURE 4.7. Irradiation Temperature Distribution of 4T Compact Tension Specimens at the Fracture Planes. Temperatures were calculated for the gamma heat generation shown over the axial length at the capsule with a mixture of 50% He - 50% Ar in the gas gap.

be required. Comparison of Plane B for two different gamma heat values, represents the extremes on which the gap design must be based. At the higher gamma heat rate only a 0.002-in. gap is allowable to obtain temperatures in the range desirable. Larger temperature gradients are also exhibited for higher values of gamma heat. A 0.030-in. gap at the high gamma heat rate yielded temperatures exceeding the melting point of the material. In addition, the calculations are based on a constant value of gamma heat over the length of the capsule, a case which is unrealistic. However, the results indicate that irradiation of 4T compact tension specimens is feasible if the gamma heating is not too high. Therefore, accurate gamma heat generation experiments for the positions being considered for capsule irradiations in the ETR are currently being conducted.

Base Material Studies of HSST ASIM A 533-B

Base material studies of unirradiated HSST material (Plate 02 of A 533-B) during the last quarter included fracture toughness and tensile testing. The unirradiated specimen testing at PNL on base plate material is oriented toward providing a cursory examination of mechanical properties for comparison with the data of other HSST investigators providing characterization of base plate material.

The measured fracture and tensile properties are shown in Figure 4.8 as a function of test temperature; the specific data are tabulated in Tables 4.11 and 4.12. Both the fracture toughness and tensile data are comparable to that of other investigators in the HSST program; however, some small differences may be observed in the tensile data. The present values of yield strength reported in Table 4.11 are slightly lower than those reported by ORNL (ref. 1, p. 22) and by Westinghouse (ref. 1, p. 58). Total elongation values in this present data and in the Westinghouse data are comparable, while the ORNL

Neg 0693069-4

FIGURE 4.8.

Fracture Toughness and Yield Strength as a Function of Temperature for ASTM A 543, Grade B, Class I Steel from HSST Plate 02. Closed points on the yield curve represent 0.2% offset yield strength values. Closed points on the fracture curve were invalid K_{Ic} determinations, based upon the ASTM criteria a or $B < 2.5 (K_{Ic}/\sigma_{ys})^{1/2}$. All the specimens were from the quarter through center positions, measuring properties in the longitudinal (rolling) direction (i.e., fracture specimens were RW and tensile specimens were R).

TABLE 4.11 *Tensile Properties of A 533, Grade B, Class I Steel From HSST
Plate 02, Longitudinal Specimens*

Specimen Ident.	Test Temperature, °F	Test Temperature, °C	Plate (a) Position	Yield Strength, psi x 10 ⁻³		Ultimate Strength, psi x 10 ⁻³	Total Elongation, %	Reduction In Area, %
				Upper	Lower			
02GA 194	-250	-156	7/19	103.8	97.7	119.1	24.3	60.6
02GA 221	-250	-156	9/19	101.2	97.3	127.9	24.5	44.1
02GA 147	-250	-156	5/19	107.6	101.3	136.1	24.2	60.7
02GA 171	-250	-156	6/19	108.2	100.4	132.6	24.6	60.2
02GA 195	-150	-101	7/19	83.7	81.6	108.8	18.0	66.5
02GA 196	- 50	- 45	7/19	70.8	70.2	93.8	19.6	50.6
02GA 197	75	24	7/19	67.5	65.4	86.5	17.5	68.3
02GA 223	76	24	9/19	67.2	66.4	85.7	17.5	71.0
02GA 198	275	135	7/19	-	-	79.9	15.0	35.8
02GA 225	475	246	9/19	-	-	80.5	11.6	65.4
02GA 175	475	246	6/19	-	-	86.2	15.6	58.9
02GA 199	475	246	7/19	-	-	83.0	15.0	63.6
02GA 200	575	300	7/19	-	-	80.8	17.4	66.0

a. Plate position fractions of plate thickness at specimen axis.

TABLE 4.22. *Fracture Toughness, K_{Ic} , of A 533-B Steel From HSST Plate 02, Measured with IT Compact Tension Specimens, RW Orientation at 4/9T Plate Position*

Specimen Ident.		Test Temperature, °F	°C	Crack Length a, in.	Yield Strength, psi x 10 ³	K_Q , ksi√in.	$2.5\left(\frac{K_Q}{\sigma_{ys}}\right)^2$	K_{Ic} , ksi√in.
02GA 28		-250	-156	0.974	105	31.2	0.220	31.2
02GA 5		-185	-121	1.005	91	39.8	0.478	39.8
02GA 6		-185	-121	1.014	91	43.3	0.566	43.3
02GA 8		-135	- 93	1.039	83	41.0	0.610	41.0
02GA 25		-135	- 93	0.972	83	49.7	0.896	49.7
02GA 47		-100	- 73	0.927	78	53.0	1.518	
02GA 26		-100	- 73	0.994	78	68.0	1.900	
02GA 48		- 50	- 45	0.942	72	60.2	1.74	
02GA 27		- 50	- 45	0.918	72	68.6	2.26	

material exhibited slightly higher values of total elongation. The variations between the data are most probably due to variations in plate properties.

Weldment Study of ASIM A 533-9

The weldment currently being studied is the HSST program weldment section 51A that had been submerged arc welded from HSST program plate 01. The original size was 30-in. long by 12-in. thick. The investigation is using both 1-in. thick compact tension specimens (IT CTS) and 1/2-in. double cantilever beam (DCB) specimens to determine the toughness gradient through the weldment. The CTS are oriented with the fracture plane parallel to the weld (and perpendicular to the plate surface) and the notches are at different positions in the weld and HAZ. The DCB specimens are oriented with the fracture plane perpendicular to the weld (and plate surface) so that the successive propagation events assess the toughness gradient. These specimen orientations are schematically depicted in Figure 4.9.

Neg 0693069-5

FIGURE 4.2. Macrograph Through the Thickness of the Submerged Arc Weldment Depicting the CTS and DCB Specimens to be Used in Determining the Toughness Gradient Through the Weldment. 1X, 5% Nital etch.

The metallurgical areas for location of the notches in the CTS are indicated in Figure 4.10. Probably the most important areas are the weldment (A), the weld-dilution region or weld-side of the fusion line (B), and the region of austenitic grain growth or HAZ-side of the fusion line (C). Four IT CTS from area A and four from area B were charged into the M-3 loop for irradiation during cycles 103 and 104 to fluences of 1 to 2×10^{19} n/cm² at approximately 540 °F. Between cycles 103 and 104, four specimens from area C will be charged into the M-3 loop. Similarly positioned unirradiated specimens will also be tested.

Results of unirradiated 1/2-in. DCB tests at -256 °F (-160 °C) and -278 °F (-172 °C) are depicted in Figure 4.11. The results are from three specimens at each temperature. The typical dimensions for the specimens were gross width of 0.500 in., double-beam height of 1.000 in., net width of 0.183 in., notch radius of 0.001 in., and length of 7 in. All the specimens were from quarter-thickness through center positions. The critical stress intensity at crack initiation and arrest (K_c and K_a) were calculated from the loads and crack length using the expression(10,11)

$$K = \frac{4 P^L}{h B B_n} (3 x^2 + 1) \quad ,$$

in which

$$x = \frac{a}{h} + 0.6$$

P = critical load

h = beam height, 0.500 in.

B = gross width, 0.500 in.

B_n = net fracture width, 0.183 in.

a = crack length from point of load application.

Neg 0693069-3

FIGURE 4.10. Locations of the Areas of Interest in the Weld Metal and HAZ of the Submerged Arc Weldment. 12X, 5% Nital etch.

Neg 0693069-7

FIGURE 4.11. *Distribution of the Critical Fracture Toughness for Initiation and Arrest Through the Base, HAZ, and Weld Metal as Determined by 1/2-In. DCB Specimens at -256 and -278 °F. The closed points at -278 °F are all from one specimen which yielded low values consistently.*

The crack lengths were both measured and calculated from the experimental compliance calibration for this specimen determined by Hoagland. (12)

The critical stress intensity ($\text{ksi}\sqrt{\text{in.}}$) data for crack initiation and arrest in the base metal and weld are summarized below:

Temperature	<u>BASE</u>		<u>WELD</u>	
	K_{IC}	a	K_{IC}	K_{a}
-256 °F	40	34	47	30
-278 °F	36	28	38	27

The principal feature of the data is that the weld metal has a higher initiation toughness but a lower arrest toughness than the base material. Such a behavior indicates that the weld metal is more rate sensitive. At -256 °F, the exclusion of any data in the HAZ is indicative of low toughness in this region; however, the initiation and arrest toughness in the HAZ at -278 °F are not particularly low. The closed points at -278 °F are all from one specimen tested during a period of high relative humidity so that the cryostat badly iced. This condition was probably responsible for the consistently low values. Limits for valid K_{IC} data are indicated on the right side of Figure 4.11. These were determined for a yield strength of 110,000 psi using the following criteria:

- $B \geq 2.5 (K_{\text{IC}}/\sigma_{\text{ys}})^2$ (See Reference 13 at the end of Section 4.) giving a K_{IC} maximum of $49 \text{ ksi}\sqrt{\text{in.}}$;
- $B \geq 4 (K_{\text{IC}}/\sigma_{\text{ys}})^2$ (See Reference 14 at the end of Section 4.) giving a K_{IC} maximum of $39 \text{ ksi}\sqrt{\text{in.}}$

Strain-Aging Embrittlement

The intent of this study is to determine the harmful effects of irradiation and thermal aging upon a vessel that has been

subjected to plastic strain at the roots of cracks, flaws, or notches. Such plastic strain will result from both the original hydro-proof test and from normal service operation. This present study is relevant to vessel integrity by simulating the above conditions in precracked specimens.

Strain-aging effects are pertinent to fracture initiation, rather than propagation and arrest, because only at the crack tip do the plastic strains and blunting occur. However, they may continually degrade fatigue-crack resistance. Also, the strain and blunting conditions cannot be identically reproduced in a bulk material. Thus, these effects have meaning within the realm of fracture mechanics; and one may expect the simulation of the load conditions in a precracked specimen to accurately predict vessel behavior.

A diagrammatic representation of the experimental work is shown in Figure 4.12. The material is the same as that used in the base metal study, HSST Program plate 02GA. The fracture specimens are IT CTS from 1/4 through 3/4 thickness positions oriented in the RW direction. After fatigue crack preparation, the stress intensities are applied to strain the metal at the crack root. After irradiation or thermal aging the extent of embrittlement will be assessed by comparing the K_{IC} - versus - temperature behavior of these specimens with the behavior of specimens not aged.

The temperature of stress intensity application corresponds to the typical proof test and operating temperatures. The stress intensities are within the range of operating and proof test belt-line stress levels on elliptical surface flaws 0.5 to 1.0 in. deep.

This present strain-aging study may be differentiated from warm prestressing on the basis that the latter is to supposedly produce beneficial effects by crack blunting in a material that

STRAIN AGING INITIAL STUDY

Neg 0693069-1

FIGURE 4.12.

Diagrammatic Representation of the Study to Determine the Extent of Embrittlement in the Plastic Strain Region at a Crack Tip Produced by Neutron and Thermal Aging Subsequent to Precracking

has been (irradiation) embrittled. The study of warm prestressing will not begin until the effects of strain-aging embrittlement are known.

Presently, 15 precracked specimens have been strained under the conditions described in Table 4.13. The extent of plastic deformation occurring at the crack tip is just as important as the stress intensity level and temperature. Therefore, the notch opening was measured at the front of the specimen to determine the K-level at which deviation from linear elastic behavior was observed and the extent of permanent notch opening displacement. The precision of the strain gauge extensometer used at 100 °F was better than 0.0001 in., while that of the LVDT used at 550 °F was about 0.001 to 0.002 in. There was no deviation from linear elastic behavior at 100 °F below a K-level of $40,000 \text{ psi}\sqrt{\text{in.}}$. During loading from 40,000 to 60,000 psi the indicated plastic displacement occurred, but no more occurred during the 1-hr hold. At 550 °F, the load-displacement record indicated that there was not an appreciably greater deviation from linearity during loading than for the 100 °F case, but that additional plastic displacement occurred during the hold. These specimens will be thermally aged at 550 °F for about 30 days, followed by testing.

Tensile specimens have also been plastically strained and are being irradiated and thermally aged to assess the effects of strain-aging embrittlement on flow properties. This information will permit a more lucid analysis of the strained metal at the crack tip in the fracture specimens. The tensile specimens have been plastically strained 1% and 10% at 100 °F. Seven specimens from each condition were charged into the M-3 loop for irradiation during cycles 103 and 104.

TABLE 4 13 Conditions of Stress Intensity Application to Precracked IT Compact Tension Specimens. Hold time of 1 hr.

Specimen No.	Plate Position	K-Level, psi/in.	Temperature, °F	Crack Length, in.	Load, lb	Plastic Displacement, in.
OZGA 13	1/2-3/4	35,000	100	1.041	4839	<0.0001
OZGA 15	1/2-3/4	35,000	100	1.038	4867	<0.0001
OZGA 29	1/2	35,000	100	0.909	5155	<0.0001
OZGA 35	1/2-3/4	35,000	100	1.035	4876	<0.0001
OZGA 56	1/2-3/4	35,000	100	1.007	5097	<0.0001
OZGA 14	1/2-3/4	60,000	100	1.039	8321	0.0004
OZGA 33	1/2-3/4	60,000	100	0.987	9008	0.0007
OZGA 30	1/2	60,000	100	1.033	8408	0.0010
OZGA 34	1/2-3/4	60,000	100	1.023	8518	0.0005
OZGA 36	1/2-3/4	60,000	100	1.015	8648	0.0007
OZGA 53	1/2-3/4	35,000	550	1.015	5029	0.001(a)
OZGA 31	1/2	35,000	550	0.971	5379	0.001(a)
OZGA 54	1/2-3/4	60,000	550	0.951	9479	0.006(a)
OZGA 55	1/2-3/4	60,000	550	0.972	9980	0.002(a)
OZGA 32	1/2	60,000	550	1.017	8613	0.002(a)

a. Extensometry at 550 °F for the 1-hr hold time was not sufficiently reliable to provide a more accurate plastic displacement measurement.

Irradiation Effects in Zircaloy-2

The research to be reported here was part of the Canadian Cooperative Program at PNL which was brought to a close at the end of FY-1969. The objectives of this study were to:

- Extend the knowledge of irradiation damage accumulation in Zircaloy-2 at 280 °C to a fluence of 1.5×10^{22} n/cm² (E > 1 MeV) by characterizing both the tensile behavior and the microstructure.
- Determine whether voids could be formed.
- Determine if differences in the microstructure existed after irradiation at 60 °C and 280 °C.

Irradiations at 60 °C were conducted in ETR core positions while the 280 °C irradiations were in the G-7 Hot Water Loop.

Transmission electron microscopy was employed using 0.020-in. thick samples taken from the undeformed tab ends of tensile specimens. The samples were first dimpled and then electropolished in a bath of four parts ethyl alcohol and one part perchloric acid at -40 °C.

Figure 4.13a shows the typical microstructure of Zircaloy-2 after irradiation to a fluence of 1×10^{21} n/cm² at 60 °C. The damage is in the form of small clusters 40 to 75 Å in diameter with an average diameter of 50 Å. If a foil thickness of 1000 Å is assumed, then the density of 1.5×10^{16} /cm³. Denuding of clusters adjacent to grain boundaries was not observed. Due to surface roughening, the clusters could not be identified by dark field techniques.

The microstructure of similar samples irradiated to 1.4×10^{21} n/cm² at 290 °C is shown in Figure 4.13b. The damage can be divided into two components, defect clusters and resolvable dislocation loops. The average size of the clusters has increased slightly as a result of the higher irradiation

Neg 7126B

A

Neg 7147A

B

181,000X

FIGURE 4.13.

Microstructure of Zircaloy-2 After Irradiation to 1.0×10^{21} n/cm² at 60 °C (A) and After Irradiation to 1.4×10^{21} n/cm² at 290 °C (B)

Neg 7087C

A

Neg 7296A

B

FIGURE 4.14.

Microstructure of Zircaloy-2 After Irradiation to 1.0×10^{22} n/cm² at 290 °C (A) and After Annealing 500 °C for 1 hr (B)

temperature. The diameters of the loops ranged from 125 to 175 Å with an average diameter of 135 Å. The total density of clusters and loops is approximately the same as the density of the clusters present after the 60 °C irradiation. The plane of the larger loops was determined to be (1120). The identity of the loops could not be established because of their small size. Some interaction between loops was inferred, but denuding adjacent to boundaries was not detected.

Figure 4.14a shows that damage in Zircaloy-2 irradiated to 1.0×10^{22} n/cm² at 290 °C is difficult to characterize. The size of the clusters and loops has increased resulting in overlapping of their images. Under these conditions a meaningful density value cannot be made. Some interaction of loops has taken place as short dislocation segments are present. High magnification studies did not disclose the presence of voids.

Annealing of the irradiated samples in vacuum at 400 °C for 1 hr produced little discernable difference in the microstructure. However, annealing at 500 °C for 1 hr does result in a drastic microstructural change. Figure 4.14b shows that a dislocation network has replaced the original damage configuration. A low density of clusters and loops are scattered about the network. Presumably the network results from the growth and interaction of loops during annealing. The network and other defect clusters are completely removed after annealing at 600 °C. Voids were not observed after any of the annealing treatments.

Tables 4.14 and 4.15 list the tensile properties of the irradiated material and unirradiated thermal controls, respectively. Cold work levels of 20 and 40% are represented in addition to the annealed condition. All the tensile data in Table 4.14 was taken from specimens irradiated in the ETR-G-7 loop at 280 °C and tested in the range of room temperature to 750 °C.

TABLE 4.14. *Tensile Properties of Zircaloy-2 Irradiated
in the ETR-G-7 Loop*

Specimen	Cold Work, %	Test Temp, °C	0.2% Yield Strength, psi	Ultimate Strength, psi	Ultimate Strain, %	Total Strain, %	Reduction of Area, %
GEII-20-17-2	0	300	76,200	79,700	1.3	1.7	14.7
GEH-20-17-3	0	500	27,800	30,300	1.5	8.3	85.6
GEH-20-17-4	0	650	4,200	5,600	2.4	38.2	99.5
GEH-20-17-5	0	750	1,000	2,800	34.6	80.7	99.5
GEH-20-17-7	20	Rm	121,500	122,000	1.1	2.5	26.8
GEH-20-17-9	20	500	29,200	31,800	1.3	6.8	86.6
GEH-20-17-10	20	650	6,700	9,600	6.0	24.9	99.5
GEH-20-17-11	20	750	1,300	3,900	20.5	45.0	99.5
GEH-20-17-13	40	Rm	131,100	134,700	1.7	2.5	13.3
GEH-20-17-15	40	500	31,100	34,800	2.0	9.6	87.2
GEH-20-17-16	40	650	8,700	10,900	4.8	18.2	99.5
GEII-20-17-17	40	750	2,100	3,700	16.2	56.4	99.5
GEII-20-28-1	0	Rm	98,600	98,600	1.1	2.6	33.6
GEH-20-28-3	0	400	61,500	61,700	1.2	3.7	63.4
GEH-20-28-4	0	500	30,400	34,000	1.5	6.3	73.4
GEH-20-28-5	0	650	6,500	7,600	2.5	23.1	93.7
GEH-20-28-12	20	Rm	113,900	114,800	1.3	1.4	17.2
GEH-20-28-8	20	300	82,500	87,300	1.6	2.2	20.8
GEII-20-28-9	20	400	58,200	63,400	2.2	9.0	73.1
GEH-20-28-10	20	500	23,200	29,800	2.3	15.4	84.5
GEH-20-28-11	20	650	2,000	2,800	6.2	48.9	99.5
GEH-20-28-13	40	Rm	108,400	112,300	1.8	2.0	16.8
GEH-20-28-14	40	300	78,000	80,900	1.9	3.3	32.2
GEH-20-28-15	40	400	58,600'	62,700	1.7	7.8	71.6
GEH-20-28-16	40	500	27,900	30,300	2.5	13.7	82.6
GEH-20-28-17	40	650	4,200	5,300	2.0	12.5	99.5

TABLE 4.15. *Tensile Properties of Zircaloy-2
Thermal Controls*

Specimen	Cold Work, %	Test Temp, °C	0.2% Yield Strength, psi	Ultimate Strength, psi	Ultimate Strain, %	Total Strain, %	Reduction of Area, %
GEH-20-17-1A	0	Rm	54,500	63,800	12.2	29.9	57.7
GEH-20-17-1B	0	300	18,500	26,300	11.9	24.8	77.7
GEII-20-17-2A	0	500	10,800	14,700	7.7	55.0	92.6
GEH-20-17-2B	0	600	6,500	8,200	7.1	52.5	99.5
GEH-20-17-3A	20	Rm	69,800	76,200	3.9	15.5	58.7
GEH-20-17-3B	20	300	42,900	45,500	1.0	1.6	68.0
GEH-20-17-4A	20	500	21,500	25,300	2.2	23.4	93.7
GEH-20-17-4B	20	600	9,200	12,000	2.7	37.5	99.5
GEH-20-17-5A	40	Rm	80,800	85,500	2.7	9.7	53.2
GEH-20-17-5B	40	300	50,800	53,400	1.4	6.0	65.6
GEH-20-17-6A	40	500	20,900	27,200	1.3	22.6	88.0
GEII-20-17-6B	40	600	9,500	11,000	2.4	47.6	99.5

In general, irradiation resulted in the usual yield strength increases and associated ductility losses. A significant result, however, is that the damage state which affects the tensile properties of Zircaloy-2 shows no evidence of saturation. The yield strength and ductility values reported here compare favorably with extrapolations of property versus fluence curves published by Bement.⁽¹⁵⁾ This observation suggests that growth of clusters and loops affects the strength and ductility beyond exposures at which the number of such defects has saturated.

