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PROGRESS RELATING TO CIVILIAN APPLICATIONS
DURING MAY, 1957

by

Russell W. Dayton
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3

TABLE OF CONTENTS

	<u>Page</u>
REPORTS RELATED TO CIVILIAN APPLICATIONS ISSUED DURING MAY, 1957	5
A. DEVELOPMENT OF MATERIALS FOR HANFORD REACTORS	7
Thermal Conductivity of Uranium	7
Mechanical Properties of Zirconium and Zircaloy 2	7
Zirconium Cladding	8
Extrusion Cladding of Round Bars	8
Preparation of Aluminum, Molybdenum, and Zirconium Single Crystals	10
B. DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS	11
Extrusion Cladding of Flat Plates	11
Study of the Liquidus in Aluminum-Uranium Alloys	12
Preparation of Aluminum-Uranium Alloys	12
Development of a Mechanical Extensometer	13
C. PLANT ASSISTANCE TO MCW	15
Investigation of Uranium Oxides	15
The Electrical Properties of Uranium Dioxide	17
The Corrosion Resistance of Selected Stainless Steels	18
D. PROCESSING OF FEED MATERIALS	21
Salt-Bath Development	21
Galling Problems in Compacting of Magnesium-Green Salt Blends	22
E. GENERAL FUEL-ELEMENT DEVELOPMENT	25
CERAMICS	25
Refractory Fuel Elements	25
Characterization of Sinterable Oxide Powders	27
METALLURGY	27
Uranium Alloys for High-Temperature Application	27
Preparation and Properties of Low-Melting Alloys	28
Compartmentalized Fuel Elements	28
CORROSION	29
Mechanism of Uranium Corrosion	29
F. STUDIES OF ZIRCONIUM-URANIUM ALLOYS	31
Heat Treatment and Mechanical Properties of Zirconium-Uranium Alloys	31
Corrosion of Zirconium-Uranium Alloys	31
Development of Corrosion-Resistant 70 w/o Uranium Alloys	33
Radiation Stability of Zirconium-Uranium Alloys	33
G. CORROSION STUDIES OF ZIRCONIUM	35
Corrosion Studies of Zirconium and Zirconium Alloys	35
H. REACTOR-MATERIALS DEVELOPMENT	37
Enameling of Special Metals	37
Oxidation-Resistant Niobium Alloys	37
Uranium-Titanium Alloy Studies	39
Vapor Deposition of Molybdenum Coatings on Stainless Steel Tubing	39

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

TABLE OF CONTENTS
(Continued)

	<u>Page</u>
I. PHYSICAL METALLURGY	43
PROPERTIES OF URANIUM COMPOUNDS	43
Preparation, Fabrication, and Physical Properties	43
Reaction With Gases	44
Heat-of-Formation Measurements	44
INVESTIGATION OF THE TENSILE TRANSITION IN ALPHA URANIUM	45
CONSTITUTION OF URANIUM ALLOYS	46
Uranium-Base Ternary Alloys	46
High-Temperature X-Ray Diffraction Examination of the Uranium-Carbon System	46
STUDY OF BONDING FUNDAMENTALS	46
NIOBIUM-GAS REACTIONS	47
J. CORROSION PROBLEMS ASSOCIATED WITH THE RECOVERY OF SPENT REACTOR FUEL ELEMENTS	49
The Zircex Process	49
The Darex Process	50
K. EVALUATION OF A REFLECTOR-CONTROLLED HETEROGENEOUS BOILING REACTOR	51
Critical Experiment	52
Simulator Studies	52
L. STUDIES OF SODIUM-TANTALUM COMPATIBILITY AT ELEVATED TEMPERATURES	55
Tantalum-Sodium Compatibility Studies	56
The High-Temperature Mechanical Properties of Tantalum	58
Weldability of Tantalum for High-Temperature Systems	58
Determination of Oxygen in Sodium	59
M. DEVELOPMENTAL STUDIES FOR THE PWR	61
Reactor Flow Studies	61
X-Ray Photometric Examination of Fuel Elements	61
High-Strength Materials for Pressurized-Water In-Pile Tubes	62

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5 and 6

REPORTS RELATED TO CIVILIAN APPLICATIONS
ISSUED DURING MAY, 1957

- BMI-1181 "Progress Relating to Civilian Applications During April, 1957", by Russell W. Dayton and Clyde R. Tipton, Jr.
- BMI-1183 "Study of Aluminum Alloys Containing up to 45 w/o Uranium", by Norman E. Daniel, Ellis L. Foster, John A. DeMastry, Arthur A. Bauer, and Ronald F. Dickerson

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A. DEVELOPMENT OF MATERIALS FOR HANFORD REACTORS

F. R. Shober

Equipment is being constructed to measure the thermal conductivity and electrical resistivity of irradiated uranium rods clad in Zircaloy 2, with NaK as the heat-transfer medium. Creep testing is continuing on Zircaloy 2 and Zircaloy 3A at elevated temperatures.

Electropolished uranium cores were successfully clad with zirconium by the hot-hydrostatic-pressing technique both at 1200 and at 1350 F. An exploratory program has been undertaken to obtain information relative to the extrusion cladding of solid cylindrical uranium bars with aluminum.

The strain-anneal technique is being used to prepare single crystals of aluminum, molybdenum, and zirconium for studies at HAPO. Special alloys have been melted and fabricated as part of a metallurgical assistance program.

Thermal Conductivity of Uranium

H. W. Deem and C. F. Lucks

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

Shop work has been completed on all parts of the apparatus and related equipment, except the lead-shield pouring form. Completion of this assembly is expected early in June.

Completed parts are being checked and fitted to facilitate final assembly when the shop work is finished. Assembly of the apparatus and preliminary testing should be completed by the end of June.

Mechanical Properties of Zirconium and Zircaloy 2

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

The differences observed in the elevated-temperature creep properties of various lots of Zircaloy 2 are being investigated. The creep strength of Zircaloy 3A and the burst strength of seamless Zircaloy 2 tubing are being determined at elevated temperatures.

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A-2

Several specimens were tested for periods greater than 2000 hr at 650 F to compare creep rates established in 1000 hr with rates determined from longer time tests. The results of these tests and others are shown in Table A-1. The creep rates established at longer times are very similar to those for shorter times. The total deformations, the major portion of which occurs during the primary stage of creep, are different. These differences have been attributed to material variations. Creep testing of Zircaloy 3A is continuing.

Welded zirconium and Zircaloy 2 tubes have been received and will be prepared for burst-strength tests over the temperature range from room temperature to 650 F.

Zirconium Cladding

F. R. Shober and R. F. Dickerson

Optimum conditions are being determined for cladding zirconium to nickel-plated and electropolished uranium by a hot-hydrostatic-pressing technique. Conditions producing the better bonds between nickel-plated uranium and zirconium cladding are a 10,000-psi pressure for 1/2 hr at 1350 F and a 10,000-psi pressure for 6 hr at 1200 F.

The quality of the bonds was determined by metallographic examination, a peel test, and water quenching. The metallographic examination showed the extent of diffusion, the peel test gave a relative indication of the adherence of the cladding to the core, and the water quenching tested the ductility and strength of the bond. The samples were water quenched from 1335 F after 20 min at temperature. The better bonds withstood four separate quenching tests before cracking was observed. The bond area was examined under a microscope after each quench.

Two electropolished cores were sealed in zirconium cans and pressurized at 10,000 psi for either 4 hr at 1350 F or 6 hr at 1200 F. Metallographic examination of these specimens showed a good metallurgical bond throughout the area examined. These are to be water quenched after heat treatment to further evaluate the bond. Shorter times at both 1350 and 1200 F will be investigated for two additional electropolished cores.

Preparations are being made to can and pressurize two nickel-plated and two electropolished cores in zirconium cans under the best conditions to obtain a sound bond. These bonded specimens will be sent to Hanford for additional testing and evaluation.

Extrusion Cladding of Round Bars

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

Preparations are being made for an exploratory program to obtain information relative to the extrusion cladding of solid, cylindrical uranium bars approximately 1.3 in. in diameter by 8 in. long. The extrusion tools for this work are being constructed and are expected to be completed in 2 weeks.

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

Lot	Specimen	Temper- ature, F	Stress, psi	Time of Test, hr	Deformation at Indicated Time, per cent										Load Off	Creep Rate ^(a) , per cent per hr
					Load On	50 Hr	100 Hr	500 Hr	1000 Hr	1500 Hr	2000 Hr	3000 Hr	Total			
Zircaloy 2																
1	--	450	21,000	1000	0.48	0.915	0.93	0.96	0.98	--	--	--	--	--	0.00040	
2	34-3	450	21,000	2517.4	0.59	2.45	2.465	2.49	2.49	2.495	2.495	--	2.495	2.37	<0.00001	
2	31-7	550	22,500	2161.5	2.65	4.60	4.62	4.64	4.65	4.658	4.658	--	4.658	3.679	<0.00001	
2	40-7	550	17,500	1000	0.50	1.13	1.15	1.337	1.65	--	--	--	--	--	0.00063	
1	--	650	9,000	3462	0.032	0.06	0.072	0.072	0.81	0.090	0.091	0.091	0.091	--	<0.00001	
2	3-2	650	9,000	3770.3	0.06	0.09	0.1	0.117	0.117	0.117	0.117	0.117	0.117	0.085	<0.00001	
1	--	650	12,500	1000	0.17	0.42	0.428	0.448	0.46	--	--	--	0.46	--	0.000024	
2	3-1	650	12,500	3350 ^(b)	0.15	0.23	0.245	0.282	0.335	0.342	0.350	0.380	0.385	--	0.00003 ^(c)	
1	--	650	17,500	1000	0.54	0.655	0.675	0.711	0.742	--	--	--	--	--	0.00006	
2	34-2	650	17,500	2489.8	1.385	1.54	1.55	1.594	1.63	1.65	1.686	--	1.705	--	0.000056 ^(c)	
2	34-4	650	17,500	1000	1.37	1.575	1.615	1.707	--	--	--	--	1.707	--	0.000047	
2	1-1	650	22,500	1082.1	6.0	6.96	7.00	7.35	7.92	--	--	--	8.02	7.90	0.0008	
Zircaloy 3A																
--	3-4	300	25,000	1156.0	0.25	2.80	3.37	5.05	6.05	--	--	--	6.35	6.07	0.0017	
--	3-3	475	18,500	1181.1	1.0	2.13	2.16	2.17	2.19	--	--	--	2.19	1.94	0.00002	

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

(b) Tests in progress.

(c) Based on creep deformation between 1000 and 2000 hr.

A-3

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A-4

Initially, an attempt will be made to clad a steel bar because of its greater high-temperature strength over that of uranium. Subsequently, uranium bars with attached steel end plugs will be used. It is anticipated that these preliminary tests will reveal any major problems which may be related to the cladding of fuel elements of this particular shape and size.

Preparation of Aluminum, Molybdenum,
and Zirconium Single Crystals

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

The preparation of single crystals of high-purity aluminum, molybdenum, and zirconium for irradiation studies has been undertaken. Several aluminum single crystals have been grown from thin sheet by the strain-anneal method and material to grow 0.50-in. -diameter rounds has been fabricated.

High-purity aluminum rod (99.99 w/o) has been procured and fabricated into standard 0.505-in. tensile bars. Samples have been submitted for chemical analysis. Single crystals will be grown in the gage section of the tensile bars by the strain-anneal process.

A modified Andrade furnace is being constructed for growing molybdenum single crystals. Molybdenum rods for this purpose have been fabricated. Furnace construction is partially completed.

Reference has been found to the growth of single crystals of alpha zirconium by the strain-anneal method. High-purity crystal-bar zirconium has been obtained and a chemical analysis of the bar stock performed. The growth of single crystals of alpha zirconium by the strain-anneal method will be attempted.

