UNCLASSIFIED

Report No. BMI-1213
C-25 Metallurgy and Ceramics
(M-3679, 20th Ed.)

Contract No. W-7405-eng-92

PROGRESS RELATING TO CIVILIAN APPLICATIONS
DURING JULY, 1957

by

Russell W. Dayton
Clyde R. Tipton, Jr.

Classification cancelled (or changed to **UNCLASSIFIED**)
Memorandum from the Chief,
by authority of *Branch, dated 3-30-60*
by *JJ* T.E. date *4-7-60*

August 1, 1957

RESTRICTED DATA

This document contains restricted data as defined in the Atomic
Energy Act of 1954. Its transmission or disclosure of its contents
in any manner to an unauthorized person is prohibited.

BATTELLE MEMORIAL INSTITUTE
505 King Avenue
Columbus 1, Ohio

UNCLASSIFIED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

CONFIDENTIAL

3

TABLE OF CONTENTS

	<u>Page</u>
REPORTS RELATED TO CIVILIAN APPLICATIONS ISSUED DURING JULY, 1957	5
A. DEVELOPMENT OF MATERIALS FOR HANFORD REACTORS	7
Thermal Conductivity of Uranium and UO ₂	7
Mechanical Properties of Zirconium Alloys	8
Zirconium Cladding	8
Preparation of Aluminum, Molybdenum, and Zirconium Single Crystals	8
B. DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS	13
Extrusion Cladding of Flat Plates	13
Study of the Liquidus in Aluminum-Uranium Alloys	14
Preparation of Aluminum-Uranium Alloys	14
Development of a Mechanical Extensometer	15
C. PLANT ASSISTANCE TO MCW.	17
Investigation of Uranium Oxides	17
The Electrical Properties of Uranium Dioxide	18
The Corrosion Resistance of Selected Stainless Steels	19
Gas-Metal Studies	19
E. GENERAL FUEL-ELEMENT DEVELOPMENT	21
Development of Dispersed UC and UN Fuel Elements	21
Fabrication of Cermet Fuel Elements	22
Fueled Zirconium Hydride Moderator	23
F. STUDIES OF URANIUM AND URANIUM-ALLOY FUELS	25
Radiation Stability of Uranium-Zirconium Alloys	25
Development of Gamma-Phase Uranium Alloys	25
Preparation of Uranium-Niobium Alloys	26
Mechanism of Aqueous Corrosion	26
H. REACTOR MATERIALS AND COMPONENTS	29
Oxidation-Resistant Niobium Alloys	29
Stabilization of Uranium Dioxide	32
High-Temperature High-Pressure Solid-State Studies	32
Rubbing Surfaces in Sodium Environments	33
Basic Studies of Pressure Bonding	33
Vapor Deposition of Molybdenum Coatings on Stainless Steel Tubing	35
I. PHYSICAL RESEARCH	37
Preparation, Fabrication, and Physical Properties of Uranium Compounds	37
Solid-State Study of the Ternary System Uranium-Nitrogen-Carbon	39
Structure of UC ₂	39
Study of Bonding Fundamentals	40
Niobium-Hydrogen Reactions	44
Migration of Hydrogen in Zirconium Hydride	49
J. CORROSION PROBLEMS ASSOCIATED WITH THE RECOVERY OF SPENT REACTOR FUEL ELEMENTS	51
The Zircex Process	51
The Darex Process	52
Fluoride-Volatility Process	53
K. EVALUATION OF A REFLECTOR-CONTROLLED HETEROGENEOUS BOILING REACTOR	55
Critical Experiment	55
Simulator Studies	56

CONFIDENTIAL

DECLASSIFIED

CONFIDENTIAL

4

TABLE OF CONTENTS
(Continued)

	<u>Page</u>
L. STUDIES OF SODIUM-TANTALUM COMPATIBILITY AT ELEVATED TEMPERATURES	57
Tantalum-Sodium Compatibility Studies	57
High-Temperature Mechanical Properties of Tantalum	58
Weldability of Tantalum for High-Temperature Systems	59
Determination of Oxygen in Sodium	60
M. DEVELOPMENTAL STUDIES FOR THE PWR.	61
Reactor Flow Studies	61
X-Ray Photometric Examination of Fuel Elements	62
High-Strength Materials for Pressurized-Water In-Pile Tubes	62
Pressure Bonding of Zircaloy 2-Clad Fuel Elements Containing Compartmented Oxide Fuel Plates	63

CONFIDENTIAL

0371228.030

CONFIDENTIAL

5 and 6

REPORTS RELATED TO CIVILIAN APPLICATIONS
ISSUED DURING JULY, 1957

BMI-1188 "An Investigation of Uranium Solid Solubility in Thorium", by Arthur A. Bauer, Frank A. Rough, and Ronald F. Dickerson.

BMI-1192 "The Reaction of Steam With Uranium and With Various Uranium-Niobium-Zirconium Alloys at High Temperatures", by Alexis W. Lemmon, Jr.

BMI-1193 "The Influence of Lubrication on the Compactability of Magnesium-Green Salt Blends for Bomb Reduction", by Stan J. Paprocki, Ronald J. Carlson, and Edward G. Smith, Jr.

BMI-1195 "New Beta Heat-Treating Salt Baths for Reducing Hydrogen Pickup by Uranium Rods", by Lawrence L. Lortscher, Karl A. Sense, and Robert B. Filbert, Jr.

BMI-1201 "Progress Relating to Civilian Applications During June, 1957", by Russell W. Dayton and Clyde R. Tipton, Jr.

CONFIDENTIAL

DECLASSIFIED

A-1

A. DEVELOPMENT OF MATERIALS FOR HANFORD REACTORS

F. R. Shober

Equipment is being assembled and tested to determine the feasibility of measuring the thermal conductivity and electrical resistivity of irradiated uranium rods clad with Zircaloy 2 with NaK as the heat-transfer medium. Design has begun on apparatus to measure the thermal conductivity of UO_2 . A comparison of the creep properties at elevated temperatures of Zircaloy 2 and Zircaloy 3A is continuing. A 5000-hr creep test at 12,500 psi and 650 F on Zircaloy 2 produced a minimum creep rate comparable with that obtained from shorter time tests at the same temperature and stress.

Determination of the burst strengths of welded zirconium and Zircaloy 2 tubes at room and elevated temperature is continuing. The soundness of bonds formed between zirconium and electropolished uranium by hot hydrostatic pressing is being evaluated by water quenching from 730 C. Growth of single crystals of zirconium by the strain-anneal method is being attempted.

Thermal Conductivity of Uranium and UO_2

H. W. Deem and C. F. Lucks

Uranium

The effect of irradiation on the thermal conductivity of uranium is to be studied. Exploratory work will determine the feasibility of making thermal-conductivity and electrical-resistivity measurements on uranium rods that are clad with Zircaloy 2 with NaK as the heat-transfer medium. The steady-heat-flow method will be used to measure the thermal conductivity of these specimens.

The vacuum system, constant-temperature water bath, and various fixtures have been completed.

Work is now in progress on a guard air-metering system, and a constant-voltage power supply. The lead for radiation shielding has been poured. The completed apparatus should be ready for a test run by the end of August.

 UO_2

A program directed toward making thermal-conductivity measurements of irradiated UO_2 is being undertaken.

A longitudinal-flow steady-state comparative method, capable of remote operation, is being considered.

It is planned to complete the design and begin construction of the apparatus and order the necessary auxiliary components during the next work period.

CONFIDENTIAL

A-2

Mechanical Properties of Zirconium Alloys

F. R. Shober, L. L. Marsh, and J. A. VanEcho

The determination of the elevated-temperature creep properties of Zircaloy 2 and Zircaloy 3A has been continued. Several additional burst tests have been made at room temperature and 300 F on welded Zircaloy 2 tubes.

The creep properties of the present lot of Zircaloy 2 and Zircaloy 3A are being determined for times up to 2000 hr. The stress level has been chosen in most instances to limit the total creep deformation to less than 2.0 per cent. The results of the creep tests are shown in Table A-1. A specimen in test at 650 F and 12,500 psi has been discontinued after 5000 hr. The creep rate for this longer test, 0.000025 per cent per hr, compares quite well with the creep rate of test which was under load for 1000 hr. Total deformations obtained during specific time intervals are not wholly in agreement. Additional results of burst tests on welded Zircaloy 2 tubes at room temperature and 300 F are given in Table A-2.

Zirconium Cladding

F. R. Shober and R. F. Dickerson

Cladding zirconium to electropolished uranium by hot hydrostatic pressing is continuing. This investigation is to be carried out to determine the time and temperature conditions to produce a metallurgical bond between the two materials.

Examination of an electropolished uranium specimen clad with zirconium and pressurized at 10,000 psi at 1350 F for 2 hr showed a good continuous metallurgical bond. The bond is similar to that obtained from a specimen pressured for 1 hr at the same temperature and pressure. The soundness of the bonds of the clad specimens are being evaluated by water quenching from 730 C.

Additional specimens are being pressurized under conditions to produce a sound bond. These specimens when completed will be sent to Hanford for testing and evaluation.

Preparation of Aluminum, Molybdenum, and Zirconium
Single Crystals

J. A. DeMastry, F. R. Shober, and R. F. Dickerson

The preparation of high-purity aluminum, molybdenum and zirconium single crystals for irradiation studies is continuing.

CONFIDENTIAL

037726A.1030

TABLE A-1. CREEP PROPERTIES OF ANNEALED ZIRCALOY 2 AND
ZIRCALOY 3A AT ELEVATED TEMPERATURES

Lot	Specimen	Temperature, F	Stress, psi	Time of Test	Deformation at Indicated Time						Load Off	Creep Rate(a), per cent per hr
					Load On	50 Hr	100 Hr	500 Hr	1000 Hr	Total		
<u>Zircaloy 2</u>												
1	6-25	300	25,000	1005.0	0.385	1.50	1.65	2.17	2.45	2.50	--	0.0005
2	20-1	300	25,000	1076.3	0.25	3.10	3.40	4.0	4.25	4.283	4.044	0.0005
2	31-2	300	25,000	1000.0	0.23	2.90	3.15	3.60	3.87	3.87	--	0.00054
1	6-10	450	23,000	1000.0	0.30	-	2.05	2.13	2.15	2.15	--	0.00002
2	1-5	450	23,000	1007.8	0.10	3.68	3.74	3.76	3.773	3.773	3.550	0.000025
2	1-6	450	20,000	1080.4	0.25	2.35	2.38	2.40	2.413	2.413	2.265	0.000025
2	1-7	450	20,000	300(b)	0.25	1.68	1.695	--	--	--	--	--
2	20-3	550	20,000	716	2.10	3.02	3.03	3.035	--	3.035	2.89	--
2	15-5	550	17,000	50(b)	0.38	1.065	--	--	--	--	--	--
2	15-3	550	15,000	1756(b)	0.16	0.47	0.475	--	--	--	--	--
2	34-7	550	14,000	125(b)	0.16	0.375	0.382	--	--	--	--	--
2	15-40	550	12,500	150(b)	0.15	0.22	0.223	--	--	--	--	--
2	3-1	650	12,500	5009.0	0.15	0.23	0.245	0.282	0.335	0.433	0.336	0.000025(c)
2	14	650	12,500	1029.1	0.10	0.15	0.155	0.192	0.220	0.220	0.113	0.000028
2	15-1	650	15,000	850(b)	0.35	0.65	0.658	0.703	--	--	--	--
<u>Zircaloy 3A</u>												
--	3-11	300	20,000	225(b)	0.17	1.24	1.37	--	--	--	--	--
--	3-12	300	17,500	150(b)	0.14	0.75	--	--	--	--	--	--

(a) Based on creep deformation between 500 and 1000 hr.

(b) Tests in progress.

(c) Based on creep deformation between 4000 and 5000 hr.

CONFIDENTIAL

CONFIDENTIAL

A-3

TABLE A-2. BURST STRENGTH OF WELDED ZIRCALLOY 2 TUBES AT
ROOM AND ELEVATED TEMPERATURES

Specimen	Material	Nominal OD, in.	Measured Wall Thickness, in.	Measured OD, in.	Measured Wall Thickness, in.	Cold Work, per cent	Burst Pressure, psi	Burst Stress, psi (a)	Circumferential Elongation, per cent	Remarks
<u>Room Temperature</u>										
541-1	Zirconium	1.5	0.60	1.507	0.065	10	10,000	105,795	38.6	Fractured in weld
541-2	Ditto	1.5	0.60	1.506	0.0648	10	9,900	104,635	31.8	Ditto
<u>300 F</u>										
483-1-3	Zircaloy 2	1.5	0.035	1.504	0.039	10	4,750	87,715	18.6	Fractured in weld
483-1-4	Ditto	1.5	0.035	1.503	0.038	10	4,750	89,125	18	Ditto
541-4	Zirconium	1.5	0.060	1.506	0.065	10	6,650	70,335	2.1	Small hole in weld

(a) Calculated by Barrow's thin-wall formula, $S = \frac{Pd}{2t}$ where S = hoop stress, P = internal pressure, d = inside diameter, and t = wall thickness.