Comparison of the irradiated data and the unirradiated control data reveals a consistency in the temperature at which the damage begins to disappear. The electron microscope examinations showed a significant difference in the microstructure following an anneal at 500 °C. At a test temperature of 500 °C, the very large yield strength increases seen at low temperatures were partially eliminated. Further correlation between properties and microstructure are evident in the apparent removal of all damage by annealing or testing at 600 °C and above.

FUEL SUBASSEMBLY MATERIALS - T. T. ClaudsonEBR-II Irradiations - P. K. McDaniel

The EBR-II irradiations program is to provide coordination and direction for the timely accomplishment of fast reactor irradiations on structural materials. Structural materials irradiation experiments are in various stages of development: some in design, some under construction, and others under irradiation in the EBR-II. The status of these experiments are described.

A metallurgical variables experiment (BNWL-B-40) is in the process of being assembled for insertion in EBR-II about October 1, 1969. This pin will be a replacement pin in sub-assembly X-018. Its goal exposure is 4×10^{22} n/cm² ($E > 0.1$ MeV). The objectives of this experiment are to provide information concerning the degree to which irradiation-induced swelling in Type 304 and Type 316 SS can be controlled by simple heat treatments and to establish a relationship between cold work (less than 25%) and irradiation-induced swelling as a function of irradiation temperature.

At the end of run No. 36 (8-1-69) two structural sub-assemblies (X-019 and X-067) were discharged from EBR-II. Sub-assembly X-019 consists of eight A-type experiment pins from PNL, the remainder of pins belong to General Electric and United Nuclear Corporation. The objective of these experiments was to achieve an exposure between 1×10^{22} and 1×10^{23} n/cm² fast fluence in an environment of bulk sodium and helium at temperatures of 700 to 850 °F. The accumulative fluence was 3.8×10^{22} n/cm² (total). Postirradiation testing of these materials (Types 304 and 316 SS) will provide data for evaluation of the effect of irradiation on design values in support of the LMFBR Program.

Subassembly X-067 consisted of 7 pins: four pins from PNL, two from Argonne National Laboratory and one from General Electric. The four PNL pins contain specimens for a study of the effect of fast reactor irradiations on Type 304 and 316 SS weldments in support of the FFTF pressure vessel and fuel ducts.

The accumulative fluence was 9×10^{21} n/cm² (total). An updated summary of active and discharged experiments to the end of run No. 36 is given in Tables 4.16 and 4.17.

Damage Analysis - J. L. Straalsund, H. R. Brager, and R. L. Fish

The objectives of this effort are to establish the radiation-induced swelling characteristics of FFTF alloys, relate fast-reactor induced substructural changes in microstructure to corresponding changes in mechanical properties, and provide a basis for extrapolation and interpolation of data using both microscopic modeling and empirical approaches.

Recent experiments concerning the effects of fast-reactor irradiation on stainless steels have demonstrated that a considerable volume change occurs during neutron bombardment. (16) Unlike volume changes in fuel materials, swelling in structural alloys and pure metals is due to the condensation of irradiation-produced excess vacancies in the form of three dimensional cavities. United Kingdom investigators have found as much as 6.8% volume change in 316 SS irradiated to 7.8×10^{22} n/cm² total fluence. Volume changes of this magnitude, if not taken into account, are sufficient to cause distortion in reactor components.

Maximum target fluences for FFTF are as high as 10^{24} n/cm². Since such a high fluence cannot be attained in a reasonable time using currently available reactors, design data for high exposures will be based on extrapolation using physically realistic swelling models. The approach taken in this study

TABLE 4.16 Summary of experiments in the EBR-II

Subassembly Designation	Reactor Position	Date Charged	Estimated Discharge Date	Type of Specimens	Material	Accumulative (b) Fluence, n/cm ²	Goal Fluence, n/cm ²
X-020	6B5	1-13-67	July 69	28 Tensile 8 Uniaxial Creep	304 SS 316 SS	3.5 x 10 ²²	1.9 x 10 ²²
X-021(a)	2D1	2-27-67	Aug 70	501 Tensile or Uniaxial Creep 65 Biaxial Creep	304 SS, 316 SS, 348 SS, 321 SS Ni Base Alloys (c)	8 x 10 ²²	8.8 x 10 ²²
X-027	4B3	1-21-67	Aug 70	5 Tensile 3 Uniaxial Creep	304 SS	3.9 x 10 ²²	5 x 10 ²²
X-041	7C4	7-24-68	May 70	713 Tensile or Uniaxial Creep 74 Biaxial Creep	304 SS, 316 SS, 321 SS, 348 SS	1.7 x 10 ²²	3 x 10 ²²
X-057	2B1	2-24-69	March 71	577 Tensile or Uniaxial Creep 164 Biaxial Creep 58 Density	304 SS, 316 SS, (c) 321 SS, 348 SS, Ni Base Alloys	1.9 x 10 ²²	6.0 x 10 ²²

a. Original space available with two single pin interchanges

b. Accumulative to end of run 33, (E > 0.1 MeV).

c. Variety of other alloys.

TABLE 4.17. *Summary of Experiments Discharged from EBR-II*

Subassembly Designation	Reactor Position	Date Charged	Date Discharged	Type of Specimen	Material	Accumulative ^(d) Exposure, n/cm ²
X-009 ^(a)	4A2	3-24-66	11-15-66	18 Tensile 10 Uniaxial Creep	304 SS	2.0×10^{22}
X-010 ^(a)	7F3	3-24-66	4-24-69	18 Tensile 10 Uniaxial Creep	304 SS	6.6×10^{22}
X-014 ^(a)	2I3I	7-17-66	12-31-66	50 Tensile	304 SS	1.7×10^{22}
X-018	2B1	12-6-66	2-25-69	198 Tensile or Uniaxial Creep	304 SS, 316 SS, 348 SS, 321 SS	4.4×10^{22}
X-019	6D2	1-13-67	July 69	64 Tensile 24 Uniaxial Creep	304 SS 316 SS	2.5×10^{22}
X-021 ^(b)	2D1	2-27-67	9-16-68	127 Tensile or Uniaxial Creep	304 SS, 316 SS ^(c) Ni Base Alloys	3.2×10^{22}
X-021A ^(b)	2D1	9-27-68	11-16-68	86 Density 16 Biaxial Creep 14 Tensile 10 Uniaxial Creep	316 SS ^(c) Ni Base Alloys	5.0×10^{21}
X-022	7C4	2-27-67	6-15-68	595 Tensile or Uniaxial Creep 65 Biaxial Creep	304 SS, 316 SS, 348 SS, 321 SS	1.1×10^{22}
X-030 ^(a)	6E1	10-10-67	3-6-68	5 Tensile 3 Uniaxial Creep	304 SS	5.2×10^{21}
X-067	4F2	1-5-69	Sept 69	204 Tensile or Uniaxial Creep 20 Fatigue	304 SS, 316 SS, Weldment	0.9×10^{22}

a. Mark A pins irradiated on space available basis, a22 other pins are B-7 type.

b. Single pin interchange.

c. Variety of other alloys also included.

d. $E > 0.1$ MeV.

is to develop swelling data at fluence levels that can be obtained in reasonable times and to use this data to develop the required swelling models.

Density measurements on sections from an EBR-II safety rod thimble were reported with the corresponding exposures in a previous quarterly report.⁽¹⁷⁾ Further consideration of the data available indicates that the specimens were at a different position with respect to the midplane than had been reported. The corrected data are indicated in Table 4.18.

TABLE 4.18. *Irradiation Induced Swelling in EBR-II Safety Rod Thimble*

Distance From Midplane, in.	Fluence x 10^{-22} (n/cm ² , E > 0.1 MeV)	Temp, °F	Swelling $\Delta\rho/\rho$, %
20.8	0.36	872	0.14
15.0	1.28	852	0.41
10.0	2.62	826	0.93
8.0	3.46	810	1.55
5.5	4.80	786	2.44
3.0	5.89	756	3.09
0.5	6.40	732	2.41
- 2.0	6.14	718	1.53
- 4.0	5.57	710	0.87
- 9.0	2.62	700	0.47
-14.8	1.34	700	0.27
-19.8	0.601	700	0.11

Immersion density measurements have recently been completed on AISI ^ 316 tensile specimens that had been irradiated in the EBR-II (X-018 Subassembly) to a maximum fluence of 3.0×10^{22} n/cm² (E > 0.1 MeV) at temperatures ranging from 700 to 1400 °F. The initial results are given in Table 4.19. These results

TABLE 4.19. *Irradiation Induced Swelling in Solution
Treated 316 Stainless Steel*

Density, (ρ), g/cm ³	Volume Change(a) ($-\Delta\rho/\rho$), %	Distance from Midplane, in.	Estimated ^(b) Temperature, °F	Fluence(c) n/cm ² x 10 ⁻²² (E > 0.1 MeV)
7.974	-0.05	19.7	700	0.29
7.972	-0.02	19.7	700	0.29
7.964	+0.07	14.1	760	0.72
7.957	+0.17	12.2	920	1.0
7.959	+0.14	12.2	920	1.0
7.948	+0.28	10.3	965	1.26
7.946	+0.30	10.3	1045	1.26
7.925	+0.57	6.6	1150	1.98
7.946	+0.30	6.6	1240	1.98
7.919	+0.64	2.8	1255	2.88
7.950	+0.26	2.8	1370	2.88

- a. The volume change was obtained by using a value of 7,970 g/cm³ for the original density. This value was determined from material that may have been from a different heat.
- b. These temperatures represent tentative results from heat transfer calculations. Consequently, the absolute values may be in error by 100 to 200 °F.
- c. The maximum fluence was obtained by converting the net exposure (10,736 MWd/ton) to neutron fluence (E > 0.2 MeV) and using calculated flux profiles to obtain relative values.

indicate that at temperatures above about 900 °F the temperature dependence of swelling may be less than predicted by the swelling equation suggested in the previous quarterly report. In Figure 4.15, the swelling values have been normalized with respect to fluence by dividing by $(\Phi t \times 10^{-22})^{1.7}$, where Φt is the fluence. As may be seen from the plot, the data

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FIGURE 4.15. *Temperature Dependence of Swelling Indicated by Initial Results on Solution Treated AISI Stainless Steel*

indicate that maximum swelling occurs near 1050 $^{\circ}\text{F}$ and none occurs above approximately 1400 $^{\circ}\text{F}$. For purposes of comparison, swelling values predicted by the swelling equation suggested in the last quarterly report have also been normalized and plotted on the figure. The two curves agree reasonably well at temperatures below 900 $^{\circ}\text{F}$, but at higher temperatures the data from Table 4.19 exhibit less temperature dependence.

At present, the temperatures and volume changes given in Table 4.19 must be considered tentative. In order to determine the volume change, it was necessary to assume an initial density of 7.970 g/cm³. This value was based upon that of a 316 specimen from a heat different from the irradiated material. The temperatures represent initial results from heat transfer calculations and, because of the complex geometry associated with the specimens, absolute values may be in error by 100 to 200 °F. Review and refinement of these calculations to evaluate and reduce these errors must be completed before the data can be used with confidence.

In a previous quarterly report,⁽¹⁷⁾ an attempt was made to estimate the kinetics for interaction between voids and interstitial loops during irradiation. The essential features postulated were that voids and loops would interact by virtue of intersecting one another during growth. After contact between the loop and the void, it was assumed that the loop could climb into the void and thereby reduce the void volume. Further consideration indicates that the range of applicability of the previous analysis is restricted to values of swelling below that where significant effects of void-loop interaction may be expected to occur.

The result previously obtained was that the amount of metal swelling would be reduced to an amount:

$$S = \frac{S'}{1 + \alpha S'} \quad (1)$$

where S is the swelling to be expected if interactions were to occur, and S' is the amount of swelling with no interactions. The term α is an interaction parameter so defined that a void with volume V absorbs all of the interstitial loops that form and are present within volume αV . It was assumed that, although α may vary with neighboring void and loop sizes, it

could be considered for the bulk process. The value of a was calculated by considering that the loops had a random angular orientation:

$$\alpha = \frac{1 + 2r_L^3}{\pi r_V^3}, \quad (2)$$

where r_L is the loop radius, and r_V is the void radius. Since the average loop radius typically appears to be about twice the average void radius, reasonable values of a were considered to be about 10. With this value for a , Equation (1) predicts a significant reduction in swelling due to interactions (25%) at 2.5% observed change in volume.

A more detailed analysis of void-loop interactions indicates that, when calculating a , one must take into account the fact that the loops do not have random angular orientation but must lie on (111) crystallographic planes. When this is done and when it is also assumed that the dislocation line associated with the loop must touch the void for interaction, the following result is obtained for a :

$$\alpha = \frac{3}{2} \frac{r_L}{r_V} \quad (3)$$

Using this value of a , Equation (1) does not predict significant reductions in swelling until about 8% observed volume change (for a 25% reduction due to interactions).

In deriving Equation (1), it was implicitly assumed that the interacted regions surrounding the voids did not overlap. After significant overlap occurs, a may be expected to change with time and Equation (1) would not be applicable. The range of applicability of Equation (1) may, therefore, be estimated by considering the possibility of overlap between interacted regions. Assuming r_L is twice r_V , a simple calculation indicates that each interacted region would overlap with an

average of one other region after 0.5% swelling or volume change. Thus, Equation (1) does not predict any appreciable effect of void-loop interaction within its range of applicability. This discussion should not detract from the possibility that void-loop interactions may inhibit swelling, but is intended to show that the simplified analysis previously reported is not sufficient to describe the effect. It appears that an adequate analysis must make use of the nucleation and growth kinetics of both voids and loops; these are not known at the present.

Voids in irradiated stainless steel are normally shown in the absorption contrast condition. For this condition, the voids appear as light regions surrounded by a dark border. Since the thickness of the dark border is often a significant fraction of the void diameter, the total void volume determined by microscopy depends upon whether one uses the outside or inside edge of the dark border to measure the void diameter. Whether the border should be considered as a part of the void or part of the matrix has been a source of contention for some time. An observation of the same voids under two different contrast conditions suggests that the dark border should be considered as part of the void.

Van Landuyt et al.⁽¹⁸⁾ have considered the contrast effects from small voids when the crystal is oriented for the Bragg Diffraction condition ($S = 0$). According to their analysis, if there is no strain field associated with the void, the intensity of the beam passing through the foil at the void equals the intensity of a beam passing through a perfect foil which has the thickness of the original foil less the diameter of the void. This means that for $S = 0$, as one proceeds from the matrix into the void, the transmitted intensity changes from the background intensity to a different value depending on the void size and foil thickness. The first perceptible

change in intensity should indicate the surface of the void. Unfortunately, due to the high concentration of displacement damage, voids in highly irradiated materials are poorly delineated under the Bragg Diffraction condition. However, by annealing at moderate temperatures, a large portion of the displacement damage may be removed without significantly affecting the voids.

A foil taken from 304 SS that had been irradiated to 7×10^{22} n/cm² (total) and annealed for 1 hr at 1200 °F was observed in the absorption contrast condition (Figure 4.16) and the Bragg Diffraction condition (Figure 4.17). Using the approach of Ashby and Brown,⁽¹⁹⁾ no strain fields were observed around the voids.

The diameter of seven voids was measured to the nearest 0.1 mm on 180,000X micrographs in both the symmetrical Laue condition ($S = 0$) and in the absorption contrast condition. Within the accuracy of the measurements, the diameter of the void for Bragg contrast condition ($S = 0$) exactly equals the outside diameter of the black border surrounding the light region of the void shown in absorption contrast.

Previously reported dark field transmission electron microscopy showed that Type 304 SS irradiated at 750 °F to 7×10^{22} n/cm² (total) contained a high concentration of small precipitates ($<100 \text{ \AA}$ parameter) having more than one morphology. Examination of single-step carbon extraction replicas taken from this material clearly showed the pattern of $M_{23}C_6$ carbide. Analysis will continue in an attempt to identify other phases.

Seven irradiated austenitic stainless steel samples were sent to Dr. H. Farrar at Atomics International for helium analysis. The helium concentration was estimated to have an uncertainty of less than 10%. The results of the analyses are given in Table 4.20.

Neg 7402A

200,000X

FIGURE 4.16. *Bright Field Electron Micrograph of Type 304 Stainless Steel Postirradiation Annealed 1 hr at 1200 °F After Irradiation at 750 °C to 7×10^{22} n/cm² (Total). Arrow points to void in absorption contrast condition.*

Neg 7407C

200,000X

FIGURE 4.17. *Same Materials as in Figure 4.16 with Void in Symmetrical Laue Contrast Condition*

TABLE 4.20 Helium Analysis of Irradiated Steel

Sample No.	Irradiated In Reactor	Type of Material	PNL Sample Designation	Neutron Fluence		Measured Helium Concentration, atom ppm
				(Total)	(E > 1 MeV)	
1	EBR-II	316 SS	14G-16-5	4×10^{21}	--	0.25
2	EBR-II	316 SS	14B-16-1	8×10^{21}	--	0.58
3	EBR-II	304 SS	3X (Safety Rod Thimble)	$10^{20} - 10^{21}$	--	0.025
4	EBR-II	304 SS	5F (XGO5 Thimble)	3.6×10^{22}	9.7×10^{21}	6.9
5	EBR-II	304 SS	C (Center Rod Thimble)	3.4×10^{22}	9.1×10^{21}	9.9
6	EBR-II	304 SS	3E (Safety Rod Thimble)	7.0×10^{22}	1.9×10^{22}	16.1
7	ETR	304 SS	GEH-20-34-7	--	6×10^{21}	305.0

Samples 1 and 2 (Type 316 SS) were irradiated in Row 7 of the EBR-II in the same pin, 12 in. above and 7 in. below the reactor midplane, respectively. The ratio of neutron fluence of Samples 1 and 2 to the amount of helium produced (2.0 versus 2.3) is quite self consistent.

Samples 4, 5, and 6 (Type 304 SS) were irradiated within 2 in. of the reactor midplane in Rows 4, 5, and 3, respectively. Sample 3 was taken from the top section of the safety rod thimble about 10 in. above the top of the core and was to be the "control" specimen. The amount of helium produced in Specimens 4, 5, and 6 was fairly consistent with the ratio of the neutron fluence (2.3 versus 1.9 and 1.6 versus 2.1). However, the concentration of helium in the steel per neutron is about 3-1/2 times higher in Specimens 4, 5, and 6 (Type 304 SS) than in Specimens 1 and 2 (Type 316 SS).

The five elements which were calculated to contribute over 98% of the helium produced by fast-neutron transmutation in Type 316 SS and Type 304 SS are shown in Table 4.21.

TABLE 4.21. *Fast Neutron (η, α) Transmutation of Major Alloy Constituents*

Element	Type 316 SS ^(a)			Type 304 SS ^(a)		
	wt %	Atom He	(atom x 1 nvt)	wt %	Atom He	(atom x 1 nvt)
Ni	13.55	539	(-30)	9.36	365	(-30)
Fe	63.64	279	(-30)	70.31	304	(-30)
Cr	17.80	72	(-30)	18.51	74	(-30)
N	0.041	67	(-30)	0.029	46	(-30)
Mo	2.33	10	(-30)	0.10	0.4	(-30)
TOTAL		9.8 x 10 ⁻²⁸			8.0 x 10 ⁻²⁸	

a. Boron Content < 5 ppm for both heats of steel.