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B-1

B. DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS

R. J. Carlson

Extrusion-cladding experiments were designed to determine if the relationship of increasing extrusion pressure with time, observed when cladding plates 28 in. in length or less, would hold for longer lengths. Several steel cores 48 in. in length were clad. The pressure increased for the first 24 in. or so and then remained constant for the rest of the test. Drawings and specifications for some of the components of the right-angle extrusion-cladding assembly have been completed.

Alloys containing 12 to 50 w/o uranium are being prepared to check the position of the liquidus line of the aluminum-uranium phase diagram in this region. e

The occurrence of voids in full-scale castings of aluminum alloys of high uranium content has been noted. These voids have been attributed to turbulence during pouring and to the gross shrinkage associated with the large difference between the liquidus and solidus temperatures. Molds have been designed featuring various gating and feeding techniques.

Extrusion Cladding of Flat Plates

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

Efforts directed toward the improvement of the extrusion-cladding process were continued.

One objective is to develop a mandrel design that will minimize compression of the core slot and also produce a satisfactory cladding. A mandrel (Tip 5) was designed with a heavy center web which was intended to strengthen the tip at that point. However, the web was found to restrict the flow of aluminum to the center of the specimen so that the center rib was undersize. The contour and height of the center web will be gradually changed in an effort to improve the metal flow without reducing the support of the core slot appreciably. The results of tests with this mandrel after its first modification indicated that more metal must be removed from the center web.

In several tests, steel cores 4 ft long were clad, with the objective of determining differences in extrusion pressure when compared with the shorter lengths (28 in. or less) clad previously. In the past, a gradual increase in the extrusion pressure was observed as the end of the extrusion was approached. With the longer cores, the pressure also increases gradually, but only until about a 2-ft length is clad. Then the pressure remains constant while the remaining portion of the core is clad. This is an indication that, with the present equipment, it is necessary to extrude a length of about 2 ft before a thermal and pressure equilibrium is reached. Variations in the temperature of the core as it passes from the outside to the inside of the hot container may contribute to the initial gradual increases in pressure. Additional tests will be required to confirm this point.

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B-2

Drawings and specifications of some of the components of the right-angle extrusion-cladding assembly have been completed. These will be studied to determine if any modifications are required.

Study of the Liquidus in Aluminum-Uranium Alloys

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

An investigation to determine the liquidus line of the aluminum-uranium system between 16 and 50 w/o uranium is planned, employing thermal-analysis, differential-thermal-analysis, electrical-resistivity, and metallographic techniques. The effects of silicon and magnesium on the liquidus temperatures are also to be determined.

Alloys containing 12 to 50 w/o uranium are being prepared by induction-melting techniques. Additions of silicon up to 1 w/o will be made at each composition through use of alloys containing 50 w/o silicon and uranium and aluminum in the same proportions as the base alloys. Additions of magnesium up to 0.5 w/o will also be made to each aluminum-uranium alloy for study.

Preparation of Aluminum-Uranium Alloys

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

A program of research concerned with the development of melting and casting techniques for the production of sound, homogeneous aluminum-uranium extrusion billets containing up to 50 w/o uranium is in progress. The objective of this program is to determine the highest uranium content in an aluminum-uranium alloy that can be achieved practically and yield reproducibly sound, homogeneous cast ingots.

The occurrence of voids in full-scale castings of aluminum alloys of high uranium content has been noted. These voids have been attributed to turbulence during pouring and to the gross shrinkage associated with the large difference between the liquidus and solidus temperatures. Molds have been designed incorporating features that should give information on various gating and feeding techniques. The casting of full-size extrusion billets will begin as soon as these molds are available.

Of further and different interest is the effect of impurities on the casting properties of aluminum alloys of high uranium content. In this instance, there is interest because of the probability that increasing amounts of silicon and magnesium will be introduced into the alloy from circulating scrap.

The present program will be concerned with the effect of these impurities, in amounts up to 1 w/o, on the castability of the alloys.

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B-3 and B-4

Development of a Mechanical Extensometer

F. R. Shoher and R. F. Dickerson

An extensometer is to be developed to measure the circumferential strain of an expanding tubular specimen. It is necessary that the instrument combine a high sensitivity with a range sufficient to determine the transverse mechanical properties of uranium tubes.

Present work is concerned with designs to modify an existing extensometer to increase its reproducibility. Some consideration has been given to an extensometer which would use a linear variable differential transformer as a measuring unit.

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C-1

C. PLANT ASSISTANCE TO MCW

A. A. Bauer

The effect of processing variables on the subsequent reactivity of UO_2 made from UO_3 both before and after hydrating and leaching to remove the water-soluble nitrates is being studied. The reoxidation rate of UO_2 made from Type III UO_3 is increased by either hydration or leaching while the reoxidation rate of UO_2 made from amorphous UO_3 is decreased by hydration and generally increased by leaching the UO_3 . Leached Type III UO_3 samples when dried exhibited an amorphous X-ray pattern, whereas samples which were hydrated without leaching exhibited no change in structure. The reoxidation rate of UO_2 made from amorphous UO_3 was found to be dependent upon the prior history of the oxide.

Studies of the effect of nitrogen, added in the form of $\text{UN}_{1.5}$ powder, on the electrical properties of UO_2 have continued. The lattice constant of a series of UO_2 -base specimens showed no variation with nitrogen content. On the basis of conductivity data, it would appear that, above 0.5 mole per cent $\text{UN}_{1.5}$, the solubility of nitrogen in UO_2 is exceeded. Plans to study the effect of nitrogen added as uranyl nitrate are now being made.

Corrosion studies of Type 304 ELC stainless steel in chloride- and fluoride-contaminated nitric acid solutions have continued. Corrosion rates have shown no essential change after prolonged exposure in 5, 18, and 30 w/o nitric acid solutions. Specimens tested in 5 and 18 w/o nitric acid solutions containing 0.30 w/o chloride show no corrosion in the 18 w/o solution, but an increasing rate has been observed for the specimen exposed to the liquid 5 w/o solution. No severe attack has been observed at chloride concentrations of 0.10 w/o and lower in 45 w/o nitric acid solutions. At higher concentrations of chloride, severe attack has been observed on specimens exposed at the interface and vapor positions but not on specimens exposed in the liquid.

Analyses of fluoride-containing solutions employed in previous tests indicate no loss of fluoride during the test period.

Investigation of Uranium Oxides

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

Uranium oxides are being studied to determine how processing variables affect the reactivity of the dioxide in subsequent steps in the production of uranium. Previous reports have related reoxidation reactivity of UO_2 and hydration reactivity of UO_3 to the nitrogen content and structure of the UO_3 . Work in May has been concentrated on the effect of reducing the nitrogen content of several UO_3 preparations by leaching of the water-soluble nitrates upon the reoxidation reactivity of UO_2 .

Several samples of Type III and amorphous UO_3 were reduced to UO_2 before and after water leaching and after hydrating in such a way to minimize leaching of the UO_3 . Nitrogen contents of the UO_3 and average reoxidation rates of the UO_2 , after the first

TABLE C-1. COMPARISON OF REOXIDATION REACTIVITY OF UO_2 MADE FROM UO_3 BEFORE AND AFTER HYDRATING AND LEACHING

Initial UO_3		Reoxidation		Hydrated UO_3			Reoxidation		Washed UO_3			Reoxidation	
Denitration Temperature, C	Type	Nitrogen, w/o	Rate of UO_2 , 10^{-3} O/U per day	Hydrate ^(a) Phase	Dried ^(b) Phase	Nitrogen, w/o	Rate of UO_2 , 10^{-3} O/U per day		Washed ^(a) Phase	Dried ^(b) Phase	Nitrogen, w/o	Rate of UO_2 , 10^{-3} O/U per day	
200	Pot III	0.24	0.36	A	III	0.048	1.8		--	--	--	--	
250(c)	Pot III	0.111	0.23	A	III	0.030	2.3		--	--	--	--	
300	Pot III	0.104	0.18	A	III	0.038	0.8		G	Amorphous	0.005	3.1	
300(c)	Pot III	0.085	1.0	A	III	0.037	1.5						
350	Pot III	0.101	0.59						G	Amorphous	0.005	1.5	
350(c)	Pot III	0.103	0.85	A	III	0.041	1.3						
400	Pot III	0.103	0.26	A	III	0.037	1.8		G	Amorphous	0.005	1.7	
400(c)	Pot III	0.093	0.66	A	III	0.039	2.6						
450	Pot III	0.058	0.85	A	III	0.027	1.9		G	Amorphous	0.005	1.8	
300	Vacuum-dried amorphous	0.13	4.6	G	--	--	0.28		G	Amorphous	0.005	5.7	
350	Belt amorphous	0.24	3.9	G	--	--	0.61		G	Amorphous	0.005	5.1	
400	Vacuum-dried amorphous	0.01	7.2	G	Amorphous	--	0.56		G	Amorphous	0.005	4.0	
425	Belt amorphous	0.032	2.7	G	Amorphous	--	0.46		G	Amorphous	0.005	6.4	

C-2

(a) Hydrate Phase A gives the diffraction pattern of $\text{UO}_3 \cdot \text{H}_2\text{O}$ and Hydrate Phase G gives the diffraction pattern of $\text{UO}_3 \cdot 2\text{H}_2\text{O}$.(b) The hydrated and leached UO_3 was dried at 300 C before reduction to UO_2 .

(c) Prepared by pyrolysis of uranyl nitrate dihydrate rather than uranyl nitrate hexahydrate.

C-3

day of exposure to air at room temperature, were determined as presented in Table C-1. Phase changes in the UO_3 , as determined by X-ray diffraction, are included in Table C-1. Although hydrated samples which were dried at 300 C (without leaching) contained less nitrogen than the as-made UO_3 , leaching was seen to be a more effective method of removing nitrogen. The reoxidation rate of UO_2 made from Type III UO_3 increased by either hydration or leaching of the UO_3 , while the reoxidation rate of UO_2 made from amorphous UO_3 decreased by hydration and increased, except for one sample, by leaching the UO_3 . It is interesting to note that leached Type III UO_3 samples when dried gave an amorphous pattern, by X-ray diffraction, whereas hydration without leaching did not change the UO_3 structure, suggesting that the residual nitrogen content aids in the formation of crystalline Type III UO_3 . It is seen in the last column of Table C-1 that the reoxidation rate of UO_2 made from amorphous UO_3 samples may differ considerably, depending upon the prior history of the oxide. Thus, the reactivity of UO_2 depends not only upon structure and composition of the UO_3 , but also upon the pyrolysis method used in the initial UO_3 preparation.

The Electrical Properties of Uranium Dioxide

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

The study of the effects of nitrogen on the electrical properties of uranium oxides has been continued. Attempts to add nitrogen to the lattice of uranium dioxide were made by mixing powders of the oxide and $\text{UN}_{1.5}$. After hydrostatically pressing the mixtures, the resulting ingots were annealed at 200 C for an extended period of time to insure homogeneity. Somewhat lower room-temperature conductivities and higher activation energies were measured for these specimens.

Ignition and X-ray analyses were made on several of the nitrogen "doped" specimens in an attempt to clarify the results of the electrical measurements. The data collected from these measurements are listed in Table C-2.

TABLE C-2. IGNITION AND X-RAY ANALYSES OF NITROGEN-CONTAINING UO_2

Amount of $\text{UN}_{1.5}$ Added, mole per cent	Oxygen-to-Uranium Ratio Measured	Lattice Constant, A
0.20	2.055	5.468 ₃
0.50	2.052	5.468 ₆
3.0	2.036	5.468 ₃
5.0	2.013	5.466 ₄

An ignition analysis of the original, undoped material gave an oxygen-to-uranium ratio of 2.057. The oxygen-to-uranium ratios listed in Table C-2 were calculated on the assumption that the entire weight of the specimen was due to uranium dioxide. The difference between these values and that obtained for the original material indicates the presence of the added nitride.

