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PROGRESS RELATING TO CIVILIAN
APPLICATIONS DURING JUNE 1958

By
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July 1, 1958

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TABLE OF CONTENTS

	<u>Page</u>
REPORTS RELATING TO CIVILIAN APPLICATIONS ISSUED DURING JUNE, 1958	5
A. DEVELOPMENTS FOR ZIRCONIUM-CLAD FUEL ELEMENTS	7
Thermal Conductivity of Uranium and UO_2	7
Mechanical Properties of Zirconium Alloys	9
Development of High-Strength Corrosion-Resistant Zirconium Alloys	10
B. DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS	13
Preparation of Aluminum-Uranium Alloys	13
Aluminum Cladding	14
Development of a Natural-Uranium Fuel Alloy With Improved Corrosion Resistance	15
Literature Survey for the Appraisal of Uranium Monocarbide as a Possible Nuclear Fuel	17
C. PLANT ASSISTANCE TO MCW	19
Orientation in Uranium	19
D. PROCESSING OF FEED MATERIALS	21
Solidification of Uranium	21
Examination of Factors Affecting the Quality of Induction-Melted Uranium	22
E. DEVELOPMENTS FOR LMFRE	25
Electroless Plating	25
Vapor Plating	26
F. STUDIES OF URANIUM AND URANIUM-ALLOY FUELS	27
Radiation Stability of Uranium-Zirconium Alloys	27
Development of Gamma-Phase Uranium Alloys	27
Preparation of Uranium-Niobium Alloys	28
Mechanism of Aqueous Corrosion	29
G. FATIGUE STUDIES OF INCONEL AND INOR-8	31
Fatigue Studies of Inconel	31
Fatigue Studies of INOR-8	31
H. REACTOR MATERIALS AND COMPONENTS	33
Valence Effects of Oxide Solutions in Uranium Dioxide	33
High-Temperature High-Pressure Solid-State Studies	33
Rubbing Surfaces in Sodium Environments	34
Basic Studies of Pressure Bonding	35
Effect of Surface Preparation on Bonds Obtained During Pressure Bonding of Zircaloy-2	36
Fueled Zirconium Hydride Moderator	37
I. PHYSICAL RESEARCH	39
Preparation, Fabrication, and Physical Properties of Uranium Compounds	39
Constitution of Uranium-Base Ternary Alloys	40
Structure of UC_2	40
J. CORROSION PROBLEMS ASSOCIATED WITH THE RECOVERY OF SPENT REACTOR FUEL ELEMENTS	41
The Darex Process	41
The Sulfex-Thorex Process	42
The Fluoride-Volatility Process	43

TABLE OF CONTENTS
(Continued)

	<u>Page</u>
K. DEVELOPMENTS FOR SRE, OMRE, AND OMR	45
EVALUATION OF URANIUM MONOCARBIDE AS A REACTOR FUEL	45
Casting Techniques for the Preparation of Uranium Monocarbide	45
Irradiation-Capsule Design for Uranium Carbide	46
POSTIRRADIATION STUDIES OF SRE, OMRE, AND OMR FUEL MATERIALS	47
SRE Fuel Material	48
OMRE Fuel Elements	49
OMR Fuel and Structural Materials	49
L. STUDIES OF SODIUM-TANTALUM COMPATIBILITY AT ELEVATED TEMPERATURES	51
Tantalum-Sodium Compatibility Studies	51
High-Temperature Mechanical Properties of Tantalum	53
Weldability of Tantalum for High-Temperature Systems	54
M. DEVELOPMENTAL STUDIES FOR THE PWR	55
Reactor-Flow Studies	55
Improved Techniques for Corrosion Detection in Subassemblies and Clusters	55
Pressure Bonding of Zircaloy-2-Clad Fuel Elements Containing Compartmented Oxide Fuel Plates	56
N. DEVELOPMENTS FOR THE MGCR	59
Investigation of the Effect of Irradiation on Clad Graphite Specimens in a CO ₂ Environment	59
O. DEVELOPMENTS FOR NMSR	61
Fabrication of Urania Fuel Pellets for Loop-Test Studies	61
Fabrication of Loop-Test Fuel Pins	62
Preirradiation Measurements	62

REPORTS RELATING TO CIVILIAN APPLICATIONS
ISSUED DURING JUNE, 1958

BMI-1267 "Progress Relating to Civilian Applications During May, 1958", by
Russell W. Dayton and Clyde R. Tipton, Jr.

A-1

A. DEVELOPMENTS FOR ZIRCONIUM-CLAD FUEL ELEMENTS

F. R. Shober

Thermal-conductivity measurements made on an uranium specimen, encapsulated in Zircaloy-2 with NaK as the heat-transfer medium, indicate that thermal conductivities of encapsulated solid rods can be measured without removing them from the capsules. Thermal-conductivity values obtained from initial tests on UO_2 were lower than expected, apparently resulting from an unexpected sensitivity of the apparatus to the thermal conductivity of the material in test. Additional calibration is necessary before measurements can be made on UO_2 .