The calculations were based on the fission spectrum average (n, α) cross section of the elements tabulated by Alter and Weber⁽²⁰⁾ except for nickel where the cross section reported by Weitman and Daverhog⁽²¹⁾ was used. These calculations do not explain the difference in helium produced in Type 304 SS and 316 SS per nvt of fluence. The difference may be due to the harder neutron energy spectrum near the EBR-II core midplane in Rows 3, 4, and 5, where the 304 SS was irradiated, resulting in more (n, α) transmutations than in Row 7 locations 6 and 12 in. from the core centerline, where the Type 316 SS was irradiated.

The calculated concentration of helium produced in the Type 304 SS irradiated near the EBR-II reactor midplane is in reasonable agreement with the measured value for neutrons with $E > 1.0$ MeV. For example, Sample 6, irradiated to 1.9×10^{22} n/cm² ($E > 1.0$ MeV) is calculated to contain 15.2 atom ppm compared to the measured value of 16.1 atom ppm. However, a dramatic increase in the (n, α) cross section with "harder" fast neutrons is indicated by the data of Specimen 7. The helium concentration of Sample 7 irradiated in the ETR, a thermal reactor, is more than 20 times the calculated value of 13 atom ppm (including 3 atom ppm assuming complete boron transmutation). This result is consistent with the data of Jackson and Dahl⁽²²⁾ that the average energy of fast neutron ($E > 0.1$ MeV) in the ETR is higher than in the EBR-II.

Biaxial Stress-Rupture Studies - R. W. Barker

The objective of this program is to determine the effects of fast reactor irradiation on the biaxial stress-to-rupture properties of candidate fast reactor cladding materials.

Biaxial stress-rupture tests have been performed on RISI Type 316 SS irradiated to a nominal fluence of $\sim 1 \times 10^{22}$ n/cm² ($E > 0.1$ MeV) in subassembly X-022 in EBR-II. Most of the

irradiated samples tested were irradiated to a fluence of 0.8×10^{22} n/cm². Test temperatures were 1000, 1200, and 1400 °F. The balance of the samples were irradiated to the higher fluence (1×10^{22} n/cm²) and will be tested at temperatures of 900, 1000, and 1200 °F.

The data obtained from biaxial stress-rupture tests on unirradiated as-received AISI Type 316 SS cladding are summarized in Table 4.22. All as-irradiated test data generated to date are summarized in Table 4.23. Figure 4.18 is a plot of stress versus rupture-time for the various tests, while Figure 4.19 is a plot of rupture strain versus rupture-time for these tests. The data points are for the as-irradiated tests, while the solid lines are for the control tests.

Some irradiation hardening (strengthening) is present at 1000 °F for very short rupture times. The as-irradiated test data then appears to coincide with the as-received data at a rupture-life of about 10 hr. At 1200 °F, the irradiated points data agree with the curve formed by the as-received data. At 1400 °F, a marked decrease in rupture-life is observed.

The failure behavior of irradiated specimens is considerably different from that of the controls. Illustrations of typical bursts of unirradiated and irradiated specimens are shown in Figures 4.20 and 4.21, respectively. Both specimens were tested at 1400 °F in an argon atmosphere and had rupture times between 5 and 10 hr. The as-received specimen tears apart transversely, forming a ruptured section approximately the same length as the initial longitudinal split (Figure 4.22). The balance of the specimen is left undamaged, and from it precise diameter measurements can be made. The as-irradiated specimens do not show a tendency to tear in the transverse direction. Instead, the longitudinal split becomes more pronounced, often propagating the entire length of the specimen.

TABLE 4.22. *Biaxial Stress-Rupture Tests on AISI Type 316 SS in the As-Received Condition*

<u>Test Temperature, °F</u>	<u>Hoop Stress, psi</u>	<u>Rupture Time, hr</u>	<u>Rupture Strain, %</u>
1000	52,000	0.1 (Burst)	
	51,500	6	16.8
	48,000	55	14.0
	50,000	78	21.0
	42,000	545	10.5
1200	39,400	0.5	16.5
	31,200	6	6.2
	29,000	15	8.6
	25,500	70	4.8
	19,900	376	1.9
	23,000	880	23.5
	17,000	1236	14.4
1400	25,000	0.4	24.0
	19,800	5.5	19.2
	19,800	6.8	19.2
	17,000	16.5	13.9
	15,200	42	21.5
	9,600	300	9.1

TABLE 4.23. *Biaxial Stress-Rupture Tests on AISI Type 316 SS Irradiated to $\sim 0.8 \times 10^{22}$ n/cm² (E > 0.1 MeV) at 815 °F in EBR-11*

AS-IRRADIATED DATA

<u>Test Temp, °F</u>	<u>Irrad. Test No.</u>	<u>Hoop Stress, psi</u>	<u>Rupture Time, hr</u>	<u>Rupture Strain, %</u>
1000	19	58,000	Burst	N.R.M.
	26	54,000	2.2	N.R.M.
	27	50,000	9.6	3.8
1200	17	36,000	0.6	5.8
	12	36,000	2.1	1.9
	13	30,000	32.0	2.4
	20	25,000	20.0	0.5
	24	23,500	70.0	1.9
1400	18	23,500	0.3	N.R.M.
	21	23,500	1.4	N.R.M.
	22	23,500	8.2	1.4
	23	7,800	33.0	5.3

Exposure for this specimen is 1×10^{22} n/cm² (E > 0.1 MeV).

Neg 0693125-1

FIGURE 4.18. The Effect of Irradiation to $\sim 8 \times 10^{21} \text{ n/cm}^2$ ($E > 1.0 \text{ MeV}$) and Test Temperature on the Biaxial Stress Rupture Life of AISI Type 316 SS in Inert Gas

Neg 0693125-2

FIGURE 4.19. The Effect of Irradiation to $\sim 8 \times 10^{21}$ n/cm² ($E > 1.0$ MeV) and Test Temperatures on the Rupture Ductility AISI Type 316 SS in Inert Gas

Neg 4691313

FIGURE 4.20. *A Typical Burst Failure for AISI Type 316 SS in the As-Received Condition. Test conditions were 1400 °F, 19,800 psi stress, rupture time 6.8 hr*

FIGURE 4.21. *A Typical Burst Failure for AISI Type 316 SS
Irradiated to $\sim 8 \times 10^{21}$ n/cm² ($E > 0.1$ MeV).
Test conditions were 1400 °F, 12,000 psi stress,
rupture time 8.2 hr.*

Neg 0693098
FIGURE 4.22 Theta Parameter Master Plot of Irradiated and As-Received AISI Type 316 SS Cladding Tested in Inert Atmosphere. [Fluence is $\sim 8 \times 10^{21}$ n/cm² ($E > 0.1$ MeV)].

Because of the tendency of irradiated specimens to fail in a burst mode, opening more fully from end to end, a good evaluation of rupture strain cannot usually be made. However, where data are available, irradiation is observed to greatly reduce ductility (Figure 4.19). The lowest ductility for as-received material occurs at 1200 °F. This appears also to be the case for the irradiated material. Rupture strains at 1000 and 1400 °F test temperatures appear to be similar, both in the unirradiated and irradiated conditions. Tests for which strain measurements could not be made are identified as not readily measureable (NRM) in Table 4.23.

The length/diameter ratio for the irradiated specimens mounted with mechanical fittings varies from 4 to about 6, depending on the length of clad after removal of the fittings and effects of remote assembly such as poor placement of the sleeve (Figure 4.21). The ratio for the particular specimen shown in Figure 4.22 was 4. Work by Venard⁽²⁴⁾ has shown that a ratio as low as 3.3 is sufficient to provide valid stress-rupture data.

Stress-rupture data from as-received and as-irradiated AISI Type 316 SS specimens have been replotted using the theta parameter suggested by other investigators.⁽²³⁾ The master plot, shown in Figure 4.22, shows that biaxial stress-to-rupture data at various test temperatures can be related the theta parameter 0:

$$\theta = \log[t_r \exp (-Q/RT)]$$

where:

t_r = time to rupture (hr),

Q = activation energy for creep (cal/mole),

T = absolute temperature (°K),

R = gas constant (cal/mole-degree).

With $Q = 95,000$ cal/mole, all as-received data in Table 4.23 is represented by a single line. Excellent correlation is obtained between tests at various test temperatures.

As-irradiated data from Table 4.22 is also plotted in Figure 4.22. At low theta values, irradiation hardening results in an increase in rupture. This increase is consistent with tensile results on AISI Type 316 SS from the same irradiation experiment. At high theta values the rupture life is decreased. This is probably due to helium embrittlement at the elevated temperatures. The fractures at high temperatures appear to be intergranular as expected from the helium embrittlement process.⁽²⁵⁾ At intermediate theta values an even balance between irradiation hardening and helium embrittlement appears to exist.

Mechanical Testing of Fuel Pin Cladding - R. L. Fish

The purpose of this program is to provide a basis for evaluating the combined effects of fuel-cladding interaction, fluence and operating temperature on the postirradiation burst and stress-rupture properties of fuel pin cladding. The effort during this fiscal year will use fueled cladding specimens from PNL-1 and BNW-1 fuel pin subassemblies, and NUMEC "A" series fuel pins.

Preliminary burst tests have been conducted on 2-3/4-in. long fueled cladding specimens taken from the PNL-1 subassembly. The cladding used to fabricate these pins was annealed Type 304 SS (E lot). The positions of the respective specimens are indicated on the diagram in Figure 4.23. The results from these tests, Table 4.24, will provide guidelines for performing longer-term stress-rupture tests at 1000 and 1200 °F on specimens from other PNL-1 fuel pins. Similar burst tests will be conducted on BNW-1 and NUMEC fuel pins for the same purpose. The same two temperatures are planned for the stress-rupture testing of these latter pins.

Neg 0693268-1

FIGURE 4.23 st Specimen tions

TABLE 4.24. PNL-1 Burst Test Results

Specimen Identification	Clad OD Operating Temp, °F	Test Temp, °F	Burst Pressure, psi	Maximum Hoop Stress At Rupture, psi	Strain ($\Delta D/D$), %
"E" Lot, Unirradiated Control	---	900	7760	56,960	19.2
Plenum Region Specimen, 1-6	~850	900	8050	59,090	18.4
1-3:C ₂	840	1000	9100	66,790	6.7
1-6:C ₃	830	900	9830	72,150	9.6
1-3:G ₄	910	900	5300	38,900	
1-6:G ₂	890	900	3700	27,160	0.3
1-6:G ₁	890	900	3000	22,020	1.0

a. C₂, C₃ from above fuel midplane; G₄, G₂, G₁ from below fuel midplane.

Activity measurements made on flux wires from the PNL-1 subassembly indicate that the peak fluence experienced by these pins was about 3.5×10^{21} n/cm² (E > 0.1 MeV).

Note that each of the fueled specimens taken from above the fuel midplane failed at a significantly lower stress level than did those specimens taken from below the midplane.

A detailed analysis of the cladding microstructure, failure mode, and possible fuel-clad interaction products has been initiated to determine the reasons for this type of behavior. Representative photomicrographs of the two completely different types of failure modes are shown in Figures 4.24 and 4.25. Figure 4.24, specimen 1-6, C₃ illustrates the transgranular type of fracture characteristic of Type 304 SS irradiated under these conditions. Figure 4.25, specimen 1-6, G₁ reveals only the intergranular type of failure characteristic of the specimens taken from above the fuel midplane.

Neg E9716

50X

FIGURE 4.24. *Normal Transgranular Rupture of Specimen 1-6, C_3*

Neg E9739

250X

FIGURE 4.25. *Intergranular Failure of Specimen 1-6, G_1*

A scanning electron microscope was used to detect possible fission product penetration of the cladding or any other unusual compositional variations in the G₁ specimen. The X-ray spectral results of this effort showed no apparent fission product penetration but did reveal increased chromium content at the grain boundaries. This could indicate some type of chromium, iron carbide precipitate in this region.

Carbon extraction replicas and electron microscope transmission specimens are being prepared from both C₃ and G₁ burst specimens to determine if any microstructural differences exist between the two specimens.

A microprobe sample was also prepared from G₁ for possible detection of fission product penetration. Efforts thus far have failed to reveal any such penetration. Because of possible high humidity conditions under which this sample was prepared, another specimen is being prepared for the microprobe, with care taken to avoid the use of water and other high humidity environments.

It has been determined that all of these initial biaxial specimens have been exposed to high humidity, post-irradiation environments which in combination with irradiation conditioning may have caused the difference in the test results observed. However, the effort to determine the exact cause of these anomalous results is continuing. The investigation thus far seems to further indicate that a presently unknown corrosive agent penetrated and weakened the grain boundaries of a sensitized cladding grain structure, thus resulting in the intergranular fractures. This hypothesis is also preliminary.

Further tests will be conducted on both PNL-1 and BNW-1 fueled specimens prepared under carefully controlled humidity conditions. This will minimize any effect that moisture may have on fueled specimen behavior during the biaxial testing. Testing of NUMEC fuel pins will begin in October.

RELATED FAST REACTOR SUPPORTING STUDIESAcceptance Testing and Clad Characterization - M. M. Paxton

The objective of this program is to ensure that acceptable cladding material is obtained and used throughout the FFTF Fuel Development and Irradiation Testing Program. Under this program, lot-to-lot variations in selected mechanical properties of AISI Type 304 and Type 316 SS tubing are being determined and evaluated.

A check analysis by Lukens Steel Company has been completed on five "H" lot tube lengths, Table 4.25.

Metallography on five "H" lot tube lengths has been completed. Figures 4.26 through 4.29 are typical transverse and longitudinal photomicrographs. The grain size varies between ASTM 7 and 8.

Tensile tests on tubing lots E, F, G, and H have been completed at room temperature, 800, 1000, 1200, 1400 and 1600 °F at strain rates of 0.01 and 1.0 min⁻¹. The results on tubing lots E and F are tabulated in Tables 4.26 and 4.27 and summarized in Figures 4.30 through 4.33. Results on tubing lots G and H are tabulated in Tables 4.28 and 4.29 and summarized in Figures 4.34 through 4.37.

Slow strain-rate tensile tests, $\dot{\epsilon} = 0.01 \text{ min}^{-1}$, on tube lots E and F show the lots to be significantly stronger than the national average data at test temperatures below 1000 °F.(26) This may be a result of the slight amount of cold work, 1 to 5%, due to a final straightening pass during fabrication. At this same strain rate, the ultimate strength and yield strength values at 1200 and 1400 °F are comparable to the national average data and fall below the average at 1600 °F.(26)

TABLE 4.25. *Chemical Analysis on Lot Tubing of 316 Stainless Steel*

Vendor Analysis (wt%):

Heat No.	C	Mn	P	S	Si	Ni	Cr	Mb	B	N ₂	Cu
20276	0.05	1.56	0.012	0.013	0.58	13.34	17.67	2.42	0.0004	0.049	0.11
20276	0.05	1.56	0.010	0.011	0.56	13.32	17.73	2.50	0.0008	0.052	0.11
20276	0.05	1.59	0.013	0.015	0.58	13.29	17.67	2.42	0.0009	0.055	0.12
20276	0.05	1.56	0.012	0.014	0.53	13.29	17.73	2.41	0.0008	0.050	0.11
20276	0.05	1.55	0.011	0.013	0.53	13.30	17.67	2.50	0.0008	0.052	0.12

Check analysis by Lukens Steel Company (wt%):

Tube No.	C	Mn	P	S	Si	Ni	Cr	Mb	B	N ₂	Cu
H060	0.043	1.48	0.018	0.018	0.49	13.33	17.95	2.37	0.0015	0.044	0.18
H135	0.046	1.48	0.017	0.108	0.50	13.33	17.95	2.39	0.0015	0.044	0.18
H157	0.045	1.48	0.016	0.017	0.50	13.33	17.95	2.37	0.0015	0.046	0.18
H234	0.047	1.48	0.017	0.018	0.50	13.33	17.95	2.39	0.0015	0.040	0.18
H240	0.045	1.48	0.017	0.018	0.50	13.33	17.95	2.39	0.0015	0.043	0.18

Neg 469-578A

80X

Neg 469-578B

195X

Neg 469-578C

390X

Neg 469-578D

780X

FIGURE 4.26. *Tubing, Lot H Mill Annealed 316 Stainless; Transverse Section of Tube Length H-135. Etch: 30% glycerin, 20% HCl, 10% HNO₃.*

Neg 469-577A

80X

Neg 469-577B

195X

Neg 469-577C

390X

Neg 469-577D

780X

FIGURE 4.27. *Tubing, Lot H Mill Annealed 316 Stainless;
Longitudinal Section of Tube Length H-135.
Etch: 30% glycerin, 20% HCl, 10% HNO₃.*

Neg 469-580A

80X

deg 469-580B

195X

Neg 469-580C

390X

Beg 469-5800

780X

FIGURE 4.28. *Tubing, Lot H Mill Annealed 316 Stainless; Transverse Section of Tube Length H-157. Etch: 30% glycerin, 20% HCl, 10% HNO₃.*

Neg 469-579C

390X

Neg 469-5790

780X

FIGURE 4.29. *Tubing, Lot H Mill Annealed 316 Stainless; Longitudinal Section of Tube Length H-157. Etch: 30% glycerin, 20% HCl, 10% HNO₃.*

TABLE 4.26. *Tensile Properties, Tube Lot E*

Specimen No.	Strain Rate, min ⁻¹	Test Temp, °F	0.2% YS, psi	UTS, psi	Uniform Elong, %	Total Elong, %
ET-1	0.01 ↓	800	22,120	73,980	40.0	43.2
ET-2		800	20,930	74,410	41.5	44.3
ET-3		800	21,690	73,980	39.9	43.6
ET-4		1000	18,980	68,050	40.5	44.1
ET-5		1000	18,640	67,880	38.5	42.4
ET-6		1000	19,240	68,730	41.2	45.2
ET-7		1200	17,970	47,460	25.5	33.1
ET-8		1200	16,530	50,000	26.5	33.1
ET-9		1200	15,850	48,200	28.0	35.5
ET-10		1400	14,240	28,640	19.1	35.1
ET-11		1400	17,120	30,250	12.1	42.5
ET-12		1400	15,760	28,050	15.5	42.6
ET-13		1600	10,680	13,390	--	--
ET-14		1600	10,590	12,970	7.5	36.6
ET-15		1600	9,660	12,540	--	--
ET-16	1.0 ↓	R.T.	41,100	96,780	72.7	87.8 } 86.3
ET-17		R.T.	42,370	98,310	73.0	
ET-18		R.T.	42,800	98,730	75.9	
ET-19		R.T.	47,460	94,240	52.8	85.7 } 85.3
ET-20		R.T.	47,420	94,240	53.7	63.7
ET-21		R.T.	44,750	90,850	54.4	65.9
ET-22		800	24,070	68,140	37.1	42.3
ET-23		800	24,070	68,140	36.9	41.8
ET-24		800	23,390	68,810	36.2	42.3
ET-25		1000	19,320	60,340	35.5	41.1
ET-26		1000	21,360	65,080	33.0	39.7
ET-27		1000	22,030	62,710	34.9	39.2
ET-28		1200	18,980	54,240	29.8	35.0
ET-29		1200	23,050	54,240	30.6	35.2
ET-30		1200	18,640	54,580	29.9	35.6
ET-31		1400	16,270	38,980	26.3	45.6
ET-32		1400	20,000	39,320	27.0	45.7
ET-33		1400	16,950	40,680	25.7	46.5
ET-34		1600	15,250	25,290	20.5	60.7
ET-35		1600	16,270	25,760	19.3	57.6
ET-36		1600	15,930	25,930	19.6	58.8

TABLE 4.27. *Tensile Properties, Tube Lot F*

Specimen No.	Strain Rate, min ⁻¹	Test Temp, °F	0.2% YS, psi	UTS, psi	Uniform Elong, %	Total Elong, %
FT-1	0.01	70	41,530	98,310	70.6	89.1
FT-2	↓	70	51,270	102,970	70.4	84.7
FT-3		70	47,630	103,050	64.8	76.9
FT-4		800	23,560	69,410	35.8	41.4
FT-5		800	23,310	68,810	35.2	40.1
FT-6		800	23,560	64,580	30.4	32.6
FT-7		1000	22,460	65,250	33.1	41.9
FT-8		1000	25,420	65,250	34.9	39.7
FT-9		1000	23,220	65,420	35.0	40.8
FT-10		1200	19,490	44,490	24.9	46.2
FT-11		1200	20,760	43,900	21.6	47.5
FT-12		1200	20,080	44,240	26.9	48.8
FT-13		1400	13,560	22,360	17.8	60.0
FT-14		1400	14,660	23,470	13.8	57.8
FT-15		1400	13,560	22,800	18.0	61.0
FT-16		1600	8,310	10,590	12.6	40.5
FT-17		1600	8,470	10,680	--	--
FT-18		1600	8,470	10,340	17.1	46.1
FT-19	1.0	R.T.	47,200	89,920	49.2	64.1
FT-21	↓	R.T.	---	---	--	63.0
FT-22		R.T.	53,810	97,710	52.0	60.1
FT-26		800	28,730	65,760	31.6	35.6
FT-27		800	33,220	66,690	31.3	36.4
FT-28		800	30,340	65,760	33.0	36.8
FT-30		1000	20,250	57,540	30.4	35.9
FT-31		1000	22,460	57,880	29.3	34.6
FT-32		1000	20,340	58,310	30.3	35.2
FT-33		1200	20,590	50,680	26.9	31.6
FT-34		1200	---	---	--	31.1
FT-35		1200	19,320	51,690	26.8	31.6
FT-37		1400	16,610	36,780	23.3	48.8
FT-39		1400	18,640	37,120	20.4	51.4
FT-40		1400	16,950	34,750	25.8	52.0
FT-41		1400	16,950	36,100	24.4	52.6
FT-42		1600	14,240	21,020	17.5	68.4
FT-43		1600	14,920	22,030	19.1	63.3
FT-45		1600	14,920	33,220	20.0	65.8

Neg 0693251-7

FIGURE 4.30. 0.2% Offset Yield Strength and Ultimate Tensile Strength Versus Test Temperature of Tube Lot E, Mill Annealed. 304 Stainless Steel.