The independence of the lattice constants on the measured oxygen-to-uranium ratios of these specimens would seem to indicate that nitrogen was not incorporated in the lattice of UO_2 even though it was present in the specimens. The lattice constants

listed in Table C-2 are consistent with an oxygen-rich structure of UO_2 . However, a close study of the powder diagrams failed to reveal the expected second phase of $\text{UN}_{1.5}$ in even the specimen containing 5 mole per cent $\text{UN}_{1.5}$. Since nitrogen would decrease the lattice constant of UO_2 ($a = 5.4704 \text{ \AA}$) in a manner similar to oxygen, and since a second phase was not found in these specimens, it might be tentatively concluded that at least part of the nitrogen did enter the lattice.

This conclusion is consistent with the measured electrical conductivity of the specimens. It will be recalled that the conductivity of the specimens decreased with nitrogen content up to about 0.5 mole per cent $\text{UN}_{1.5}$. The decrease in conductivity was larger than a simple dilution effect would produce. Above 0.5 mole per cent $\text{UN}_{1.5}$, the conductivity was relatively insensitive to the nitrogen content, indicating perhaps that the solubility limit of nitrogen in UO_2 was exceeded. In the range of solubility the differences in lattice constant may not be great enough to determine with precision.

It may be that these results are not more conclusive because of the method used in doping with nitrogen. It is conceivable that the anneal time was not sufficient to allow complete diffusion of nitrogen into the crystals and that, for the higher nitrogen contents, most of the nitride was distributed in the grain boundaries. It is now planned to add uranyl nitrate to the oxide and to decompose the nitrate during an anneal to effect the nitrogen doping.

The Corrosion Resistance of Selected Stainless Steels

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

Investigations were continued during May on the corrosion of Type 304 ELC stainless steel by chloride- and fluoride-contaminated nitric acid solutions such as might occur during the recovery of nitric acid from uranium processing plants.

The prolonged-exposure tests have now extended to 3000 hr in the case of the 5.0 and 4500 hr in the case of the 18.0 and 30.0 w/o nitric acid solutions. The corrosion rates have remained essentially constant during the last several 500-hr periods. The corrosion rate of the specimen exposed at the interface position in 30.0 w/o acid solution containing 0.30 w/o chloride appears to be leveling off to a value of 1 to 2 mils per month.

Additional tests in 5.0 and 18.0 w/o nitric acid solutions containing 0.30 w/o chloride have now run for 1000 hr. No corrosion has been observed with the specimens exposed in the 18.0 w/o solution but, in the case of the liquid specimen exposed in the 5.0 w/o solution, the corrosion rate is increasing with continued exposure and has reached 0.77 mil per month.

It has been shown by means of short-term tests that even small concentrations of fluoride (0.001 and 0.005 w/o) will promote corrosion of specimens exposed in 18.0 w/o nitric acid solutions boiling at atmospheric pressure. The measured corrosion rate has not exceeded 3 mils per month and practically no corrosion of the specimen exposed in the vapor phase has been observed.

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Analyses of some of the solutions remaining from previous 48-hr tests with fluoride contaminations of 0.01 w/o in 5.0 and 30.0 w/o acid solutions and 0.05 w/o contaminations in 18.0 w/o acid solutions have shown no loss of fluorides during the test periods. These solutions were arbitrarily selected from periods which gave the highest and lowest corrosion rates and no correlation was found between the corrosion rate and the amount of fluoride remaining in the solution.

Chloride additions to 45.0 w/o nitric acid solutions boiling at 250 mm of mercury absolute pressure are being studied. After completing five 48-hr exposure periods, no severe specimen attack has been observed at chloride concentrations of 0.10 w/o or lower. However, at higher concentrations (0.30 and 0.50 w/o) of chloride, severe attack has been observed during some of the exposure periods on the specimens which are exposed at the interface and vapor position, but not those exposed in the liquid. This may be related to the fact that some chloride is lost from solutions of these higher concentrations during the exposure period, thus lowering the chloride concentration in the liquid but increasing the concentration of chloride or oxidation products of chlorine in the vapor.

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D-1

D. PROCESSING OF FEED MATERIALS

E. L. Foster, Jr.

Studies being conducted for the National Lead Company of Ohio (NLO) are in feed-materials processing.

The detrimental effect of hydrogen in uranium-slug fuel material was the motivation for a study of salt-bath development. Two salt-bath systems, 50 w/o sodium chloride-50 w/o potassium chloride and 50 w/o sodium carbonate-50 w/o potassium chloride, were selected for study. Results from tests devised to differentiate between the effectiveness of these two baths indicated that uranium heat treated in the all-chloride bath contained less hydrogen than uranium heat treated in the chloride-carbonate bath. Further, weight loss of uranium and corrosive attack of the salt on the containers was slight. Comparisons are to be made of the hydrogen contents of uranium specimens treated in the all-chloride bath at Battelle and uranium specimens treated in the Li_2CO_3 - K_2CO_3 production salt bath at NLO.

Densification of bomb-reduction charges, by briquetting in a closed die, is currently limited by magnesium galling which occurs between the ram and the die wall, impeding repeated operation.

It has been concluded that if adequate lubrication is maintained, galling can be reduced. Punch clearance was found to play a significant role in the ejection operation, depending principally upon magnesium buildup. If compacting was done in a tapered section (10 min) of the die, slight galling did not impede the operation. However, occasionally magnesium buildup was great enough to cause the punch to stick at 1-mil clearance. The 1-mil clearance value was found to be satisfactory, as almost no powder buildup was noted on the punch or disk surfaces.

Salt-Bath Development

L. L. Lortscher, K. A. Sense,
and R. B. Filbert, Jr.

Hydrogen pickup by uranium rods heat treated in pilot-plant-scale beta heat-treating salt baths is being studied. Previous laboratory work showed that two salt mixes (50 w/o potassium chloride-50 w/o sodium chloride and 50 w/o potassium chloride-50 w/o sodium carbonate) resulted in lower hydrogen pickup than the beta heat-treating salt mix used by NLO at Fernald. The planned experimental program, which consists of operating three KCl - NaCl and two KCl - Na_2CO_3 salt baths containing different contaminating salt mixes for 3 weeks each, was completed, and the final hydrogen pickup and uranium-weight-loss data reported in BMI-1181. Operation of the KCl - NaCl salt bath containing 1.6 w/o Li_2CO_3 and 4.9 w/o K_2CO_3 is being continued through June, so that hydrogen-pickup data can be obtained during this period of increasing humidity. The extended program was planned so that a direct comparison could be obtained between the KCl - NaCl pilot-plant-scale salt bath at Battelle and the Li_2CO_3 - K_2CO_3 production salt bath at NLO.

The extended research program consists of heat treating two specimens in the KCl-NaCl pilot-plant-scale salt bath at Battelle and two specimens in the Li_2CO_3 - K_2CO_3 production salt bath at NLO each day. Six specimens are cut from an 18-in. section of a uranium rod that has been through the alpha heat treatment at NLO. The two additional specimens are analyzed for hydrogen without receiving a beta heat treatment so that the actual hydrogen pickup of the other four specimens can be determined. Ambient humidity is being recorded throughout the test and two salt-bath samples per week are analyzed for hydroxyl ion content. All hydrogen analyses are being conducted by NLO.

Galling Problems in Compacting of Magnesium-Green Salt Blends

S. J. Paprocki, R. J. Carlson, and E. G. Smith

The compacting of briquettes of uranium tetrafluoride (UF_4) and magnesium has been offered as a means of improving the efficiency of the bomb-reduction process. Small-scale and production-size experimental runs at FMPL have confirmed this premise. However, during compacting, a serious problem has become evident. Magnesium seizing occurs on the die-cavity surface and is so severe that only one compact can be pressed before punch movement is hindered. The severity is such that the desired mass production cannot be attained.

The elimination of this problem has been the object of a program of study which has proceeded along two lines: punch clearance and lubrication.

Pressing has been done in a cylindrical die with a 10-min taper in the area of final compacting. The portion of the program devoted to punch clearance indicates that the tapering of this section lessens the criticality of clearance in that seizing by magnesium in the compacting area is not aggravated by the continuing contact from punch movement during compact ejection. However, on occasion, the magnesium buildup has been great enough to cause the punch to stick.

The evaluation of lubricants has continued with emphasis being placed on the ability of the lubricants to prevent seizing. In the procedure followed, the die surfaces were wiped with a specific lubricant. After three compacts were made, the cycle was repeated till prohibitive seizing occurred. The results are presented in Table D-1.

The ability of lubricants to prevent seizing varied greatly. Mineral oil and Dag 217 were the most satisfactory. However, even these would not prevent magnesium buildup for more than five successive compacts. It should be noted that both mineral oil and Dag 217 possess a high viscosity and remain evident on the die surfaces for longer times.

The completion of the lubrication study concludes the planned program and a topical report containing the detailed results is being prepared.

D-3 and D-4

TABLE D-1. LUBRICANT PERFORMANCE IN MAGNESIUM-GREEN
SALT BLEND COMPACTING TESTS

(Each lubricant was applied after every third press)

Lubricant	Number of Compacts Pressed Before Seizing
Ceremul "C"	12
Molybdenum disulfide	5
Camphor and acetone	1
Stearic acid	6
Oleic acid	6
Paraffin and benzene	2
Salicylic acid	1
Benzoic acid	2
Oildag	6
Vermiculite	1
Mineral oil	18
Zinc stearate	5
Dag 217	27 ^(a)

(a) Slight seizing. An additional 51 compacts were pressed with no prohibitive buildup and compacting was discontinued.

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E-1

E. GENERAL FUEL-ELEMENT DEVELOPMENT

F. A. Rough

Research and development work on ceramic, metal-alloy, and cermet-type fuel elements, sponsored by the Reactor Division of the AEC, is reported in this section.

It is reported that the uranium-loss rate in UO_2 graphite fuel elements which were heated at 2000 F in flowing high-purity nitrogen was greatly reduced after a 48-hr heating period.

Because of a change in emphasis in the over-all alloy-development programs, the research on the alloys for high-temperature-reactor application and the low-melting castable alloys is being discontinued. Tensile data are reported for uranium alloys containing 7 to 15 w/o niobium.

Progress is reported on the successful fabrication of compartmentalized fuel plates which contain uniformly dispersed normal UC and UN in a matrix of stainless steel.

In the investigation of the mechanism of uranium corrosion it was shown by experimental evidence that the valence of uranium in oxide produced at 100 C is lower than that produced at 200 C.

CERAMICSRefractory Fuel Elements

G. B. Engle, M. J. Snyder, J. F. Quirk, and A. J. Roese

Nuclear fuel elements utilizing the refractory materials BeO and graphite as matrices for UO_2 are being investigated. Current studies are on the diffusion of uranium in densely sintered BeO and the evaluation of UO_2 -graphite elements for fission-product retention and uranium losses.

BeO

Five UO_2 -BeO diffusion couples, prepared by coating finely ground UO_2 onto half the length of the bore of a dense BeO tube, were heated in hydrogen for 48 hr at 2800 F. During the heating, portions of the UO_2 coating peeled off, indicating that the UO_2 did not adhere firmly enough to provide a reliable UO_2 element for the diffusion couple.

However, to gain experience in preparing specimens, a chord section was removed by grinding away a portion of the tube wall to produce a flat extending the length of the tube. The ground tubes, with a minimum wall thickness of about 0.021 in. at the

center of the flat, were placed in contact with alpha-sensitive photographic plates for 6 days. Examination of the plates showed no significant radiation tracks as compared with areas exposed to standards containing 1, 5, or 10 w/o UO_2 .