The creep and rupture strengths of 15 per cent cold-work Zircaloy-2 are being determined at 290, 345, and 400 C. The total deformations associated with the points of initiation of third-stage creep are approximately 1.0 per cent for tests at 290 and 345 C, and 1.5 per cent at 400 C. Corrosion tests in 300 C water and hot-hardness tests of high-strength zirconium alloys containing molybdenum, niobium, and tin indicate that tin is an effective addition with respect to corrosion and strength at 300 C.

Thermal Conductivity of Uranium and UO_2

C. F. Lucks and H. W. Deem

The effect of irradiation on the thermal and electrical conductivities of uranium and on the thermal conductivity of uranium oxide is being studied.

Uranium

Thermal-conductivity and electrical-resistivity measurements are to be made on uranium, uranium-1.5 w/o zirconium, and Zircaloy-2 specimens before and after irradiation to measure the effects of irradiation. The specimens to be measured were clad with Zircaloy-2 and NaK was used as the heat-transfer medium. Thermal-conductivity measurements are being made using a steady-heat-flow comparative method with the specimen assemblies protected by a vacuum.

Thermal-conductivity measurements were made on unclad rods of the three materials. The observed values are shown in Table A-1.

Thermal-conductivity measurements of clad specimens of the three materials were started. Temperatures of the specimens were measured by thermocouples flash welded to the cladding. A solid section of the Zircaloy-2 rod was used as the comparative standard. These measurements showed that, with good balances between specimen and guard, thermal conductivities of the clad specimens can be measured to ± 5 per cent of the values shown in Table A-1. This indicates success of the proposed measuring method.

TABLE A-1. THERMAL CONDUCTIVITY OF UNIRRADIATED URANIUM, URANIUM-1.5 w/o ZIRCONIUM AND ZIRCALOY-2

Temperature, C	Thermal Conductivity ^(a) , w/(cm)(C)		
	Uranium	Uranium-1.5 w/o Zirconium	Zircaloy-2
20 ^(b)	0.24 ₀	0.22 ₆	0.12 ₆
100	0.25 ₅	0.24 ₀	0.13 ₄
200	0.27 ₅	0.26 ₀	0.14 ₅
300	0.30 ₀	0.28 ₅	0.15 ₆
400	0.32 ₅	0.31 ₀	0.17 ₀
500	0.35 ₅	0.34 ₀	0.18 ₄
600	0.39 ₀	0.37 ₀	0.19 ₉
700	0.42 ₅	0.40 ₅	0.21 ₅
800	0.46 ₀	0.44 ₅	0.23 ₁

(a) Error estimated to not exceed ± 5 per cent.

(b) Extrapolated.

A-3

During the next month additional thermal-conductivity measurements will be made on the clad specimens, and an attempt will be made to find out why electrical-resistivity values have shown excessive scatter.

Uranium Oxide

An apparatus for making thermal-conductivity measurements on UO_2 , both before and after irradiation has been completed. A steady-state absolute method is being used. Briefly, accurately measured power is introduced into the top part of the specimen and the heat flows through the specimen into a heat sink. Compensated thermocouples at known positions measure the thermal gradients. The thermal conductivity of the specimen will be calculated from the heat flow, cross-sectional area, and the thermal gradients. Guarding to prevent stray heat flows has been provided.

Although the apparatus calibrated well with fused quartz as a standard, the values for UO_2 were lower than expected. Inconel, owing to its higher thermal conductivity, was then used as a reference material. Reliable thermal-conductivity values for the Inconel are available. Measured thermal conductivities of Inconel were about half of what they should have been.

A change was made in the design and materials used in the specimen heater. A relatively high thermal-resistance path was established between the specimen and guard heaters.

The apparatus will be thoroughly calibrated before any more work is done on UO_2 . Three materials will be used for the calibration work: fused quartz, Inconel, and a titanium alloy. Relative to the anticipated thermal conductivity of UO_2 , the fused quartz has a lower conductivity, Inconel a higher conductivity, and the titanium alloy, over a reasonable temperature range, about the same conductivity.

Work is in progress on the titanium alloy, and the complete calibration of the apparatus will continue.

Mechanical Properties of Zirconium Alloys

F. R. Shober and J. A. VanEcho

The creep properties of 15 per cent cold-worked Zircaloy-2 are required for the design of reactor components for operation at elevated temperatures. Of special interest is the deformation associated with the initiation of third-stage creep as a function of stress. Testing falls into two categories, short-time rupture tests and tests up to 10,000 hr in duration. The metallurgical stability of the cold-worked material under load at elevated temperatures for these periods of time is being investigated.

Three short-time stress-rupture tests have been completed, one each at 290, 345, and 400 C. The total deformations at the points of initiation of third-stage creep are approximately 1.0 per cent for tests at 290 and 345 C, and 1.5 per cent at 400 C. The magnitude of the deformation at this transition is apparently insensitive to temperature changes or to changes in stress at 290 and 345 C. However, it appears that the deformation is influenced by the increase in temperature to 400 C. There are 11 test units in operation and of these, 8 have been at test temperature and load for approximately 6000 hr. Small total deformations, less than 0.5 per cent, and low secondary creep rates, <0.00001 per cent per hr, are characteristic of these tests.