CONFIDENTIAL

A-4

CONFIDENTIAL

A-5 and A-6

Single crystals of high-purity aluminum (99.99 per cent) have been grown by the strain-anneal method in thin sheets and standard 0.505-in. -diameter tensile bars. These single crystals have been shipped to HAPO.

The construction of a modified Andrade furnace has been completed. Preparation of molybdenum single crystals will be attempted using this furnace.

The growth of high-purity zirconium single crystals has been initiated. Several 0.50- and 0.125-in. -diameter tensile bars have been prepared. These bars have been strained and are now being annealed.

B-1

B. DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS

R. J. Carlson

A new lot of M276 SAP material end plugs has been tested under various extrusion-cladding conditions. The end plugs, attached to steel cores, were clad without failure at extrusion speeds of 8 and 36 in. per min and at pressures of 34,000 and 54,000 psi, respectively. The plugs normally increased in length about 3 per cent. In one case, where the plug was preheated before cladding, it increased in length about 75 per cent without failure. This was unexpected, since the ductility of the M276 material is supposed to be very low at elevated temperatures. Additional tests will be made to determine the effect of speed and plug temperature on cladding-to-plug bonding. Construction of the "right-angle" extrusion-cladding assembly has begun.

A number of castings have been made containing 45 w/o uranium and utilizing a steel mold rotating at approximately 700 rpm. The evaluation of the castings has not been completed, but preliminary examination indicates that the best material was obtained from a casting which was poured at 2450 F into a steel mold at 850 F. Chemical analysis and radiographic examinations are being made of these ingots exhibiting the lesser degrees of visible porosity. Future work will be concerned with the evaluation of gating and feeding techniques in static molds and with the effects of variables encountered during centrifugal casting.

Extrusion Cladding of Flat Plates

R. J. Fiorentino, C. J. Slunder, and A. M. Hall

Efforts directed toward the evaluation of variables involved in the extrusion-cladding process were continued. One important variable is the design of the mandrel tip, which appears to have an effect on the extrusion pressure. A four-port mandrel was constructed with the intention that it might require less extrusion pressure than that obtained with the six-port mandrels used in the past. The four-port mandrel was made with a blunt-nosed tip and no reduction in the extrusion pressure was observed. However, the contour of the tip will be changed gradually to a more streamlined shape and additional experiments will be made to determine the effect of shape on extrusion pressure.

A new lot of M276 SAP material made into T-joint end plugs was tested under various conditions. The end plugs, attached to steel cores, were clad without failure at extrusion speeds of 8 and 36 in. per min and at pressures of about 34,000 and 54,000 psi, respectively. Radiographic examination of the clad specimens showed that the length of the plugs increased about 3 per cent. In one case where the plug was preheated before being clad, the length increased about 75 per cent, but with no apparent failure. This was unexpected since the M276 material is reported to have rather low ductility at high temperatures. Additional tests will be made with M276 and perhaps M355 SAP end plugs to determine the effect of extrusion speed and plug temperature on cladding-to-plug bonding.

CONFIDENTIAL

B-2

Construction of the "right-angle" extrusion-cladding assembly has begun. Approximately 10 weeks will be required to complete the tools. Meanwhile, auxiliary equipment needed for this assembly will be purchased and tests with the present "coaxial" assembly will be continued.

Study of the Liquidus in Aluminum-Uranium Alloys

V. W. Storhok, A. A. Bauer, and R. F. Dickerson

The investigation to determine the liquidus line of the aluminum-uranium system between 16 and 50 w/o uranium is continuing. Also being investigated are the effects on the system of magnesium additions up to 0.5 w/o and of silicon additions up to 1 w/o.

Preliminary differential thermal analysis confirms the eutectic temperature at 640 C. To date, insufficient data have been obtained to detect any effect silicon and magnesium may exhibit. However, metallographic examination of alloys close to the eutectic composition seems to indicate that magnesium additions have a coarsening effect on the UAl_4 particles while silicon additions produce finer particles.

Additional data are being obtained and should be available shortly.

Preparation of Aluminum-Uranium Alloys

N. E. Daniel, E. L. Foster, and R. F. Dickerson

Aluminum-uranium alloys containing up to 50 w/o uranium are of interest as a possible fuel for low-temperature water-cooled reactors. Therefore, a program of study concerned with the melting and casting of these alloys is now in progress. The primary objective of this program is the refinement of melting and casting techniques capable of producing sound homogeneous extrusion billets. It is also desired that the limit of uranium concentration that can be incorporated into such an ingot by conventional air-melting techniques be determined.

During July, a number of castings containing 45 w/o uranium have been made utilizing a steel mold rotating at approximately 700 rpm. All of these melts were poured at approximately the same temperature except for the last of the series. The first two castings of this series were made from remelt stock which had been alloyed at 2350 F and allowed to solidify in the graphite crucible. After solidifying, the material was reheated to 2550 F and poured through a graphite pouring cup with a 3/4-in. orifice. The first casting was poured into a cold mold and the succeeding castings were poured into heated molds. The evaluation of the castings has not been completed; therefore, the extent of segregation and porosity within the castings has not been fully determined, but the visual examination seems to indicate that the best material was obtained from the last casting, which was poured at 2450 F into the steel mold at 850 F. Chemical analyses and radiographic examinations are being made of those ingots exhibiting the lesser degrees of visual porosity.

CONFIDENTIAL

CONFIDENTIAL

B-3 and B-4

Further work will be concerned with the evaluation of gating and feeding techniques in static molds and with the effects of some of the variables encountered during centrifugal casting.

The effects of magnesium and silicon on the casting properties of the alloys is the subject of a related study. For this purpose, melts in the form of 1-in.-diameter rounds containing up to 50 w/o uranium and up to 1 w/o of the specified impurities are being prepared. The preparation of the major portion of these alloys is now completed. Those still to be made are the alloys containing 40 w/o or more of uranium.

Development of a Mechanical Extensometer

F. R. Shoher and R. F. Dickerson

The development of a mechanical extensometer of sufficient sensitivity and range to determine the transverse mechanical properties of uranium tubes is being continued. Design of an extensometer has been finalized and construction will be started.

C-1

C. PLANT ASSISTANCE TO MCW

A. A. Bauer

Studies of structural and compositional variations in the oxides of uranium and their effect on UO_2 reactivity have continued. Specimens have been prepared to determine whether the nitrogen in UO_3 is retained in hydrogen-reduced UO_2 . X-ray and microscopic examination of the UO_3 particles, containing various amounts of nitrogen, and of the UO_2 particles subsequently produced showed that there were variations in both structure and color in the oxide particles. Additional studies of these differences are planned and a microchemical method for nitrogen analysis is being investigated.

Plans have been made for studying the effect of impurity ions of different valencies on the electrical properties of UO_2 . However, no experimental work was performed this month.

The corrosion of Type 304 ELC stainless steel in chloride- and fluoride-contaminated nitric acid solutions has continued to receive study. Corrosion has been slight in 5, 18, and 30 w/o nitric acid solutions under long-term tests when the chloride content is 0.10 w/o or less. Chloride concentrations to 0.25 w/o are now being studied. In a 38 w/o nitric acid solution containing 0.25 w/o chloride, corrosion has been negligible after three 48-hr exposures.

Material and equipment are being assembled for a determination of hydrogen-permeation rate through molten magnesium fluoride slag.

Investigation of Uranium Oxides

D. A. Vaughan, J. R. Bridge, and C. M. Schwartz

The study of the structural and compositional variations in uranium oxides is being continued. The purpose of this investigation is directed toward increasing metal production and improving metal quality. Previous work showed that the structural and compositional differences in UO_3 that are created during the pyrolysis of uranyl nitrate hexahydrate are carried through the hydrogen-reduction step and influence the reoxidation reactivity of the UO_2 . In the current program, emphasis will be directed toward a study of the UO_2 and its hydrofluorination to UF_4 .

Experiments this month were designed to determine whether nitrogen that is retained in UO_3 may be retained through the hydrogen-reduction step. Uranium trioxide containing 0.02 (Sample 1), 0.08 (Sample 2), and 0.59 w/o (Sample 3) nitrogen was prepared by methods described in BMI-1205. These samples were reduced to UO_2 in hydrogen at 600 C. Preliminary X-ray diffraction and optical microscopical examinations were made of the UO_3 and the UO_2 . A microchemical test for nitrogen content in UO_3 and in UO_2 is being investigated.

C-2

The X-ray examination revealed the 0.02 w/o nitrogen UO_3 (Sample 1) to be amorphous, the 0.08 w/o nitrogen UO_3 (Sample 2) to be Type III, and the 0.59 w/o nitrogen UO_3 (Sample 3) to be a mixture of amorphous and a small amount of Type III. After reduction and exposure to air at room temperature all three preparations gave the UO_2 pattern. However, the two made from amorphous UO_3 contained a second cubic structure that has a slightly smaller lattice parameter and gave broader diffraction lines. The X-ray diffraction studies of the UO_3 and UO_2 will be continued.

Microscopic investigation of the UO_3 and the subsequent UO_2 powders revealed several interesting facts. It was seen that none of the powders was entirely homogeneous, but a mixture of red and yellow particles, in the case of UO_3 , and a mixture of black and brown particles in UO_2 . A primarily yellow Type III UO_3 (Sample 2) which contained some red particles reduced to a brown UO_2 which contained some black particles. The red amorphous UO_3 (Sample 1), containing some yellow, yielded black UO_2 , with some brown particles. Sample 3, an amorphous plus Type III UO_3 , contained more yellow particles than Sample 1, and the UO_2 made from Sample 3 contained more brown than UO_2 made from Sample 1. These differences are to be investigated further during the next period.

A microchemical method for the analysis of nitrogen in UO_3 has been investigated. The method appears to be sensitive to less than 0.02 w/o nitrogen. An evaluation of its possibilities for quantitative analysis is being made. In the case of UO_2 , solubility in the reagents may limit the use of the same test. Additional studies of the nitrogen-analysis methods will be made during the next period.

The Electrical Properties of Uranium Dioxide

J. W. Moody, R. K. Willardson, and H. L. Goering

No experimental work was undertaken on this project during July; however, the work for the future was planned in detail. The immediate objective of the program is to finish the study of the effects of nitrogen on the electrical properties of UO_2 . Some work utilizing uranyl nitrate as a doping agent remains to be done. Following this, a systematic study of the effects of metallic impurities will be undertaken. It is planned that UO_2 will be "doped" with the specific impurities CaO (Ca^{+2}), ZrO_2 (Zr^{+4}), and MoO_3 (Mo^{+6}). All these oxides form solid solutions and/or compounds with UO_2 so that they should serve as excellent doping agents. The choice of doping agents will allow the effects of the valence of impurities in UO_2 to be assessed. It is planned to prepare specimens containing 0.5, 1, and 5 a/o of the impurity ions. The actual doping will be accomplished by mixing intimately the oxide powders prior to pressing and sintering. The density of the specimens will be measured and the effects of the impurities on the sintering process will be correlated with the electrical properties.

C-3 and C-4

The Corrosion Resistance of Selected Stainless Steels

C. L. Peterson, W. C. Baytos, F. W. Fink, and R. S. Peoples

Investigations have been continued on the corrosion of Type 304 ELC stainless steel by nitric acid solutions contaminated with chlorides and fluorides in a manner similar to solutions that might occur during the recovery of nitric acid from uranium-processing plants.

Specimens have been exposed for 4500 hr in 5.0, 6000 hr in 18.0, and 5500 hr in 30.0 w/o nitric acid solutions boiling at reduced pressures. No indication of serious attack has been observed with chloride concentrations of 0.10 w/o or less. Tests have been started recently with chloride concentrations in the range of 0.15, 0.20, and 0.25 w/o, since these represent about the maximum range of chloride concentrations that would be considered in the operation of the nitric acid reconcentrator at Weldon Springs. The specimen exposed in the liquid phase of 5.0 w/o nitric acid bearing 0.15 w/o chloride has a corrosion rate of 0.4 mil per month following 500 hr of exposure. While this is not an excessive rate, it is considerably higher than any that have been measured in the presence of lower chloride concentrations.

Three 48-hr exposure periods have been completed with specimens exposed to 38.0 w/o nitric acid solutions containing chloride contaminations up to 0.25 w/o. So far, there is no sign of any severe attack at these concentrations.