Since separation of the UO_2 film during heating tends to vitiate the experiment, efforts will be made to prepare reliable diffusion couples from the sintered BeO tubes.

Graphite

In evaluating UO_2 -graphite fuel elements for possible use in a nitrogen-cooled power reactor, experiments were continued on fission-product retention and uranium losses.

In studies to determine uranium losses from a UO_2 -graphite element when heated in a flowing nitrogen atmosphere, the heating period was extended to 93 hr on the sample from the run in April. The sample was heated at 2000 F in flowing high-purity nitrogen. The additional data along with the data obtained previously are shown in Table E-1.

TABLE E-1. URANIUM LOSS FROM A UO_2 -GRAPHITE ELEMENT
HEATED IN FLOWING NITROGEN AT 2000 F

Period, hr	Uranium Loss Rate, $\mu\text{g}/(\text{cm}^2)(\text{hr})$
0-28	0.53
28-48	0.57
48-68	0.034
68-93	0.020

Note: Samples consisted of a 30-mil-diameter UO_2 rod imbedded in a cylindrical graphite matrix by coextrusion with the graphite mix. The element was heated previously at 2300 F for 4 hr to carbonize the binder.

The uranium-loss rate was relatively high during the first 48 hr, but was sharply reduced during the last 45 hr.

The sample will be heated for an additional period to determine whether the saturation effect observed after 48 hr continues beyond 93 hr. The sample then will be dissected and a cross-sectional area examined. The surface also will be examined by X-ray diffraction.

A new lot of UO_2 -graphite elements is being fabricated with about 2.7 w/o of 200 to 300- μ UO_2 nodules. The elements consist of nodules dispersed in a graphite matrix core, coextruded with another UO_2 -free graphite mix. These elements will be examined for fission-product retention and uranium loss.

Previous data on the amount of fission gas evolved from UO_2 -graphite elements heated at 2000 and 2200 F were doubtful because of the possible loss of xenon due to distortion and devitrification of the quartz capsules during heating. To determine the permeability of quartz to xenon, 20 quartz capsules were filled to 0.1 atm with xenon.

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E-3

These capsules will be checked after heating at 2000 and 2200 F for loss of xenon due to diffusion through the capsule walls.

Characterization of Sinterable Oxide Powders

J. F. Quirk and N. Mosley

Research to determine the basic properties affecting the sinterability of BeO is being continued, with further efforts being directed to define the effect of minor impurities. Emphasis in May was on the preparation of six BeO samples to be used in an experiment aimed at correlating impurity content and sintering behavior. Some sintering trials have been made on this group of samples and analytical work is under way, but no significant results are available as yet.

METALLURGY

Uranium Alloys for High-Temperature Application

W. E. Murr, A. A. Bauer, and R. F. Dickerson

Elevated-temperature properties of certain binary uranium alloys are being obtained as part of an evaluation of the materials for use as power-reactor fuels. Uranium alloys containing 7, 10, and 15 w/o niobium appear to offer promise as fuels in the operating-temperature range of 1200 F.

Table E-2 lists tensile properties of uranium-7, 10, and 15 w/o niobium alloys at 550 and 1000 F. The alloys were transformed at 1110 F prior to testing. The tensile strength of all three compositions is approximately equal at 550 F, but at 1000 F niobium additions produce increased elevated-temperature strengths. Increased niobium content is also shown to benefit the yield strength of all three compositions at both testing temperatures.

TABLE E-2. TENSILE PROPERTIES OF URANIUM-NIOBIUM ALLOYS

Nominal Niobium Content (Balance Uranium), w/o	Testing Temperature, F	Tensile Strength, psi	Yield Strength, psi	Elongation, per cent
7(a)	550	114,500	86,000	8.1
7(a)	1000	14,100	6,800	46.2
10(a)	550	113,100	107,000	6.0
10(a)	1000	17,800	9,600	49.6
15(b)	550	113,500	110,000	6.4
15(b)	1000	20,500	14,200	41.5

(a) Heat treated for 5 hr at 1110 F and air cooled.

(b) Heat treated for 24 hr at 1110 F and air cooled.

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A second phase of this program has been concerned with the irradiation and subsequent inspection of uranium alloys containing 10 and 20 w/o niobium, or 15 w/o zirconium. However, a decision has been made to terminate this program at the present time, and capsules awaiting loading into MTR have been recalled and will be retained for possible future loading.

A topical report is in progress which will summarize the development work of this program. Included in the report will be elevated-temperature mechanical properties of alloys of uranium-3.5 through 12 w/o molybdenum and uranium-3 through 20 w/o zirconium, and the physical and mechanical properties of uranium-7 through 15 w/o niobium alloys.

Preparation and Properties of Low-Melting Alloys

C. K. Franklin, A. W. Hare, W. E. Murr, A. A. Bauer,
and R. F. Dickerson

The program concerned with the experimental development and evaluation of a low-melting castable alloy for possible use as a power-reactor fuel has been discontinued because of a change in emphasis in the over-all alloy program.

This program had been concerned with the study of a uranium-5 w/o chromium-1 w/o molybdenum alloy. As-cast pins of this alloy were prepared and a number of pre-irradiation tests were conducted to determine the physical properties of the alloy and how these properties varied with temperature. Irradiation capsules containing test specimens of this alloy were also prepared. These have been retained for future use in the event that interest in this alloy is revived.

A topical report summarizing the preparation of the irradiation-test specimens and noting the results of the physical-properties studies is being prepared.

Compartmentalized Fuel Elements

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

Preparation of irradiation specimens containing 27 volume per cent UN or UC dispersed in stainless steel and clad with Type 347 stainless is in progress. This phase of the program includes preparation and encapsulation of specimens of each composition which are to be irradiated at 1500 F to uranium-235 burnups of 5, 10, 15, and 20 a/o.

UN powder has been prepared by the nitriding of fully enriched uranium metal. Fully enriched UC has been prepared by arc-melting techniques and is being crushed to the correct particle size in an argon-filled dry box.

Additional dummy specimens have been prepared using natural UN and UC. Pack designs have been completed which will give final specimens with a 1 by 1/2 by 0.050-in. core.

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Detailed metallographic examinations of the first dummy packs have been completed. The UC and UN appear to be uniformly dispersed; however, there are some defects on the core-to-cladding edge bonds. Additional dummy specimens are now being roll clad in a new pack design which should correct this deficiency.

CORROSION

Mechanism of Uranium Corrosion

J. B. Schroeder, 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 corrosion-resistant alloys for reactor fuels.

Additional measurements were made to determine the accuracy of the mean-valence analysis. Two samples of $\text{UO}_{2.03}$ were used as standards. The valence determined for the standards was 3.96 and 3.97, which indicates that the measured valence is probably accurate to at least ± 0.09 . Two samples of oxide produced at 100 C were then run. These samples were made in a container which allowed the oxide to fall into a cool (25 C) zone as soon as it became detached from the metal. The valence of these samples was 3.14 and 3.15. These values are considerably lower than those reported in BMI-1181 (3.69 and 3.72) for samples which remained in the hot zone. This indicates that oxidation continues after the oxides have left the metal and supports the hypothesis that Type II or III uranium is formed as an intermediate phase.

The same procedure was also used to determine the mean valence of oxide produced at 200 C in the wet-argon system. The volume of two fractions was found to be 4.23 and 4.40. A thermal analysis of the same sample showed the valence to be 4.22, while the valence of a different sample was determined to be 4.40. The results thus show that, as might be expected, the valence of uranium in oxide produced at 100 C is lower than that produced at 200 C. An X-ray diffraction pattern of the oxide produced at 200 C showed that the sample was largely UO_2 with a very small crystallite size.

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F-1

F. STUDIES OF ZIRCONIUM-URANIUM ALLOYS

F. A. Rough

Research on the fundamental properties, corrosion resistance, and radiation resistance of zirconium-uranium alloys is reported in this section.

Interpretation of the TTT curves and the determination of the mechanical properties of zirconium-uranium alloys with various heat treatments are being completed. The corrosion behavior of alloys containing 7 through 70 w/o uranium in water at 500, 550, 600, 640, and 680 F has been determined and a topical report is being prepared.

Data are reported on the effects of heat treatment and ternary additions to the corrosion resistance of zirconium-70 w/o uranium alloy.

Completion of the study of the effects of radiation on zirconium-22 and 50 w/o uranium alloys has been delayed. Initial work on the zirconium-22 w/o uranium alloy has been completed, and long-term irradiations are now being initiated on the zirconium-50 w/o uranium alloy.

Heat Treatment and Mechanical Properties of Zirconium-Uranium Alloys

D. L. Douglass, L. L. Marsh, and G. K. Manning

Beta decomposition of zirconium-uranium alloys containing 0.094 and 0.29 w/o oxygen is being studied by interrupted quenching and the determination of TTT curves.

Heat treatments have been completed. Interpretation of the microstructure will be made as soon as metallographic preparation has been completed. TTT curves will be constructed for each alloy on the basis of an analysis of the microstructures.

The room-temperature mechanical properties of zirconium-uranium alloys with up to 70 w/o uranium after various heat treatments are being determined. These data are presently being analyzed with respect to microstructure. These results are also being combined with those in the literature for zirconium-uranium alloys, and a composite picture on the effect of microstructure on mechanical properties should evolve.

Corrosion of Zirconium-Uranium AlloysW. Chubb, W. E. Berry, R. S. Peoples,
and R. F. Dickerson

One phase of the program on the properties of zirconium-uranium alloys is the study of their corrosion behavior in high-temperature water. Alloys being evaluated consist of 7 through 70 w/o uranium additions to crystal-bar, sponge, and high-oxygen-sponge zirconium.

Corrosion tests of bare samples in water at 500, 550, 600, 640, and 680 F have been concluded. A topical report is being prepared on the corrosion behavior of bare alloys in high-temperature water.

A program is under way to determine the effect of heat treatment on the corrosion behavior of Zircaloy 2-clad zirconium-uranium alloys.

Three zirconium-uranium alloys made from sponge zirconium and clad with Zircaloy 2 have been heat treated in six different ways and submitted for corrosion testing in 680 F water. Packs containing zirconium-22 and 50 w/o uranium cores were de-fected by shearing to expose the core; packs containing 70 w/o uranium cores were de-fected by drilling a 0.040-in hole through the cladding. Cross sections of the fuel elements containing 22 or 50 w/o uranium cores are being exposed to follow the progress of corrosion in the core and in the alloy diffusion layers at the core-cladding inter-face. Fuel elements with zirconium-70 w/o uranium cores contain a 0.040-in. - diameter defect drilled halfway through the core. Samples were given the following postcladding heat treatments:

- (1) None (as roll-bonded at 800 C)
- (2) 800 C, 1/2 hr, water quench
- (3) 400 C, 10 min, air cool
- (4) 650 C, 1 hr, air cool
- (5) 900 C, 1 hr, air cool
- (6) 800 C, 1/2 hr, slow furnace cool.

Samples have been exposed 24 days in 680 F water. Cracking at the core-cladding interface has occurred on samples containing 22 w/o uranium cores which were water quenched from 800 C or were aged 10 min at 400 C after roll bonding. Cracking occurred at the bond line and in the core of a water-quenched 50 w/o uranium sample. The cladding on a duplicate sample bulged from attack at the bond line. A 50 w/o uranium sample which was air cooled from 900 C also exhibited bulging from attack at the bond line.

Of the samples containing 70 w/o uranium cores, only the as-roll-bonded sample is unattacked after 24 days of exposure. A blister approximately 1/2 in. in diameter has formed at the defect on the sample which was air cooled from 650 C. The water-quenched sample was completely oxidized and the cladding ruptured within 3 days of exposure. The sample which was slow cooled from 800 C was removed from test after 9 days of exposure because of general oxidation of the core outward from the defect. The samples which were aged 10 min at 400 C or were air cooled from 900 C failed from oxidation of the core at faulty Zircaloy 2-to-Zircaloy 2 bond lines at the edges of the samples.