Neg 0693251-8

FIGURE 4.31. *Uniform and Total Elongation Versus Test Temperature of Tube Lot E, Mill Annealed 304 Stainless Steel*

Neg 0693251-5

FIGURE 4.32. 0.2% Offset Yield Strength and Ultimate Tensile Strength Versus Test Temperature of Tube Lot F, Mill Annealed 304 Stainless Steel

Neg 0693251-1

FIGURE 4.33. Uniform and Total Elongation Versus Test Temperature of Tube Lot F, Mill Annealed 304 Stainless Steel

TABLE 4.28. Tensile Properties, Tube Lot G

Specimen No.	Strain Rate, min ⁻¹	Test Temp, °F	0.2% YS, psi	UTS, psi	Uniform Elong, %	Total Elong, %
GT-1	0.01 ↓ 1.0 ↓	R.T.	42,970	93,310	64.9	81.4 } 79.8 76.4 81.8
GT-2		R.T.	41,530	93,390	64.1	
GT-3		R.T.	42,200	91,530	68.2	
GT-4		800	27,800	81,440	39.3	
GT-5		800	26,530	78,560	39.7	41.2
GT-6		800	27,710	80,850	38.5	40.4
GT-7		1000	24,580	74,750	33.8	37.4
GT-8		1000	23,310	74,580	31.8	36.2
GT-9		1000	24,240	76,270	36.6	41.3
GT-10		1200	20,930	52,370	18.8	45.0
GT-11		1200	22,030	54,240	17.6	42.9
GT-12		1200	20,340	53,810	22.1	45.4
GT-13		1400	17,540	29,240	11.5	60.6
GT-14		1400	16,950	28,640	11.9	58.3
GT-15		1400	16,440	28,140	12.7	58.3
GT-16		1600	10,850	13,900	9.1	47.9
GT-17		1600	12,030	14,070	9.0	46.4
GT-18		1600	11,440	14,150	9.4	46.0
GT-19	1.0 ↓	R.T.	47,460	93,560	51.6	61.0
GT-20		R.T.	48,140	93,560	46.7	59.8
GT-21		R.T.	46,780	92,200	44.1	58.0
GT-22		800	25,760	71,020	32.8	37.4
GT-23		800	27,800	70,170	34.0	36.7
GT-24		800	28,810	72,200	34.2	37.2
GT-25		1000	24,070	66,780	30.8	33.2
GT-26		1000	25,080	66,780	30.7	33.3
GT-27		1000	---	---	--	33.4
GT-28		1200	22,370	60,680	28.6	34.0
GT-29		1200	20,680	58,310	28.5	33.2
GT-30		1200	20,680	58,310	25.9	32.0
GT-31		1400	19,660	43,220	23.9	48.1
GT-32		1400	19,660	42,370	32.3	48.5
GT-33		1400	21,360	43,730	22.5	47.6
GT-34		1600	12,370	27,120	19.0	65.9
GT-35		1600	18,640	28,310	17.3	70.0
GT-36		1600	15,930	27,120	17.3	61.5

TABLE 4.29. Tensile Properties, Tube Lot H

Specimen No.	Strain Rate, min ⁻¹	Test Temp, °F	0.2% YS, psi	UTS, psi	Uniform Elong, %	Total Elong, %
HT-1	0.01 ↓ 1.0 ↓	70	42,800	92,800	57.5	75.6
HT-2		70	44,070	93,220	55.9	72.6
HT-3		70	46,610	98,310	58.3	70.9
HT-4		800	26,690	78,140	37.7	41.7
HT-5		800	25,850	77,970	39.5	42.6
HT-6		800	25,850	79,240	37.3	39.9
HT-7		1000	24,150	74,410	35.6	38.6
HT-8		1000	23,730	76,270	36.3	40.7
HT-9		1000	23,730	75,420	--	--
HT-10		1200	21,780	52,540	21.4	39.1
HT-11		1200	20,930	51,020	22.3	38.6
HT-12		1200	21,100	51,100	24.5	40.6
HT-13		1400	17,540	29,490	9.5	54.3
HT-14		1400	17,540	28,640	16.8	56.8
HT-15		1400	17,800	28,470	12.6	63.7
HT-16		1600	12,030	14,320	4.3	46.4
HT-17		1600	10,680	14,150	8.6	46.2
HT-18		1600	8,310	14,830	10.2	47.0
HT-19	1.0 ↓	R.T.	46,780	87,800	45.5	57.6
HT-20		R.T.	46,780	88,140	44.0	56.8
HT-21		R.T.	51,530	93,560	46.1	57.4
HT-22		800	---	---	--	36.8
HT-23		1000	23,050	65,420	29.5	33.1
HT-24		1000	20,680	66,780	29.9	32.3
HT-26		1000	25,760	67,120	30.6	33.3
HT-27		1200	23,730	60,510	30.0	39.7
HT-28		1200	22,370	57,800	30.4	33.3
HT-31		1400	21,020	41,360	20.0	48.4
HT-34		1400	26,440	68,470	44.2	47.1
HT-35		1400	20,000	41,690	21.6	46.2
FIT-36		1400	19,320	40,680	22.2	46.5
HT-37		1600	20,170	28,640	15.7	64.8
HT-38		1600	19,660	28,640	15.9	64.4
HT-39		1600	18,980	28,470	15.8	63.6
HT-42		1200	23,730	59,660	27.7	33.1
HT-40		800	28,810	71,530	33.4	36.7
HT-41		800	30,170	73,390	32.9	36.7

Neg 0693251-6

FIGURE 4.34. 0.2% Offset Yield Strength and Ultimate Tensile Strength Versus Test Temperature of Tube Lot G, Mill Annealed 316 Stainless Steel

Neg 0693251-4

FIGURE 4.35. Uniform and Total Elongation Versus Test Temperature of Tube Lot G, Mill Annealed 316 Stainless Steel

Neg 0693251-2

FIGURE 4.36. 0.2% Offset Yield Strength and Ultimate Tensile Strength Versus Test Temperature of Tube Lot H, Mill Annealed 316 Stainless Steel

Neg 0693251-3

FIGURE 4.37. Uniform and Total Elongation Versus Test Temperature of Tube Lot H, Mill Annealed 316 Stainless Steel

Tubing lots G and H do not differ significantly from the national average data with regard to ultimate tensile strength and 0.2% offset yield strength at the slow strain rate.⁽²⁶⁾ The total elongations for these two lots are slightly below the national average values (2 to 3%) at all test temperatures except room temperature.⁽²⁶⁾ At this temperature, BNW data were approximately 10% higher.

At all temperatures, the expected increase in 0.2% yield with increasing strain rate was observed. Below 1100 °F, the ultimate tensile strength was found to decrease with increasing strain rate, while above 1100 °F the ultimate strength increased with increasing strain rate. In all cases the uniform elongation showed similar strain-rate dependency as the ultimate strength.

The total elongation of tube lot E followed the same trends as ultimate strength while tube lots F, G and H exhibited decreasing ductility with increasing strain rate below 1500 °F and increasing ductility with strain rate above 1500 °F. Tensile tests at 10.0 min^{-1} on all four lots are now in progress.

The anomalous reduced rupture ductility previously reported⁽²⁷⁾ concerning the F-1 creep test has not been resolved. Photomicrographs do not reveal any significant grain size variation that might have caused this. A second possibility is that the area of reduced ductility was slightly cold worked, 1 to 5%, due to a final straight'ening pass during fabrication. This slight amount of cold work would reduce the ductility and be very difficult to discern metallographically.

Tensile Studies - A. L. Ward

The objective of this program is to provide mechanical property and metallurgical stability data on LMFB vessel and core structural components in direct support of the FFTF in order to define adequate lifetime performance under anticipated FTR service conditions.

Tensile property data is available from tests of AISI 304 SS specimens irradiated in EBR-II subassembly X-022. The testing schedule covers various test temperatures, irradiation temperatures, and fluences. Table 4.30 lists the data and the associated irradiation and test conditions. For comparison, as-solution-annealed values are included where available. As indicated in Table 4.30, some estimated and some experimentally-determined fluence values are included. The experimentally determined values are based upon the results of the dosimetry experiment conducted by the Irradiation Effects Section of Metallurgy and Ceramics Department. Irradiation temperatures listed are values which were calculated using the best available gamma heat and specimen position information.

The specimens irradiated at 870 °F to a total fluence of $\sim 8 \times 10^{21}$ n/cm² showed moderate strengthening, as evidenced by yield strength increases and associated ductility losses. The material irradiated at 770 °F to $\sim 6 \times 10^{21}$ n/cm² showed very large yield strength increases and ductility losses. It would appear that the effects of the lower temperature more than compensates for the slight decrease influence. However, the magnitude of the yield strength increases is quite large for a relatively small difference in irradiation temperature.

Tests on one set of specimens irradiated to $\sim 4 \times 10^{21}$ n/cm² at 725 °F in a single pin show no change in properties except for reduced elongation at higher test temperatures (1100 to 1200 °F). However, specimens from a different pin apparently irradiated to the same fluence at the same temperature show considerable yield strength increases. The present information on the effect of pin location on fluence and temperature does not appear to explain the discrepancy. This problem will be studied further.

In general, the results reported here are consistent with those given previously for AISI 316 SS irradiated under nearly identical conditions.

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In general, the results reported here are consistent with those given previously for AISI 316 SS irradiated under nearly identical conditions.

TABLE 4.30. *Tensile Properties of Selected AISI 304 Stainless Steel Specimens Irradiated in the EBR-11 Subassembly X-022*

Specimen Ident	Test Temp, °F	Irrad Temp, °F	Total Fluence, n/cm ²	P.E.L., psi	0.2% Yield, psi	Ultimate, psi	Uniform Elong, %	Total Elong, %	R.A.,
9G24-4 Sol. Ann.	500 500(a)	735 ---	$\sim 4 \times 10^{21}$ ---	26,800 ---	38,600 $\sim 18,000$	66,900 $\sim 62,000$	23.1 --	31.0 ~ 45	72.8 --
14G24-3 Sol. Ann.	700 700	725 ---	$4.6 \times 10^{21(b)}$ ---	12,000 8,900	20,200 21,300	63,200 65,000	32.4 36.3	37.7 41.5	73.6 67.2
9G14-5 Sol. Ann.	900 900	735 ---	$\sim 4 \times 10^{21}$ ---	20,500 8,100	31,900 19,000	60,900 62,000	23.1 35.9	28.6 41.7	64.2 69.1
14G14-3 Sol. Ann.	1000 1000	725 ---	$4.01 \times 10^{21(b)}$ ---	9,800 7,300	16,400 17,500	54,500 55,000	32.9 32.6	38.9 37.1	64.0 70.7
14G14-4 Sol. Ann.	1100 1100	725 ---	$4.01 \times 10^{21(b)}$ ---	12,200 7,300	14,500 15,000	44,600 46,000	20.1 27.7	22.4 33.3	74.8 42.3
14G14-5 Sol. Ann.	1200 1200	725 ---	$4.01 \times 10^{21(b)}$ ---	9,800 8,500	15,000 16,800	33,000 35,000	6.8 18.0	8.9 24.4	17.1 22.8
10E24-1 Sol. Ann.	500 500(a)	770 ---	$\sim 6 \times 10^{21}$ ---	44,400 ---	55,500 $\sim 18,000$	75,000 $\sim 62,000$	14.8 --	21.4 ~ 45	70.4 --
10E24-2 Sol. Ann.	700 700	770 ---	$\sim 6 \times 10^{21}$ ---	36,600 8,900	54,900 21,300	72,300 65,000	14.6 36.3	21.5 41.5	65.8 67.2
10E14-1 Sol. Ann.	800 800	770 ---	$\sim 6 \times 10^{21}$ ---	32,500 8,900	46,500 20,000	67,000 63,000	15.3 35.5	23.1 40.1	67.5 67.5
10E14-2 Sol. Ann.	900 900	770 ---	$\sim 6 \times 10^{21}$ ---	32,600 8,100	50,000 19,000	64,900 62,000	18.2 85.9	25.7 41.7	67.2 69.1
9E14-3 Sol. Ann.	1200 1200	760 ---	$\sim 6 \times 10^{21}$ ---	21,800 8,500	34,700 16,800	35,500 35,000	1.2 18.0	7.2 24.4	15.4 22.8
13B84-4 Sol. Ann.	900 900	870 ---	$\sim 8 \times 10^{21}$ ---	16,000 8,100	23,700 19,000	59,300 62,000	23.8 35.9	27.2 41.7	39.8 69.1
13B14-1 Sol. Ann.	1000 1000	870 ---	$\sim 8 \times 10^{21}$ ---	21,800 7,300	28,900 17,500	54,200 55,000	16.6 32.6	21.1 37.1	44.7 70.7

a. Extrapolated values from higher temperature tests.

b. Based upon dosimetry experiment.

Uniaxial Creep-Rupture Studies - A. J. Lovell, J. F. Bates

The objectives of this program are to evaluate the effects of fast neutron irradiation and environment on the pre- and postirradiation uniaxial creep-rupture properties of candidate vessel and core structural materials for LMFBR and to establish the usable materials limits to assure their safe and reliable performance.

Work is in progress to evaluate the effects of aging on rupture-life and creep rate of 304 and 316 SS. The data are presented in Tables 4.31 and 4.32 with comparisons to solution-treated control data where available.

The specimens were aged 3045 hr at temperatures ranging from 800 to 1400 °F. Figures 4.38 and 4.39 present the results of these tests graphically. As indicated by the dashed line, there appears to be a maximum stress above which the specimen will break on loading. This maximum stress appears to decrease as the aging temperature of the specimens increases. Metallography has shown that carbide formation is more predominant at the higher aging temperatures. This can be immediately seen from Figure 4.40, which contains micrographs of 316 SS aged at 800, 1100, and 1400 °F. Note the increased carbide formation of the 1400 °F age. This reduction in rupture life at low rupture times is more noticeable for the 1400 °F age condition than for the other ages investigated thus far. Another interesting result is that at stresses above 50,000 psi, 316 SS specimens aged at 900 °F exhibited longer rupture lives than the control specimens. From the existing data, it appears that at all test temperatures, for both 304 and 316 SS, the rupture life of the aged specimens may be the same as that for the control specimens at low stresses.

TABLE 4.31. Uniaxial Creep-Rupture Data on Aged 304 Stainless Steel

Spec No.	Aging Temp (°F) for 3045 hr	Tested at		Creep Rate, in./in./hr		Rupture Time, hr	Total Elongation, %
		Temp, °F	Stress, ksi				
9304	800	1000	50	0.002	(0.00174)	26.0 (25.1)	31.8 (27.7)
9313	800	1000	40	On Test		On Test	On Test
9306	800	1100	45	0.0515	(0.023)	2.25 (5.2)	30.58 (30.2)
9326	800	1100	37	0.0113		9.4	30.2
9314	800	1100	30	0.00065	(0.000506)	106.6 (200)	19.45 (19.3)
9280	900	1000	50	0.00133	(0.00174)	26.5 (25.1)	30.5 (27.7)
9261	900	1000	45	0.0000482	(0.00047)	56.7 (81.3)	20.96 (20.62)
9229	900	1000	45	0.000284	(0.00047)	116.0 (81.3)	17.33 (20.62)
9046	900	1000	40	0.00012	(0.00011)	342.0 (336.7)	17.24 (18.17)
9106	900	1100	45	0.075	(0.023)	1.65 (5.2)	35.9 (30.2)
9178	900	1100	35	0.0040	(0.0030)	31.3 (40.0)	25.3 (25.5)
9194	900	1100	28	0.00041	(0.00033)	192.8 (241.4)	16.0 (17.42)
9332	1000	1000	50	On Test		On Test	On Test
9331	1200	1000	50	On Test		On Test	On Test
9162	1400	1000	50	---	(0.00174)	~35 sec (25.1)	26.6 (27.7)
9128	1400	1000	45	0.0023	(0.00047)	48.95 (81.3)	36.9 (20.62)
9211	1400	1000	35	0.00021	(0.0000189)	1023.2 (1506.1)	37.2 (11.6)
9283	1400	1100	45	---	(0.023)	~30 sec (5.2)	29.6 (30.2)
9285	1400	1100	35	0.00924	(0.0030)	23.7 (40.0)	43.38 (25.5)
9269	1400	1100	28	0.00118	(0.00033)	220.7 (241.4)	52.53 (17.42)

() = Solution Treated Control.

TABLE 4.32. Uniaxial Creep-Rupture Data on Aged 316 Stainless Steel

Spec No.	Aging Temp (°F) for 3045 hr	Tested at		Creep Rate, in./in./hr		Rupture Time, hr	Total Elongation, %
		Temp, °F	Stress, ksi				
9315	800	1000	60	0.000134	(0.0002)	124.1 (106.0)	30.57 (35.0)
9305	800	1000	55	0.000071	(0.000091)	206.0 (263.1)	25.15 (25.4)
9317	800	1000	50	On Test	(0.000030)	On Test(432.9)	On Test (18.2)
9318	800	1200	32	0.0071		30.25	40.89
9319	800	1200	25	0.000685	(0.00063)	193.4 (374.1)	
9320	800	1200	20	0.000114		On Test	On Test
9336	900	1000	62	On Test		On Test	On Test
9278	900	1000	60	0.000131	(0.0002)	184.5 (106.0)	31.56 (35.0)
9260	900	1000	55	0.000027	(0.000091)	461.3 (263.1)	25.4 (25.4)
9228	900	1000	55	0.0000891	(0.000091)	414.0 (263.1)	21.86 (25.4)
9157	900	1000	50	0.000014	(0.000030)	450.8 (432.9)	22.1 (18.2)
9226	900	1000	45	0.000024	(0.0000117)	989.0 (1032.0)	15.7 (15.4)
9047	900	1200	30	0.0055	(0.0024)	53.9 (95.0)	66.0 (49.19)
9082	900	1200	25	0.0004	(0.00063)	419.5 (374.1)	51.1 (65.7)
9335	1000	1000	58	On Test		On Test	On Test
9334	1200	1000	55	On Test		On Test	On Test
9129	1400	1000	55	0.00087	(0.000091)	67.5 (263.1)	38.9 (25.4)
9282	1400	1000	50	0.000433	(0.000030)	377.7 (432.9)	42.22 (18.2)
9333	1400	1200	38	On Test		On Test	On Test
9242	1400	1200	35	0.012	(0.0096)	22.9 (24.9)	58.2 (50.0)
9123	1400	1200	28	0.0047	(0.0010)	63.9 (168.4)	64.4 (50.9)
9243	1400	1200	22	0.00071	(0.00017)	460.2 (1294.2)	71.2 (34.7)

() = *Solution Treated* (1950 °F/1 hr) *Control*.