Gas-Metal Studies

J. B. Bodkin, M. J. Trzeciak, and M. W. Mallett

An investigation of the hydrogen-magnesium fluoride slag relationships is being made as part of the program of evaluating factors which influence the hydrogen content of ingot uranium. The present phase involves the determination of the permeation rate of hydrogen through molten magnesium fluoride slag.

Several designs for the experimental system were made and evaluated. Materials and equipment are being assembled for the experiments.

E-1

E. GENERAL FUEL-ELEMENT DEVELOPMENT

F. A. Rough and D. C. Carmichael

Fuel-element development programs sponsored by the AEC Division of Reactor Development are reported in this section.

The development of dispersed UC or UN fuels is continuing and the preparation of samples for irradiation is in progress. These fuels were selected on the basis of fabrication studies which indicate that superior microstructures can be obtained compared with uranium dioxide dispersions.

Studies of cermet-type fuels have begun with emphasis on the fabrication of samples containing 60 to 90 volume per cent of UO_2 or other ceramic compounds.

Studies of a fueled zirconium hydride moderator have been initiated. Details of the program are still being worked out, but will include irradiation of samples as well as basic study of the structure of the complex hydrides.

Development of Dispersed UC and UN Fuel Elements

S. J. Paprocki, D. L. Keller, and G. W. Cunningham

Methods of fabricating flat-plate fuel elements containing cores of UC or UN dispersed in stainless steel have been established and reported previously. These methods will be used in this phase of the program to fabricate specimens. The main objective of the present program is to obtain information concerning the resistance to irradiation of these materials. Irradiation specimens will contain cores of 24.2 w/o UN or 24.2 w/o UC dispersed in Type 302B stainless steel and clad with Type 347 stainless steel. Specimens of each type will be irradiated to 10, 15, and 20 a/o uranium-235 burnup.

In addition, a limited number of mechanical tests will be run on similar specimens.

Irradiation specimens are being fabricated by the following procedure:

- (1) Blend powders 2 hr in Kelly V-type blender: (a) 24.2 w/o UN (minus 200 plus 270 mesh), minus 200-mesh Type 302B stainless, and 1/2 w/o camphor-alcohol solution; (b) 24.3 w/o UC (minus 100 plus 150 mesh), minus 200-mesh Type 302B stainless, and 1/2 w/o camphor-alcohol solution.
- (2) Press compacts at 50 tsi.
- (3) Clean the Type 347 stainless packs: (a) degrease in acetone or tri-chloroethylene, (b) wash in hot Alconox-water solution, (c) rinse in cold

CONFIDENTIAL

E-2

water, (d) rinse in 180 F hot water, (e) rinse in cold water, and (f) pat dry with paper towels.

- (4) Place core compacts in the picture-frame pack and weld covers and evacuating stem in place under an argon atmosphere.
- (5) Evacuate packs and degas at 600 F.
- (6) Seal the evacuated packs by forging the stem closed.
- (7) Hot roll packs: (a) UN-containing specimens are rolled at 2000 F to a 0.070-in. final thickness. (b) UC-containing specimens are rolled at 1800 F to a 0.065-in. final thickness. Both materials are rolled from a hydrogen muffle. Packs are reduced 30 per cent in thickness on the first pass and 20 per cent in thickness on each subsequent pass.
- (8) Anneal hot rolled plates for 2 hr at the rolling temperature.
- (9) Vapor blast plates to remove the thin oxide layer.
- (10) Cold roll plates to a final size of 0.060 in. ± 0.002 in., which gives a ratio of 7.4 to 1 for the total reduction in thickness.
- (11) Radiograph plates.
- (12) Machine irradiation specimens to 0.060 by 0.688 by 1.531 in. (core size is 0.050 by 0.5 by 1 in.).
- (13) Radiograph plates.

After the specimens have been fabricated, they will be subjected to a boiling nitric acid solution to determine whether there are any cladding defects. In addition, one specimen out of five will be examined metallographically.

Fabrication of Cermet Fuel Elements

S. J. Paprocki, D. L. Keller, and G. W. Cunningham

Techniques for fabricating cermet fuel elements are being investigated. In addition to the advantages of high fuel loadings and possible service at moderate to high temperatures, these fuel elements should have improved thermal conductivity and resistance to thermal shock compared with a pure ceramic fuel. Fuel materials which are to be considered include UO_2 , UC, and UN. Matrix materials include stainless steel and one or more refractory alloys. Fuel loadings to be investigated are in the range 60 to 90 volume per cent. Fabrication techniques will include preparation of cores by sintering and hot-pressing procedures and cladding of cores by hot-pressing and pressure-bonding techniques. A limited number of physical tests will also be run.

CONFIDENTIAL

037028.030

E-3 and E-4

One series of specimens containing 60, 75, and 87 volume per cent of UO_2 dispersed in stainless steel were pressed at 60 tsi and sintered 1.5 hr at 2200 F in hydrogen. After sintering, the UO_2 was leached out of the specimens by a nitric acid solution and an examination of the stainless steel skeletons was made. The 60 volume per cent UO_2 specimen had a stainless skeleton which was both continuous and uniform. The 75 volume per cent UO_2 specimen had a stainless skeleton which was continuous but somewhat segregated. The 87 volume per cent UO_2 specimen had a discontinuous skeleton. Density measurements indicated that porosity increased with increasing UO_2 contents.

Additional specimens are being prepared to determine the effect of compacting pressure and powder particle size on the density of sintered compacts. The hydrogen furnace is being altered so that specimens can be sintered at the melting temperature of stainless or at higher temperatures if desired.

Fueled Zirconium Hydride Moderator

H. E. Bigony and J. B. Vetrano

The object of this investigation is to determine the feasibility of using the hydride of a uranium-zirconium alloy as a combination fuel-moderator for a nuclear reactor. Although no definite operating specifications are being assumed, it is felt that this concept will have its greatest utility in a small, compact, gas-cooled power reactor operating at greater than 1000 F.

In order to evaluate the system, it is desirable to irradiate two different sets of encapsulated specimens of the material. One set of specimens would have a hydrogen/zirconium atomic ratio of about 0.8 to 1.0 and the other set would have a hydrogen/zirconium ratio of about 1.6 to 1.7. The unhydrided alloy will contain 1 w/o enriched uranium. Both specimens would be run to about a 5 per cent burnup at about 1500 F.

The uranium concentrations have been chosen as typical of the amounts required for criticality in a 2-ft-diameter by 2-ft-long cylindrical core. The hydrogen concentrations correspond with two different alloys of markedly differing mechanical properties.

It will also be necessary to determine the nature of the phases present in the hydrided alloys by using such techniques as dissociation-pressure measurements, electron fluorescence, and high-temperature X-ray diffraction.

F-1

F. STUDIES OF URANIUM AND URANIUM-ALLOY FUELS

F. A. Rough and A. W. Hare

An irradiation-test capsule containing zirconium-22 w/o uranium-alloy specimens is being prepared for long-term irradiation at the MTR.

Research is being continued on the development of corrosion-resistant uranium alloys but no new data or conclusions are reported this month.

The initial results of a literature survey on the current status of uranium-niobium alloys is reported in this section. This work will precede studies to perfect techniques for the production of sound homogeneous alloys.

It is reported that further investigations are being continued to determine the mechanism of aqueous uranium corrosion.

Radiation Stability of Uranium-Zirconium Alloys

A. W. Hare, A. E. Austin, A. A. Bauer,
E. L. Foster, and R. F. Dickerson

Irradiation of a temperature-controlled capsule containing two specimens of uranium-50 w/o zirconium is scheduled to begin on August 5, 1957. It is planned to irradiate the specimens at a temperature of about 650 C to a burnup of 2 total a/o.

As a follow-up to irradiation studies which previously have been made on zirconium-22 w/o uranium alloys, three specimens of this alloy representing three different heat treatments will be irradiated at a temperature below 400 C to a burnup of at least 2 total a/o. This irradiation will be done to determine if the microstructure of this alloy is an important factor in its radiation stability.

Development of Gamma-Phase Uranium Alloys

V. W. Storhok, A. A. Bauer, and R. F. Dickerson

A program has begun which has as its objective the development of a corrosion-resistant uranium alloy. Ternary and quaternary stabilized gamma-phase alloys are being sought for this purpose. Alloying elements which should stabilize the gamma phase and which are being investigated are chromium, molybdenum, niobium, ruthenium, vanadium, and zirconium.

A series of binary, ternary, and quaternary alloys containing 55 to 90 w/o uranium and with additions of the above elements is being prepared for a preliminary study which should eliminate any obviously inadequate alloy. This screening study will

consist of a determination of the effect which these additions have on the temperature of gamma decomposition, a brief study of the transformation kinetics of each alloy, and corrosion tests of the gamma-quenched alloys in high-temperature water. From the results of these investigations, an attempt will be made to establish the trend that these alloying additions have on alloy behavior. This may eliminate some additions and provide direction to changes in composition which may result in an improved alloy. Hot-hardness measurements will be obtained on all alloys as a measure of high-temperature strength and as an aid in the fabrication of these alloys.

On the basis of the initial results, additional compositions will be prepared for study.

Preparation of Uranium-Niobium Alloys

E. L. Foster and R. F. Dickerson

The results of investigations performed to date on certain uranium-niobium alloys indicate that the entire alloy system has great possibilities for use as reactor fuels. However, before any such complete study of the system can be planned, it is essential that a complete knowledge of all work being done by other investigators be obtained and that techniques are developed for the preparation of reproducible high-purity homogeneous alloys in every composition range.

In order that a knowledge of the current status of the alloy system can be obtained, a complete search of the literature has been initiated. The results of this search will be summarized in a report.

A review of the literature to date shows that much of the reported data have been obtained from alloys of questionable quality. This has been caused primarily by the use of highly contaminated niobium and by difficulties encountered in obtaining homogeneous alloys. The initial step in this study concerned with alloy preparation will be to determine the availability of niobium of consistently high purity. It is essential to the program that such material be available.

Once a supply of suitable material is obtained, studies will be initiated to perfect techniques for the production of sound homogeneous alloys.

Mechanism of Aqueous Corrosion

J. B. Schroeder, G. A. Westenbarger, C. M. Schwartz,
H. D. Hannan, P. D. Miller, and R. S. Peoples

The mechanism of aqueous uranium corrosion is being studied in order to develop more corrosion-resistant alloys for use as reactor fuels.

An objection to the proposed theory of the hydroxide corrosion mechanism, which is based on lower valence states of uranium, has been raised on the basis of results

F-3 and F-4

obtained when uranium is cathodically charged with hydrogen. An investigation of the effect of hydriding upon the corrosion resistance of uranium has been initiated. Experiments are under way to determine the optimum conditions for forming UH_3 in 0.05 N H_2SO_4 . Results obtained so far at 50, 60, and 70 C and 20 v for 6 hr show both alpha and beta UH_3 in addition to UO_2 .

Work has also been started to determine the amount of uranium which goes into solution in water during corrosion at 100 C.

Work has continued on corrosion reactions above 100 C. A program is now under way to attempt to identify the composition of the corrosion products at various stages during the reaction.

H-1

H. REACTOR MATERIALS AND COMPONENTS

F. A. Rough and A. W. Hare

Data are reported on the oxidation resistance of niobium-25 a/o vanadium, -10 a/o molybdenum, and -45 a/o zirconium alloys.

Research has been initiated on the stabilization of UO_2 at elevated temperatures and under oxidizing conditions. Experimental work has begun; however, no data are available this month.

An investigation of the effects of high pressure and high temperature on the reaction between UO_2 and BeO for the purpose of forming reactor materials for elevated-temperature applications has begun.

It is reported that a program concerned with the factors which control the friction and wear behavior of materials to be used in an environment of sodium is being investigated. Considerable emphasis for the preliminary work will be placed on the design of effective experimental equipment.

Studies have been initiated to better understand the phenomena involved in pressure bonding. The program has been designed so that correlations will be made between theoretical and experimental results to determine the validity of mathematical models for predicting bonding considerations.

Results of the work concerned with niobium coating small-bore stainless steel tubing in a modified moving-temperature-gradient coater are reported in this section. It is estimated that coating costs could be reduced by a factor of about 10 with a modest increase in the present size of the coating apparatus.

Oxidation-Resistant Niobium Alloys

W. D. Klopp, C. T. Sims, and R. I. Jaffee

The objective of this investigation is to determine the factors which influence the rate and mechanism of oxidation of niobium and niobium-base alloys. Continuous-weighting oxidation tests are currently being conducted on niobium and niobium alloys in dry and wet air at 1000 C.

Results from one test on pure niobium and three tests on niobium alloys are given in Table H-1. The pure-niobium sample had been polished to a metallographic finish before oxidizing, in contrast to previous niobium samples, which had been abraded to a 600-grit finish before testing. This sample oxidized approximately linearly, but at a rate one-third that of previous samples. The lower rate may result from the smoother starting surface, allowing a thicker subscale to build up before breaking down into the nonprotective outer scale. Tests on pure niobium are continuing in order to investigate the effects of surface preparation and oxygen content of the metal sample.