Microsections will be made on the failed samples to ascertain the progress of attack. Tests will be continued on the remaining samples.

Metallographic examination of the core-cladding interface in control samples has disclosed no contamination at the interface and no lack of bonding that can be attributed to the roll-cladding operation. In no case did a sample show incomplete cladding on both sides of the core. In five out of eighteen control samples, incomplete cladding was observed on one side of the core. This intermittent or occasional lack of bonding may be

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related to pipes, seams, and other types of defects in the core alloys. Further examination of the control samples will be dependent upon the results of the corrosion tests reported above.

Development of Corrosion-Resistant 70 w/o Uranium Alloys

V. W. Storhok, A. A. Bauer, W. E. Berry, and R. S. Peoples

The investigation directed toward improving the corrosion resistance of the zirconium-70 w/o uranium alloy is continuing. The effect of heat treatment of the base alloy and the effect of ternary additions of molybdenum, niobium, silicon, and titanium to the base alloy are being studied for this purpose. The ternary additions were made with the objective of either tying up the free alpha uranium found in the 70 a/o alloy or of stabilizing the gamma phase.

Zirconium-uranium-niobium alloys have been prepared and are being corrosion tested bare in 680 F water. However, metallographic examination showed structures normally not associated with these alloys and heat treatments. A chemical analysis is being run to determine whether an error was made in the preparation of charges for melting. Meanwhile, additional alloys containing 3 and 5 w/o niobium are being prepared. Corrosion tests of the other bare ternary alloys have been completed and the results reported (BMI-1161 and BMI-1173).

Binary and similar ternary alloys are being clad with Zircaloy 2 and will be corrosion tested using pinhole defects to study clad-alloy corrosion resistance.

Radiation Stability of Zirconium-Uranium Alloys

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

Research on the effects of radiation on zirconium-22 and 50 w/o uranium alloys has continued. The chemical burnup determinations which were begun in April on irradiated specimens of the zirconium-22 w/o uranium alloy have not been completed to date. A metallographic examination of three specimens of the zirconium-22 w/o uranium alloy is planned. However, because of the large backlog of metallographic work at the hot-cell facility, the investigation of these specimens has not been initiated.

The temperature-controlled capsule which contains two specimens of zirconium-50 w/o uranium alloy of two different heat treatments is at the MTR awaiting available reactor test space. Under the present schedule, the capsule is listed for insertion into the reactor on June 24, 1957.

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G. CORROSION STUDIES OF ZIRCONIUM

R. S. Peoples

Long-term corrosion studies pertaining to the development of corrosion-resistant zirconium and zirconium-base cladding alloys have been continued in elevated-temperature water and steam.

Major effort is directed toward a study of Zircaloy 2 and modified Zircaloy 2 compositions.

Corrosion Studies of Zirconium and Zirconium Alloys

W. K. Boyd and R. S. Peoples

Corrosion data for intentionally contaminated Zircaloy 2 based on approximately 2 years of exposure in 680 F degassed water may be summarized as follows:

- (1) In the range 500 to 1500 ppm, hydrogen does not affect the corrosion properties of Zircaloy 2.
- (2) Oxygen contents in excess of 6000 ppm destroy corrosion resistance in 680 F water.
- (3) A complete loss of corrosion resistance is experienced with nitrogen contents of 200 ppm or higher.

In other studies, the samples of seven different melts of Zircaloy 2 continue to exhibit excellent corrosion resistance after more than 3-1/2 years of exposure to 600 F degassed water. Specimens evince a black film, no indication of breakaway, and corrosion rates slightly in excess of $5 \text{ mg}/(\text{dm}^2)(\text{month})$.

Zirconium Committee Program

Extended-exposure data are being obtained in 680 F degassed water and 750 F steam for modified Zircaloy 2 compositions, i. e., 0.25 to 0.50 w/o tin plus 0.03 to 0.50 w/o iron and/or nickel, with and without additions of nitrogen and aluminum.

After up to 616 days of exposure, weight gains are of the same order as those tabulated in BMI-1176. Data continue to indicate that:

- (1) A slow cool from 775 C or water quenching from 900 C does not significantly affect corrosion resistance of tin-iron or tin-nickel alloys.
- (2) As the nitrogen content is increased from 70 to 300 ppm, or if the aluminum level exceeds 200 ppm, corrosion resistance of the alloys is markedly decreased.

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- (3) Sponge-base alloys containing 0.2 to 0.5 w/o tin plus 0.03 to 0.5 w/o nickel are somewhat more resistant to 750 F steam than are similar tin-iron alloys. However, for 680 F water exposures, the reverse is true.

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H-1

H. REACTOR-MATERIALS DEVELOPMENT

F. A. Rough

Reactor-materials studies supported by the AEC Reactor Development Division are reported in this section along with one program of assistance to Argonne National Laboratory.

No new data are available on the enameling of uranium.

Additional oxidation tests of niobium-vanadium and niobium-molybdenum alloys have been carried out at 1000 C. The results are parabolic in most cases.

Tensile data are presented for uranium-titanium alloys containing up to 6 a/o titanium and having various heat treatments. A correlation is being made of the microstructures of the alloys and characteristics of the various treatments with mechanical properties.

Thin coatings of molybdenum have been produced on the inside of small-bore stainless steel tubes by vapor deposition; but attempts to build up thicker coatings met with experimental difficulties. Preliminary experiments have also been made to deposit niobium on these small tubes.

Enameling of Special Metals

B. W. King, M. C. Brockway, and E. D. Dietz

No experimental work was done on the enameling of special metals in May.

Oxidation-Resistant Niobium Alloys

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

The oxidation behavior of binary niobium-base alloys shown earlier by intermittent-weighing air-oxidation tests to have better oxidation resistance than pure niobium are being investigated by more sensitive continuous-weighing air-oxidation tests. These tests, which are performed on 50-mil annealed strip samples, have confirmed the improved oxidation resistance of niobium binary alloys with molybdenum, titanium, and vanadium, and have shown that many of the more promising alloys oxidize according to the parabolic time law, while pure niobium oxidizes linearly.

Oxidation tests at 1000 C have been completed on niobium-molybdenum and niobium-vanadium alloy samples shown in Table H-1. The behavior of niobium-vanadium alloys in dry air is almost identical with the behavior in wet air reported in

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TABLE H-1. OXIDATION DATA FOR BINARY NIOBIUM ALLOYS TESTED IN AIR AT 1000 C

Sample	Composition, a/o	Parabolic Rate Constant, (mg/cm ²) ² /hr	Transition to Linear Rate, hr	Linear Rate Constant, mg/(cm ²)(hr)	Weight Gain in Indicated Time, mg per cm ²					
					1 Hr	2 Hr	3 Hr	4 Hr	5 Hr	6 Hr
<u>Reaction of Niobium Alloys With Dry Air</u>										
O-30(a)	100Nb	--	0.1	30.0	30.0	61.3	89.8	114.2	136.9	158.6
O-51	Nb-5V	147.3	--	--	11.0	16.4	20.5	23.7	26.6	29.2
O-43(a)	Nb-7.5V	97.7	--	--	9.5	14.0	17.2	19.8	22.2	24.2
O-47	Nb-10V	63.4	--	--	7.5	10.8	13.4	15.6	17.5	19.3
O-49	Nb-12.5V	42.8	--	--	7.3	9.9	11.9	13.4	14.8	15.9
O-53	Nb-2.5Mo	220.0	--	--	12.6	19.3	24.2	28.4	32.3	36.0
O-45	Nb-5Mo	133.0	--	--	10.0	15.1	18.7	22.0	25.1	28.2
O-55	Nb-7.5Mo	39.3	--	--	7.2	9.7	11.5	13.0	14.5	15.6
O-57	Nb-10Mo	61.2	0.6	3.95	8.1	12.1	16.0	19.7	23.3	27.1
<u>Reaction of Niobium Alloys With Wet Air</u>										
O-54	Nb-2.5Mo	226.8	--	--	14.0	20.3	25.2	29.5	33.5	37.1
O-46(a)	Nb-5Mo	133.0	--	--	8.8	14.2	18.2	21.7	24.8	27.8
O-56	Nb-7.5Mo	43.0	--	--	7.6	10.2	12.2	13.9	15.3	16.6

(a) Data from previous test included for comparison.

H-3

BMI-1181. Alloys containing 5 to 12.5 a/o vanadium oxidize entirely according to the parabolic time law, the rate decreasing with increasing vanadium content. It is also planned to prepare and study niobium alloys with 15, 17.5, and 25 a/o vanadium. The latter composition has been hot rolled to strip and will be evaluated first; it is expected that evolution of gaseous V_2O_5 will be a major factor in the oxidation rate.

Niobium-molybdenum alloys also oxidize parabolically, and show a decrease in rate with increasing molybdenum content similar to the niobium-vanadium alloys. No significant difference between dry- and wet-air oxidation could be detected. A transition from parabolic to linear oxidation behavior was noted at 10 a/o molybdenum. The linear rate was about one-tenth the rate for pure niobium. The reason for the transition from parabolic to linear is not immediately apparent, but may be associated with the formation of MoO_3 . At low alloy concentrations, both vanadium and molybdenum are believed to dissolve substitutionally in the Nb_2O_5 scale.

Future work on this project will include:

- (1) Continuation of continuous-weighing air-oxidation tests on binary niobium alloys containing chromium, molybdenum, tungsten, vanadium, and zirconium.
- (2) Comparison of the oxidation behavior of metallographically polished pure niobium with previous results obtained on niobium abraded with silicon carbide metallographic paper.

Uranium-Titanium Alloy Studies

D. L. Douglass, L. L. Marsh, Jr., and G. K. Manning

The phase equilibria of uranium-titanium alloys on the uranium side of the compound U_2Ti are being investigated. The mechanical properties of alloys martensitically transformed from the high-temperature solid solution have been determined. A correlation between properties and microstructure will be made.

Results of tensile tests are listed in Table H-2.

All alloys showed a very low proportional limit as compared to the yield strength (0.2 per cent offset). Specimens of all tested tensile bars are being prepared for metallographic examination.

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 on the inside and outside of small-diameter Type 304 stainless steel tubing for ANL is continuing.

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TABLE H-2. TENSILE DATA FOR MARTENSITIC URANIUM-TITANIUM ALLOYS

Titanium Content (Balance Uranium), a/o	Heat Treatment	Tensile Strength, psi	0.2 Per Cent Offset Yield Strength, psi	Elongation, per cent	Reduction in Area, per cent
0.59	Quenched from 800 C	191,000	93,000	3.8	3.6
1.14	Ditto	198,000	128,000	1.7	3.2
2.10	Ditto	112,000	55,000	7.5	9.1
2.87	Ditto	137,000	57,000	11.5	10.5
3.67	Ditto	159,000	71,000	3.2	4.0
4.50	Ditto	196,000	133,000	1.0	2.5
5.75	Ditto	174,000	98,000	1.0	1.1
0.59	Quenched from 800 C, tempered 1 hr at 300 C	179,000	114,000	1.5	0.78
1.14	Ditto	171,000	122,000	1.0	1.2
2.10	Ditto	111,000	50,500	6.5	8.6
2.87	Ditto	136,000	60,000	9.5	10.8
3.67	Ditto	169,000	76,500	3.5	3.9
4.50	Ditto	213,000	121,000	4.5	5.5
5.75	Ditto	170,500	100,000	1.0	0.92
0.59	Quenched from 800 C, tempered 1 hr at 400 C	173,000	129,500	0.5	5.2
1.14	Ditto	178,000	168,000	--	5.2
2.10	Ditto	128,000	56,000	10.5	10.6
2.87	Ditto	140,000	65,000	9.0	8.7
3.67	Ditto	165,000	81,000	3.0	3.8
4.50	Ditto	200,000	147,000	0.5	0.65
5.75	Ditto	167,000	112,000	1.0	0.65
0.59	Quenched from 800 C, tempered 1 hr at 500 C	171,000	156,000	1.0	1.2
1.14	Ditto	167,000	166,000	0.5	0.9
2.10	Ditto	147,000	56,500	19.5	19.2
2.87	Ditto	153,000	61,300	11.0	11.4
3.67	Ditto	139,000	84,000	0.5	0.9
4.50	Ditto	156,000	146,000	0.5	0.4
5.75	Ditto	166,000	116,000	1.0	1.3

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H-5 and H-6

In prior work, adherent, uniform coatings were applied to the outside of 20-in. - lengths of the small-bore tubing, but uniform coatings could not be obtained over the inside surface of tubing of this length. A coater operating on a new principle for coating the inside of small-bore tubing was devised, built, and tested.