Neg 0693280-2

FIGURE 4.38. Effect of Aging on the Creep-Rupture Properties of AISI Type 304 Stainless Steel

Neg 0693280-1

FIGURE 4.39. *Effect of Aging on the Creep-Rupture Properties of AISI Type 316 Stainless Steel*

Neg 469-973C
Aged 800 °F/3045 hr

Neg 469-978C
Aged 1100 °F/3045 hr

Neg 469-995C
Aged 1400 °F/3045 hr

FIGURE 4.40. *Microstructures of Solution-Treated 316 Stainless Steel, CANEL Heat, After Aging. (Original magnification 1000X)*

Specimens aged at 1400 °F also exhibit an increase in creep-rate when compared to control specimens. This is shown in Figures 4.41 and 4.42 and in Tables 4.31 and 4.32. The observed differences in creep rates are expected to continue at lower stresses.

Another objective of the creep-rupture program is to provide information on the behavior of irradiated AISI Type 304 SS at relatively low stresses (below 25,000 psi) for relatively long periods of time (~100,000hr). Should the "theta parameter" prove successful in this application, then fewer long-term tests will be required.

A plot correlating stress-rupture data using the theta parameter ($\theta = t_r \exp \frac{-\Delta H}{RT}$) is shown in Figure 4.43. Both the solution-treated control specimens and those irradiated at 900 °F to a fluence of $\sim 1 \times 10^{22}$ n/cm² fit a linear plot for the stresses and temperatures investigated. From previous data,⁽²⁸⁾ it appears the curves will bend down at lower stresses.

Data are tabulated in Table 4.33 for creep-rupture tests on welded specimens. There are four weld processes being investigated: Sub-Arc, Stick-Electrode, TIG, and MIG. From results obtained thus far, it appears that the Stick-Electrode-type weld will produce specimens having longer rupture lives than the other three processes (see Figure 4.44). However, there is some question about where the longer-term, lower-stress tests will fall due to the scatter of the higher-stress Stick-Electrode data. The minimum creep-rate data for the welds fall in a scatter band around the solution-treated control data (see Figure 4.45). There is some indication that at lower stresses the weld specimens will have lower creep rates than the solution-treated control.

Neg 0693280-7

FIGURE 4.41. Effect of Aging on the Creep Rate of AISI Type 304 Stainless Steel

Neg 0693280-6

FIGURE 4.42. Effect of Aging on the Creep Rate of AISI Type 316 Stainless Steel

Neg 0693280-5

FIGURE 4.43. "Theta Parameter" Plot of Creep-Rupture Test of Type 304 Stainless Steel

TABLE 4.33. *Uniaxial Creep-Rupture Data on Stainless Steel Welds at 1100 °F*

Spec No.	Weld Type	Material, 304 SS Base in all cases	Stress, ksi	Rupture Time, hr	Creep Rate, in./in./hr	Total Elong, %
9290	Sub-Arc	308 SS Filler	45	0.33	0.0236	21.69
9295			40	1.00	0.0038	28.44
9316			30	48.8	0.0021	15.2
9296			25	285.0	0.00021	10.57
9307			23	589.0	0.000051	4.8
9330	Stick- Electrode	16-8-2 Weld Metal	45	7.2	0.0127	
9327			40	49.9-45.3	0.000978	
9353			35	38.5	0.00085	
9404			30	On Test		
9364			25	On Test		
9383	TIG Weld	308 SS Weld Metal	45	Broke on Loading		
9329			40	4.4	0.0122	
			35	On Test		
9359			30	~205.0	0.000162	
	MIG Weld	308L SS Weld Metal	45	Broke on Loading		
9328			40	3.2	0.0382	
9361			30	38.1	0.00178	
9362			25	~270.0	0.000158	
9384			18	On Test

Neg 0693830-1

FIGURE 4.44. Stress Versus Time to Rupture for Specified Weldments at 1100 °F

Neg 0693830-2

FIGURE 4.45. *Creep Rate Versus Stress for Specified Weldments at 1100 °F*

Transient strain has been determined for 316 SS. The purpose of this phase of the program was to provide a means whereby transient strain of irradiated ($900\text{ }^{\circ}\text{F} - \sim 1 \times 10^{22}\text{ n/cm}^2$) AISI Type 316 stainless steel could be obtained as a function of initial stress. Transient strain is defined here as being the value of strain obtained when the second stage of the creep curve is extrapolated to zero time. This value is broken down into two components and tabulated in Table 4.34. The two components are ϵ_0 , which includes the elastic and time-independent plastic strain (due to the inaccuracy involved in taking this value from the results of the creep test, this value was taken from tensile tests for both the irradiated and control specimens), and ϵ_p , the additional plastic strain obtained from the creep curve. These symbols are explained graphically in Figure 4.46. Data was obtained for test temperatures ranging from 1000 to 1400 $^{\circ}\text{F}$ and is shown graphically in Figure 4.47. These show that the irradiated specimens had a substantial reduction in transient strain at the higher stresses.

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TABLE 4.34. *Transient Strain Data of AISI Type 316 Stainless Steel Irradiated at 900 °F to $\sim 1 \times 10^{22}$ n/cm²*

Spec No.	Tested at:		ϵ_{o-irr}	ϵ_{o-stc}	ϵ_{p-irr}	ϵ_{p-stc}	ϵ_{t-irr}	ϵ_{t-stc}
	Temp, °F	Stress, ksi						
14B36-3	1000	55	0.095	0.230	0.0002	0.00115	0.095	0.231
14B36-4	1000	50	0.060	0.173	0.0012	0.0000	0.061	0.173
14B66-1	1000	45	0.040	0.130	0.0067	0.0000	0.047	0.130
14B36-5	1000	40	0.025	0.096	0.0034	0.00133	0.028	0.097
14B46-3	1000	30	0.015	0.073	0.0022	0.0000	0.017	0.073
14B56-3	1100	45	0.037	0.141	0.0028	0.0138	0.040	0.155
14B46-1	1100	40	0.016	0.107	0.0000	0.0115	0.016	0.119
14B56-1	1100	36	0.007	0.085	0.0085	0.0124	0.016	0.097
14B56-5	1100	30	0.002	0.055	0.0051	0.0222	0.007	0.077
14B36-2	1200	30	0.002	0.048	0.00667	0.0089	0.009	0.057
14B56-2	1200	28	0.000	0.036	0.0031	0.0115	0.003	0.048
14B46-2	1200	25	0.000	0.022				
14B66-2	1200	22	0.000	0.009	0.0058	0.0058	0.006	0.015
14B46-4	1400	17	0.000	0.005	0.0027	0.0000	0.003	0.005
14B66-3	1400	15	0.000	0.000	0.0013	0.0045	0.001	0.005
14B66-4	1400	12	0.000	0.000	0.0067	0.0067	0.007	0.007
14B66-5	1400	10	0.000	0.000	0.0024	0.00115	0.002	0.003

stc = solution treated (1950 °F/1 hr) control

Neg 0693280-4

FIGURE 4.46. *Creep Curve Showing Initial, Plastic, and Transient Strains (ϵ_o , ϵ_p , ϵ_t , respectively where ϵ_o includes elastic and time-independent plastic strain, and ϵ_p is the time-dependent plastic strain)*

Neg 0693280-3

FIGURE 4.47. Effect of Irradiation on Transient Strain in AISI Type 316 Stainless Steel

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FAST REACTOR DOSIMETRY AND DAMAGE ANALYSIS

R. E. Nightingale

EBR-II DOSIMETRY TEST

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Results of the analysis of the EBR-II dosimetry test^(1,2) have been summarized in two technical papers.^(3,4) Results not previously reported^(5,6,7) or updated are discussed here.

Reaction Rates

The recent EBR-II dosimetry test required the measurement of approximately 1200 reaction rates from 830 activation monitor foils.^(1,2) Reaction rate data for Row 2 - midplane used in references 2 and 3 are the results of a cooperative effort between eight different laboratories.

Correlation of the data provided by the eight laboratories who analyzed replicate foils located in the three core center positions provides an accuracy estimate of the data. Table 5.1 indicates the estimated accuracies achieved for the foil reactions used for SAND-II reduction.⁽⁸⁾ These measurements along with associated low to high power ratios,⁽⁷⁾ axial and radial plots,⁽⁹⁾ and data from previous spectral studies⁽¹⁰⁾ were then correlated for use with the remaining data since a majority of the foils located in 25 reactor positions were analyzed by no more than one laboratory.

A number of anomalous results, although only a small percentage of the total, were initially found through the use of axial plots. Most were found to be errors in calculation. Two laboratories were found to be using erroneous branching ratios in two separate instances. Results from two laboratories appeared to be biased, and one laboratory subsequently found a 5% bias in the initial calibration of their counting system. These problems confirm the need for a continuing "Round Robin" laboratory calibration program^(11,12,13)

TABLE 5.1. *Estimated Accuracies Achieved for the Foil Reactions Used for SAND-II Reduction*

Foil Reaction	Participants	Foils Reported	Participants Used (a)	Foils Used (b)	Reaction Rate (c)	Standard Deviation (%)	Standard Error of Mean. (%)
$^{235}\text{U}(\text{n},\text{f})^{140}\text{Ba}, ^{137}\text{Cs}$	3	9	3	5	2.90×10^{-9}	5.7	2.2 (d)
$^{238}\text{U}(\text{n},\text{f})^{140}\text{Ba}, ^{137}\text{Cs}$	3	3	3	3	1.81×10^{-10}	4.9	2.8
$^{237}\text{Np}(\text{n},\text{f})^{140}\text{Ba}$	3	4	3	3	1.26×10^{-9}	1.6	1.2
$^{232}\text{Th}(\text{n},\text{f})^{140}\text{Ba}$	2	2	2	2	4.74×10^{-11}	10.4	7.4
$^{238}\text{U}(\text{n},\gamma)\text{Np}^{239}$	1	1	1	1	3.08×10^{-10}	<7	<5.0 (e)
$^{54}\text{Fe}(\text{n},\text{p})^{54}\text{Mn}$	7	12	6	6	3.75×10^{-11}	2.6	1.1
$^{58}\text{Fe}(\text{n},\gamma)^{59}\text{Fe}$	5	7	5	5	8.76×10^{-12}	5.4	2.4
$^{58}\text{Ni}(\text{n},\text{p})^{58}\text{Co}$	7	13	6	6	3.11×10^{-11}	2.5	1.0
$^{46}\text{Ti}(\text{n},\text{p})^{46}\text{Sc}$	5	7	4	4	4.80×10^{-12}	1.0	0.5
$^{47}\text{Ti}(\text{n},\text{p})^{47}\text{Sc}$	3	5	3	3	3.82×10^{-12}	12.1	7.0
$^{48}\text{Ti}(\text{n},\text{p})^{48}\text{Sc}$	3	5	3	3	1.21×10^{-13}	3.8	2.2
$^{45}\text{Sc}(\text{n},\gamma)^{46}\text{Sc}$	1	1	1	1	3.08×10^{-11}	<11	<7 (e)
$^{63}\text{Cu}(\text{n},\gamma)^{64}\text{Cu}$	2	3	2	2	4.35×10^{-11}	12.5	8.9
$^{27}\text{Al}(\text{n},\alpha)^{24}\text{Na}$	1	1	1	1	2.75×10^{-13}	<8	<5 (e)
$^{23}\text{Na}(\text{n},\gamma)^{24}\text{Na}$	1	1	1	1	1.08×10^{-12}	<11	<8 (e)
$^{59}\text{Co}(\text{n},\gamma)^{60}\text{Co}$	2	2	2	2	3.14×10^{-11}	5.5	3.9
$^{197}\text{Au}(\text{n},\gamma)^{198}\text{Au}$	2	2	2	2	4.68×10^{-10}	8.9	6.3

(a) Analysis of variance using F statistics and Scheffe's contrast test used to determine reliability and bias whenever numbers of participants and reported foils were sufficient. (14)

(b) Total number of single and/or mean values (where more than one foil was reported for a single foil reaction by the participant) used for calculation of reaction rate, standard deviation, and standard error of the mean. (15)

(c) Fissions/sec/nucleus or dps/nucleus (saturated activity) of target element.

(d) Error values for the fission foils do not include fission yield error. Since both ^{140}Ba and ^{137}Cs were utilized for the ^{235}U and ^{238}U foils, the overall foil standard deviation and standard error for these reactions would suggest the fission yield error contribution is small.

(e) Values estimated by participant.

The participants used various types of calibrated gamma counting systems such as ionization chambers, NaI(Tl), and the Ge(Li) detectors in conjunction with multichannel pulse height analyzers. The high activities encountered, as great as 10^{12} dpm/foil of the desired isotope, required most participants to dissolve the foils and count suitable aliquots. One laboratory was able to count all foils directly.

The monitor reaction rates from 20 positions were used to generate a two dimensional (r,z) plot. Plots for the $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ and $^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$ reactions, shown in Figure 5.1, illustrate features of all the plots. The $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ illustrates the distribution from reactions responding to neutrons of energy >1 MeV, whereas the $^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$ illustrates the distributions from a reaction responding to neutrons of both low and high energies. The reaction rate isopleths are useful to determine the proper sample size for individual flux monitors for a specified irradiation and to estimate relative activation levels at different locations, especially outside the core.

Multiple-Foil Derived Fluxes

In the 31F dosimetry test, the accuracy of calculations and the use of flux monitors for determining flux levels and fluences has been investigated by the multiple foil activation method. ^(8,16,17) Combinations of seventeen foil reaction rates, ^(4,9) and different input spectra, listed in the first column of Table 5.2, were used to generate flux-spectral solutions for a Row 2 - midplane location in a structural subassembly.*

* *The results presented here are based on more current reaction rate and foil reaction evaluated energy-dependent cross-section data than that used in references 2, 6, and 7.*

Neg 0693172-2

FIGURE 5.1. Isopleths of Saturated Activity Levels for the $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ and $^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$ Reactions on an R, Z Grid of the EBR-II Reactor

TABLE 5.2. Integral Flux Comparison - EBR-II Row 2 Midplane^(a)

Input Spectral Approximation Based On	Solution ^(b)		SAND-II Sol'n Integral Flux Values $\Phi(E)$ (n/cm ² -sec) Above Specified Energies (MEV) $\times 10^{-15}$					
	K	SPD	10 ⁻³ ^(c)					
			0.1	0.5	1.0	2.0	3.0	
Monte Carlo Code ^(d) (MC) (17 Foils)	26	3.1	2.01	1.78	0.933	0.535	0.232	0.105
Diffusion Code ^(e) (DC) (17 Foils)	26	3.1	2.07	1.81	0.916	0.531	0.236	0.104
Transport Code ^(f) (TC) (17 Foils)	28	3.0	2.06	1.82	0.915	0.538	0.236	0.103
Transport Code ^(f) (4 Fission Foils)	12	2.9	2.11	1.85	0.909	0.528	0.237	0.119 ^(d)
Transport Code ^(f) (13 Non-Fission Foils)	50	2.0	2.07	1.83	0.955	0.566	0.238	0.103
Average Values (AV)			2.06	1.82	0.926	0.540	0.236	0.104
Standard Deviation %			± 1.7	± 1.4	± 2.0	± 2.8	± 0.97	± 0.92
Constant Input ^(e) (CI) (17 Foils)	33	2.6	2.01	1.78	0.943	0.524	0.229	0.101
Absolute Deviation ^(f) Between (AV) and (CI) %			2.4	2.2	2.8	2.8	3.0	2.9
$\frac{\text{Measured (AV)}}{\text{Predicted}}$			0.87	0.87	0.85	0.89	0.86	0.76

(a) Determined for a stated power of 50 MW, Reference 21.

(b) The convergence criterion used was that the change between solution standard percent deviation (SPD) between successive iterations had stabilized to less than 1%, see Reference 16. The SPD is defined as:

$$SPD^{(K)} = \left[\frac{1}{n-1} \sum_{i=1}^n \left(\frac{A_i^{(K)} - A_i^m}{A_i^{(K)}} \right)^2 \right]^{1/2}$$

where "n" is the number of foil reactions, A_i^m is the measured reaction rate for the i^{th} foil, and $A_i^{(K)}$ is a calculated value using the solution differential flux at the K^{th} cycle of iteration and the energy-dependent cross section for the i^{th} foil. In general, iteration beyond a stability limit of 1% produces little change in the integral flux.

(c) The integral flux below 10⁻³ MeV is negligible compared to that above this energy.

(d) This value was not used in obtaining the average value because of the lack of high energy foil coverage.

(e) Constant above 10⁻² MeV, with a form proportional to energy below this energy, since there are few neutrons below 10⁻² MeV.

(f) Defined as 100[$|\Phi(E) - \bar{\Phi}(E)| / \bar{\Phi}(E)$], where $\bar{\Phi}(E)$ is the average value (AV).

In Table 5.2, differences in integral flux solutions (indicated by the standard deviations, Row 7) based on the most reasonable input spectra [from diffusion,⁽¹⁸⁾ transport,⁽¹⁹⁾ and Monte Carlo⁽²⁰⁾ code calculations] define approximate limits for the shape of the true solution. The solution based on a constant spectrum input gives a further estimate of the accuracy of this shape (indicated by the absolute deviations, Row 9). These results indicate that the iterative integral flux solution is dependent on the accuracy of the measured activities and energy dependent cross sections, but is largely independent of the input spectral form.

The present and previous studies, references 16 and 17, suggest an absolute uncertainty estimate of $\sim \pm 10\%$ (1 standard deviation) for the measured average integral flux values (AV), Table 5.2. These values are compared in the last row of Table 5.2 with theoretical predictions based on diffusion code results, reference 18, with an energy release value of 200 MeV/fission as suggested by Kirn.⁽²¹⁾ Since the reactor physics calculation does not account for local perturbation effects (which might be as high as $\sim 10\%$ to $\sim 20\%$ since the structural assembly used in the dosimetry test contains no fuel), the agreement of the measured and predicted values to within $\sim 15\%$ helps to validate the stated accuracy for the multiple foil technique for this dosimetry test.

Preliminary results from similar studies for different axial locations in Rows 2, 4, 6, and 7 indicate agreement between multiple foil and predicted total integral flux values to within $\sim 15\%$ in the central core region. At distances up to ~ 70 cm above reactor midplane, some multiple foil results are 2 to 4 times greater than reactor physics calculations. This is not surprising since these regions are beyond the accepted range of applicability of the calculations.

It is concluded that localized perturbation effects in core, and large uncertainties associated with calculations for edge- and out-of-core locations, make necessary the use of single and/or multiple foil flux monitors in experimental sub-assemblies for accurate flux level and fluence estimates.

Spectral Averaged Cross Sections

Results from a number of fast reactor tests⁽¹⁰⁾ indicate that the previously reported^(22,23) EBR-II core spectral-averaged cross sections, $\bar{\sigma}_c$, for the $^{54}\text{Fe}(n,p)$ and $^{58}\text{Ni}(n,p)$ reactions should be reduced by about 25 and 30%, respectively, to obtain consistency with fission reactions.

The expression used to obtain measured values of spectral averaged cross section, $\bar{\sigma}_m$, is

$$\bar{\sigma}_m = \frac{A_m}{\Phi} \quad (1)$$

where A_m and Φ are the measured reaction rate and total value of integral flux, respectively. For the EBR-II run 31F, Row 2 - midplane, dosimetry test, values of A_m with assigned error limits are given in Table 5.1. A value of $\Phi = 2.06 \times 10^{15}$ n/cm²-sec, Table 5.2, was used to obtain the measured values of $\bar{\sigma}_m$, Table 5.3.* The absolute uncertainties associated with these $\bar{\sigma}_m$'s are estimated to be $\pm 10\%$ to 15% (1 standard deviation); i.e., the combined estimated absolute uncertainties in the values of A_m and Φ . Relative uncertainties of ratios of one $\bar{\sigma}_m$ to another are much smaller.