TABLE H-1. AIR-OXIDATION DATA FOR NIOBIUM AND NIOBIUM ALLOYS AT 1000 C

Sample	Composition, a/o	Humidity ^(a) , per cent	Parabolic Rate Constant, (mg/cm ²) ² /hr	Transition to Linear Rate, hr	Linear Rate Constant, mg/(cm ²)(hr)	Weight Gain, mg per cm ² , After Time Indicated					
						1 Hr	2 Hr	3 Hr	4 Hr	5 Hr	6 Hr
O-30 ^(b)	100 Nb	0	--	0.1	30.0	30.0	61.3	89.8	114.2	136.9	158.6
O-68 ^(c)	100 Nb	0	462	0.7	10.5	21.6	32.2	43.8	54.2	64.2	73.2
O-60	Nb-25V	100	--	--	228	(d)					
O-58	Nb-10Mo	100	92.1 ^(e)	2.7	2.61	8.7	13.0	16.2	19.1	21.8	24.3
O-67	Nb-45Zr	0	467	--	--	22.8	31.5	38.2	43.8	48.2	52.2

(a) Humidity at 20 C.

(b) Data from previous test on abraded niobium included for comparison.

(c) Surfaces were metallographically polished.

(d) Sample completely oxidized in 61 min.

(e) Parabolic rate changed from 67.8 to 92.1 (mg/cm²)²/hr at 0.7 hr.

CONFIDENTIAL

H-2

CONFIDENTIAL

H-3

The niobium-25 a/o vanadium and -10 a/o molybdenum alloys oxidized in wet air in essentially the same manner as in dry air. The vanadium alloy oxidized rapidly to Nb_2O_5 and V_2O_5 , the reaction being accelerated by the presence of liquid V_2O_5 . Since vanadium alloys containing up to 12.5 a/o vanadium oxidize in a parabolic manner, a transition from protective to nonprotective scale occurs between 12.5 and 25 a/o vanadium. Alloys containing 15 and 17.5 a/o vanadium are being prepared to investigate this intermediate region.

Niobium-10 a/o molybdenum formed a protective scale initially, but, at 2.7 hr, a transition to linear behavior occurred. Alloys containing 2.5 to 7.5 a/o molybdenum form scales which are protective for at least 6 hr. The breakdown of the protective scale at 10 a/o molybdenum may be associated with the formation of volatile MoO_3 . This test completes the present series of niobium-molybdenum alloys.

Niobium-45 a/o zirconium oxidized in a parabolic manner, with weight gains lower than previously tested niobium-10, 25, and 35 a/o zirconium alloys. Weight-gain data indicate that intermediate additions of zirconium (10 and 25 a/o) increase the oxidation rate of niobium, but large additions (35 and 45 a/o) cause the formation of protective scales and effect an improvement over the behavior of pure niobium. Additional tests will be conducted on these alloys in wet air at 1000 C.

Alloys containing 20, 25, 30, and 35 a/o chromium, and 1, 5, 10, and 25 a/o tungsten are being prepared by arc melting. These will be fabricated into strip, annealed, and oxidation tested in the same manner as the niobium-titanium, -vanadium, -molybdenum, and -zirconium alloys.

The continuous-weighing oxidation apparatus is currently being revamped to make it automatic recording. A 0.0005-in. wire will be fastened to one end of the balance beam, and changes in sample weight during oxidation measured by the change in impressed voltage across the fine wire. The voltage change will be graphed automatically on a recording millivoltmeter.

Future work plans include:

- (1) Continuation of tests at 1000 C on niobium-zirconium alloys
- (2) Fabrication and oxidation testing of niobium-vanadium, -chromium, and -tungsten alloys as described above
- (3) Oxidation testing of vacuum-degassed and oxygen-saturated niobium to investigate effects of oxygen content
- (4) Preparation and oxidation testing of niobium-cerium and -lanthanum alloys
- (5) Testing of niobium-alloy strips at 1200 C in dry air for comparison with behavior at 1000 C.

CONFIDENTIAL

H-4

Stabilization of Uranium Dioxide

W. B. Wilson and C. M. Schwartz

The purpose of this project is to explore possibilities of stabilization of the fluorite structure in UO_2 by the application of principles of crystal physics. UO_2 will oxidize to U_3O_8 when heated in an oxidizing atmosphere. The transition to U_3O_8 involves a volume expansion of the order of 30 per cent and results in a volatile oxide being formed which is not suitable for the temperatures being tentatively proposed for advanced reactors. Protection of the fuel by cladding techniques does not necessarily result in eliminating the problem, since failures will occur and the volume expansion rapidly accelerates the failure.

The concept to be generally applied to stabilization will be to react UO_2 with divalent and trivalent materials in such amounts that the valence change of uranium from plus 4 to plus 6 will be compensated. The zone theory of solids by which the valence electrons of all solids are free to move throughout the structure suggests, theoretically, that such valence adjustments are possible.

The present project will evaluate the performance of UO_2 under oxidizing conditions at elevated temperatures to establish a comparison basis. Stabilization will be attempted by reacting UO_2 in hydrogen to form solid solutions with selected divalent and trivalent cation oxides. Experimental work has been initiated in the evaluation of the behavior of UO_2 under oxidizing conditions and in the preparation of the solid solutions. Chemical analyses, weight-change data, X-ray diffraction parameter determinations, and electrical and optical properties will be utilized in determining the behavior of the solid solutions after exposure to oxidizing conditions at elevated temperature.

High-Temperature High-Pressure Solid-State Studies

W. B. Wilson and C. M. Schwartz

High-temperature high-pressure studies will be initiated to investigate the feasibility of forming reactor materials suitable for use at elevated temperatures. The general effect of both high temperature and high pressure is to form phases which have high cohesive energy and high bond symmetry, both of which are desirable from the standpoint of use at elevated temperature.

It is proposed to first investigate the effects of high pressure and high temperature on the reaction of various oxides with uranium oxide. In particular, the reaction between UO_2 and BeO will be investigated. These oxides have been selected for initial studies for two reasons; they are thermodynamically stable and their normal pressure-temperature phase diagrams have been studied in some detail.

Present work has been devoted to arriving at suitable die designs for the pressure apparatus. It is tentatively proposed to construct two die designs. The first of

CONFIDENTIAL

037000.000

H-5

these will probably be a modification of the Bridgman flat-anvil design with an external heater. The second unit will be designed for work at higher pressures and temperatures. As it now stands, a composite of Bridgman's flat-anvil design together with Hall's* design is planned.

Rubbing Surfaces in Sodium Environments

R. W. Endebrook, J. H. Stang, and W. H. Goldthwaite

A program of research has been initiated to explore factors which govern the friction and wear behavior of materials that may be useful for nonhydrodynamic fluid-immersed rubbing-surface members (for example, the sealing surfaces of face-type pump-shaft seals and the sliding parts of valves) in high-temperature (500 to perhaps 1500 F) sodium flow systems. It is the intent of the program to produce a better understanding of the parameters which restrict the performance of such rubbing surfaces and to establish basic information which will define and extend their working limits. It is visualized that this information can be useful not only in solving components problems presented by sodium-cooled reactors currently being developed but also can be applied to problems posed by future higher temperature systems.

Past experience with sodium-immersed rubbing surfaces, both at Battelle and at other sites, has indicated that, generally, the temperature limit for their reliable performance is about 500 F. At higher temperatures, the rubbing compatibility deteriorates and failure results. Several factors are suspected to be responsible for this behavior. Among these are corrosion phenomena, the degree of surface wetting, and the degree of volatilization of the sodium lubricating film at local overheated spots. It seems very likely that the stability of this lubricating film is related to the latter two factors.

According to present plans, considerable attention in this research will be devoted to investigating such relations and in devising fundamentally sound methods and techniques which can improve the stability. For example, ways and means of achieving an optimum degree of surface wetting may constitute an important area of investigation. During August, further emphasis will be placed on designing an experimental technique which can be useful in isolating some of the effects described above.

Basic Studies of Pressure Bonding

S. J. Paprocki, E. S. Hodge, S. D. Beck, and D. C. Carmichael

An investigation has been initiated which is directed toward a better quantitative understanding of the phenomena involved in pressure bonding, a cladding technique employing high hydrostatic pressures at elevated temperatures to accomplish bonding between metal surfaces. Battelle has been investigating this process for the past several years, and successful applications have been made in the cladding of fuel,

*Hall, H. T., "Chemistry at High Temperature and High Pressure", paper from High Temperature - A Tool for the Future, Stanford Research Institute, Menlo Park, California, p 161.

H-6

control, and moderator components. In this technique, the components are fabricated to final dimensions and assembled into a can which is evacuated and sealed. The assembly is placed in a high-pressure autoclave containing an internal heater and subjected to helium pressure up to 10,000 psi at elevated temperatures for a period of several hours to effect bonding. With this technique it is possible to clad brittle materials which cannot be fabricated by conventional techniques, bond components with complicated shapes, and bond in one operation complete subassemblies at relatively low cost.

This study has been initiated with the objective of understanding the basic phenomena of pressure bonding and determining quantitatively the important parameters affecting the process. With such information it should be possible to estimate the optimum conditions of time, temperature, pressure, and initial state of the bonding components required for fabricating any set of materials and fuel-element design. To pursue these objectives the study will proceed with closely related experimental and analytical investigations.

Analytical Studies of Pressure Bonding

In this phase of the program, the nature of pressure bonding will be described analytically from several viewpoints, each of which may or may not account for part or all of the important effects observed. To attempt to determine the predominant viewpoint, each one will be considered in sequence, and, whenever it is possible, a mathematical model will be constructed and used to predict results for specific materials and operating parameters.

The first major question under investigation is to determine the criteria under which the surfaces come into sufficiently intimate contact that bonding can take place. To simplify the analysis, the yielding of mating surfaces is being treated initially as equivalent to the yielding of a thick-walled cylinder under uniform pressure on the exterior surface. During July, cases were considered using the Mises yield criterion to determine the pressures needed to produce plastic flow at the inner surfaces.

The results thus far indicate that the pressures required to close the hole by plastic flow are proportional to the yield strength in simple shear. Furthermore, they imply that unless the yield strength in shear becomes very low at high temperatures, a very large pressure is required to reduce the hole size an appreciable amount. In the future, it is planned to apply experimental values of material properties together with pressure-bonding results with these materials in an effort to determine the applicability of this simplified model to the pressure-bonding process.

Experimental Studies of Pressure Bonding

This phase of the investigation is being directed toward the utilization of experimental data to achieve sufficient knowledge of the bonding phenomena to permit the prediction with some degree of assurance of the desired surface treatment and the range of temperature, pressure, and time required for bonding based on the physical properties of the materials.

H-7

Pressure-bonding experiments will be specially designed to evaluate each of the viewpoints postulated in the analysis. Specimens will be prepared which represent a variety of materials, properties, and geometries and will be pressure bonded under various conditions. A correlation will then be made between theoretical and experimental results to determine the validity of the mathematical models, and a determination will be made of the feasibility of predicting bonding parameters based on such considerations.

Vapor Deposition of Molybdenum Coatings
on Stainless Steel Tubing

C. F. Powell, D. M. Rosenbaum,
R. B. Palmer, and I. E. Campbell

Development of processes for vapor depositing molybdenum and niobium on the inside and outside of small-diameter Type 304 stainless steel tubing for ANL is continuing.

In prior work, adherent, uniform molybdenum coatings were obtained over the outside, but not over the inside, of 20-in. lengths of tubing. The deposition of niobium on the inside and outside of the small-bore tubing was tried in a moving-temperature-gradient coating unit. Black, powdery coatings were obtained at 850 C in this unit, and bright, adherent, but nonuniform niobium coatings were obtained at 1000 C. Following burnout of this coating unit, it was modified to provide a larger NbCl_5 capacity to permit a sharper temperature gradient to be obtained in it, and to permit seven of the small-bore tubes to be coated simultaneously on the inside only (instead of one tube on the inside and outside).

Work during July was concerned with niobium coating several sets of the small-bore tubing in the modified moving-temperature-gradient coater, and with examination of the coatings.

Niobium deposition was carried out by hydrogen reduction of NbCl_5 vapor at a total pressure of 5 to 7 mm mercury and a temperature of 1000 C. The passage of the temperature gradient was from the gas-inlet end to the exit end, the temperature of the specimens dropping from 1000 C to 200 to 250 C after passage of the gradient. The specimen tubes were coated in batches of seven. The usual undercoating of catalytically deposited nickel could not be used in making these coatings because of its low melting point (880 C) due to the presence of phosphorus in the nickel. One bundle of tubes which was given the nickel undercoating and then niobium coated at 1000 C welded inseparably together because of fusion of the nickel layers.