Work during May was devoted to continued operation of the new coating unit, to microscopic examination of some of the coatings produced, and to completion and testing of a moving-temperature-gradient coating unit.

The principle of operation of the new coating unit was described in BMI-1181. Microscopic examination of some of the coatings obtained with this unit during work in April showed that they were uniform from one end of the tube to another, both inside and out, but that they were less than 0.1 mil thick. Continued attempts to build up heavy coatings with long plating periods resulted only in plugging of the coating tube with lower molybdenum chlorides. Proper operation of the unit apparently depends upon critical adjustment of the gas-flow rates.

Deposition of niobium on the inside and outside of a 20-in. -length of the small-bore tubing was tried in the newly completed moving-temperature-gradient coating unit. In this unit, part of the specimen is heated uniformly to deposition temperature, while the remainder is heated to a temperature only slightly above the temperature of the halide salt vaporizer. The transition zone between the two temperature regions is moved slowly from one end of the tube to the other, so that a uniform coating is obtained throughout the tubing. The first run in this unit, in depositing niobium, was not entirely successful, since part of the tapped resistor winding on the specimen-heater section burned out before the end of the run. A thin niobium coating was obtained on either end of the specimen, both inside and out.

Work on depositing niobium on the Type 304 stainless tubing specimens with both types of coating units will be continued.

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I-1

I. PHYSICAL METALLURGY

F. A. Rough

Research in physical metallurgy and ceramics sponsored by the Research Division of the AEC is reported in this section.

The preparation of UBe_{13} samples has begun for measurement of physical properties. Reaction of the uranium silicides with oxygen or nitrogen is reported. Generally, the silicides react slower with nitrogen than with oxygen.

Study of the tensile transition in alpha uranium is continuing.

In work on the constitution of uranium alloys, study of uranium-molybdenum-zirconium alloys is continuing, but the investigation of the uranium-carbon system by high-temperature X-ray diffraction is complete.

Additional data are reported for the bonding of gold needle points and plane gold surfaces at 700 and 800 C. Attempts to establish a correlation between electrical-resistance measurements and bond area in these tests indicate that resistance does not distinguish between areas of contact and areas of metallurgical bond.

In the study of niobium-hydrogen reactions, data have been obtained confirming earlier equilibrium data. Reaction-rate and diffusion experiments are also in progress.

PROPERTIES OF URANIUM COMPOUNDSPreparation, Fabrication, and Physical Properties

W. D. McKee and M. J. Snyder

UBe_{13} was prepared by induction melting a stoichiometric mixture of UH_3 and beryllium powder under an argon atmosphere. The melted sample had an apparent specific gravity of 4.69, compared with a theoretical (X-ray) crystal density of 4.37 per cm^3 . Further evaluation by metallographic and X-ray diffraction examination is under way. Preliminary results from these examinations indicate the sample will be satisfactory for physical-property measurements.

In sintering a powder prepared from an arc melt of the composition UAl_3 , it was found that UAl_3 dissociates markedly at 1250 C in a vacuum of 8×10^{-5} mm of mercury, losing aluminum. In view of this and its relatively low uranium content, no further study of this compound is planned.

Attempts to tin the silicides for thermal-conductivity measurements have been unsuccessful to date. This work will be continued.

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Reaction With Gases

W. M. Albrecht and M. W. Mallett

The reactions of several common gases with certain uranium compounds are being studied.

Additional reaction-rate data were obtained between sintered compacts of U_3Si_2 , USi_2 , and USi_3 and nitrogen. The reactions followed the parabolic law at 450, 400, and 700 C, respectively.

The relative reactivities of the various uranium silicides with oxygen and nitrogen are summarized in Table I-1. It is seen in Table I-1 that, with the exception of U_3Si_2 , the reactivity of the silicides with both gases increased with the uranium content of the compounds. U_3Si_2 appears to react slower than either USi or USi_2 .

TABLE I-1. COMPARISON OF WEIGHT-GAIN DATA FOR THE REACTION OF URANIUM COMPOUNDS WITH OXYGEN AND NITROGEN

Compound	Weight Gain After 20 Min in Indicated Atmosphere, μg per cm^2	
	Oxygen at 300 C	Nitrogen at 500 C
USi_3	66	182
U_3Si_2	70	330
USi_2	146 ^(a)	500
USi	1270	2550
UAl_2	(b)	--

(a) Weight gain at 250 C.

(b) Burned nonisothermally.

Although most of the uranium silicides burned nonisothermally in oxygen at 400 C or lower, weight-gain data could be easily obtained for the nitrogen reactions at 500 C, indicating that the reaction of the silicides with oxygen is faster than with nitrogen at the same temperatures. The UAl_2 burned nonisothermally in oxygen at 300 C.

Heat-of-Formation Measurements

J. W. Droege, A. W. Lemmon, Jr., and R. B. Filbert, Jr.

Heats of formation of refractory uranium compounds are being determined as part of the program on properties of these materials. The work has so far been confined to the development of methods for the measurement of the heat of combustion of USi_2 . The heat of formation of USi_2 may be found by subtracting its heat of combustion from that of its constituent elements.

One method has involved the use of an oxygen combustion bomb. Thus far, it has been possible to obtain only about 80 per cent combustion in this bomb and it has not

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I-3

been possible to determine the extent of combustion accurately. A technique involving grinding of the fused combustion product and igniting at 900 C for 4 days was insufficient for completing the oxidation. One expedient for obtaining complete combustion has met with some success. In one experiment, NH_4Cl was mixed with the sample in an effort to limit the reaction temperature to a level below the fusion point of silica. Oxidation appeared to be nearly complete. The NH_4Cl or its oxidation products corroded the walls of the bomb, however. Since the use of NH_4Cl would necessitate extensive measures for protecting the bomb from attack and for determining the extent of oxidation of the NH_4Cl , this method will not be used unless no other can be found.

Experiments with the peroxide fusion bomb have been directed toward obtaining complete reaction of the USi_2 sample with the $\text{Na}_2\text{O}_2\text{-KClO}_4$ fusion mixture. The splashing of sample onto the cold walls of the fusion cup was greatly reduced by placing a small nickel disk on top of the fusion mixture. This helps to keep the reaction confined to the lower part of the cup. By increasing the ratio of sample to peroxide, a higher temperature and more complete reaction has been obtained. About 5 per cent of the sample remains unreacted, however. Because of the difficulties in determining the exact amount of unreacted sample, further efforts will be made to obtain complete reaction.

Samples of UC, UN, and UAl_2 are at hand. While work continues on USi_2 , some combustions will be carried out on these other materials, with which less difficulty is to be expected.

INVESTIGATION OF THE TENSILE TRANSITION IN ALPHA URANIUM

L. L. Marsh and G. K. Manning

The flow and fracture behavior of uranium is being investigated over the temperature interval defining the tensile transition. The variables being examined are grain size and strain rate. In addition, the metallography of plastic deformation as influenced by these variables is also being examined.

It has been observed that increasing the grain size and the strain rate shifts the transition curve to higher temperatures. The effect of both these variables on the modes of plastic deformation indicates that with larger grain sizes and/or greater strain rates the amount of deformation by twinning increases at the sacrifice of slip. These observations are presently being interpreted as being the result of impurity and grain-size interaction on the slip process. The progressive inhibition of slip with decreasing temperature results in the initiation of twinning as the major deformation process.

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I-4

CONSTITUTION OF URANIUM ALLOYSUranium-Base Ternary Alloys

M. S. Farkas, A. A. Bauer, and R. F. Dickerson

Determination of the delta-phase relationships of the uranium-zirconium-molybdenum ternary system is progressing with study of specimens that lie on a line joining uranium-31.5 a/o molybdenum and uranium-74 a/o zirconium.

Results of X-ray diffraction and metallographic examination of a group of specimens show that the solid solubility of zirconium in the delta uranium-molybdenum phase is very limited.

High-temperature X-ray diffraction investigations are now being performed to clarify the nature of structures found by room-temperature X-ray methods and to more accurately define phase boundaries. Indications are that transitional phases are being developed as the result of incomplete transformation.

Determination of the phase relationships of the uranium-rich corner of the uranium-zirconium-silicon system is continuing by metallographic and X-ray diffraction methods. Metallographic examination coupled with differential-thermal-analysis results show that the uranium-silicon compounds are stable over a large region of the area being studied. The solubility of silicon in the uranium-zirconium-base alloys was found to be less than 2 a/o.

High-Temperature X-Ray Diffraction Examination
of the Uranium-Carbon System

W. B. Wilson, A. E. Austin, and C. M. Schwartz

Experimental work has been essentially concluded, and the results are being summarized for publication.

STUDY OF BONDING FUNDAMENTALS

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

Experiments have been continued for determining the mechanism for the formation and growth of metallic bonds in the solid state. A modified single-asperity-type bonding experiment has been adopted for this work in which junctions are established between gold needle points and plane gold surfaces under various experimental conditions. Rupture strengths of the junctions are then measured. In May experiments were conducted at 700 and 800 C.

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I-5

A 100-mg initial load was applied and immediately removed; contact then was maintained at zero load for periods up to 60 min. Average test results are shown below, together with data obtained previously at 600 C.

Temperature, C	Rupture Load ^(a) , g, After Indicated Time, min						
	0	2	5	10	20	40	60
600	1.2	--	--	2.0	--	3.4	4.0
700	4.3	6.4	7.7	10.5	11.1	14.0	14.9
800	4.0	7.0	9.75	12.8	18.0	22.8	25.0

(a) Load values have been corrected to room temperature.

Logarithmic plots of rupture load versus time yield straight-line relationships. That is, the slope $\frac{d \ln L}{d \ln t}$ is constant for any given temperature. The slopes for 600, 700, and 800 C are 0.27, 0.28, and 0.38, respectively. The slope of 0.38 at 800 C would be expected, if volume diffusion is the process by which growth of the bond area proceeds. The lower slope values of 0.27 and 0.28 are not as applicable to the volume-diffusion mechanism.

Electrical-resistance measurements of contact areas showed that the bond area is a power function of time. That is, contact area is proportional to t^n , where n is about 0.1. The strength data above indicate that bond strengths are also proportional to t^n , where n lies in the range 0.27 to 0.38 for tests between 600 and 800 C. The change in contact area with time as measured electrically does not parallel the change in bond strength with time. It appears that electrical-resistance measurements cannot distinguish between the mechanical-contact area and the metallurgically bonded area. This idea is supported by the results of modified hot-hardness measurements conducted on gold. A 1/16-in.-diameter steel-ball indenter and a 500-g load were used in these tests. Loads were maintained up to 100 min. Measurements were made of the projected impression areas and plotted against time on a logarithmic scale. A linear relation resulted with a slope in the range 0.1 to 0.17. These measurements of rate-of-area change are very similar to results of electrical-resistance measurements, in which the logarithmic slope was about 0.1. This agreement between experimental methods shows that electrical-resistance measurements probably indicate only the mechanical-contact area, but not the metallurgical-bond area.