Development of High-Strength Corrosion-Resistant Zirconium Alloys

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

Zirconium alloys, because of their excellent corrosion resistance in water, strength, and low thermal-neutron cross section, are excellent cladding materials for reactor components exposed to water at elevated temperatures. Development of stronger zirconium alloys will provide material for reactor components for operation in an intermediate temperature range. It is believed that alloys containing additions (less than 5.0 w/o of molybdenum, niobium, and/or tin) can be developed which will have good corrosion life in 300 C water and a yield strength twice that of Zircaloy-2 at 300 C. These alloys should have expansion coefficients, nuclear properties, and thermal conductivities similar to Zircaloy-2.

Nine series of alloys are being developed. They consist of ternary zirconium-base alloys containing 2.0, 3.0, and 4.0 w/o tin plus 0 to 2.0 w/o molybdenum, ternaries containing 2.0, 3.0, and 4.0 w/o tin plus 0 to 3.0 w/o niobium, and quaternary alloys containing 2.0, 3.0, and 4.0 w/o tin plus 0.5 to 2.0 w/o molybdenum and 1.0 to 3.0 w/o niobium. The above-mentioned alloys are being screened by corrosion tests 1000 hr in duration in high-purity 300 C water, and by room-temperature hardness measurements. A maximum weight gain of 34 mg per dm^2 and a minimum room-temperature hardness of 250 have been set as limiting parameters for acceptable alloys. Hot-hardness measurements are being made over the 200 to 900 C temperature range on alloys containing the maximum and minimum alloying additions. Alloys which show the best combination of properties will be selected for further physical- and mechanical-property measurements.

Sheet specimens were prepared by arc melting and hot rolling at 850 C from a helium-atmosphere furnace, and are being tested in the annealed condition (4 hr at 700 C, furnace cooled). The corrosion specimens were prepared by shearing three pieces 1 by 3/4 by 0.070 in. from the fabricated sheet and machining approximately 0.003 in. from each surface and pickling an additional 0.002 in. from the surfaces (45 volume per cent HNO_3 + 5 volume per cent HF + 50 volume per cent H_2O pickle solution).

A-5

The majority of the zirconium-2 w/o tin corrosion samples have been in test 1000 hr (6 weeks). Table A-2 lists those alloys having the better corrosion properties in 300 C water after 2 and 4 weeks of exposure. One of the more corrosion resistant alloys, zirconium-2.0 w/o tin-0.5 w/o molybdenum, has a hot hardness of 140 at 300 C. This represents an increase of 40 DPH numbers over Zircaloy-2 at 300 C.

TABLE A-2. CORROSION DATA ON NINE SERIES OF ZIRCONIUM ALLOYS

Alloy Composition (Balance Zirconium), w/o	Weight Gain for Indicated Exposure, mg per dm ²		Remarks
	672 Hr	1000 Hr	
2.0Sn-0.5Mo	17	18	Shiny black, no evidence of flaking
2.0Sn-0.5Mo-1.0Nb	--	25	Shiny black, no evidence of flaking
2.0Sn-1.0Mo-2.0Nb	26	34	Slightly milky, no evidence of flaking
2.0Sn-1.0Mo-2.0Nb-0.1Fe-0.05Ni	22	26	Black, no evidence of flaking
2.0Sn-1.0Mo-3.0Nb	21	24	Slightly milky, no evidence of flaking
2.0Sn-2.0Mo-2.0Nb	37	50	Milky, slight flaking
2.0Sn-1.0Mo-0.1Fe-0.05Ni	24	25	Shiny black, no evidence of flaking
1.5Sn	22	26	Shiny black, no evidence of flaking
1.5Sn-0.1Fe-0.05Ni	23	25	Shiny black, no evidence of flaking

Two-week corrosion data on the zirconium-3.0 w/o tin alloys indicate that several of these alloys fall within the limits of the screening parameters. In general, the corrosion rates are slightly lower for the zirconium-3.0 w/o tin alloys than for zirconium-2.0 w/o tin alloys with similar additions, indicating a beneficial effect of an increase in tin content. Hot-hardness data for the zirconium-3.0 w/o tin alloys indicate that there is a definite increase in hardness at higher temperatures. It appears that the addition of 3.0 w/o tin has increased the hardness 25 to 35 DPH numbers at 300 C. The increase in hot hardness does not appear to be dependent upon the other alloying additions. The main effect of the molybdenum and niobium addition is a more rapid lowering of the hardness at temperatures above 550 C. The hot hardness at 800 and 900 C confirms the choice of a rolling temperature.

A-6

Two-week corrosion data for the zirconium-4.0 w/o tin alloys are not sufficient to indicate which alloys in this group will show promise. Hot-hardness measurements on alloys of this group are in progress.

All corrosion tests on specimens of the zirconium-3.0 w/o tin and zirconium-4.0 w/o tin alloys will be completed by the second week in July. It appears that several of these alloys will meet the requirements set up for this screening operation.