The expression used to obtain calculated values of spectral averaged cross section is

$$\bar{\sigma}_c = \frac{\int_0^\infty \phi(E) \sigma(E) dE}{\int_0^\infty \phi(E) dE} \quad (2)$$

* *Phi is not a directly measured quantity; however, multiple foil measurements yield a reasonable estimate of Φ .*

TABLE 5.3 Measured ($\bar{\sigma}_m$) and Calculated ($\bar{\sigma}_c$) Spectral Averaged Cross Sections for EBR-II Row 2 - Midplane

Reaction	$\bar{\sigma}_m$ (mb) (a)	$\bar{\sigma}_c$ (mb) (b)			
		Iterative (c) Solution	Diffusion (d) Code	Monte (e) Carlo	Reference (22)
$^{235}\text{U}(n, f)$	1408	1367	1357	1383	--
$^{238}\text{U}(n, f)$	87.9	91.5	90.7	86.1	98.2
$^{237}\text{Np}(n, f)$	612	621	628	615	--
$^{58}\text{Fe}(n, \gamma)$	4.25	4.30	4.28	4.42	--
$^{54}\text{Fe}(n, p)$	18.2	18.0	20.8	18.5	24.1
$^{58}\text{Ni}(n, p)$ (f)	24.8	24.9	28.4	25.6	35.4
$^{46}\text{Ti}(n, p)$	2.33	2.35	2.97	2.62	3.8
$^{47}\text{Ti}(n, p)$	4.28	4.25	4.92	4.48	--
$^{48}\text{Ti}(n, p)$	0.0587	0.0557	0.0641	0.0660	--
$^{59}\text{Co}(n, \gamma)$	15.2	14.7	14.3	16.0	--
$^{45}\text{Sc}(n, \gamma)$	15.0	15.5	15.3	16.7	12.2
$^{63}\text{Cu}(n, \gamma)$	21.1	20.9	21.8	27.1	25.5
$^{197}\text{Au}(n, \gamma)$	227	223	229	251	--
$^{23}\text{Na}(n, \gamma)$	0.524	0.523	0.508	0.553	--
$^{232}\text{Th}(n, f)$	23.0	22.0	20.3	20.9	--
$^{238}\text{U}(n, \gamma)$	150	146	147	156	--

(a) Calculated from equation 1.

(b) Calculated from equation 2, using the indicated form of $\phi(E)$ and the current SAND-II evaluated energy-dependent cross-section library tape.

(c) Solution based on the transport input, Table 5.2.

(d) Reference (18)

(e) Reference (20); below 10^{-2} MeV, the transport shape has been used to extend the Monte Carlo results for these comparisons. This may be a cause for the higher values for the (n, γ) reactions.

(f) A calculated value of 28 mb has recently been reported by Kampouris. (24)

where $\sigma(E)$ is the energy-dependent cross section and $\phi(E)$ is the neutron differential flux. The SAND-II evaluated cross sections, $\sigma(E)$, for the reactions listed in Table 5.3 have been adjusted by SAND-II analysis and/or unfolding⁽²⁵⁾ based on reaction rate measurements in a number of fast reactor environments. (2)

Adjusted energy dependent cross sections and $\phi(E)$'s based on an iterative solution, a diffusion code calculation, and a Monte Carlo code calculation were used with equation (2) to obtain values of $\bar{\sigma}_C$, Table 5.3. Values for $\bar{\sigma}_C$ using previously reported, but unadjusted $\sigma(E)$'s⁽⁸⁾ are given in the last column for a $\phi(E)$ based on a transport code calculation.⁽²²⁾

Comparison of $\bar{\sigma}_C$ values for Fe and Ni in the last column with values in the other columns provides the basis for the suggested 25 to 30% reductions.

It is concluded that the use of measured values, $\bar{a}_m$, is preferred because of the smaller uncertainty associated with the measured values of total integral flux and interaction rates. In the absence of measured values, however, energy-dependent $\sigma(E)$'s are available, that can be used to obtain reasonably reliable calculated values, $\bar{\sigma}_C$, if the spectral form, $\phi(E)$, is well defined. Such measured and calculated values of spectral averaged cross section from the ³¹F dosimetry test are now being used to interpret individual flux monitor wires such as ⁵⁴Fe(n,p) used in structural and fuel subassemblies irradiated in EBR-II.^(26,27)

Single-Foil Derived Fluxes

Tables 5.2 and 5.3 show the results of multiple-foil derived flux levels and spectral averaged cross sections, respectively. Single-foil derived total flux levels and spectral averaged cross sections have been reported by Jackson and

and Ulseth⁽²²⁾ and Dudey.⁽²³⁾ These previously reported values are given in Table 5.4 together with the most recent multiple-foil results.

Flux/MW data previously reported (references 22 and 23) have been updated on the basis of the measured values of $\bar{\sigma}_m$, Table 5.3. Comparison* of the measured flux/MW values in column 6 and the ratio of measured high to low power values, column 9, shows that for Row 2 - midplane the difference of $\sim 59\%$ previously found between high and low power flux/MW values, reference (22), has been reduced to $\sim 7\%$.

FAST-REACTOR NEUTRON SPECTRA AND FOIL-ACTIVATION CROSS SECTION EVALUATION

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Previously reported^(17,28,29) inconsistencies between activation detector and n-p scattering/time-of-flight (T-O-F) measurements of the thermal-neutron-induced ^{235}U fission spectrum prompted a comparison of such measurements in the core center and on the surface of a bare ^{235}U assembly, referred to as Godiva by Grundl and Hansen.⁽³⁰⁾ For the present study, Neill et al.,⁽³¹⁾ made T-O-F measurements, and we made multiple foil measurements of the core and surface spectra of the APFA-III - Godiva.^{††}

Measured reaction rates with estimated absolute uncertainties of 3 to 10% (1 standard deviation) were obtained for ~ 20 reactions for the core center and surface locations. These new results are in good agreement with the high precision relative measurements of Grundl and Hansen.⁽³⁰⁾ [The estimated accuracy of the new results is supported by recent measurements in the Experimental Breeder Reactor - II (EBR-II).⁽⁴⁾]

* *Direct comparison is complicated by the uncertainties associated with local perturbation effects, which may be as high as 10% to 20%.*

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TABLE 5.4 Comparison of EBR-II Low and High Power Flux Measurements

Reactor Run	Reaction and Test	Measured Flux/MW ^(a)	Reported Spectral Averaged Cross Section, mb	Test Cross Section, mb ^(b)	Measured Flux/MW ^(c)	Calculated Flux/MW ⁽¹⁸⁾	Ratio Col. 6//	Ratio Low Power to High Power
25A	Low Power (20 kW) ⁽²²⁾							
	⁵⁴ Fe(n, p)	5.00 x 10 ¹³	24.1 ⁽²²⁾	18.2	6.62 x 10 ¹³			
	⁵⁸ Ni(n, p)	4.80 x 10 ¹³	35.4 ⁽²²⁾	24.8	6.85 x 10 ¹³			
	Avg	4.90 x 10 ¹³			6.74 x 10 ¹³	4.86 x 10 ¹³	1.39	
25-24	High Power (45 MW) ⁽²²⁾							
	⁵⁴ Fe(n, p)	3.29 x 10 ¹³	24.1 ⁽²²⁾	18.2	4.36 x 10 ¹³			1.59
	⁵⁸ Ni(n, p)	2.89 x 10 ¹³	35.4 ⁽²²⁾	24.8	4.13 x 10 ¹³			
	Avg	3.09 x 10 ¹³			4.24 x 10 ¹³	4.86 x 10 ¹³	0.872	
31E	Low Power (84 kW) - This Test							
	14 Foils ^(d) SAND-II			Multiple Foils	4.42 x 10 ¹³	4.86 x 10 ¹³	0.909	
31F	High Power (50 MW) - This Test							
	14 Foils ^(d) SAND-II			Multiple Foils	4.14 x 10 ¹³	4.86 x 10 ¹³	0.852	1.07
	High Power - Dudey ⁽²³⁾							
	⁵⁴ Fe(n, p)		26.0 ⁽²³⁾	18.2				
	⁵⁸ Ni(n, p)		36.0 ⁽²³⁾	24.8				
	Avg	4.9 x 10 ¹³			6.74 x 10 ¹³			

(a.) Based on the cross sections in column 4.

(b.) Table 5.3

(c.) Based on the cross sections in column 5.

(d.) The reactions used for flux estimates in this table are ²³⁵U(n, f), ²³⁸U(n, f), ²³⁷Np(n, f), ²³²Th(n, f), ⁵⁴Fe(n, p), ⁵⁸Ni(n, p), ⁴⁶Ti(n, p), ²⁷Al(n, α), ²³⁸U(n, γ), ⁵⁸Fe(n, γ), ⁴⁵Sc(n, γ), ⁶³Cu(n, γ), ¹⁹⁷Au(n, γ) and ²³Na(n, γ). Other reactions were studied at high power, but for high/low power comparisons only the reactions common to both tests were selected for this analysis.

Using the SAND-II code,^(8,16) the reaction rates were used together with different input spectral approximations to obtain iterative integral flux solutions for comparison with the time-of-flight results. In Table 5.5, the small deviation of individual solution results from the average for all results (indicated by the percent of standard deviation, 4th from last row) based on different input forms indicates that a single average solution can be defined that depends on the accuracy of the measured reaction rates and reaction energy-dependent cross sections, but little on the input spectral form.* Comparison of the integral fluxes above specified energies for the average solution with Neill's 0° angular T-O-F results,** in the last row of Table 5.5, shows agreement to within ~5%. (There are no T-O-F data for comparison above 11 MeV.)

Similar results have been obtained at 1/2 in. above the core surface, where fission reaction rate measurements obtained by track^(33,34) and radio-chemistry⁽³⁵⁾ counting techniques, provide further validation of the estimated accuracy of the measured reaction rates. These results show disagreement between the multiple foil, Neill's T-O-F, and previously reported photo-plate data,⁽³⁷⁾ which is similar to that found for measurements in the fission spectrum.^(17,28,29)

Measured values of spectral averaged cross section, $\bar{\sigma}_m$, were obtained by use of the expression

$$\bar{\sigma}_m = \frac{A_m}{\Phi}, \quad (1)$$

where A_m is the measured reaction rate and Φ = the average absolute value of total flux, Table 5.5. Calculated values, $\bar{\sigma}_c$, were obtained by use of the expression

* Such uniqueness is consistent with that found for measurements in the fission spectrum,⁽¹⁷⁾ and more recently for measurements in EBR-II.^(2,32)

** The 0° angular and scalar fluxes are equal at core center.

TABLE 5.5 Sand-II Absolute Integral Flux Solution Comparison APFA-III Core Center

Run No.	Input Spectral Form ^(b)	Foil Reactions Used	Solution ^(c) K	SPD	Average ^(d) Energy, MeV		Integral Flux Values Above Specified Neutron Energies ^(e) (MeV) x 10 ⁻¹⁰ (n/cm ² -sec)							
					>0.01	>0.4	0.01	0.1	0.4	0.9	1.4	3.0	6.0	11.0
1	1	20 ^(f)	41	6.1	1.96 ^(a)	1.99	2.792 ^(a)	2.771 ^(a)	2.751	2.228	1.471	0.5012	0.06531	0.001963
2	2	20	33	5.1	1.54	1.83	3.632	3.576	2.955	1.969	1.406	0.5145	0.06567	0.001947
3	3	20	22	4.1	1.67	1.94	3.248	3.137	2.735	1.969	1.432	0.5099	0.06416	0.001938
4	4	20	12	5.1	1.62	1.93	3.372	3.237	2.755	1.988	1.446	0.5082	0.06319	0.001939
5	4	17 ^(g)	19	5.1	1.61	1.92	3.407	3.277	2.777	1.995	1.449	0.5062	0.06406	0.001939
6	5	20	14	5.1	1.57	1.86	3.526	3.438	2.893	2.005	1.420	0.5164	0.06378	0.002070
7	6	20	9	5.1	1.55	1.87	3.570	3.499	2.856	1.942	1.416	0.5211	0.06261	0.002051
8	7	20	6	5.1	1.56	1.90	3.501	3.402	2.802	1.938	1.409	0.5209	0.06280	0.002040
9	7	14 ^(h)	22	4.1	1.55	1.98	3.362	3.246	2.550	1.800	1.356	0.5161	0.06437	0.002070
Average Value ^(a)					1.58	1.91	3.452	3.352	2.786	1.982	1.423	0.5127	0.06399	0.001995
Percent Standard Deviation					±2.9	±2.8	±3.7	±4.5	±4.1	±5.6	±2.3	±1.3	±1.6	±3.0
Normalized Average Value					1.58	1.91	1.000	0.9710	0.8071	0.5742	0.4122	0.1485	0.01854	0.0005779
Normalized T-O-F Value (Ref. 5)					1.56	1.88	0.9995	0.9713	0.8033	0.5515	0.3964	0.1457	0.01957	-
Ratio (NAV)/(T-O-F)					-	-	1.000	0.997	1.005	1.041	1.040	1.019	0.9474	-

TABLE 5.5. (Contd)

- (a) Excluded from average because of excessive deviation from the mean; the extreme form of the input spectral form caused this excessive deviation.
- (b) Number 1 is based on a $1/E$ form between 10^{-10} and 18 MeV. Number 2 is based on an E form between 10^{-10} and 18 MeV. Number 3 is based on a constant form between 10^{-10} and 18 MeV. Number 4 is based on a constant form above 10^{-2} MeV, and an E form below. Number 5 is a Watt fission form.⁽³⁶⁾ Number 6 is the Godiva Zeakage spectrum above 10^{-5} , zero below.⁽³⁷⁾ Number 7 is Neill's time-of-flight (T-O-F) data (0° angular flux), Reference 31.

- (c) The convergence criterion used was that the change between the solution standard percent deviation (SPD) between successive iterations had stabilized to less than 1%, see Reference 16. The SPD is defined as:

$$SPD^{(K)} = \left[\frac{1}{n-1} \sum_{i=1}^n \left(\frac{A_i^{(K)} - A_i^m}{A_i^{(K)}} \right)^2 \right]^{1/2}$$

where "n" is the number of foil reactions, A_i^m is the measured reaction rate for the i^{th} foil, and $A_i^{(K)}$ is a calculated value using the solution differential flux at the K^{th} cycle of iteration and the energy-dependent cross-section for the i^{th} foil. In general, iteration beyond a stability limit of 1% produces little change in integral flux.

- (d) Defined as:

$$\bar{E}(>E) = \int_E^\infty E \phi(E) dE / \int_E^\infty \phi(E) dE$$

- (e) The energy values have been selected to correspond to those used by Grundl, References 28 and 30, and the magnitudes of the fluxes are for a stated APFA-III reactor power of 130 watts.⁽³⁸⁾ The fraction of the flux below 10^{-2} MeV is estimated to be $<0.05\%$.

- (f) Fission and nonfission foils.

- (g) Nonfission foils only were used.

- (h) Run with the more reliable foil reaction cross-sections: i.e., $^{235}\text{U}(n,f)$, $^{238}\text{U}(n,f)$, $^{238}\text{U}(n,\gamma)$, $^{197}\text{Au}(n,\gamma)$, $^{23}\text{Na}(n,\gamma)$, $^{58}\text{Ni}(n,p)$, $^{54}\text{Fe}(n,p)$, $^{56}\text{Fe}(n,p)$, $^{27}\text{Al}(n,p)$, $^{32}\text{S}(n,p)$, $^{27}\text{Al}(n,\alpha)$, $^{24}\text{Mg}(n,p)$, $^{127}\text{I}(n,2n)$, $^{115}\text{In}(n,n')$.

$$\bar{\sigma}_c = \int_0^{\infty} \sigma(E) \phi(E) dE / \int_0^{\infty} \phi(E) dE \quad (2)$$

where $\sigma(E)$ is the energy-dependent cross-section* and $\phi(E)$ is the T-O-F or photo-plate measured spectrum. In core center, good agreement is obtained between these measured and calculated values in contrast to findings for the core surface and the fission spectrum. (17,28,29)

In conclusion, the agreement in core center between measured and calculated values of $\bar{\sigma}_m$ and $\bar{\sigma}_c$ establishes increased confidence in current microscopic cross-section data and multiple foil results. On the other hand, the consistency found in core center between T-O-F and multiple foil derived flux-spectra and spectral averaged cross sections, and the corresponding lack of such consistency for core surface and fission spectrum measurements, (17,28,29) suggests that previous photo-plate measurements appear to have underestimated the mean energy of the Godiva leakage and fission spectrum.

DAMAGE FUNCTIONS AND DATA CORRELATION

W. N. McElroy, R. L. Simons, and L. D. Blackburn

Previous reports (7,40, 41) discussed the development and study of a method for generating energy-dependent damage functions for the correlation and prediction of neutron-induced property changes. The method is being applied to additional sets of mechanical property data.

Preliminary results are given in Figure 5.2 for a damage function, $G(E)$, for 10% residual total elongation (TE) for 304 SS irradiated and tested at ~ 450 °C. The solid curve represents, perhaps, the most reasonable of a number of possible solutions for the empirically derived damage function.

(39)
* *Current SAND-II evaluated cross-section library.*

Neg 0694012-5

FIGURE 5.2. $G(E)$ Solution for a Residual 10% Total Elongation for 304 SS

The dash-dot curve represents an unmodified Kinchin and Pease input model plus contributions from (n,γ) and (n,α) reactions. The form of the input model below $\sim 10^{-2}$ MeV is represented by the total (n,γ) cross section for iron, assuming ~ 8 displacements per (n,γ) reaction. Except for the resonances, the approximate $E^{-1/2}$ shape below $\sim 10^{-2}$ MeV also represents the (n,α) cross-section for any boron reactions that produce helium.

The dashed curve represents a solution based on a constant input form; i.e., all neutrons are initially considered to be equally damaging. Above $\sim 10^{-2}$ MeV, this solution falls approximately midway between the envelope bounds of the other solutions, while below this energy it falls nearer the upper bound. It represents, therefore, a damage function that assigns a high weight to low energy neutrons.

The high energy ($\sim 10^{-2}$ to 5 MeV) part of the $G(E)$ is fairly well defined in both shape and magnitude and shows a significant departure from the unmodified Kinchin and Pease input model with the (n,γ) and (n,α) contributions. Though the $G(E)$ is not as well defined below $\sim 10^{-2}$, it shows a significant departure from the unmodified input model. The solution bounds are sufficiently small to conclude tentatively that low energy neutrons (below ~ 0.1 to 1 MeV) contribute significantly to damage of 304 SS.

The departure of the envelope of the derived $G(E)$'s from the Kinchin and Pease input form results from the use of a single fixed slope straight line fit through each experimental property change data point (See the dashed lines in Figure 5.3). The single fixed slope approach was used because a grouping of experimental data by EBR-II spectral hardness, Figure 5.3, indicated that it was a reasonable and conservative method of extrapolation and interpolation of the existing experimental data to the 10% TE level.

EXPERIMENTAL DATA - TOTAL ELONGATION VERSUS NEUTRON FLUENCE

Neg 0694012-4

FIGURE 5.3. Experimental Data - Total Elongation Versus Neutron Fluence

The experimental data in Figure 5.3 indicate that the softer EBR-II spectra (such as 53, 54, 55, and 57 in Figure 5.3) require less fluence to cause a reduction in TE to the 10% level. Since this is not necessarily consistent with previous findings, a recheck and review of all experimental neutron spectral, fluence, specimen irradiation and test temperatures, and property change data for TE are in progress. In addition, data for other property changes such as uniform elongation, ultimate tensile strength, yield strength, and void volume change are being prepared and/or considered for analysis. This additional work will establish whether or not these preliminary results are qualitatively valid.

COMPUTER SIMULATION OF RADIATION DAMAGE

Cascade Formation and Annealing

D. G. Doran

The DEFECT computer code is used to study configurations of point defects in atomic lattices. A study⁽⁴²⁾ was begun some time ago of the sensitivity of certain vacancy and interstitial computations with DEFECT to selected variations of interatomic potentials representing copper and iron. Additional computer runs this quarter completed this work and a paper has been prepared for publication. The principal result is that a new understanding has been achieved of the effects of small discontinuities in the potential; such discontinuities at the maximum range of the potential are not uncommon in simulation work. Furthermore, the difficulties associated with formulating a potential that unambiguously involves only certain neighbor shells in the presence of large microscopic strains is emphasized.

Simulation of short term annealing of displacement damage is continuing. A new aid to visualizing the pre- and post-anneal defect configurations has been developed in collaboration with the Process Simulation and Analysis Section of BNW.

FIGURE 5.4. A 5 keV Displacement Spike in a-Fe. This is an example of a very compact spike. The direction $[3,2,1]$ is indicated by the arrow; the take-off point of the PKA is at the origin. Edge length of the box is 25 half-lattice constants (36 Å). Vacancies and interstitials are represented by o's and x's, respectively. A hybrid computer was used to produce the CRT Displays.

Top: 0 °K simulation with CASCADE-CLUSTER code.

Center: Room temperature simulation -- spike in top photo after short term, low temperature anneal with ANNEAL code.

Bottom: Elevated temperature simulation -- spike in top photo after short term, elevated temperature anneal with ANNEAL code.

A program has been written for a hybrid computer that provides an oscilloscope display of the defect configuration as viewed from any angle or apparent distance. By varying the angle slightly, stereo pairs are easily made. An example is shown in Figure 5.4.