Experiments will be continued in the present direction. In addition, a topical report will be prepared to summarize the results of the past year's work.

NIOBIUM-GAS REACTIONS

W. M. Albrecht, and A. Levy

A fundamental investigation to determine the kinetics and the mechanism of the reaction of hydrogen with niobium is being made.

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I-6

A determination of the temperature-pressure-composition equilibrium diagram of the niobium-hydrogen system was continued. Additional data have been obtained at intermediate temperatures, 350, 450, and 550 C, in the 0.1 to 0.6 hydrogen-to-niobium atomic ratio composition range. These data are in good agreement with the equilibria data previously reported.

Reaction-rate experiments have been made to produce a solid solution having a concentration of hydrogen-to-niobium of 0.05. Rates were determined in the range 500 to 800 C. The reaction initially followed the linear rate law at all temperatures. Further rate data are being obtained.

An experiment to determine the diffusion of hydrogen in niobium at 600 C has been completed. The analysis of the hydrogen concentration gradient prepared in a cylindrical specimen shows that the time (about 10 min) used to prepare the gradient was too long. However, an approximate diffusion coefficient of 2×10^{-5} cm² per sec was calculated from the data. Because it is not experimentally feasible to prepare diffusion gradients in less than about 10 min, future experiments will be made at temperatures lower than 600 C.

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J-1

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

C. L. Peterson, P. D. Miller, E. L. White, E. B. Friedl,
O. M. Stewart, W. D. Beasley, and R. S. Peoples

Investigations of corrosion problems associated with the recovery of spent reactor fuel elements have been continued.

Cyclic hydrochlorination tests simulating the Zircex process are in progress. Studies have been under way related to an understanding of the mechanism of the corrosion of titanium in HNO_3 -HCl systems.

Darex process dissolution studies are showing favorable results for titanium, Type S-816, and Haynes 21.

The Zircex Process

In the Zircex process for reclaiming spent fuel elements, hydrochlorination is used to volatilize the zirconium cladding or alloy. Dissolution of the residue, followed by solvent extraction, is used to recover the uranium.

During May, the investigations of candidate materials of construction for the hydrochlorination and chloride-stripping processes have been continued.

The cyclic tests are now in progress. In these tests, specimens are alternately exposed to the condition of hydrochlorination at 600 C and then to the possible boiling nitric acid solutions which might be used for dissolution. By the end of May, ten cycles had been completed. Each cycle consists of approximately 11 hr at 600 C, followed by 2 hr in the boiling solutions. The corrosion of the specimens was measured following the second, fifth, and tenth cycles. Only Haynes 25, Type S-816, and Type S-590 are performing well under all dissolution conditions. Haynes 21, which did so well in the elimination tests, is more heavily attacked by the hydrochlorination part of the cycle than the three materials above. Carpenter 20, Type 304ELC stainless steel, Illium R, and Inconel are showing poor resistance.

Over 14 weeks of exposure has been completed for specimens exposed in the 5.0 M HNO_3 , 500-ppm chlorine elimination test. So far, the corrosion rates have remained essentially constant at fairly low values and have shown no sudden increases.

It has been shown previously that whenever a titanium specimen failed in the 15 M HNO_3 -HCl Zircex elimination tests, no nitrate could be detected in the liquid. This has been further substantiated by tests in azeotropic HCl containing small nitrate additions. Here, the presence of 0.1 M HNO_3 inhibited attack.

A series of experiments is under way to find which of the possible ions or decomposition products are necessary to inhibit corrosion in the liquid and in the vapor phases

J-2

of titanium specimens exposed to boiling azeotropic HCl. So far, it has been shown that N_2O and NO afford no protection in either place. Chlorine protects the specimens in the vapor but increases the corrosion in the liquid. Nitrogen dioxide affords protection in both places, providing the most inhibition in the vapor phase. The tests with NOCl have been inconclusive and will be run again. Uranium ions in solution provide protection in the liquid phase.

Titanium specimens are still fairly resistant to simulated Zircex chloride-stripper solutions after 12 weeks of exposure. Welded specimens show no adverse effects after 5 weeks.

The Darex Process

In the Darex process, a dilute solution of nitric and hydrochloric acids (dilute aqua regia) is used to dissolve fuel elements containing stainless steel. Following dissolution, the chlorides are stripped from the solution with concentrated nitric acid and the uranium is then recovered by solvent extraction.

The exposure of the specimens in the presence of dissolving stainless steel has been continued. A total of 1453 hr of exposure in the continuous dissolver and 1205 hr in the batchwise dissolver has been completed. To date, no serious attack is occurring.

The tantalum specimen which has been exposed in boiling dilute aqua regia while impressed with an external emf has shown a decided embrittlement, but no great increase in hydrogen content. The titanium specimen from a similar experiment shows metal loss in the form of scaling, but no embrittlement or excessive hydrogen pickup.

Titanium specimens have been exposed for 12 weeks to simulated chloride-stripper solutions. No corrosion of consequence has been observed. This is true also of the welded specimens of titanium, which also present a crevice for testing. These have been exposed for 5 weeks.

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K-1

K. EVALUATION OF A REFLECTOR-CONTROLLED HETEROGENEOUS BOILING REACTOR

J. W. Chastain

This work is directed toward evaluating a reflector method of control for a boiling-water heterogeneous reactor. The control system utilizes the height of the water reflector in an annular tank surrounding the reactor core as a means of controlling the reactor. The annular reflector tank is intimately connected with the core and also to an external surge tank so that the reflector height is changed by variations in the steam pressure in the core vessel. The change in reflector height, in turn, causes the reactor to operate at a new stable power level, this new level being determined by power-output demand. This control method may permit long-time, unmanned operation of boiling reactors for military applications.

An analysis of such a system, using an electronic reactor simulator, indicated the method was feasible for transient control and may even provide sufficient control to take care of burnup and poison override. To further investigate the feasibility of this control scheme, two experiments were devised. The first of these is a full-scale mock-up of the hydraulic system, driven by the reactor simulator. The second experiment is a critical-assembly mock-up of the core and reflector of a typical boiling-water heterogeneous reactor. This report deals only with the critical-assembly work.

The critical assembly is being used to measure the reactivity worth of the reflector for various moderator and reflector conditions. These conditions include a "cold"-water case, a hot-nonboiling case, and the boiling power-producing case. The information obtained will be used as input data for the hydraulic mock-up to determine the dynamics of the system.

In the critical experiment the water of the moderator and reflector was simulated by using polyethylene. The polyethylene may be voided by drilling holes or making saw cuts to duplicate higher temperature water or the voids produced in boiling. This system is safer in operation than a voided-water system since the voids in the plastic cannot collapse accidentally as could those in the water if the heaters or air bubbles failed.

During May, results on the reactivity worth of the reflector measured for a hot-nonboiling case were used in the reactor-simulator studies. Operation was stable, although some oscillations occurred when the maximum slope on the reflector-worth curve was used.

The experiments on the hot-nonboiling case were run during May and should be completed in June. Calculations to determine the void distribution for the first boiling case were carried out. Experiments on this case should be started during June.

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K-2

Critical Experiment

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

The experimental results obtained in this experiment will be used to evaluate the reflector system of control so that the feasibility of such a system in practice may be determined. Further use of the results of the critical experiments, as input data to the reactor simulator for the determination of the long- and short-term transient behavior of the system, will be made.

The main effort during the month of May was directed toward the experimental setup for the hot-nonboiling case. The experiments have begun and should be completed early in June.

Certain compromises in the radial-reflector design had to be made because of time considerations. Since sufficient polyethylene blocks voided to simulate water at 417 F and 300 psi could not be received in a reasonable length of time, it was decided to build only the inner radial reflector and the inner 2 in. of the 6-in. outer radial reflector from voided polyethylene blocks. The remainder of the outer radial reflector was constructed from normal solid polyethylene. This reduced the amount of plastic needed and insured that the experiments could be continued without delay. It is not believed that the 4 in. of solid polyethylene used in place of the voided material originally intended for the outer reflector will substantially affect the results.

In addition to the work done on the hot-nonboiling case, some work was done on setting up for the power-producing boiling case. Calculations were made to give the void distribution in the core for an assumed power of 1.5 megawatts. The results of these calculations are shown in Figure K-1. The flux plots made in the "semihot" case (plastic voided to represent hot water in the moderator and axial reflectors) were used to obtain a first iteration to the boiling power distribution. Polyethylene strips are now being voided nonhomogeneously for the moderator and upper axial-reflector region (equivalent to the header region in the power reactor) in accordance with these calculations.

It is expected that the boiling power-producing experiments with a nonhomogeneous void distribution will be started near the latter part of June.

Simulator Studies

B. B. Gordon and J. J. Stone

During May the information received from the critical-assembly experiment concerning reflector worth was incorporated into the control-system simulation. The greatest slope on the reflector-worth curve occurred at the center of the curve and would correspond to the operating point of a new reactor. The minimum slope occurred at the extreme upper end and corresponds to the operating point of an old reactor. The system was subjected to disturbances at each of these operating points. A low value of

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K-3

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FIGURE K-1. CALCULATED VOID CONTOURS

Zero inlet subcooling, 1.5-megawatt power,
300-psi pressure, and 10-in. reflector position.

Note: The flux distribution used to calculate this
case was obtained with a voided moderator
and axial reflector but solid radial reflector.

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K-4

damping was used for these tests. The system was stable at both operating points. Some undesirable oscillations did occur at the operating point corresponding to a new reactor. It is anticipated that more damping will eliminate these oscillations.

During the next period the same test will be made using a greater amount of damping. The tests will also be carried out using a 1-in. section of pipe between the simulated reflector tank and the surge tank.

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L-1

L. STUDIES OF SODIUM-TANTALUM COMPATIBILITY AT ELEVATED TEMPERATURES

J. H. Stang

LAMPRE I is a fast reactor being developed at Los Alamos Scientific Laboratory. The fuel alloy in this reactor will be contained in a small all-tantalum tube-to-shell heat exchanger. Sodium will flow at high velocities through the tubes and serve as the coolant. Peak sodium temperatures will be in the neighborhood of 1200 F. Research is being conducted to develop information pertinent to this situation wherein tantalum is exposed to high-temperature sodium. Metallurgical programs are also in progress to evaluate the high-temperature mechanical properties of tantalum and its weldability.

During May, two polythermal flow-loop experiments were completed in the investigation of the effect of high-temperature (a peak of 1200 F) sodium environments on tantalum. The weight losses of tantalum specimens exposed for 8, 11, and 15 days in sodium which contained two levels of oxygen, < 10 and 80 ppm, were quite small (equivalent to about 1 mil of metal removed per year). Metallographic examinations of exposed specimens have revealed that the minor attack which occurs is localized at the surface rather than being intergranular.

A considerable fund of data is now on hand from 1200 F creep tests of annealed tantalum from sintered material. However, this tantalum has an oxygen content (about 200 ppm) higher than that which will be present in the core of LAMPRE I after a certain period of operation owing to the strong gettering action of the fuel mixture. Consequently, future creep testing will concentrate on the evaluation of low-oxygen material. One test along this line already completed indicated that thermally degassed (and, consequently, large-grained) material is considerably weaker than the high-oxygen sintered material.

Welding studies during May were devoted to making small tantalum tube-to-header (3/16 in. in OD by 15-mil wall tubing and 20- and 30-mil sheet) assemblies. While no difficulty was encountered in welding the tubes to the thin header sheets, the final welding operation, which consists of welding the headers to an outer cylindrical can (also fabricated of thin sheet), proved to be troublesome. The factors involved, however, are minor and can probably be eliminated by the use of a holding jig now under construction.