B-1

B. DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS

R. J. Carlson

Centrifugal-casting techniques for the production of aluminum-35 w/o uranium extrusion billets are being investigated. Various pouring temperatures and mold speeds have been utilized. It was found that a mold speed of 700 rpm is near the optimum for the above composition. Metallographic examinations of the aluminum-35 w/o uranium alloys containing various ternary additions indicate that, of the additions investigated to date, palladium and germanium appear most promising as compound-particle refiners.

The pressure-bonding parameters of time, temperature, and pressure necessary to obtain optimum bonding of nickel-plated uranium slugs with aluminum have been determined. Additional uranium slugs are being prepared and will be shipped to another site for evaluation.

The corrosion testing of natural uranium containing a series of 17 different alloying additions indicates aging may be detrimental to corrosion resistance. To determine if this is so an additional corrosion test will be performed on selected alloys in both the 900 C furnace-cooled and the aged conditions of heat treatment.

A literature survey is being conducted with the object of ascertaining the merits and disadvantages of uranium-monocarbide relative to other nuclear fuels. An additional goal of this survey will be to suggest those areas where the technology would benefit most from further experimental effort.

Preparation of Aluminum-Uranium Alloys

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

A number of research and production reactors now in use utilize a plate-type fuel element composed of an aluminum-uranium alloy core clad with aluminum. These fuel elements have proved satisfactory for use in low-temperature water-cooled water-moderated reactors. A continued interest in reactors of this type is reflected in present efforts to prepare more satisfactory fuel alloys and core configurations. One fuel element now being considered consists of an aluminum-35 w/o uranium alloy tube with both the inside and outside surfaces clad with aluminum. To facilitate the fabrication of such an element, it is desirable to cast the fuel in the form of a hollow right cylinder suitable for extrusion cladding with aluminum. For this purpose a homogeneous, sound, nonporous extrusion blank is necessary. Previous work at Battelle has indicated that a major difficulty in melting and casting the 35 w/o uranium alloy is porosity. This porosity is attributed to the evolution of hydrogen and metal shrinkage during solidification. Another difficulty is segregation due to the density differences between the phases present and to the large freezing range of the alloy.

The increase in uranium concentration from 25 to 35 w/o has necessitated a re-evaluation of the variables encountered in centrifugal casting. For this purpose, a

number of melts have been poured at 2200, 2300, and 2400 F into a steel mold rotating at 300, 550, 700, and 900 rpm. The steel mold had a variable wall thickness: 1/2 in. thick at the rear and tapered to a 1/8-in. thickness at the forward end. By using this type of mold, it was possible to displace the porous zone toward the thin section of the mold. The evaluation of these melts has not been completed. However, it is apparent that with the techniques employed a pouring temperature of 2300 F or more is required, and that a mold speed of 700 rpm is at or near optimum. Samples for analysis have been obtained from the best ingots.

A movable pouring spout has been designed and should be constructed in the near future. When this mechanism is available, efforts will be made to deposit the molten metal in the mold in such a manner as to induce solidification in a longitudinal direction, and thereby increase the yield of sound material from an ingot. Further refinements will be made in the melting technique which may afford more protection to the material during melting and while pouring. These refinements should aid in producing more homogeneous castings containing less porosity.

Concurrently with the casting program, there is in progress a study concerned with the improvement of the fabricating and casting characteristics of alloys of interest. It is believed that improved fabricability can be obtained either through the retention of UAl_3 in the alloys or by a refinement of the size of the massive UAl_4 particles found in simple binary alloys. It has been reported that the addition of 3 w/o silicon to the aluminum-48 w/o uranium alloy will suppress the UAl_3 - UAl_4 reaction to such an extent that no UAl_4 is found in the cast material. Therefore, a series of 1-in. -diameter by 8-in. -long melts containing selected ternary additions have been prepared and are being evaluated by metallographic examination, X-ray diffraction, and thermal analysis. The metallographic examination of these melts indicates that, among the additions examined to date, palladium and germanium are at least as effective as silicon in retaining the UAl_3 . Furthermore, the particle sizes of the compound in the alloy containing palladium appear to be smaller than those in the alloy containing silicon. The ingot containing germanium is not sound, containing many gas holes. This alloy will be remelted in an effort to improve its soundness.

Future work will be concerned with continued investigation of the variables encountered in casting the aluminum-35 w/o uranium alloy by centrifugal-casting techniques, and with the evaluation of the ingots already made. The small melts containing ternary additions will be examined by lineal analysis to determine the amount of compound present in those alloys that microscopic examinations indicate contain less compound than the binary alloy containing the same amount of uranium. The compounds observed in those alloys offering promise will be identified by X-ray diffraction. Another series of melts is planned in an effort to refine the UAl_4 particle size without retention of UAl_3 . These melts will utilize oxide additions just prior to casting.

Aluminum Cladding

S. J. Paprocki, E. S. Hodge, C. B. Boyer,
and C. C. Simons

The cladding of internally and externally cooled fuel elements consisting of a nickel-plated uranium core clad with aluminum is being investigated. The process

B-3

employed to obtain a metallurgical bond between the core and cladding is pressure bonding, a technique which incorporates gas pressure and temperature.