Small-bore tubing can be coated internally from end to end in the present apparatus, but the coating uniformity depends upon maintaining a constant rate of vaporization of NbCl_5 into the hydrogen carrier gas. This can be done by increasing the vaporizer temperature slowly throughout the deposition period, but the rate of increase as well as the initial (or final) vaporizer temperature must be determined empirically. Exact control has not yet been established, though the desired temperature range has

H-8

been bracketed. Consequently, the tubes so far coated have had slightly heavier coatings on either the inlet or the exit end. One group of specimens, which were submitted for testing, had an average coating thickness of about 0.5 mil, with the thickness varying from about 0.35 mil at one end to 0.7 mil at the opposite end. Subsequent batches of coated tubes have shown less variation than this. There was less than 10 per cent variation in coating thickness between the coatings on individual tubes in a single bundle.

In the present apparatus, the average coating thickness obtained has ranged from 0.5 to 0.6 mil for an 8-hr coating period to about 1.4 mil for a 16-hr coating period. These coatings were very smooth and adherent, though somewhat brittle. The ductility can undoubtedly be improved by purification of the NbCl_5 and the hydrogen used. The minimum deposition efficiency ranged from 68 to 75 per cent (based on the amount of NbCl_5 vaporized).

It has been estimated from current costs that the small-bore tubes could be niobium coated for about \$20 to \$23 each in apparatus similar to that used in the present bench-scale investigation. A modest increase in the size of the apparatus should bring the cost down to about \$2.50 per coated tube, exclusive of the cost of the tube itself.

Work for the coming month will be devoted to improving coating uniformity and ductility, and to the preparation of ten or more niobium-coated tubes to be submitted for testing.

I-1

I. PHYSICAL RESEARCH

F. A. Rough

Various programs of physical research supported by the AEC Division of Research are reported in this section.

The determination of properties of uranium compounds is continuing, and routine progress is reported. Initial reports are included covering studies which have begun on the system uranium-nitrogen-carbon, and on the structure of uranium dicarbide. Research is continuing on the constitution of ternary uranium alloys.

In the study of bonding fundamentals, the Kuczynski-type equation has been shown to be of considerable value, although not precisely applicable. Modifications of the equation are being made.

The results of temperature-pressure-composition equilibria studies in the ranges 100 to 900 C, 10 to 1000 mm of mercury, and atomic ratios of hydrogen to niobium of 0.01 to 0.85 are presented and interpreted. Emphasis is now being directed to a determination of the kinetics of the niobium-hydrogen reaction.

A new program on the migration of hydrogen in zirconium hydride has begun. The ultimate objective is to be able to predict hydrogen-concentration patterns in zirconium hydride or zirconium under any assumed thermal-gradient pattern.

Preparation, Fabrication, and Physical Properties
of Uranium Compounds

A. B. Tripler, Jr., and M. J. Snyder

The fundamental properties of some refractory uranium compounds which are potential nuclear fuels are being determined.

The study of the preparation and fabrication of UBe_{13} was continued. Hydrostatically pressed compacts of UH_3 and beryllium powder were reacted and sintered by heating in argon at 1500 C. The resultant material had a density of 90 per cent of the theoretical UBe_{13} density. However, the material showed small amounts of one or two extraneous phases which have not been identified as yet. In an attempt to homogenize and densify further, the material was ground to pass 200 mesh, and will be resintered. Fused specimens (102.5 per cent of theoretical density) have also been prepared; they too show extraneous phases.

The Brush Beryllium Company has studied the oxidation resistance of UBe_{13} . Brush specimens were prepared by mixing uranium and beryllium powders, cold pressing, and sintering in an argon atmosphere at 1400 C. A density 95 per cent of theoretical was reported. X-ray diffraction analysis detected only a single phase, UBe_{13} . No metallographic examination was reported. The following weight increases were observed on heating in a stream of dry air (100 cm³ per min) at 2300 F.

I-2

5 hr	45 mg per cm ²
25 hr	218 mg per cm ²

The Brush specimens followed the linear rate law in air at 2300 F. No data were given by Brush for lower temperatures. Lower temperature reactions with oxygen and nitrogen and reactions with H₂O vapor will be studied in this investigation.

Sintering conditions for UC₂ were investigated. Density was increased from 75 to 93.5 per cent of theoretical by increasing the sintering temperature from 1650 to 1850 C and by decreasing the average particle size from greater than 20 μ to about 5 μ . A small amount of a 2 w/o solution of Carbowax 400 in CCl₄ was used as a binder in pressing the UC₂ compact prior to sintering. The first UC₂ specimen broke into many pieces during rapid cooling (1850 C to room temperature in 1 hr) after sintering. Another specimen broke into three pieces during slower cooling (1850 C to room temperature in 48 hr). No phase changes over this temperature range are indicated in the literature. The apparent thermal fragility will be studied further by making thermal-expansion measurements. Examinations will be made on the UC₂ fragments to determine purity.

A technique was developed for electroplating nickel on USi which appears to be satisfactory for preparing thermal-conductivity specimens. The method is an adaptation of the method recommended for plating on uranium*. It consists of the following steps:

- (1) Polish specimen with 240- and 400-grit metallographic paper
- (2) Clean anodically in hot alkaline cleaner
- (3) Rinse
- (4) Dip in 1-1 HNO₃ at room temperature for 6 min
- (5) Rinse
- (6) Treat anodically in H₃PO₄-HCl solution (same composition given by Gray) at 80 F for 10 min at 25 amp per ft²
- (7) Rinse and remove powdery residue from Step (6) by rubbing on 240-grit paper
- (8) Clean anodically as in Step (2)
- (9) Rinse
- (10) Dip in 1-1 HNO₃, room temperature, for 4 min

*Gray, Allen, "Electroplating of Tuballoy Handbook", GT-2116.

I-3

(11) Rinse

(12) Nickel plate 15 min; nickel bath is same as recommended by Gray.

The nickel plate was uniform and appeared adherent. Tin was flowed over the nickel plate. No bubbles or discontinuities appeared, indicating excellent contact between the tin and the base metal. The tinned specimen was easily joined to a tinned piece of Armco iron, as would be done in the thermal-conductivity apparatus.

Some progress was made in electroplating nickel on USi_3 , but complete coverage of the USi_3 surface with nickel has not been achieved. Attempts to coat USi_3 with tin or 50-50 solder by the so-called "loaded-wheel" technique (described in BMI-1201) were not successful. Further work will be done with an alloy especially designed for coating glass.

A vacuum induction furnace is being readied for fabrication of specimens for thermal-conductivity testing.

Solid-State Study of the Ternary System Uranium-Nitrogen-Carbon

A. E. Austin and C. M. Schwartz

Study has been started of the uranium-nitrogen-carbon system to determine stability of uranium carbide solid solutions with uranium nitrides and/or ternary phases. The uranium-nitrogen system is quite similar to the uranium-carbon system and solid solution between UN and UC has been reported. Structure considerations indicate possible solid solution at high temperature of UC_2 with UN_2 under high nitrogen pressure.

Solid-solid reactions between compounds of the binary systems and solid-nitrogen reactions will be studied. The dissociation pressure-temperature dependence of selected compositions will be investigated. Adequate amounts of binary phases are being prepared.

Structure of UC_2

A. E. Austin and C. M. Schwartz

The study of UC- UC_2 systems has disclosed the transformation of UC_2 from tetragonal to face-centered-cubic structure. There were anomalies observed in the thermal expansion of the low-temperature tetragonal phase of UC_2 . The structure of UC_2 requires clarification for correct interpretation of its thermal behavior and for understanding the type of carbon bonding in relation to other carbides. Therefore, a study of the crystal structure of UC_2 has been started. Neutron-diffraction data will be obtained to aid in the determination of the carbon-atom positions.

Study of Bonding Fundamentals

J. B. Melehan, F. C. Holden, H. R. Ogden, and R. I. Jaffee

The purpose of this program is to determine the mechanism of the solid-state bonding process and to study the manner in which the process is affected by experimental variables. The bonding model considers the bonding reaction as being separable into two stages. The first is the instantaneous adhesion at points of metallic contact. The second stage represents a time-dependent growth of the interface between the contacting surfaces. The degree of bonding can be expressed either in terms of area (A) or in terms of the adhesion force (F). Therefore, these two criteria should show the separate effects of the instantaneous and time-dependent stages of bonding. Contact area and bond strength (or adhesion force) may be expressed as follows:

$$A = A_o + A_t$$

$$F = F_o + F_t,$$

where A_o and F_o are, respectively, the instantaneous bond area and the bond strength formed on initial contact of the bonding surfaces. A_t and F_t are, respectively, the time-dependent increments of bond area and bond strength.

The simulated single-asperity system now under investigation consists of an electropolished gold needle point in contact with a flat gold surface. The normal adhesion force has been measured under controlled conditions of temperature, time, and load. Results obtained from these experiments are summarized in Table I-1. Graphical presentation of these data reveals that an exponential breaking force-time relation exists having the form:

$$F = F_o + Ct^n,$$

where F_o is the instantaneous adhesive force, C is a constant at constant temperature, t is the time of bonding, and n is a constant having numerical values between 0.27 and 0.38. The form of the exponential term is similar to that derived by Kuczynski* for the sintering of spherical metal powder particles to flat metal plates. This derivation was based on the assumption that volume diffusion is the primary mechanism of bond-area growth between the sphere and the plate. The theoretical relation was also verified with some success by Kuczynski. It relates the bond growth to fundamental physical properties of the metal and experimental conditions as follows:

$$x = \left[\frac{40 \sigma \delta^3 a^2 D_v t}{kT} \right]^{1/5},$$

where

x = radius of area of contact

σ = surface tension at sintering temperature

*Kuczynski, G. C., "Self-Diffusion in Sintering of Metallic Particles", Transactions AIME. Journal of Metals, 185, 169-178, (February, 1949).

CONFIDENTIAL

TABLE I-1. NORMAL BREAKING FORCE OF SINGLE-ASPERITY GOLD CONTACTS UNDER VARIOUS CONDITIONS OF BONDING

Temperature, C	Breaking Force ^(a) , g, In Indicated Time, min							
	0	2	5	10	20	30	40	60
600 ^(b)	1.3	--	--	2	--	3.3	--	4.0
650 ^(c)	2.8	5.5	7.0	6.5	8.3	--	11.0	11.5
700 ^(c)	4.0	6.5	8.8	10.5	11.0	--	14.0	15.0
800 ^(c)	3.8	7.0	9.8	12.8	18.0	--	22.8	25.0

(a) All values of normal breaking force corrected to room-temperature values.

(b) Preload of 400 mg; 10-mg bonding load maintained for duration of test.

(c) Preload of 100 mg; zero bonding load maintained for duration of test.

I-5

CONFIDENTIAL

I-6

 δ = interatomic distance a = radius of spherical particle D_v = coefficient of volume diffusion t = time of sintering k = Boltzmann's constant T = temperature of sintering.

From this it follows that the contact area may be expressed as:

$$A = \pi \left[\frac{40 \sigma \delta^3 a^2 D_v t}{kT} \right]^{2/5}.$$

The adhesion force is

$$F = S\pi \left[\frac{40 \sigma \delta^3 a^2 D_v t}{kT} \right]^{2/5} \quad (I-1)$$

where S is the tensile strength. In the current work the specimen geometry is similar to that used in Kuczynski's work. However, instead of a spherical particle in contact with a flat surface under zero applied load, a needle with a spherical point is brought into contact with a flat surface under an initial load which forms a finite initial area of contact. Therefore, there is a measurable instantaneous adhesion in addition to the time-dependent increase in strength. Equation (I-1) can be modified to include the instantaneous-adhesion term as follows:

$$F = S\pi x^2 \\ = S\pi \left[\left(\frac{40 \sigma \delta^3 a^2 D_v t}{kT} \right)^{1/5} + x_0 \right]^2, \quad (I-2)$$

where x_0 is the radius of the contact area formed under the initial applied bonding load. It may be seen that the exponent (n) approaches the theoretical value of 0.4, and thus lends support to the theory that, in the absence of creep induced by external forces, volume diffusion is the rate-controlling process in the growth of bond. However, further analysis of the data shows that the magnitude of experimental increments of breaking force are much greater than those predicted by Equation (I-2). For example, at 700 C the increment of breaking force after the first 10 min of bonding is more than 100 per cent, whereas the predicted increment, determined by appropriate substitution into Equation (I-2), is found to be negligible in comparison with the zero time strength of 4 g. Similar discrepancies occur in varying degrees under all of the presently reported experimental conditions. This means that, in the general equation $F = F_0 + Ct^n$ above,

the constant C is not equivalent to the factor $S\pi \left[\frac{40 \sigma \delta^3 a^2 D_v}{kT} \right]^{2/5}$. Of the several

I-7

constants included in the latter term, the one which is most probably in error is the quantity a , the radius of curvature of the needle point. The specimen geometries considered in the theoretical treatment and in the experiments are not identical. The theoretical expression was derived for a system consisting of a small sphere of radius a in contact with a flat surface without external load. The initial area of contact thus was assumed to be zero. In the experiments, contact was made between a rounded needle point and flat surface under an applied load. The resulting deformation of the needle point increases the radius of curvature above that of the undeformed needle point. The effective radius of curvature required to bring agreement between the predicted strength increments and the experimental values would be about two orders of magnitude larger than that of the undeformed needle tip. That is, the radius of curvature would be 0.1 cm compared with the average value of 0.001 cm, for the original needle tip. No direct measurements have been made on the degree of needle-point deformation during loading.