Results of one run made during May on the gettering technique development for the determination of oxygen in sodium were very encouraging. Using titanium as the getter at 650 C, the indicated oxygen content of a charge of test sodium was 32 ppm; using the amalgamation method, the level was 40 ± 10 ppm. In future runs, higher gettering temperatures will be used to improve the gettering efficiency. A few modifications of the apparatus are now being made to permit higher temperature operation.

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L-2

Tantalum-Sodium Compatibility Studies

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

The behavior of tantalum exposed to 1200 F peak-temperature sodium is being investigated using forced-convection flow-loop apparatus. The loops being employed are fabricated of Type 316 stainless steel; the tantalum being exposed is present as a stack of small specimens immersed in the sodium stream flowing through the heated leg of each system. Cold-trapping and gettering (by hot zirconium foil) circuits are present in all loops to achieve close control of the level of oxygen in the test fluid.

During May, two flow-loop experiments were completed. Table L-1 presents a summary of the available data from these experiments (Tests 2 and 3) and also includes similar information from the first experiment in this series; the latter was conducted during April. Noteworthy trends indicated by these results include the following:

- (1) The specimen weight losses increase markedly with the oxygen content of the fluid. However, even the highest weight loss sustained is equivalent to only about 1 mil per year of metal removed from the exposed surfaces.
- (2) The weight losses vary approximately as the temperature of the immediate sodium environment when the oxygen level is high. With low oxygen levels, weight loss-temperature correlations are not evident.
- (3) In every case thus far examined, the tantalum specimens lost a sizable fraction of their original oxygen content as a result of exposure to sodium. This is evident directly from thermal-diffusion analyses of test pieces and, indirectly, from hardness data.

Two other findings from available posttest analyses should be mentioned. First, metallographic examinations of a number of exposed specimens have revealed that very slight surface roughening occurs as a result of the corrosion attack; intergranular penetration has not been found. Secondly, there is no evidence that the tantalum which is removed from the hot-zone specimens is transported and collected in cold-zone regions. As has been mentioned in previous reports, removable stainless steel sleeves are present in the cold zones of the test loops to facilitate inspections for mass-transfer deposits. Thus, the question of the whereabouts of the removed tantalum is now unanswered; to gain a better understanding of this situation, spectrographic techniques are being employed to determine if the removed material remains in the test fluid.

Further loop tests are under way with the sodium containing 20 ppm and < 10 ppm (continuously gettered) of oxygen. In addition, one loop assembly has been slightly modified to accommodate tantalum specimens for subsequent mechanical-property evaluations. Based on data thus far accumulated, the specimens involved will be degassed during the contemplated exposures; however, they should not suffer grain-growth effects such as are inevitable during very high-temperature degassing procedures.

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L-3

TABLE L-1. SUMMARY OF DATA FOR TANTALUM SPECIMENS EXPOSED TO 1200 F PEAK-TEMPERATURE SODIUM IN LOOP TESTS COMPLETED TO DATE

Specimen	Oxygen in Sodium, ppm	Exposure Time, days	Estimated Average Temperature of Specimen, F	Weight Loss of Specimen, mg per in. ²	Oxygen in Tantalum, ppm		Tantalum Hardness, VHN	
					Before Test	After Test	Before Test	After Test
<u>Test 1</u>								
A-7	<10 ^(a)	15	1200	3.1	118	88	211	161
A-4	<10 ^(a)	15	1000	5.7	118	28	211	173
A-2	<10 ^(a)	15	500	0.3	118	94	211	207
<u>Test 2</u>								
B-8 ^(b)	80 ^(a)	11	1200	11.1	113	31	176 ^(c)	135
B-7	80 ^(a)	11	1200	10.5	118	48	170	105
B-5	80 ^(a)	11	1100	6.9	118	46	170	106
B-4	80 ^(a)	11	900	1.6	118	52	170	108
B-2	80 ^(a)	11	750	2.0	118	66	170	107
<u>Test 3</u>								
D-6	<10 ^(d)	8	1200	0.2	118	(e)	170	(e)
D-5	<10 ^(d)	8	1150	0.2	118	--	170	--
D-4	<10 ^(d)	8	1100	0.2	118	--	170	--
D-8 ^(b)	<10 ^(d)	8	1050	1.0	113	--	155	--
D-3	<10 ^(d)	8	1000	0.2	118	--	170	--
D-2	<10 ^(d)	8	800	1.6	118	--	170	--

(a) Both hot and cold traps were frozen out of the system during the exposure.

(b) These specimens were from rolled arc-cast sheet, all others were from sintered-bar tubing.

(c) Lower original hardness as a result of annealing.

(d) Zirconium-foil hot trap was left in system during entire test.

(e) Data not yet available.

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L-4

The High-Temperature Mechanical Properties of Tantalum

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

Creep testing of annealed tantalum sheet (208 ppm oxygen) from sintered material was continued in May. Extrapolation of results now on hand from 1200 F experiments in helium using loads from 20,000 to 32,000 psi suggests that about a 0.0001 per cent per hr minimum creep rate should occur at the 13,000 to 14,000-psi stress level and about 0.00001 per cent per hr at the 10,000-psi level. It is also indicated that these same stress levels should result in about 5 per cent and 2 per cent total deformation, respectively, in 10,000 hr. A plot of stress-rupture life shows the following:

<u>Rupture Life, hr</u>	<u>Stress, psi</u>
10	29,000
100	26,000
1000	23,000

Additional tests with this material at 10,000-, 12,500-, and 15,000-psi levels are now in progress.

Emphasis in future testing will be concentrated on low-oxygen-content tantalum obtained by two methods, thermal degassing and exposure to purified sodium in the flow loops being run to obtain tantalum-sodium corrosion data (discussed in the preceding subsection). Thermal degassing results in large grains while sodium degassing should retain the original grain size. Creep testing of thermally degassed tantalum sheet (oxygen content reduced from 208 ppm to about 2 ppm) having large grains has been started. A test specimen loaded at 10,000 psi ruptured in about 21 hr.

Arc-cast tantalum test material rolled down to 0.030-in. sheet has been received. Analysis of one sample shows relatively high oxygen (260 ppm), carbon (320 ppm), and nitrogen (280 ppm). It is planned to expose a few specimens of this material to sodium prior to creep testing. However, a few tests with it will also be made in the annealed condition (as-received oxygen content) to provide correlation data with the results obtained with the annealed sintered material, as described above.

Weldability of Tantalum for High-Temperature Systems

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

This investigation is principally concerned with the weldability of a small all-tantalum tube-to-header assembly. Work during May has consisted mainly of fabricating tantalum tube-to-header assemblies. The materials used in this work were 20- and 30-mil virgin arc-cast tantalum sheet and a new batch of 3/16 in. in OD by 15-mil wall arc-cast tantalum tubing. While welding the tubes to the headers was accomplished, some difficulty has been encountered in making the final assembly (header-to-wrapping jacket) welds because of warpage caused by the prior welding on the assembly. This warpage has caused poor fitting of the pieces and consequently burn-through has occurred during welding. It is believed that the difficulty from warpage can be alleviated

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L-5 and L-6

by mounting the weldments rigidly during the welding operation. A jig which features a collet for mounting purposes is now being built.

Future work on this program will consist of fabricating a number of complete miniature tube-to-header assemblies. Upon completion, these specimens will be tested for soundness by helium leak-detection methods.

Determination of Oxygen in Sodium

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

A new and particularly sensitive method for the determination of oxygen in sodium is being developed. It involves the extraction of oxygen from the sodium by a getter metal which in turn is analyzed for increased oxygen content by vacuum-fusion techniques.

During May, gettering experiments were made at 650 and 690 C. At 650 C, 10-mil titanium sheet was in contact with filtered sodium for 2-1/2 hr. Under these conditions the titanium picked up 0.0032 w/o oxygen from the sodium. This sodium had previously been analyzed by the amalgamation method and found to contain 0.0040 ± 0.0010 w/o oxygen. Although analyses by the two methods are in substantial agreement, additional data from higher temperature runs are needed to determine if the gettering reaction is complete. Analytical results for the gettering test at 690 C are incomplete.

In several attempts to heat the gettering bomb with the existing furnace, it was found that the maximum temperature which could be attained in the test chamber proper was about 700 C. Rapid heat transfer to the exposed portions of the bomb outside the furnace prevented higher reaction-zone temperatures which would be desirable to increase the gettering efficiency. An auxiliary furnace is being added to the system to heat some of these portions and thus reduce this heat loss. A cobalt rod which can be used for magnetic stirring at the elevated temperatures has been obtained. However, it appears that before this new rod can be successfully used, the amount of magnetic flux to it must be increased through minor alterations.

It is planned that the next experiments will be made with niobium and zirconium to determine the gettering properties to these metals.

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M. DEVELOPMENTAL STUDIES FOR THE PWR

R. W. Dayton

Additional flow studies in the PWR model are to be made, to study the effects of lower plenum design on mixing and on flow distribution. Preparations for this work have been started.

Further studies of X-ray photometric inspection of fuel elements are to be made. The object of the present study is to develop a production-type instrument.

A literature study has been started that is aimed at evaluating materials suitable for constructing an in-pile loop using pressurized water.

Reactor Flow Studies

L. J. Flanigan and H. R. Hazard

This project was reopened early in May, with the objective of studying the effects of lower plenum design on mixing and on flow distribution in Core 2 of the PWR reactor. Air-flow studies will be conducted in the quarter-scale flow model of the PWR reactor which has been used for previous studies. The model is to be altered to simulate one or more configurations being considered for Core 2, and the effects of flow-baffle design on mixing and core-flow distribution are to be investigated.

An aluminum casting for a core extension for the quarter-scale model has been ordered, and a new flow baffle has already been received. The perforations in this baffle have not yet been designed, however.

All openings in the model were repaired so as to be airtight, all instrumentation was checked for tightness, and the model was painted.

The measuring equipment for determining sulfur dioxide concentrations was set up and work started on improving the gaseous-tracer technique for studying mixing.

In June, work will be completed on the tracer-gas injection system and the core extension and flow baffle will be completed and installed. It is estimated that the flow-distribution and mixing studies can be started late in June.

X-Ray Photometric Examination of Fuel Elements

J. B. Schroeder and C. M. Schwartz

As assistance to WAPD, a previous study showed that it would be feasible to examine Zircaloy 2-clad UO_2 fuel pins for weld porosity, and clad alloy fuel elements

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M-2

for uranium segregation and core location, by X-ray photometry to the accuracy required by the given specifications. The present work is aimed at developing an instrument capable of making these tests in a production plant.

A very intense X-ray source, with a small focal-spot size, is required to attain the desired testing rate. Commercially available equipment has been compared from the standpoint of source intensity. The Muller MG-150 constant-potential unit appears to be the best suited for this purpose.

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

F. H. Lyon

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

Several materials of possible interest for this application have been selected and their high-temperature properties are being studied. The materials selected for this preliminary screening are the following:

Alloy steels

"17-22A" S
SAE 4340 and 4130

Chromium steels

AISI 410
AISI 414
AISI 422
Croloy 16-1

Precipitation-hardening stainless steels

AM-350
AM-355

Superalloys

A-286
Discaloy 24
W-545
Inconel "X"

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Mechanical properties available at Battelle have been collected for four of the above steels:

"17-22A" S

A-286

Discaloy 24

Inconel "X"

Collection of information for the other materials is in progress.

A literature search is in progress to obtain information which is pertinent to welding the listed materials to themselves and to 18-8 stainless steel.

During the next report period it is expected that progress will continue at an accelerated pace and that corrosion information on the materials will be collected and included in the survey.

RWD:CRT/all

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