The pressure-bonding parameters of time, temperature, and pressure were varied to determine the conditions necessary for obtaining an optimum bond. Specimens bonded 1-1/2 hr at 750 F and 5000 psi showed complete lack of bonding. Optimum bonding was obtained in 1 hr at 950 F and 10,000 psi. Under this condition of bonding, two distinct phases were observed at the nickel-uranium interface. These phases have not been identified; however, it has been observed that the strength of the bond is increased significantly with the formation of the second uranium-rich phase. Decreasing bonding time from 1 hr to 5 min at 950 F and 10,000 psi had very little effect on the depth of the nickel-uranium diffusion; however, this was insufficient time for the formation of the second uranium-nickel phase to go to completion. Although the bond obtained by these conditions was fairly strong, the strength was inconsistent over the length of the specimen.

Additional uranium slugs are being prepared by bonding 1 hr at 950 F and 10,000 psi.

Development of a Natural-Uranium Fuel Alloy
With Improved Corrosion Resistance

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

A high-uranium fuel alloy possessing improved corrosion resistance in 300 C water is being sought. This alloy must possess better corrosion resistance than a uranium-2 w/o zirconium alloy but have a thermal-neutron cross section no greater than that of a 4 w/o zirconium alloy.

Corrosion tests conducted on 17 compositions are reported in Table B-1. These tests were conducted on specimens measuring 1/4 by 1 by approximately 0.06 in. Alloys of similar heat treatments were corrosion tested together. The specimens were sealed in an autoclave, which was then flushed with argon and evacuated several times. Heating to 300 C took place in about 2 hr, after which preheated water was admitted. Corrosion took place at 300 ± 10 C for 15 min. Cooling was effected by the release of steam, followed by a cold-argon flush. When the autoclave temperature reached 204 C it was water quenched.

Although the variables of all three corrosion tests were closely controlled, it is possible that slight variations in test conditions could appreciably affect the results because very rapid corrosion rates are involved. Therefore, comparison between alloys tested at different times is not expected to be as valid as when they are tested together. The results indicate that for the heat treatments investigated, certain ternary additions to the uranium-2 w/o zirconium alloy increase its resistance to corrosion. The additions most effective were 0.33 w/o niobium, 0.16 w/o ruthenium, 0.05 w/o platinum, and 0.46 w/o aluminum. Also, an alloy of uranium containing 1 w/o zirconium and 0.50 w/o niobium showed comparatively good corrosion behavior. For alloys in the aged condition, uranium-2 w/o zirconium-0.33 w/o niobium performed roughly twice as well as the uranium-2 w/o zirconium alloy. In general, the alloys furnace

TABLE B-1. CORROSION RESULTS OBTAINED ON URANIUM ALLOYS TESTED 15 MIN IN 300 C WATER

Sample	Alloy Composition (Balance Uranium), w/o	Corrosion Rate ^(a) for Alloy Given Indicated Heat Treatment, mg/(cm ²)(hr)		
		1 Hr at 900 C, F.C.	3 Min at 800 C, W.Q.; 6 Hr at 690 C, W.Q.; 1 Day at 450 C, F.C.	1 Hr at 880 C, A.C.; 1 Day at 450 C, F.C.
	Biscuit uranium		4040 ^(b)	4760 ^(c)
1	2Zr	1700	3068	>3660 ^(d)
2	4Zr	2080	3284	3160
3	5Zr	2040	2888	2812
4	2Zr-0.04V	2100	3184	4200
5	2Zr-0.33Nb	1710	1496	2328
6	2Zr-0.047Ni	1690	3160	4040
7	2Zr-0.03Ti	1760	2128	3640
8	2Zr-0.14Mo	2250	3120	2992
9	2Zr-0.05Pt	1360	1544	2440
10	2Zr-0.07Cr	1760	2616	3782
11	2Zr-0.015V-0.015Si-0.01Mo	2230	3128	Disintegrated
12	0.015V-0.15Si-0.01Mo	2640	Disintegrated	Disintegrated
13	2Zr-0.46Al	1400	1916	3120
14	1Zr-0.5Nb	1950	1568	2348
15	2Zr-0.15Nb-0.05Mo	3110	2128	2600
16	2Zr-0.16Ru	1360	1300	3216
17	0.04Cr-0.04V-0.1Nb-0.2Zr	2240	Disintegrated	Disintegrated

(a) Based on original area.

(b) As hot rolled.

(c) 1 hr at 720 C, water quenched plus 1 day at 450 C, furnace cooled.

(d) Disintegrated; maximum corrosion rate based on actual specimen thickness before corrosion test.

B-5 and B-6

cooled from 900 C exhibited lower corrosion rates than those that were aged at 450 C. To determine if aging actually does lower the corrosion resistance of these alloys, an additional corrosion test will be performed on alloys showing the best performance, the alloys to be tested in both the 900 C furnace-cooled and the aged conditions of heat treatment.

Literature Survey for the Appraisal of Uranium
Monocarbide as a Possible Nuclear Fuel

J. B. Melehan, A. A. Bauer, and R. F. Dickerson

A literature survey is being conducted to obtain information pertinent to the science and technology of uranium monocarbide in order to determine its merits and disadvantages relative to other nuclear fuels. An examination and evaluation will be made of published research data and of data from research now in progress on such aspects as physical, chemical, and mechanical properties, techniques of preparation and fabrication, and irradiation behavior. This includes information concerning the relationship between radiation effects and properties, dimensional stability, fuel-element design, allowable burnup, and the compatibility of uranium carbide with various heat-transfer media.