The equation $x = \left[\frac{40 \sigma \delta^3 a^2 D_v t}{kT} \right]^{1/5}$ incorporates geometrical approximations of the contact model which were made to simplify the mathematics of the derivation. One result of these simplifications is that the expression is accurate only for values of the ratio $\frac{\text{radius of contact area}}{\text{radius of curvature}}$ which are much smaller than unity. The approximation becomes less valid as the bond growth continues, and as the contact area is enlarged. In the present experiments, the initial contact area under preload conditions gave values of the above ratio about equal to one. It is apparent that even in the early stages of bonding the Kuczynski equation is a rather inaccurate approximation to the experimental conditions.

Preliminary experiments have been conducted to study the effect of creep on contact area. Measurements of the hot hardness of gold have shown a transient effect with a general mathematical area-time relation of the form: $A = A_0 + Bt^n$, where A_0 is the area of the hardness impression at zero time, B is a temperature-dependent constant, t is time, and n is a constant. Values found experimentally for n were in the range 0.12 to 0.16 for tests in the range of 400 to 800 C. Electrical-resistance measurements were made at the bonded contacts, and contact areas were calculated from these measurements. Although the absolute magnitudes of these areas are uncertain, the area-time relation also is of the form: $A = A_0 + Bt^n$, with values of n equal to about 0.1.

Experiments in which needles were bonded to mechanically polished surfaces indicated that the degree of cold work is a significant factor. In general, bond strengths are markedly increased when cold-worked surfaces are present.

Future work will be directed toward deriving an expression to describe the observed bonding behavior more accurately. The Kuczynski equation, although not precisely applicable to the present experimental situation, has been shown to be of considerable value. Modifications of the equation to better approximate the experimental conditions are being made. Additional work also is needed to determine the effects of creep and cold work on bond growth, and experiments are in progress to study these factors.

Niobium-Hydrogen Reactions

W. M. Albrecht, M. W. Mallett, and W. D. Goode

An investigation of the niobium-hydrogen system was started in Fiscal Year 1957, and is continuing. To date, a study has been made of the temperature-pressure-composition equilibria in the niobium-hydrogen system. A summary of the experimental results is given below.

The equilibria of the niobium-hydrogen system were determined in the ranges 100 to 900 C, 10 to 1000 mm of mercury, and atomic ratios of hydrogen to niobium of 0.01 to 0.85. Determinations were made with three different samples of 0.03-cm niobium sheets and one of 0.3-cm-diameter rod. Figures I-1 and I-2 show isotherms for all the equilibrium data. Figure I-1 is a plot of equilibrium pressure against the hydrogen-to-niobium ratio while Figure I-2 is a logarithmic plot of the same variables. The 300 C isotherm (Figure I-1) is invariant at about 8 mm of mercury, indicating the presence of a two-phase region. No other invariant pressures were obtained for any of the isotherms at 350 C and above. Therefore, the results show that nearly all of the equilibrium data were obtained in a single-phase region.

Room-temperature X-ray diffraction patterns were obtained and metallographic examinations were made of several niobium specimens having compositions of hydrogen to niobium of 0.09, 0.20, and 0.68. These samples were quenched from the respective equilibrium temperatures. The sample with the 0.09 hydrogen-to-niobium ratio showed primarily a body-centered-cubic niobium structure, alpha phase, with a lattice parameter, $a_0 = 3.308 \pm 0.002$ kX. A very small amount of a second phase was also detected. Two phases were found in the specimen with the 0.20 hydrogen-to-niobium ratio, an alpha niobium phase ($a_0 = 3.312 \pm 0.002$ kX) and another body-centered-cubic structure, beta phase, with $a_0 = 3.44$ kX. The specimen containing 0.68 hydrogen to niobium contained primarily the beta phase, with $a_0 = 3.42$ kX, and a small amount of alpha phase, with $a_0 = 3.308 \pm 0.002$ kX. These data are in agreement with published results of various room-temperature X-ray investigations. If all the X-ray data are considered, the two-phase region appears to extend from 0.08 to 0.7 hydrogen to niobium at room temperature. Apparently, the temperature of the critical point where the two phases, alpha and beta, become identical lies somewhere between 300 and 350 C, as seen in Figures I-1 and I-2. Above the critical point, S-shaped isotherms were obtained. In equilibrium diagrams S-shaped isotherms are not unusual in single-phase regions immediately above the critical point.

Figure I-2 shows that the isotherms from a logarithmic pressure-composition plot were straight at concentrations up to a 0.1 hydrogen-to-uranium ratio. At higher hydrogen contents, the S-shaped curvature of the isotherms is quite evident. A plot of square root of equilibrium pressure against composition deviated from Sieverts law. However, it would not be expected that the equilibrium solubilities necessarily follow a square root of pressure dependency since the published X-ray investigations show lattice expansions in niobium as the hydrogen content increases. This constitutes another variable affecting equilibrium solubility.

I-9

FIGURE I-1. ISOTHERMS IN THE NIOBIUM-HYDROGEN SYSTEM

FIGURE I-2. PLOT OF LOGARITHMIC ISOTHERMS IN THE NIOBIUM-HYDROGEN SYSTEM

I-11

To put the equilibrium in the niobium-hydrogen system on a sound basis, it is necessary to consider the equilibrium of formation of various products (solid solutions) having definite hydrogen-to-niobium ratios. Therefore, various isopleths, logarithm of equilibrium pressure against reciprocal temperature, were plotted in Figure I-3. It is seen that a series of straight lines was obtained which increase in slope with hydrogen content. The equation for these lines is

$$\log_{10} P_{\text{mm}} = - \frac{A}{T \text{ (K)}} + B.$$

The values of the constants, A and B, in this equation and the heats of solution for the various compositions are given in Table I-2. It is seen that the heat of solution increases with hydrogen content. This increase in the heat of solution may be related to the expansion in the niobium lattice.

TABLE I-12. THERMODYNAMIC DATA FOR VARIOUS NIOBIUM-HYDROGEN COMPOSITIONS

Hydrogen-to-Niobium Ratio	Constants in $\log_{10} P_{\text{mm}} = - A/T + B$		ΔH , cal per mole
	A	B	
0.01	3509	4.92	16,050 $\pm$ 380
0.02	3628	5.54	16,600 $\pm$ 170
0.05	3674	6.30	16,810 $\pm$ 170
0.10	3693	6.83	16,900 $\pm$ 190
0.20	3908	7.49	17,880 $\pm$ 240
0.50	4131	8.33	18,900 $\pm$ 450
0.60	4503	9.21	20,610 $\pm$ 190
0.70	5093	10.80	23,300(a)

(a) Calculated from two points.

Further work is necessary to define the niobium-hydrogen system. Elevated-temperature X-ray studies will help to determine the extent of the two-phase region. Also, the lattice changes at the elevated temperatures will be observed. Further equilibrium studies at low pressures (>10 mm of mercury) will help establish the phase boundaries. However, with the portion of the equilibrium diagram established in this study, the kinetics of the reaction of hydrogen with niobium can be investigated. Accordingly, the rate of formation of solid solutions of known hydrogen compositions will be determined. The equation for the various isopleths shown in Figure I-3 will be used to define the experimental conditions. Rate experiments will be made at various equilibrium pressures to produce a constant composition over a temperature range.

CONFIDENTIAL

I-12

FIGURE I-3. REPRESENTATIVE ISOPLETHS FOR THE NIOBIUM-HYDROGEN SYSTEM

CONFIDENTIAL

03712281030

I-13

Migration of Hydrogen in Zirconium Hydride

J. B. Vetrano

It is well known that the capacity of zirconium for absorbing hydrogen by solution or compound formation is dependent upon the temperature and pressure of the experiment. The exact capacity under a given set of conditions may be determined from an inspection of the phase diagram, which is fairly well established. In general, zirconium has a larger capacity for hydrogen at lower temperatures and higher pressures. It naturally follows that if a hydrogen-containing bar of zirconium were subjected to a thermal gradient, there would be a shifting of the contained hydrogen until each portion of the bar had reached its saturation solubility at its particular environmental condition. A pile up of hydrogen would be expected in the cooler regions at the expense of hydrogen loss at the warmer regions. The mechanism or driving force for this hydrogen migration and the extent to which the migration would occur are not completely understood. The object of this program is to obtain information which will lead to a better understanding of this phenomenon, and eventually allow prediction of the hydrogen-concentration patterns in zirconium under any supposed thermal-gradient pattern.

The program to meet this objective has been planned to cover a 2-year span, and will encompass three separate phases. The first phase will involve the determination of the patterns of migration as a function of time, temperature gradient, and original composition. The second phase will consist of a determination of the diffusion coefficients of hydrogen in beta zirconium and in the delta hydride as a function of temperature, and the third phase will be concerned with finding a mathematical relationship between the diffusion coefficients and the final steady-state hydrogen-concentration pattern.

The migration patterns will be determined by subjecting clad bars of zirconium hydride to predetermined thermal gradients for a given time, quenching, and finally analyzing for hydrogen content as a function of position in the specimen. The 1/2-in. - diameter by 6 to 12-in. -long specimens will be prepared by hydriding hafnium-free zirconium to the desired initial concentration and homogenizing. The hydrogen used will be generated from the decomposition of uranium hydride. This gives a convenient and extremely pure source of hydrogen. The hydrided specimens will then be slipped into stainless steel tubes containing molybdenum liners, and the ends closed by welding on end caps. This entire assembly will be pressure bonded to achieve a metallurgical bond and to eliminate voids between the core and the cladding. The purpose of the molybdenum liner is to prevent loss of hydrogen through the cladding. These clad hydride specimens will then be subjected to linear thermal gradients in an evacuated furnace for a given time and then withdrawn and quenched. The extent of the hydrogen migration will be determined by sectioning the sample and analyzing the sections for hydrogen content.

Concurrent with this effort, the diffusion coefficients of hydrogen in beta zirconium and in the delta hydride will be determined as a function of temperature. The diffusion coefficient of hydrogen in beta zirconium can probably best be measured by a gradient technique. Using this method a cylindrical sample of zirconium will be hydrided just to the beta-phase and then homogenized. Additional hydrogen will then be

CONFIDENTIAL

DECLASSIFIED

I-14

introduced for a predetermined length of time to prepare a suitable hydrogen-concentration gradient, and then quenched to room temperature. The ends of the cylinder will then be machined off and discarded. The rest of the cylinder will be machined radially into layers of equal weight and these layers analyzed for hydrogen content. The thin outer layer of delta hydride which will probably be formed will be discarded, and the diffusion coefficient in the beta phase determined from the average hydrogen concentrations of each layer, the average radii of the layers, and the time of diffusion. It is possible that there is a composition as well as temperature dependence of the diffusion coefficient within the single beta-phase region, and although the concentration range in the beta phase is relatively small, this phenomenon may be detected. Therefore, it is planned to run some of the specimens at varying pressures in order to pick up any composition dependence.

The gradient technique is not applicable to the determination of the diffusion coefficient of hydrogen in delta-phase zirconium hydride since it is difficult to machine this material into layers for hydrogen analysis. A moving-boundary method would probably be best suited for this particular problem. Using this method, a zirconium specimen will be hydrided until it is at a composition corresponding to the point where the beta-delta-phase boundary line crosses the particular temperature of the experiment. It will then be homogenized, and additional hydrogen introduced for a controlled time. The movement of the phase boundary during this time will be determined by quenching the specimen and examining it metallographically. The diffusion coefficient at this particular temperature can be determined from a knowledge of the rate of movement of the boundary.

Once the pattern of migration has been established and the diffusion rate of hydrogen in zirconium hydride of varying composition is measured, an attempt will be made to interpret the measured migration patterns in terms of the predicted equilibrium concentrations and the varying isothermal diffusion rates.