The simulation of displacement damage due to (n,γ) recoils continues. Additional 400 eV CASCADE-CLUSTER and ANNEAL computer runs have been made to improve the statistics. Recognizing that the CASCADE code was not originally designed for such low energies, a comparison has been made between present results and the earlier low energy work of Erginsoy et al.⁽⁴³⁾ which utilized a more appropriate code. The latter code (GRAPE) handled multibody collisions whereas CASCADE treats all collisions as binary ones. Although Erginsoy et al. did not publish complete cascade configurations,* a dominant feature of their results is the frequency with which replacement chains appear. These chains serve to dissipate energy and to isolate interstitials from the vacant lattice sites left behind.

Such chains are inhibited in CASCADE, presumably by the binary collision model employed, resulting in a somewhat greater number and higher density of defects. The higher density causes a more rapid annihilation rate upon annealing, however, so that the number of defects in the annealed distributions is about the same whether the spike was produced by CASCADE or GRAPE. The higher defect density from CASCADE also results in excessive vacancy clustering relative to GRAPE. For example, 1000 eV spikes from CASCADE and GRAPE,* both produced by a PKA moving initially in the [531] direction, were given a simulated low temperature anneal (interstitial motion only) for 500 time steps. Four ANNEAL runs using different random number sequences were performed for each spike with the following results: The CASCADE spike started with 14 single

* For comparison purposes, cascades were synthesized from composites of published PKA trajectories computed with GRAPE.

interstitials (I_1) and ended with an average total of 5.75 interstitials per spike including di- and tri-interstitials. The GRAPE spike started with only 8 I_1 s, but ended also with an average of 5.75 interstitials per spike and a very similar interstitial cluster size distribution. The vacancy distribution of the CASCADE spike included initially a tetravacancy (V_4), a V_5 , plus 5 V_1 s; after annealing, vacancy clusters up to V_5 were still present. In contrast to this, the GRAPE spike started with 8 V_1 s and ended with an average of 5.75 V_1 s per spike.

The averaged results from the short term (250 time steps), low temperature simulated annealing of twenty 400 eV spikes in α -iron are presented in Table 5.6. In view of the above observations, vacancy clustering appears negligible.

Of greatest interest is a comparison of the annealing of the low energy spikes with that of much higher energy spikes reported previously.⁽⁶⁾ This is because the present investigation was prompted by the discovery from empirical studies of damage correlation (ductile-brittle transition temperature) that thermal neutrons seem to be contributing more to the damage state, relative to faster neutrons, than can be accounted for with present models of displacement damage. Since these models did not account for annealing, the possibility exists that annealing enhances the role of thermal neutrons as producers of damage through (n,γ) recoils.

Interstitial clustering is much less frequent at the lower energy, as would be expected, and annihilation is somewhat greater. The net result is that 41% of the pre-anneal interstitials remain after annealing, seven-eighths of which are mobile (I_1 or I_2). The 41% figure is presumably low, however, because of the apparent overestimate of displacements by CASCADE at low energies. The GRAPE program produced an average of 4 displacements at 400 eV compared with 5.9 from CASCADE.

TABLE 5.6. *Distribution of Defects in Clusters After Short-Term, Low-Temperature Annealing Simulation of 400 eV Cascades*

<u>n</u>	<u>Average Number V_n Clusters Per Spike</u>		<u>Average Number I_n Clusters Per Spike</u>	
	<u>Pre-Anneal</u>	<u>Post-Anneal</u>	<u>Pre-Anneal</u>	<u>Post-Anneal</u>
1	2.0	1.6	5.7	1.85
2	0.55	0.25	0.10	0.10
3	0.55	0.10	0	0.05
4	0.15	0	0	0.05
5	0.05	0	0	0
6	0.05	0	0	0
Total Defects	5.9	2.4	5.9	2.4

Assuming that the difference would disappear upon annealing, as in the 1000 eV example cited above, the fraction of interstitial-surviving annealing would be raised from 41 to $\sim 60\%$. Previous work on 5 and 20 keV spikes resulted in $\sim 48\%$ of the interstitials remaining after the same number of time steps; about one-third of these were mobile. There is thus a three-fold increase in the number of mobile interstitials contributed by (n,γ) recoils relative to high energy spikes as the result of short term annealing.

The result is not directly applicable to the empirical results noted above, however, because in that work a Kinchin-Pease model with a 25 eV displacement energy was employed, giving 8 displacements produced by a 400 eV recoil. Since this is double the GRAPE code result, the factor of three reduces to one and one-half.

A relative increase in mobile defects may be important because of their possible interactions with impurities.

Impurities are known to have important effects on the ductile-brittle transition temperature in ferritic steels.

Structure Damage in Imperfect or Strained Lattices

G. E. Russcher

The new computer code, SLIDES (Strained Lattice Irradiation Damage Experiment Simulation), is being developed to simulate lattice structure damage in materials with imperfect or strained fcc structure exposed to the high fluence fast reactor irradiation environments.

A re-evaluation of the program design has resulted in three significant modifications. The first modification is the incorporation of a new target selection routine that evaluates even weak interactions in proper sequence. Although the moving atom may not closely approach a target atom, weak interactions can change its direction. Each atom that the moving atom passes, within the range of the interatomic potential, is now treated as the subsequent target. In addition, the selection of a target is based on the exact locations of both the moving atom and target atom, not simply on their nearest lattice site locations. This target selection system eliminates unnecessary calculations of interatomic distances and atomic locations. This is important since target selection is repeated many times in the simulation of a displacement cascade.

Another improvement to the SLIDES code is the capacity for simulating a many-body interaction between a moving atom and two or more atoms situated symmetrically with respect to its path. This refinement is necessary in order to describe the movement of atoms along crystallographic directions. In focusing or channeling interactions, the probable paths of the moving atoms pass through groups of two to four lattice atoms. These atoms must respond in concert to the passage of the

moving atom, absorbing energy from it and in some cases also becoming displaced. The simulation of these many body interactions should bring the description of energy transfer along crystallographic directions into closer agreement with experiments.

The third modification to the SLIDES code is less complete than the others, because the necessary information is not yet available. Computer simulation results show that the orientation of an interstitial in an fcc lattice influences its interactions with neighboring interstitials and vacancies. The necessary criteria to describe these processes cannot be incorporated into the computer code until the appropriate interaction energies are determined. In order to use this forthcoming information, the SLIDES program is being modified to accept the orientation of defects in the description of irradiation damage production.

Programming of the SLIDES code is progressing in two phases: compilation and debugging. The compilation phase is about 90% complete. The only subroutine yet to be compiled is the OUTPUT subroutine which prints out the results of a problem and prepares magnetic data tapes. However, some interface problems still exist between already compiled subroutines.

The debugging phase has begun. Test problems are being run to demonstrate that the computations are self-consistent and compatible with the program as a whole.

INTERACTIONS OF CARBON IMPURITY ATOMS WITH A TILT GRAIN BOUNDARY

R. E. Dahl, Jr., J. R. Beeler, Jr., and R. D. Bourquin

Small concentrations of interstitial impurity atoms can modify the properties of metals. Of these impurities, which include boron, nitrogen, hydrogen, carbon, and others, carbon is perhaps the most important, particularly for the iron-based alloys. This exploratory study was initiated to obtain a

qualitative understanding of the effects on carbon atoms on a grain boundary and to obtain data on the relative energy required to place and move carbon impurity atoms in the vicinity of a grain boundary. The results should be pertinent to the effects of other interstitial impurities. A major objective of this study was to demonstrate the applicability of the computer experimentation with polycrystalline metals to specific problems involving precipitation, annealing, or strengthening.

Effects of impurity atoms on a grain boundary are of interest because ordering or disordering of the boundary could affect the strength and fracture toughness of the grain boundary and therefore the material. The stress required for sliding could be altered significantly. Structural perturbations could also affect grain boundary migration by pinning the boundary, thus affecting rates and mechanisms for recovery and recrystallization.

The carbon studies were made using the computer code GRAINS for a cell containing 1275 atoms arranged in seven (100) planes. The grain boundary in a (010) plane extended through the cell near its center. Carbon atoms were inserted in or near the center (100) plane to eliminate boundary effects on atom movements. Insertion at nonequilibrium positions assured some migration was necessary.

Single Carbon Interstitials

Carbon atoms were inserted individually at several locations in and near the 6° grain boundary to determine their relative effects on the grain boundary structure and to determine the regions where interactions between the grain boundary and impurity atoms could occur. The most striking results were changes in the structure and energy of the grain boundary. The structure of the grain boundary (Compare Figures 5.5 and 5.6.) became very well ordered. Apparently the carbon atom bonding drew the neighboring iron atoms together. Those iron atoms formed iron-iron bonds and considerable relaxation took place

THIS PLOT IS OF AN FCC LATTICE VIEWED IN THE (100) PLANE
 GRRIN BOUNORRY AT X= 11. IS FOR A ROTATION OF 6.00 DEGREES
 THIS PLANE IS: Z = 4

Neg 0693245-16

FIGURE 5.5. A 6° Tilt Grain Boundary in γ -Iron
 [Center (100) plane].

THIS PLBT IS OF AN FCC LATTICE VIEWED IN THE (100) PLANE
 GRAIN BOUNDARY RT X = 11. IS FOR A ROTATION OF 6.00 DEGREES
 THERE ARE 1 INTERSTITIALS IN THIS LATTICE
 THIS PLANE IS: Z = 4

Neg 0693245-17

FIGURE 5.6. A 6° Tilt Grain Boundary with a Carbon Interstitial Atom in a Misfit Region Plane Z = 4.

in the entire configuration, as shown by the absence of misfit regions in the planes above and below that containing the carbon atom. The addition of the carbon atom in the grain boundary hole reduced the energy of the cell some 2.6 eV. Approximately 1.6 eV of this change can be attributed to the relaxation of the lattice into a more stable configuration. The ordering or "healing" of the grain boundary would certainly strengthen the grain boundary and facilitate transmission of stresses and dislocation movement from grain to grain.

Energy and structural changes from the addition of carbon atoms 1 and 2 octahedral positions away from the boundary in a close-packed $[0\bar{1}1]$ direction (i.e., at coordinates 10, 10, 4 and 9, 11, 4, respectively) were almost identical to changes from the addition of a carbon atom to a perfect crystal. Strains caused by carbon atoms located in a normal or near normal octahedral site are too small to influence the structure. Thus, at only one-half unit cell distance from the boundary, carbon atoms appear to be in solution in a perfect lattice, that is, as if there were no interaction between the grain boundary and the impurity atom.

If the carbon atom was added one plane into the A grain in a $[0\bar{1}0]$ direction from the grain boundary hole, a very slight ordering resulted. The change in energy was approximately 0.1 eV greater than that observed for the perfect crystal experiment, as shown in Figure 5.7. The carbon atom forced atom 402 slightly into the misfit region and caused a slight ordering of the structure. The carbon atom moved to a position slightly away from a normal octahedral position. If a carbon atom was added at the edge of the grain boundary hole (10.9, 9, 4), the energy change was about 1.55 eV or about 0.25 eV greater than that for the perfect crystal experiment. The energy of the carbon atom in this position, -0.82 eV, was slightly higher than the -0.9 eV value in the perfect crystal

Neg 0694144-7

FIGURE 5.7. *Changes in Energy Caused by Carbon Interstitial Atoms in and near a 6° Tilt Grain Boundary.*

experiment. The carbon atom moved from the point of insertion into the grain boundary hole about 0.4 Å. No ordering of the structure was apparent. If a carbon atom was inserted midway between two (100) planes at coordinates (11.5, 10, and 4.5), which apparently was somewhat removed from the equilibrium position, the carbon atom moved to coordinates (11.97, 9.96, and 4.72). The mid-point between two (100) planes was found to be a saddle point, and migration towards a hole lying in or near a (100) plane is energetically favorable. The energy change was slightly less than that for the perfect crystal.

The most favored position for carbon atoms was in the center of the misfit region where they could interact with atoms of both grains; there is no analogous position in a free surface. Clearly, the configurational energy of the system was substantially lower with the carbon atom located between the boundary plane and the first interior plane than with the carbon atom either on the edge of the misfit region or between the second and third planes. Thus, carbon would also segregate either into the misfit region, or between the first and second plane, giving a carbon-rich region similar to that near a free surface.

Di- and Tri-Interstitials in a Grain Boundary Hole

The capacity of a grain boundary to accommodate interstitial atoms is of interest since carbide precipitates and cementite, Fe_3C , occur preferentially in grain boundary regions. The mechanisms of formation for these particles are largely undetermined as are the carbon concentrations required to significantly alter the physical and mechanical properties of polycrystalline specimens.

Experiments were conducted in which a second and then a third carbon impurity atom were added into the grain boundary hole. Each experiment was begun with the strain field of the

experiment in which the carbon atom was added at 12, 9, 4. Each of the subsequent carbon atoms was added at exactly the same location and were allowed complete freedom of motion so that they were forced to seek equilibrium positions. One notes that a large change in energy (Figure 5.8) or structural ordering occurred upon the addition of the first carbon atom to the misfit region. The energy change accompanying the addition of the second and third carbon atoms were approximately equal and slightly less than that observed in the perfect crystal experiment. Some relaxation of the crystal could be postulated. However, it seems certain that the tri-interstitial did not cause a disordering or an extensive strain field which extended into the grains, and thus the tri-interstitial which assumed the shape of a platelet was easily accommodated in the misfit region of the grain boundary. The shape of the tri-interstitial can be determined from the coordinates presented in Figure 5.9. This is seen to be a triangular configuration or platelet which lies in the same (100) plane. The irregularity of the triangle formed is due to the semi-fluid characteristics of the carbon cluster which results from the hard sphere potential function employed. Thus, the carbon atoms are constrained to assume the shape of the void since no attractive interaction of the carbon atoms is possible with this function. The effects upon the grain boundary structure with successive carbon additions are seen in Figure 5.10. No excessive strains are apparent and the lattice seems to easily accommodate the planar cluster.

Carbon Migration in a 6-Degree Tilt Boundary

Migration of impurity atoms in a grain boundary is a mechanism for the growth of impurity clusters and perhaps for precipitation. In order to determine the energy barriers which must be surmounted in migrating from one grain boundary hole to the next, a series of experiments were conducted in which the

Neg 0694144-9

FIGURE 5. 8. *Changes of the Cell Energy upon the Addition of Carbon Atoms into a "Bad Fit" Region 60° Boundary [Common (100) Plane]*

Neg 0694144-16

FIGURE 5.9. Configuration of Di- and Tri-Interstitial Clusters in Grain Boundary "Hole"

THIS PLOT IS OF AN FCC LRTTICE VIEWED IN ME (100) PLRNE
 GRAIN BOUNDARY AT X= 11. IS FOR A ROTRTION OF 6.00 DEGREES
 MERE RRE 1 INTERSTITIALS IN MIS LRTTICE
 MIS PLANE IS: Z = 4

Neg 0693245-8

FIGURE 5.20. *6° Grain Boundary with Three Carbon Interstitials in "Bad Fit" Region*

the carbon atom was placed at increments along the [100] direction between the two grain boundary holes. The interstitial migration energy plot, Figure 5.11, is very irregular. The low-energy position apparently lies at a point one-fourth the distance between the two grain boundary holes, and thus the center of the hole apparently does not coincide with a (100) plane. The midpoint appears as a definite saddle point and an energy barrier to migration of 1.2 eV in one direction and approximately 1.0 eV in the opposite direction. The migration energy plot is quite asymmetric, perhaps due to the characteristic fcc stacking. A metastable position is seen about 70% of the distance along the direction vector. The tortuous migration path of the carbon atom apparently follows the nooks and crannies of the grain boundary structure.

Neg 0694144-4

FIGURE 5.11. Carbon Migration Energy in a 6° Boundary $\langle 100 \rangle$ in γ -Fe.

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RADIATION EFFECTS ON ABSORBING MATERIALS
FOR CONTROL RODS

A. L. Pitner

The first subassembly of boron carbide being irradiated in a Hanford reactor was discharged after an estimated 3% ^{10}B burnup and disassembled. Samples were in the form of 60 and 80% TD powder, and 65, 90, and 99% TD pellets. Irradiation temperatures ranged from 550 to 1130 °F. In general, a high percentage of gas release was noted, and pellet degradation was very evident, particularly in the 99% TD samples.

The subassembly was composed of fifteen capsules, each capsule containing a particular sample form. Five of the capsules were thermocoupled, and the remaining capsule temperatures were inferred from these readings. Table 6.1 summarizes the loading of the subassembly and the irradiation conditions. Most of the capsules were punctured and the internal gas pressure measured after irradiation. The table also lists the percentage of helium gas released from the $^{10}\text{B} (n, \alpha) \text{Li}^7$ reactions assuming 3% ^{10}B burnup. Burnup analysis is not yet complete, however, and it must be emphasized that the gas release values quoted are very dependent on the burnup level. In any case, the relative values for the different sample forms should give an indication of comparative sample behavior. The results appear to be consistent within themselves. Gas release for similar materials generally increases with irradiation temperature. The gas release in the pellets is inversely proportional to the sample density, but apparently independent of density in the powders. The 88.5% gas release in the 65% TD pellet is unexpectedly high. Unfortunately, the only other capsule containing a 65% TD pellet leaked water during irradiation, and consequently, no check on the high gas-release figure could be made.

TABLE 6.1. Gas Release from Irradiated Boron Carbide

<u>Sample</u>	<u>Form</u>	<u>Temperature, °F</u>	<u>Gas Release, (a) %</u>
1 - 1	60% TD powder	550	24.2
1 - 6	60% TD powder	700	40.0
1 - 7	80% TD powder	600	21.4
1 - 8	80% TD powder	605	
1 - 2	80% TD powder	655	35.7
1 - 9	80% TD powder	715	37.3
1 - 10	65% TD pellet	865	88.5
1 - 3	65% TD pellet	Leaker	
1 - 4	90% TD pellet	725	35.5
1 - 11	90% TD pellet	860	
1 - 12	90% TD pellet	1120	43.0
1 - 13	99% TD pellet	865	18.1
1 - 14	99% TD pellet	870	
1 - 5	99% TD pellet	1035	27.1
1 - 15	99% TD pellet	1130	

a. Assumes 3% ^{10}B burnup.

Although a 0.010-in. gap was allowed for expansion of the 0.240-in. diameter pellet during irradiation, some of the pellets were stuck in their holders when opened and had to be forced out. This was particularly true for the 99% TD pellets. All pellets showed considerable degradation from the irradiation. Corners were rounded, cracks were prominent, and the pellets were mostly broken into smaller particles. The surface of the 99% TD pellets was quite powdery, while the lower density pellet's surfaces chipped easily. The three pellets in capsule 1-3, which leaked water during irradiation, were fused together, and the surface appeared very eroded.

The densities of several of the particles were measured by immersion in distilled water, and the results are shown in Table 6.2. After irradiation, the pellets all showed apparent densities near the theoretical value (2.52 g/cm^3), indicating that the porosity and cracks generated during irradiation are easily accessible to water.

Two more subassemblies are continuing under irradiation at similar temperatures, and their discharges are scheduled so that data at ^{10}B burnup levels of about 10 and 15% will be obtained. In addition, another capsule assembly operating at temperatures from 500 to 750 °C is also under irradiation, and data will be obtained at similar burnup levels in this temperature range.

TABLE 6.2. Particle Density Measurement

<u>Sample</u>	<u>%TD</u>	<u>Pre-Irradiation Density (g/cm^3)</u>	<u>Post-Irradiation Density (g/cm^3)</u>
1-10	65	1.700	2.482
1-4	90	2.006	2.391
1-12	90	2.000	2.303
1-5	95	2.440	2.498
1-13	95	2.520	2.466
1-14	95	2.530	2.490
1-15	95	2.530	2.480

NUCLEAR GRAPHITETHERMAL EXPANSION - W. J. Gray

The bulk coefficient of thermal expansion (CTE) for many nuclear graphites is in the range $1 - 4 \times 10^{-6}/^{\circ}\text{C}$, while the theoretical value based on crystallite average techniques for an isotropic graphite is about $9 \times 10^{-6}/^{\circ}\text{C}$. The difference between theoretical and observed values has been attributed to Mrozowski cracks⁽¹⁾ which are produced in the graphite when it is cooled from graphitization temperature. During irradiation with fast neutrons, the bulk CTE decreases slightly for many graphites until very high doses where it increases markedly. The increase at high doses is attributed to closure of the Mrozowski cracks but the slight decrease at lower doses seems anomalous since the lattice CTE of the crystallites has been observed to remain constant for irradiation temperatures above 300°C .⁽²⁾

For other graphites, including POCO graphite, the CTE decreases by as much as a factor of 3 when irradiated at 900°C and above. This is difficult to explain if the lattice CTE remains constant. Therefore, we decided to measure the lattice CTE of a POCO graphite that had been irradiated at high temperatures.