Initial efforts have been spent in obtaining readily available information on physical and chemical properties, and in discussions with personnel who have been engaged in research involving uranium carbide. On the basis of data thus far assembled, it is not possible to draw any definite conclusions concerning the potential uses of uranium carbide. For instance, uranium carbide has a high thermal conductivity, relative to other ceramic fuel materials, shows promising resistance to failure from thermal cycling, and exhibits compatibility with liquid NaK up to 1300 F. Moreover, uranium carbide can be prepared by existing melting procedures, obtaining densities of about 98 per cent of the theoretical value. On the other hand, the material has low impact strength and requires careful handling, oxidizes readily, and enters into a hydrolysis reaction with water that results in slow disintegration in moist air or water at room temperature and in rapid decomposition in hot water.

Virtually no effort has been made to study the behavior of uranium carbide under nuclear-irradiation conditions. In the absence of definite information, it may be useful to make predictions as to the probable behavior in terms of allowable burnup, accumulation of fission products, rate of fission-gas diffusion from fuel elements at operating temperatures, and the most efficient fuel-element geometry. An additional goal of this literature survey will be to suggest those areas where the technology would benefit most from further experimental effort.

C-1 and C-2

C. PLANT ASSISTANCE TO MCW

D. A. Vaughan

An analysis is being made of the effect of preferred orientation of crystallites on the X-ray diffraction line intensity.

Orientation in Uranium

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

The terms in the intensity formula for X-ray diffraction are being evaluated with respect to the effect of changes in degree of crystallite orientation. In the present analysis three factors, (1) the number of crystals irradiated, (2) the multiplicity factor, and (3) the receiving-slit geometry, defining the segment of the diffraction cone examined, appear to be of major importance in determining the change of diffraction intensity from a given hkl plane due to preferred orientation. Since the receiving-slit geometry may be fixed at some small fraction of the diffraction cone, the spread in the orientation along the diffraction cone is considered negligible in the present study.

D-1

D. PROCESSING OF FEED MATERIALS

E. L. Foster

Two programs concerned with the quality of unalloyed uranium fuel-slug material are reported in this section. One of the programs includes the theoretical considerations involved in the solidification of unalloyed uranium castings. The other program has been devoted to the study of the factors affecting the quality (in terms of chemical composition) of unalloyed vacuum-melted uranium.

The program concerned with the study of factors affecting the quality of vacuum-melted uranium and one phase of the solidification program have been completed. Separate reports, covering the Fiscal Year 1958 research, are being prepared.

Solidification of Uranium

E. L. Foster, C. K. Franklin, B. L. Fletcher,
B. Schwartz, and R. F. Dickerson

The objective of this program is to evaluate the importance of the various factors involved in the solidification of uranium castings. The program includes two phases of study: a theoretical study of the transfer of heat from the metal, and an experimental study to establish the validity of assumptions used in the theoretical phase. These assumptions will be checked by means of data recorded during the casting of small-scale ingots. The ultimate aim is to provide a means of evaluating proposed process changes in terms of their effect upon the internal soundness of production-size uranium ingots.

In studying the heat transfer in the metal and mold, a mathematical model has been constructed. This model, consisting of 75 cells and representing a pie-shaped section of the casting and mold, has been programmed and solutions have been obtained through the use of an IBM 650 computer.

Analysis of the computed data was completed during June, and the results obtained appear logical and reasonable. An over-all solidification time of 13.2 min has been calculated for a 1270-lb ingot, this being for an initial melt temperature of 1200 C and initial mold temperatures that varied from 1000 C at the top to 100 C at the bottom of the mold. The mold-wall temperatures observed just after casting are of interest in that the inner mold-wall temperatures show a sharp peak just before separation occurs; if this same effect can be detected in the experimental work, it will be of value in determining the time of separation.

At this time it is believed that a sufficient amount of computation has been performed with the 75-cell model to establish the validity of the mathematical expressions describing it. Further minor modifications of the expressions are being postponed until the constants can be recomputed for the experimental-size ingot. This postponement is necessary so that developments, such as the determination of more accurate values for thermal properties, can be included in the first attempts at correlation.

The correspondence of the numerical solution to the real situation is the major concern at present; it is always possible that the number of cells used is inadequate to properly represent the heat flow in the casting. However, it is gratifying to note that in a study of a similar nature it was found that, for two-dimensional transient heat flow in a steel ingot, the results obtained with three reference points from the center to surface of the ingot and two reference points across the mold gave results little different from those obtained with many more points.* In the present 75-cell model, there are four points in the casting (from center to wall) and two in the mold wall.

Various physical-property measurements on uranium and graphite are being made. These values will be used in the mathematical expressions to more accurately calculate the rate of heat transfer in the casting and mold materials.

Linear-thermal-expansion measurements on uranium specimens, oriented both transversely and longitudinally in the ingot, have been completed. The uranium specimens taken from a production-size casting in the longitudinal direction showed a permanent deformation during the second heating. Because of this deformation all the data are being re-evaluated before they are presented.