J-1

J. CORROSION PROBLEMS ASSOCIATED WITH THE RECOVERY OF SPENT REACTOR FUEL ELEMENTS

C. L. Peterson, P. D. Miller, E. L. White, W. D. Beasley,
and R. S. Peoples

The study of corrosion problems associated with the various processes for the recovery of spent reactor fuel elements was continued in July.

Scouting experiments are under way in the liquid-phase Zircex evaluation program. In the cyclic tests for the vapor-phase Zircex process, both Haynes 25 and Type S-816 alloys continue to look promising after 35 cycles.

Titanium specimens are still giving encouraging results when exposed to Darex solutions.

Titanium is not seriously attacked by either Zircex or Darex synthetic chloride-stripper solutions if there is some nitrate ion remaining in these solutions.

Evaluation of materials of construction for the fluoride-volatility process will begin soon.

The Zircex Process

The volatility of zirconium tetrachloride is utilized in the Zircex process to separate uranium from zirconium cladding or alloys. Following hydrochlorination of the fuel elements, the uranium is dissolved in nitric acid, stripped of its chloride content, and extracted by means of the usual solvents. The hydrochlorination can be accomplished either in liquid or gaseous media.

Some investigations have just started to find a suitable material for containing a liquid-phase hydrochlorination process. The liquid phase, in this case, is the molten double salt, $\text{AlCl}_3 \cdot \text{NH}_4\text{Cl}$, which boils at about 425 C. Scouting experiments are being assembled in which metal specimens, together with a charge of the double salt, will be sealed in a partially evacuated glass capsule and held for several days at a temperature slightly below the boiling point of the salt. One experiment has already been conducted in a system in which gaseous HCl is passed continuously through the molten salt. Metal specimens will be exposed for short times in this apparatus (24 hr or so). The length of the test is presently limited by problems of carry-over of the vaporized salt causing plugging throughout the equipment.

The cyclic tests, which are designed to evaluate materials for a vapor-phase hydrochlorinator and include alternate exposure to liquid and gaseous media, are still under way. All of the materials started in the first series of tests have been eliminated except Type S-816 and Haynes 25. These two materials have been exposed for 35 cycles. Ten cycles have been completed with Haynes 23, 30, and 36. Both Type S-816 and Haynes 25 continue to look promising. Haynes 23 is definitely out of

CONFIDENTIAL

J-2

consideration, while Haynes 30 and 36 are borderline with indications that their corrosion rates are constantly increasing with further cycling.

In addition to the specimens that are being cycled between hot gases and boiling acid solutions, tensile specimens are being exposed to the hot-gas cycle only, to see if this atmosphere will have an adverse affect upon their physical properties. Comparison of the yield strength and ultimate strength of specimens of Type S-816 and of Inconel show little difference between specimens exposed to the hot hydrochlorinator gases and those exposed to inert atmospheres at the same temperature. Inconel is being examined as a possible material for construction of a two-vessel hydrochlorinator in the event that the acid dissolution must be carried out in another vessel. As yet, tensile specimens of Haynes 25 have not been evaluated. However, they are now being exposed and will be inspected within the next month.

Some titanium specimens, exposed in solutions simulating those from the chloride stripper in the Zircex process, were severely attacked after 16 weeks of exposure. Analysis of the solution remaining after the attack showed that the HCl which had been sweeping through the boiling solution had completely removed all the nitrate ion. As earlier work has shown, once the nitrate ion is depleted, titanium is quickly attacked in the vapors of boiling hydrochloric acid solutions. Exposure of the other tests with titanium specimens in this series was continued for 18 weeks with no sign of severe attack. Welded titanium specimens which were exposed are now being evaluated.

The Darex Process

In the Darex process, fuel elements containing stainless steel are dissolved by a dilute solution of nitric and hydrochloric acids, the chlorides are stripped by concentrated nitric acid, and the uranium is removed from the resulting nitric acid solution by solvent extraction.

A total of 2007 hr of exposure has been completed for specimens of various materials, including titanium, exposed in the presence of dissolving stainless steel. No serious attack has been observed. Operation of the batchwise dissolver has been curtailed for the present.

A repeat experiment, in which a titanium specimen is being exposed in boiling dilute aqua regia, while impressed with an external emf similar to that which it would attain during the dissolution of uranium, is now under way to determine how deleterious the scale which forms on the surface may be.

As in the case with the Zircex chloride-stripper tests, some of the titanium specimens suffered severe attack following 16 weeks of exposure in a synthetic Darex chloride-stripper solution. Again, analysis showed that all of the nitrate ion had been lost, and herein apparently lies the explanation of this attack. The other two tests were discontinued after 18 weeks with no severe attack being observed.

There is no indication of any stress-corrosion cracking or other severe attack of either stress or unstressed specimens of Type 347 stainless steel exposed to chloride-contaminated Purex acid systems at room temperature.

CONFIDENTIAL

0372291031

J-3 and J-4

Fluoride-Volatility Process

In the fluoride-volatility process, a stream of HF is passed through a molten bath of $\text{ZrF}_4\text{-NaF}$ containing zirconium-clad or alloyed fuel elements. The uranium tetrafluoride which forms is later volatilized away as the hexafluoride by passing fluorine through the melt.

Equipment is presently being assembled to evaluate possible materials for use as containers for the hydrofluorination step of this process.

K-1

K. EVALUATION OF A REFLECTOR-CONTROLLED
HETEROGENEOUS BOILING REACTOR

J. W. Chastain

This program is being carried out in order to evaluate the use of a reflector control scheme for long-time unmanned operation of a boiling reactor. The system will use the height of the water in an annular-reflector region as the means of control. This reflector will be connected to an external surge tank which is maintained at a fixed pressure. Pressure changes in the reactor vessel will cause changes in the reflector height; this in turn will tend to keep the reactor at the power required to maintain the design steam pressure. The program has two phases of concurrent effort. One phase is studying the hydraulic mock-up of the system piping. The other phase is studying the mock-up of the reactor (i.e., critical-assembly studies). These two phases together are aimed at the over-all evaluation of the stability and feasibility of the reflector control.

Critical-assembly experiments during the past months completed the hot-nonboiling mock-up. The first iteration to the nonhomogeneous voiding of the moderator plastic, which simulates the boiling power-producing-reactor case, was loaded into the core and experiments with this case were done. A second iteration is planned.

The hydraulic simulator studies continued with variations on the coupling pipes. A series of bends in this pipe was tried in an attempt to improve the stability characteristics. This proved unsuccessful. The effect on stability of continuous feed-water makeup to the system was studied. No instability was noted. Further changes are being investigated.

Critical Experiment

W. C. Ballowe, D. A. McPherson, R. G. Wilson,
F. J. Jankowski, D. A. Dingee, and J. W. Chastain

The results of this experiment will be an evaluation of the worth and effectiveness of the reflector system of control and will be further used as input data to the reactor simulator to evaluate transient behavior of the reactor system.

During the month of July, the hot-nonboiling reactor case (moderator, axial, and radial reflectors voided to have nuclear properties equivalent to water at 417 F and 300 psi) was completed. The final results of this experiment have not as yet been analyzed.

The first iteration on the nonhomogeneous-boiling case was preferentially loaded into the core in the radial direction to obtain some radial-void-coefficient information. This loading was done with the inner reflector at a 10-in. height. Power-distribution measurements, obtained from previous flux plots with the reflector at this height, were used to void the core. A flux plot was taken at the 10-in. position and the results of

K-2

this flux plot were compared to the corresponding flux plot used to calculate the void distribution. This comparison showed that the void distribution should be improved upon. Therefore, a second iteration was made and the calculated voids will soon be introduced into the core.

In addition to this work being done on the nonhomogeneous-boiling case, the voiding for the homogeneous-boiling case has begun and should be completed shortly.

Simulator Studies

B. B. Gordon and J. J. Stone

During July, the control-system mock-up was operated using four elbows inserted in the 2-in. pipe between the surge tank and the simulated reflector tank. The elbows increase the friction losses in the pipe. However, the installation of the elbows required two additional 6-in. lengths of pipe.

The adverse effect of increased inertia of the system due to the increased pipe length was greater than the damping effect of losses due to the elbows. This actually produced a slightly less stable system than a straight 2-in. pipe.

The system was checked at the same values of incremental reflector worth as in the previous runs and, in addition, at the higher incremental reflector worth corresponding to a new reactor with hot moderator and reflector. The system was unstable for this higher incremental moderator worth following a large positive step in power demand.

The effect of adding water to the system during control operation was studied. Water was added to the system at a rate of approximately 1/2 gpm with no observable disturbance of control.

During August, the system will be modified to produce damping with no increase in inertia.

L-1

L. STUDIES OF SODIUM-TANTALUM COMPATIBILITY AT ELEVATED TEMPERATURES

J. H. Stang

This section covers studies of the metallurgy of tantalum and of the tantalum-sodium combination at elevated temperatures. This research is in assistance to the development program of LAMPRE I, a fast reactor in which the plutonium-alloy fuel is contained in an all-tantalum core and is cooled by flowing sodium.

In the only sodium flow-loop experiment completed during July, tantalum specimens, which are to be used for subsequent mechanical-property determinations, were exposed for 33 days in 1200 F peak-temperature fluid; the oxygen level in the system was <10 ppm. The specimens lost an amount of weight equivalent to about 0.1 mil of metal removed from the surfaces per year. Their oxygen level was lowered as evidenced by an initial hardness of about 170 VHN and a final hardness of about 130 VHN.

In the creep-rupture experiments at 1200 F, tantalum specimens from annealed sintered material exhibited minimum creep rates of about 0.0001 and 0.00003 per cent per hr at 12,500 and 10,000 psi, respectively. Thermally degassed material from the same lot gave a minimum creep rate of 0.00005 per cent per hr at a stress of 6000 psi.

Studies concerning the effects of oxygen content on the weldability of tantalum performed during the month indicated that ingassing the metal with high amounts of oxygen (up to 560 ppm) results in weld deposits which are quite porous. This result is contrary to previous observations that higher oxygen levels have a beneficial effect on weldability. It seems probable that another element contributes to porosity but the information on hand is inadequate to identify it.

During July, emphasis in the research to develop the gettering method as an analytical tool for oxygen in sodium was placed on establishing the level of residual contamination in the test system. A "blank" run (without sodium) at 750 C, indicated that this base contamination is equivalent to about 10 per cent of the oxygen extracted from sodium which analyzed 30 ± 10 ppm of oxygen by the amalgamation method. With this blank considered, the oxygen in this sodium, according to the gettering results, averaged about 49 ppm, or approximately 50 per cent higher than indicated by the amalgamation technique.

Tantalum-Sodium Compatibility Studies

G. E. Raines, C. V. Weaver, and J. H. Stang

Tantalum specimens are being exposed to environments of high-temperature flowing sodium to obtain data pertinent to the corrosion situation. The experimental apparatus in this investigation consists of polythermal forced-convection flow loops constructed of Type 316 stainless steel. The specimens being exposed are suspended in the furnace zone of the loop systems, which operate with a peak sodium temperature of 1200 F and, normally, with a fluid-temperature cycle of 500 F. In each system, the sodium is

CONFIDENTIAL

DECLASSIFIED

L-2

purified either intermittently or continuously by cold trapping and by gettering with heated zirconium foil.

During July, an exposure of tantalum specimens (from arc-cast sheet) for posttest mechanical-property evaluations was completed. In this case, the sodium was gettered continuously during the 33-day duration of the run by maintaining a flow through the zirconium-foil trap, which was held at 1000 F. The oxygen level in this system was too low to be measured by a plugging-indicator technique, which is sensitive to about 10 ppm. It was found after the run that the hardness of all specimens was reduced from an initial value of 170 VHN to about 130 VHN. The weight losses ranged from 7.2 to 8.0 mg per specimen; this corresponds to a depth of penetration of approximately 0.1 mil per year, a rate comparable to those obtained for specimens exposed in Test 3, which was another system employing continuous gettering (information for Test 3 was presented in BMI-1189). Of the six specimens involved in the recent exposure, one will be examined metallographically and analyzed for carbon, hydrogen, nitrogen, and oxygen; as indicated above, the other five are scheduled for mechanical-property tests.

Three other long-run systems which are using conventional corrosion test specimens (cut from lengths of sintered tubing and arc-cast sheet) continued their operation during July. As described in BMI-1201, the sodium in one of these is at the 20-ppm oxygen level and, in the other two, is at the <10 ppm level. These, as well as a shorter run at the 80-ppm level about to start, are expected to terminate during August. Other plans for August include the start of an experiment in which welded mechanical-property specimens will be exposed to low-oxygen-level (<10 ppm) fluid.