The c-axis lattice CTE of POCO grade AXZ-501 graphite was measured both before and after irradiation at 1000 to 1100°C to $1.2 \times 10^{22} \text{ n/cm}^2$. The unirradiated bulk CTE of this graphite is $6.7 \times 10^{-6}/^{\circ}\text{C}$; the irradiated value is about $2 \times 10^{-6}/^{\circ}\text{C}$. The lattice CTE was measured between 25 and 425°C by observing the change in the 2θ angle of the (002) diffraction peak using Cu $K\alpha$ X-rays. The unirradiated value was found to be $27.1 \pm 1.0 \times 10^{-6}/^{\circ}\text{C}$; the irradiated value was $26.8 \pm 3.0 \times 10^{-6}/^{\circ}\text{C}$. This is well within the accepted value of $27.8 \times 10^{-6}/^{\circ}\text{C}$.⁽³⁾ Thus, the decrease in the bulk CTE is not caused by a decrease in the lattice CTE.

Since the idea of Mrozowski cracks is also incompatible with observed changes in CTE and dimensions, for POCO and Gilsonite graphites at least, it is obvious that this problem will require additional careful study.

COMPUTER CALCULATION OF GRAPHITE OXIDATION IN HTGR

R. C. Giberson

The GOP computer program has been applied in the past to the DRAGON experimental reactor and to the Fort St. Vrain Reactor, the latter being a 330 MW(e) high temperature gas cooled power reactor (HTGR). The next generation of power reactors of the HTGR type is an 1100 MW(e) design. We are currently adapting GOP to permit analysis of the 1100 MW(e) reactor.

The basic differences between the two HTGR's [330 and 1100 MW(e)], as far as the graphite chemistry calculations are concerned, are an increase in the number of "zones" and higher graphite temperatures in the higher power reactor. Five different temperature zones, each having two distinct flow rates are required to approximate the 1100 MW(e) core, making a total of 10 zones. This may be compared with the 4 zones used in the 330 MW(e) case. To accommodate this calculation, GOP has been modified now to permit multizone calculations up to a maximum of 20 zones.

The higher temperatures expected in part of the 1100 MW(e) core may make gas boundary layer diffusion an important parameter in the overall graphite chemistry. This effect (gas boundary layer diffusion) is not part of the present GOP code. Data and equations have been obtained, however, and we anticipate including this effect in the code in the future.

COBALT-60 ADDITION TO THE GAMMA IRRADIATION FACILITY

D. E. Baker

An additional 360,000 Ci of ^{60}Co were added to the gamma irradiation facility which increased the total inventory of

cobalt to 620,000 Ci. Sixty flat, plate-type elements were transferred to the 13.5 ft. deep water-filled pool facility. The cobalt elements are supported in a source holder and surround four 2-in. diameter facility tubes. Ceric sulfate dosimetry measurements were completed in a 2-in. diameter dry facility tube. The maximum dose rate was 3.87×10^7 R/hr, and the variation in dose rate over a 6-in. length was less than 5%. The core volume where the dose rates are highest is 5-1/2 in. in diameter by 16 in. long. Two of the four facility tubes are equipped with heaters and gas purge lines to perform gamma irradiation at temperatures up to 500 °C. One facility tube is being used to study radiation-induced reactions which occur in reactor systems. Modifications are being made to the apparatus external to the gamma facility for measuring the radiolytic rate of reactions of CO₂ with H₂ in the 100 to 1000 vpm range in helium.

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NONDESTRUCTIVE TESTING

G. J. Dau

EDDY CURRENT METHODS - THREE-DIMENSIONAL REPRESENTATION OF
EDDY CURRENT TEST SIGNALS - H. L. Libby,

Three and four dimensional signal space representations of typical eddy current test signals were made as an aid in planning a digital computer approach for the separation of test variables. The test specimen was a short length of 0.63 in. O.D. stainless steel tubing with 0.54-in. wall. An internal differential test coil was used with operating frequencies at 125 kHz and 250 kHz.

Measurements were made in sequence for three test conditions at each of the two operating frequencies. The test conditions were:

- Probe wobble
- A fabricated notch opening onto the inner wall of the tube
- A fabricated notch opening onto the outer wall of the tube.

The in-phase and quadrature components of the test signals were displayed on an X-Y oscilloscope. Photographs were made of the Lissajou figures that were generated as the test coil assembly was traversed past the two differently fabricated notches. Cross references were made between the signals by making measurements in sequence at two operating frequencies for three selected fixed test coil positions. This was done for both fabricated notches.

Some of the signal data obtained are shown in the three dimensional representation in Figure 8.1. The axes C1 and C2 correspond to the reactance and resistance axes, respectively, of the 125 Hz channel of the tester, and the C4 axis represents the reactance axis of the 250 Hz channel. The signal vectors L, M, and N resulting from the inside notch occupy a different

FIGURE 8.1. *Three-Dimensional Signal Space Representation of Eddy Current Tubing Test*

region of signal space than those signal vectors, P, Q, and R associated with the outside notch. The signals are shown with a common origin O. It is further noted that the signals associated with the two different notches lie approximately in two separate planes. The planes defined by OLN (inside notch) and OPR (outside notch) have been extended by analytical methods to show their line of intersection OB. Thus, the inside notch signals appear in the plane OAB and the outside notch signals appear in the plane OCB.

It is determined that the line of intersection OB of the two planes is important in establishing a decoupling or separation matrix for use in identifying the signals. In this three-dimensional example a modulation matrix [A] containing three column vectors is first established. One of these column vectors is the vector OB. A second column vector is selected from the plane OAB (exclusive of vector OB). The separating matrix is then obtained by computing the adjoint $\frac{[A]^{-1}}{|A|}$ of the modulation matrix [A].

The above is placed on a more formal basis in the following. The relative values of the signals shown in Figure 1 are:

	Vector	C ₁	C ₂	C ₄
Inside Notch	L	- 4.6	11.5	12.0
	M	-16.5	22.8	26.0
	N	-10.8	12.5	13.0
Outside Notch	P	15.5	16.0	0.4
	Q	16.4	28.3	6.0
	R	6.5	16.0	5.0

The vectors L and N and P and R are assumed to define the two respective planes. A vector B common to each of these planes is

$$B = (L \times N) \times (P \times R) = \begin{matrix} C_1 & C_2 & C_4 \\ 5070 & 4910 & 5140 \end{matrix}$$

where

$$\begin{matrix} L \times N \\ P \times R \end{matrix} = \begin{matrix} 0 & -69.9 & 66.8 \\ 736 & -74.9 & 144.0 \end{matrix}$$

THERMAL METHODS - THERMAL TRANSDUCER METHODS - D. R. Green

Thermal techniques developed under this program have been applied to several practical nondestructive testing problems, and research and development of new thermal techniques is continuing. The high speed thermal transducer has been redesigned to allow infrared readout of the thermal image that is formed during a test. A system for automatically converting part of the transducer output to thermal impedance values is being developed. This system should enable better interpretation of the meaning of the thermal image, and pave the way for automation of the data interpretation function.

A transducer using the new infrared readout has been tested on several types of specimens. A vacuum is used to hold the transducer onto the specimen and to provide enough force on the transducer foil so that it is held in intimate contact with the test specimen. The infrared readout of the transducer output is independent of the test specimen emittance, and it gives a satisfactory image of defects within the test specimen. However, the first prototype of the new transducer design lacks the flexibility required for application to a variety of test specimens. In addition to this fault, tests showed that the transducer failed to reach equilibrium rapidly enough for proper operation of the impedance conversion circuit. Redesign of the transducer will be necessary to improve its performance and allow further development of the impedance conversion circuit.

The new transducer system is based on principles developed in past studies using low-speed sinusoidal thermal wave transducers. The high speed of the new system is due to the rapidity with which the thermal image can be obtained. (Typical imaging times range from 3 to 10 sec.) When development is complete, the new system is expected to be capable of automatically testing for defects which cause a difference in heat flow and/or

density within a specimen. This will be a desirable capability in testing fuel assemblies for future reactors, since one of the ultimate limitations on reactor performance will be the inability to rapidly remove heat that is generated within the fuel element.

ULTRASONIC METHODS - DETECTION OF COLD WORK AND GRAIN SIZE IN STAINLESS STEEL USING ULTRASONIC CRITICAL ANGLE REFLECTIVITY -
F. L. Becker and R. L. Trantow

The use of ultrasonic waves incident on a liquid-solid interface at the Rayleigh critical angle is an important non-destructive testing method because it can detect changes in near-surface material properties; e.g., residual stress, neutron radiation induced embrittlement, or as in this case, cold work and grain size. The depth of penetration of such waves is approximately one wavelength or from 2 to 50 mils for the frequencies used. Although experimental evidence has shown that a drastic change occurs in the ultrasonic energy reflection factor at the Rayleigh critical angle, only recently⁽¹⁾ a mathematical wave model was developed to predict the nature of the critical angle reflection factor.

Cold work and grain size affect the serviceability of clad for FFTF fuel pins. Standard methods of evaluating these parameters are not satisfactory as they are either destructive, too slow, lack required accuracy, or cannot measure cold work and grain size independent of other parameters.

By applying the ultrasonic critical angle reflectivity technique, we have developed a nondestructive test able to detect small changes in percent cold work and grain size.

Figures 8.2 and 8.3 demonstrate the effectiveness of this method in measuring cold work and grain size. In Figure 8.2 the amplitude is shown for states of cold work in 10% increments from the fully annealed condition to 50% cold work. These

FIGURE 8.2. Reflected Amplitude Versus Incident Angle for Cold Worked 316 SS
Test was performed on flat plates at a frequency of 15MHz.

FIGURE 8.3. Critical Angle Reflection Factors for 3 Grain Sizes of 316 SS Grain sizes are ASTM 8, 10 and 12. The test frequency was 15MHz.

results show that the measured critical angle changes from 31.6° for a fully annealed condition to 35.2° for 50% cold work. The grain size is the same in each case, namely ASTM 8. Figure 8.3 shows the reflected amplitude versus incident angle data for grain sizes of ASTM 8, 10 and 12, each sample having 20% cold work. These data demonstrate two facts. First, changes in grain size are detectable due to changes in the depth of the minimum; second, changes in the grain size have little effect on the critical angle.

The knowledge gained in the above tests was applied to small diameter tubing used for FFTF fuel pin clad. The results of these tests are shown in Figure 8.4 for cold working percentages of 0, 10, and 20%. The resolution shown here approaches $\pm 2\%$ of cold work, but this could be improved by a factor of at least 2 using electronic shaping and null detection circuitry.

The laboratory apparatus used for these tests is shown in Figure 8.5. For the tubing test, the transmitting transducer was focused on a 1 mil diameter area on the very top of the tube and the angle was varied in the radial direction. The disk in the center (Figure 8.5) drives the two radius arms apart varying the incident and receiving angles accurately and simultaneously. The angular position is obtained from the geared potentiometer with an accuracy of about 0.01° . The tube was held fixed at the center of curvature with its axis perpendicular to the page.

Tests performed with this apparatus are relatively slow, approximately 2 min per test, and unsuitable for production testing. One method of eliminating this handicap would be to use sampled multiple-element receiving transducers.

When the included angle of a focused beam contains the critical angle, the characteristic minimum can be observed in

FIGURE 8.4. Critical Angle Reflection Factors for 1/4 in. Fuel Cladding Tubes with Cold Work Percentages of 0, 10, and 20%

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FIGURE 8.5. *Dual Angle Goniometer Used for These Tests*

that angle in the reflected beam. The schlieren photograph (Figure 8.6) indicates such a condition. In Figure 8.7 the entire beam lies above the critical angle and no such minimum is observed. A multiple-element transducer with 32 elements in 1/2 in. was placed in the reflected beam. Both receiver and transmitter were held fixed while each transducer element was gated on, in sequence, by a logic circuit. The entire sequence is completed in 6.25 msec or 160 times a second. For an areal resolution of 1/8 in.², a scanning velocity of 12 ft/min is obtainable. The detected signal is shown in Figure 8.8. The critical angle is indicated by the minimum in the center of the photograph.

This method shows considerable promise but will require considerably more development to become practical.

NEW TECHNIQUES - MOSSBAUER EFFECT BACKSCATTER STUDIES -

R. M. Ord

The remarkable discovery in 1958 by Dr. Mossbauer of the emission and absorption of gamma rays by nuclei without recoil has led to one of the most sharply tuned resonant systems in nature, differences of up to one part in 10^{15} can now be determined. We have developed a system which operates in the backscatter mode rather than the transmission technique employed by others. Using this system, we can make Mossbauer effect measurements in a nondestructive manner with a signal-to-noise ratio greater than reported in prior work.

The importance of this development to nondestructive analysis is highly significant because we can analyze the sample "as is." The spectral results obtained are then used to study metallurgical and chemical characteristics of the sample. Although our initial effort is only a few months old, we are able to accurately measure the chemical shift, quadruple moment and the magnetic Zeeman hyperfine

FIGURE 8.6. *Schlieren Photograph of a Focused Ultrasonic Beam*
The incident beam, on the left, contains the critical angle and the critical angle minimum is observed in the reflected beam.

FIGURE 8.7. *Same as Figure 8.6 except incident beam does not contain the critical angle.*

FIGURE 8.8. *Detected Output of a 32 Element Transducer in the Reflected Beam Shown in Figure 6 Each element corresponds to approximately .2° angular resolution.*

splitting for spectroscopic analysis by using a learn computer fit program. Studies are currently underway to determine the feasibility of applying the Mossbauer effect to detection of stress corrosion cracking and strain detection.

We have designed a Mossbauer backscatter system with a capability of detecting 130% resonance effect magnitude $\left[\frac{I - (0)}{I} - \frac{(\infty)}{I} \right]$. This effect is a function of the sample surface area and the energy of the detected photon. We used a rolled foil stainless steel sample having an elliptical scatter area of 6.46 in.². We have calculated the sample surface area for both the nonresonance count and the resonance effect magnitude. These two effects are compared for both 14.4 keV y-rays and 6.3 keV X-rays. Specifically, we have shown that for a sample surface area greater than 0.5 in.², the 14.4 keV y-rays are the most efficient. Thus,

our results indicate that counting times can be shortened from hours for transmission geometry to minutes for backscatter geometry.

The components of the Mossbauer effect setup, shown in Figure 8.9, include a 30 mCi ^{57}Co source deposited on a palladium matrix which is Doppler shifted in the constant acceleration mode by a Nuclear Science Model NS-1 spectrometer. The collimating window is a frustum of a cone cut through the shielding. The rolled foil stainless steel sample is shown placed at an appropriate angle with respect to the source and the detector. The angles θ and ϕ as indicated in Figure 8.9 were obtained by maximizing the resonant peak to background signal by trial and error. These angles turn out to be very nearly 45° . A Reuter-Stokes Krypton- CO_2 Model RSG 61 M-2 proportional counter was used as the detector. The remaining part of the system included amplifying electronics and a multi-channel analyzer. We found that the background count was reduced appreciably using depleted uranium rather than lead as the shielding material around the hot source.

Figures 8.10 and 8.11 indicate the effect of the size of sample area upon the nonresonant count $I(\infty)$ and resonance

$$\left[\frac{I(0) - I(\infty)}{I(\infty)} \right]$$

for both the 6.3 keV and 14.4 keV photons.

The nonresonance count without a sample in the field of view is approximately 380 counts per hour for the 6.3 keV X-rays and 680 counts per hour for the 14.4 keV γ -rays. (The 14.4 keV count is higher because a piece of plexiglass is placed over the collimating window when counting 6.3 keV X-rays.) Because the sample is rotated by an angle θ as shown in Figure 8.9, the exposed area on the sample due to the incoming γ -rays is an ellipse. The area of an ellipse from zero to 1.38 in.^2 (where one of the foci is placed on the center line from the source) is the region where the entire sample area is exposed to the

FIGURE 8.9. Backscatter Mossbauer Effect Configuration

source, i.e., is not eclipsed by the collimating window. The penumbra region is the area between the two ellipses indicated by points a, d and b, c in Figure 8.9. This is the proportional area shown in Figures 8.10 and 8.11 between 1.38 and 6.46 in.². The reason that the nonresonance count of the 14.4 y-rays does not increase with sample area is because the Rayleigh scattering is small compared to the ambient background. In Figures 8.10 and 8.11, only the ambient background and points A and B were measured.

The sampling time necessary for Mossbauer backscatter can be determined by statistical theory which states that one standard deviation is approximately equal to $(N)^{1/2}$ where N is the average count in each channel of the multichannel analyzer. The deviation is also equal to ϵN where ϵ is the

FIGURE 8.10. Percent Resonant Effect as a Function of Scatter Area, 302 SS Sample 0.001 in. Thick

FIGURE 8.11. Nonresonant Count as a Function of Scatter Area,
302 SS Sample 0.001 in. Thick

error that can be accepted for each channel. Since 10^4 counts are required for a 1% error, with our setup 14.2 hr are required for a 2 in. by 2 in. scatterer. However, knowing that $I(\infty)$ is parabolic and the peak is lorentzian, all the channels can be used to estimate the statistical moments and thereby reduce the counting time to about 2 min.

We found that the surface of the sample can be penetrated to a depth up to 0.0005 in. (see Figure 8.12). This was obtained by using an 0.0005-in. iron foil mounted in front of an 0.001 in. stainless steel foil. The single peak of the stainless steel can be observed between the two inner peaks of the iron hyperfine splitting lines.

In conclusion, we have shown the 14.4 keV γ photons are more efficient than the 6.3 keV X-rays because of the lower nonresonance effect. A relationship has been shown for the resonance effect magnitude as a function of sample surface area. The decrease in sample times using the backscatter geometry and appropriate shielding is significant for nondestructive testing.

REFERENCE

1. F. L. Becker and R. L. Richardson. "Critical Angle Reflectivity," J. Acoustical Soc. Am., 45, No. 3, March 1969, pp. 793-794.

FIGURE 8.12. *Sample Thickness Effect - 0.001 in. Fe in Front of 0.001 in. SS; Center Peak is the 302 SS Resonant Peak Using 14.4 keV Gamma Rays*

ATR WATER LOOP OPERATION AND MAINTENANCE

R. S. Hope

The ATR completed Core I testing on June 8, 1969. Core II startup began on June 28, 1969, and full power of 250 MW was reached on August 16, 1969. Accumulated reactor operation was 5724 MWd as of September 15, 1969. Operational goal for Core II was extended from an original 4250 MWd to 8500 MWd. This leaves a goal of 11 more equivalent full power days before shutting down for Core III. The reactor has experienced several shutdowns because of instrument malfunctions and suspected abnormalities. Nucleate boiling was suspected in two loops due to excessive reactor instrument "noise" at 190 MW. Investigations could not explain the instrument noise and conditions improved as the reactor was taken to 250 MW. Flow problems in the reactor coolant also caused a 19-day shutdown the first part of July.

A "clean loop" corrosion coupon test was inserted into the pressurized water loop for Core II. The test includes three 42 specimen holders, two of which are located at the reactor midplane and one located just above the core. The purpose of a clean loop is to determine if fission products influence the corrosion rate of Zircaloy-2 specimens. The loop is being operated at 520 °F, 2000 psig, 70 gpm, pH of 10.00 ± 0.2 and oxygen concentration between 0.5 and 3 ppm. These conditions compare with previous conditions where corrosion specimens were irradiated in the ETR. Trouble was experienced in trying to maintain oxygen concentration at a nominal 1 ppm in that it kept decreasing to less than 0.05 ppm. Low pressure demineralized water (LDW) containing 7 or 8 ppm oxygen was pumped into the loop. However, a definite feed-and-bleed rate could not be established because of irregular oxygen concentrations showing up in different water samples.

On one occasion when nucleate boiling was suspected in the pressurized water loop, the loop temperature was lowered from 520 to 200 °F. The oxygen concentration increased to 15.75 ppm. Degassing was started but did not lower the oxygen level. After 9 hr of degassing, 330 cm³ of hydrogen was added to the loop. The extra hydrogen did not decrease the oxygen content and 5 hr later, 40 ml of hydrazine was added to the loop. The oxygen decreased from a high of 19.8 ppm to 0.32 ppm. The reactor shut down 45 min later and the excess hydrogen and ammonia was degassed off.

High oxygen contents were also observed during water chemistry tests in the 1-D loop during Core I. The loop temperature was 200 °F then and the high oxygen of 15 ppm could not be degassed out. It is believed that radiolitic decomposition of the water caused the high oxygen, and recombination would not take place at such a low temperature. Pressure of the loop was 2000 psig.

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