Thermal-conductivity measurements on uranium in the longitudinal direction of a casting have been completed. These data also will not be reported until measurements are run in the transverse direction and properly evaluated. Comparison of the conductivity characteristics in the two directions should indicate if the heat flow is isotropic.

Preliminary tests on small-scale castings have been made. The establishment of experimental details is the objective of these efforts. Adequate accuracy, response time, and physical stability have been indicated for thermocouples placed in the mold and in the mold cavity. The next report will include results from the first fully instrumented small-scale experimental run.

Examination of Factors Affecting the Quality of Induction-Melted Uranium

R. W. Endebrock, E. L. Foster, and R. F. Dickerson

As an investigation of the various chemical reactions that occur during the vacuum-induction melting of uranium, this program has for its main objective the investigation of those reactions which can vary from melt to melt and in a way that could be detrimental to the cast ingot. The experimental portion of the program is being conducted in a 10-lb-capacity vacuum-melting furnace which has been specially equipped with a gas-sampling manifold to allow analyses of furnace gas by a mass spectrometer. A second phase of the program is a literature survey which is being conducted concurrently with the experimental portion of the investigation. A more complete knowledge pertaining to furnace reactions occurring during the remelt cycle should suggest a means for their control and should result in the production of uranium ingots of greater uniformity.

*Sarjant, R. J., and Slack, M. R., "Internal Temperature Distribution in the Cooling and Reheating of Steel Ingots", J. Iron Steel Inst. (London), 177, 428 (August, 1944).

D-3 and D-4

An important outcome of certain tests in which various gases were admitted to the furnace chamber containing pure uranium held at a temperature of 2500 F was the discovery of the formation of surface-protective drosses on the molten-metal surface. These drosses were formed by the action of some of these gases in the uranium-graphite system. Taking advantage of the protective nature of such drosses, an experiment was conducted for the purpose of demonstrating the existence of a water-gas reaction in the uranium-graphite system. About 1.7 liters of carbon dioxide was added to the furnace to produce such a protective dross on the molten uranium surface. Residual gas was removed and 1 cm³ of water was introduced to the furnace chamber. After the water addition, a 25 per cent increase in furnace pressure, compared at identical gas-sampling times, was observed. This increased pressure was directly proportional to the increase in residual CO in the furnace gas as determined by gas analysis, and demonstrated the water-gas reaction in the system. It demonstrated in addition a mechanism for putting large amounts of carbon in solution in the uranium. A thermodynamic treatment of such a mechanism indicated that small changes in the partial pressure of CO over the melt may result in large changes in the carbon-oxygen level of the melt. A protective cover could, therefore, be beneficial as long as it remained intact. Once broken, the large CO partial pressure that would exist prior to the disturbance would drastically affect the solution of carbon from the crucible.

A high dissolution of crucible graphite was observed in this experiment. The long period of exposure (4-1/2 hr) at high temperatures probably accounts for this effect. However, the pattern of graphite loss from the crucible suggests a mechanism of transfer that may be related to the kind and pressure of the gas over the melt and to the variation between surface and subsurface metal temperatures.

E-1

E. DEVELOPMENTS FOR LMFRE

J. McCallum

One phase of the development of the Liquid-Metal-Fueled Reactor involves selecting the best material of construction to contain a slurry of uranium bismuthide in bismuth at relatively high temperatures in a variable radiation field. Both direct-corrosion and mass-transfer phenomena are anticipated with the more common materials of construction. It is believed that a thin protective coating of molybdenum on the interior surfaces of the reactor system would considerably extend the life of the reactor loop, should these problems prove to be serious.

Two methods of preparing a protective coating of molybdenum are being investigated. One method involves electroless plating by chemical reaction of a molybdenum compound at relatively low temperatures. The other is vapor deposition. No electroless process has been established for molybdenum. However, if a suitable reaction system can be devised, coating an entire reactor loop would not appear to be an unduly complex engineering problem. In contrast, the chemistry of vapor depositing molybdenum from a halide or carbonyl is well understood. However, the temperature limitations on the process and the attendant engineering problems may require a considerable development effort before a satisfactory coating can be deposited.

Electroless Plating

J. McCallum and C. L. Faust

A study of the thermodynamics of molybdenum compounds and various reducing agents indicates several possibilities for an electroless process to deposit a molybdenum metal coating. Some of the reduction possibilities are:

- (1) Molybdenum chlorides with Grignard reagents (organic magnesium compounds known to be strong reducing agents) in an organic solvent
- (2) Molybdenum iodides with hydrazine, in aqueous solution
- (3) Molybdenum oxides or halides with hydrides, in aqueous solution.

One experiment was tried with ammonium molybdate and sodium borohydride in aqueous solution. Reduction of the molybdate to a lower valence state occurred, but no metal was observed. It appears that nonaqueous systems will provide the best approach because the formation of complex molybdates, apparently not reducible to metal, will be a problem in aqueous solutions. Effort will be concentrated on organic systems in future work.