High-Temperature Mechanical Properties of Tantalum

G. F. Huber, C. J. Slunder, J. G. Dunleavy,
and A. M. Hall

Creep testing at 1200 F on annealed (recrystallized) and thermally degassed tantalum sheet fabricated from sintered material continued during July. Table L-1 presents recent information from the experiments which have been in progress. The creep data listed for annealed material are in good agreement with those presented in BMI-1201 for higher loads (to 32,000 psi). In the case of thermally degassed material, informative data have been obtained at only two load levels, 8000 and 6000 psi; since the spread in minimum creep rate between these two is large (from 0.01 to 0.00005 per cent per hr), it was deemed necessary to make a run at 7000 psi.

A test on annealed sheet fabricated from arc-cast tantalum has been started at 15,000 psi. Analyses and hardness tests on this material after annealing in vacuum at 2650 for 8 min gave the following results:

Vickers Hardness Number (5-Kg Load)	Contamination, ppm			
	Oxygen	Hydrogen	Nitrogen	Carbon
185	214	>0.3	150	>10

Also, during August, it is planned to test the sodium-exposed specimens, discussed in the previous subsection.

L-3

TABLE L-1. CREEP STRESS-RUPTURE DATA FOR SINTERED TANTALUM SHEET TESTED AT 1200 F IN HELIUM

Stress, psi	Time in Progress, hr	Time, hr, to Reach Indicated Percentage Deformation		Total Deformation, per cent	Minimum Creep Rate, per cent per hr
		0.5	1.0		
		<u>Annealed Material^(a)</u>			
15,000	1011 ^(b)	14	46.5	2.2	0.00035 ^(c)
12,500	600	--	--	0.15	~ 0.0001
10,000	1120	--	--	0.065	~ 0.00003
<u>Thermally Degassed Material^(d)</u>					
7,000	Test just started				
6,000	1121 ^(b)	436	9000 (estimated)	0.552	0.00005

(a) Annealed in vacuum at 2650 F for 8 min; recrystallization occurred.

(b) Test discontinued at the time indicated.

(c) This test was covered in BMI-1201; however, the minimum creep rate was erroneously listed as 0.000035 per cent per hr.

(d) Heat treated in vacuum at 4500 to 4800 F for 5 hr; grain diameter over 1 mm.

Weldability of Tantalum for High-Temperature Systems

S. M. Silverstein, R. P. Sopher, and P. J. Rieppel

Previous welding studies with tantalum have indicated that some lots of material are consistently weldable while others are poor from the welding viewpoint, as evidenced by gross porosity in the weld deposit. The former has been the case with a lot of virgin arc-cast material used in the successful fabrication of several simulated tube-to-header assemblies. The latter has been observed with some heats which had a relatively low oxygen content. During July, studies were made to examine further the rôle of oxygen content on weldability.

In this recent investigation of this situation, an unweldable sample of reclaimed sintered tantalum containing about 35 ppm of oxygen was divided into three specimens; these were ingassed with 140, 280, and 560 ppm of oxygen, respectively. Bead-on-plate welds with the argon-gas-shielded tungsten-arc process were made under identical conditions on each of the specimens. Visual examinations of these welds indicated that the higher the oxygen content of the tantalum sheet, the greater the amount of porosity in the weld metal. This finding is contrary to those made in the early part of this research program. It is now believed that some other element combined with oxygen might be responsible for porosity in tantalum welds. However, from a comparison of the analyses of the different lots of material employed to date, the other element or elements cannot be identified.

L-4

Determination of Oxygen in Sodium

P. H. Goble, W. M. Albrecht, and M. W. Mallett

An investigation is being made of a method to determine oxygen in sodium by extracting the oxygen from a large quantity of the liquid metal with a small piece of a getter metal. The getter metal is then analyzed for increased oxygen content by vacuum-fusion techniques. During July, four gettering experiments were made in which 10-mil titanium sheet was heated in filtered sodium for 2 hr at 750 C. In these experiments the bomb was charged with 0.4 atm of argon, which is equivalent to about 1.5 atm at 750 C. A run in which no sodium was used was also made using a titanium getter. This was done to evaluate contamination from the walls of the bomb and from the impurities in the blanket gas. A summary of information for these experiments is given in Table L-2. The oxygen pickup by the titanium in the experiment without sodium was approximately 10 per cent of that picked up in the experiments with sodium. The difference between these values represents the oxygen picked up from the sodium.

TABLE L-2. SUMMARY OF ANALYSES FOR OXYGEN IN SODIUM USING TITANIUM AS THE GETTER AT 750 C FOR 2 HR

Run	Titanium Weight, g	Total Oxygen Pickup, w/o of titanium	Oxygen Pickup From Sodium, w/o of titanium	Sodium Weight, g	Calculated Oxygen Content of Sodium, ppm by weight
A	0.779	0.061	--	--	--
B	0.824	0.616	0.555	97	47.2
C	0.798	0.568	0.507	95	42.6
D	0.835	0.690	0.629	97	54.2
E	0.815	0.651	0.590	92	52.3
Average ^(a)					49.1

(a) The probable error is ± 3.3 ppm.

On the basis of these four experiments, the sodium analyzed 49.1 ppm oxygen with a probable error of ± 3.3 ppm. An analysis by the amalgamation method gave 30 ppm oxygen. Previous experience with the latter method indicates that with it a series of analyses has a probable error of about ± 10 ppm. Thus, the analytical results are higher by the gettering method than by the amalgamation method. The reason for this is not readily apparent. However, it should be pointed out that the indications to date are based on the results from only one blank experiment. It is planned to make another run of this kind to establish more definitely the blank level. Also, before the study is terminated, an attempt will be made to obtain some analytical data using sodium containing about 10 ppm oxygen.

M-1

M. DEVELOPMENTAL STUDIES FOR THE PWR

R. W. Dayton

Flow studies in the PWR model have been resumed. Studies of the effect of lower-plenum design on mixing are being made. The first design tested for Core 2 did not show better mixing than the previous design for Core 1.

Studies of the X-ray photometric fuel-element examination technique are awaiting the delivery of apparatus.

The collection of data on materials of construction for pressurized in-pile loops is nearly complete, and a topical report is being written.

A study has been started of the fabrication of Zircaloy 2-clad fuel plates containing UO_2 cores, made by pressure bonding. The initial work is directed at preparing samples for in-pile loop tests. Suitable techniques have not yet been found, and satisfactory samples have not yet been prepared.

Reactor Flow Studies

L. J. Flanigan and H. R. Hazard

To determine the effects of lower-plenum design on mixing and flow distribution in Core 2, air-flow studies in a quarter-scale model of the PWR are being conducted. Previously reported work includes construction and installation of a core extension and a new flow baffle, improvement of the gaseous-tracer technique for studying mixing, installation of additional static-pressure taps in the lower dome, and a preliminary core-distribution study.

In July, two four-loop core-flow-distribution tests were run. An analysis of the data showed that a number of control-orifice gaskets were leaking. All of these gaskets, which were more than a year old, were replaced with gaskets made of closed-cell sponge rubber. Three core-flow-distribution tests, at various flow rates, have been run to evaluate the leakage occurring with the new gaskets, but analyses of these data have not been completed.

Two mixing tests have been run. Both were run with three loops operating plus one simulated cold loop. In the first test, the distribution of SO_2 in the inlet was unsatisfactory, and considerable additional development of injection equipment was necessary. The second was run after refinement of the tracer-gas injection system, which reduced the variation in the measured concentrations in the inlet duct to $\pm 4\frac{1}{2}$ per cent of the average concentration. The mixing pattern obtained with the extended core arrangement is considerably different from that for Core 1, but mixing is no better.

CONFIDENTIAL

M-2

In August, any control-orifice gasket leakage found will be eliminated, a mixing study will be run with the core extension removed to increase the mixing space above the flow baffle, and an attempt will be made to improve mixing by making changes in the lower-plenum geometry.

X-Ray Photometric Examination of Fuel Elements

J. B. Schroeder and C. M. Schwartz

A nondestructive production-test unit is being developed to locate weld porosity and core segregation in reactor fuel elements. No work was planned during the month, while waiting for equipment to be delivered. An order has been placed for a fine-focus X-ray tube and power supply. Delivery is expected in September.

High-Strength Materials for Pressurized-Water
In-Pile Tubes

F. H. Lyon and H. J. Grover

The objectives of this program are to collect and evaluate pertinent physical-property information on materials suitable for pressurized-water in-pile tubes. At the completion of the work, an experimental program will be instituted, the final objectives of which are the construction and testing of a prototype tube.

The previous reports have listed the materials being considered under four general classifications: alloy steels, chromium steels, precipitation-hardening steels, and superalloys. During July, the available data on the materials in the four classifications have been supplemented by information obtained directly from producers of the materials. The question of availability of certain of the materials under consideration has also been resolved by these contacts. One of the most interesting results is the knowledge that Babcock & Wilcox is able to produce a coextruded tube. This process results in a tube with an exterior of one material and an inside surface of a different material.

Most of the information required for this phase of the program is now available with the exception of corrosion information on the specific materials and conditions desired. There are minor gaps in the physical-properties information, but the body of data permits elimination of certain of these materials. On this basis, a tentative selection of materials for work in Phase 2 has been made which includes Inconel X, AM-350, and Discaloy. In addition, the coextrusion process will probably receive some attention.

It is expected that work on Phase 1 of this program will be completed before the end of the next report period and a topical report is being prepared.

CONFIDENTIAL

0000000000

M-3

Pressure Bonding of Zircaloy 2-Clad Fuel Elements Containing
Compartmented Oxide Fuel Plates

S. J. Paprocki, E. S. Hodge, and D. C. Carmichael

A plate-type Zircaloy 2-clad fuel element containing compartmented oxide fuel is being considered for the PWR Core 2. A development program is being conducted to investigate the preparation of these elements by pressure bonding. This process utilizes a gas pressure at elevated temperatures to achieve bonding with a minimum amount of deformation and appears to be very promising for this application. The components are fabricated to final dimensions and assembled into a can which is then evacuated and sealed. The assembly is placed in a high-pressure autoclave and gas pressure at elevated temperatures is employed to bring the components into intimate contact for bonding.

The program involves two major parts, one being directed toward the development of the optimum conditions for pressure bonding and the other toward the preparation of irradiation specimens. The latter phase is considered very urgent, since the feasibility of the Core 2 fuel-element design must be established by irradiation tests to permit completion of conceptual design of Core 2. Therefore, preparation of the irradiation specimens has been started first and the best techniques available at this time are being used. A number of in-pile specimens have been produced and partial bonding has been achieved. These plates have been tested for leaks and corrosion tested in 750 F steam for 3 days. Failure of some specimens after corrosion testing has indicated that the bonding was discontinuous and efforts are now being directed toward the elimination of possible causes for the defects.

It is suspected that gas might be released from the oxide cores during pressure bonding, and could cause contamination of the Zircaloy 2 surfaces to be bonded. Cores are presently being outgassed at elevated temperatures prior to the pressure-bonding operation. In addition, the surface condition of the Zircaloy 2 used for the specimens appears to be a critical factor in determining the extent of bonding, since it has been indicated that Zircaloy 2 with a machined surface is more readily bonded than the cold-rolled, vapor-blasted, and pickled Zircaloy 2 being used for the PWR specimens. Specimens with abraded surfaces are being prepared for pressure bonding by wet polishing the mating surfaces of the Zircaloy 2 with emery paper and by wire brushing.

The extent of the reaction between the core and cladding during bonding will be observed and it will be determined whether or not it is necessary and feasible to prevent the reaction by coating the cores with a graphite coating.

The effect of using oxide cores rather than wrought cores is being investigated. Specimens with Zircaloy 2 cores are being prepared to compare with specimens containing oxide cores. Other pressure-bonding techniques which have been modified in an effort to eliminate possible detrimental effects are the substitution of stainless steel cans for Ti-Namel cans and machined Ti-Namel spacers for pickled Ti-Namel spacers. It is also planned to determine if there is an improvement in the bond when the bonding temperature is increased from the present temperature of 1550 to 1625 F.

CONFIDENTIAL

M-4

The other phase of the program will be the development of the optimum pressure-bonding process for the preparation of the Core 2-type fuel elements. Emphasis will be given to an investigation of the most practical methods for preparing Zircaloy 2 surfaces that can be successfully pressure bonded, pressure-bonding parameters, and methods for minimizing the reaction between the core and cladding. Part of this investigation has been initiated in connection with the preparation of irradiation specimens. Additional studies will be conducted to determine the allowable dimensional tolerances and the most satisfactory can and spacer materials necessary to satisfactorily bond a PWR-type element. The bond strength of the specimens prepared in these studies will be investigated by chisel, peel, bend, and intercompartmental pressure tests. Metallographic examination will be employed to determine bond integrity, dimensional control, and extent of core-cladding reaction. As a final proof test, the specimens will be corrosion tested in 750 F steam for periods of time from 3 to 60 days.

RWD:CRT/all

CONFIDENTIAL

03712291030