E-2

Vapor Plating

C. F. Powell and I. E. Campbell

An experimental program covering the vapor deposition of molybdenum on Croloy 2-1/4 has begun. Initial experiments in this program resulted in only partial reduction. Additional experiments on molybdenum deposition by the hydrogen reduction of the halides at 1020 to 1050 F will be conducted during July.

F-1

F. STUDIES OF URANIUM AND URANIUM-ALLOY FUELS

F. A. Rough and A. W. Hare

Research on uranium-alloy fuels is reported in this section. These studies are part of the AEC Reactor Development Division's research program.

Radiation Stability of Uranium-Zirconium Alloys

D. J. Hamman, A. W. Hare, A. A. Bauer,
and R. F. Dickerson

Experimental work on the radiation effects of uranium-zirconium alloys has been recessed.

Development of Gamma-Phase Uranium Alloys

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

A program to determine the effect of alloying additions of chromium, molybdenum, niobium, ruthenium, vanadium, and zirconium on the thermal stability of gamma-phase uranium alloys is being conducted. In addition, the effects of alloying on hot hardness, corrosion resistance, and transformation temperatures are being determined.

After 1848 hr in 680 F water, gamma-quenched uranium-20 w/o niobium and uranium-20 w/o niobium-5 w/o zirconium alloys are corroding at rates of 0.03 and 0.02 mg/(cm²)(hr), respectively. Of those alloys prepared, the only other alloy surviving after 1848 hr is a uranium-7.5 w/o molybdenum-2 w/o ruthenium alloy which is corroding at a rate of 0.28 mg/(cm²)(hr). Several uranium-molybdenum-base alloys recently placed on test have corrosion rates as tabulated below after 672 hr in 680 F water.

Alloy Composition (Balance Uranium), w/o	Corrosion Rate, mg/(cm ²)(hr)
7.5Mo-3Nb-2Ru	0.14
7.5Mo-2Nb-2Ru	0.08
10Mo-1Nb	0.27
10Mo-3Nb-2Ru	0.02
10Mo-2Nb-1Ru	0.15
12Mo-2Nb-1Ru	0.27

Generally, those alloys which exhibit good gamma stability also exhibit good corrosion resistance.

The effects of alloying on gamma stability, hot hardness, and transformation temperature of various uranium-molybdenum, -niobium, and -zirconium-base alloys have been previously reported.

Preparation of Uranium-Niobium Alloys

J. B. Melehan, E. L. Foster, and R. F. Dickerson

The objective of this investigation is the development of methods for melting and fabricating alloys of the uranium-niobium system. Attention has been focused on the alloys containing 20, 40, and 80 w/o niobium. Supplementary alloy property studies were also to be considered.

It was reported in BMI-1267 that heating the three alloys in air for 24 hr at 400 C, followed by 24 hr at 500 C, and finally by 24 hr at 600 C resulted in complete oxidation of the specimens. It was also reported that the 80 w/o niobium alloy was oxidized only very slightly when exposed to air at 400 C for 144 hr. In June, an experiment was conducted to determine a temperature at which oxidation of the uranium-20 and -40 w/o niobium alloys was less drastic. Specimens were exposed at 350 C for 163 hr. The data for this experiment and for the test of the uranium-80 w/o niobium alloy, begun previously, are given in Table F-1. A graphical presentation of this data would show that the oxidation rates after the first 24 hr are linear for all alloys. Slight spalling occurred from each specimen, and additional exposure would probably have resulted in a continuing linear oxidation rate. It is of interest to compare the weight gain in 24 hr at 350 C to the data reported for the 20 and 40 w/o alloys after 24 hr at 400 C (BMI-1267). The weight gains at 400 C for the 20 and 40 w/o alloys respectively are 15 and 30 times the weight gains at 350 C. After 163 hr at 350 C the total weight gain was only one-fourth or one-fifth the gain in 24 hr at 400 C.

TABLE F-1. OXIDATION OF URANIUM-NIOBIUM ALLOYS

Niobium Content (Balance Uranium), w/o	Initial Weight, g	Initial Area, cm ²	Temperature, C	Weight Gain After Exposure, g						
				24 Hr	48 Hr	72 Hr	96 Hr	120 Hr	144 Hr	168 Hr
20	16.1170	10.063	350	0.0039	0.0063	0.0084	--	0.0128	--	0.0168
40	17.7611	10.736	350	0.0017	0.0045	0.0052	--	0.0048	--	0.0096
80	8.2852	7.3011	400	0.0012	0.0039	0.0039	0.0059	0.0064	0.0077	--

F-3 and F-4

Mechanism of Aqueous Corrosion

E. Adelson, C. M. Schwartz, D. A. Vaughan,
O. M. Stewart, W. E. Berry,
P. D. Miller, and F. W. Fink

A topical report on the investigations of uranium corrosion in 100 C water and 200 C steam at atmospheric pressure is being prepared. The solid corrosion product was analyzed for phase composition and valence state; it was found to consist of (1) an oxygen-rich UO_2 , and (2) uncorroded metal particles. No hydride phase was detected, in contrast to other investigations. The differences may be explained on the basis of hydrogen overpressure in the reaction vessel.

G-1 and G-2

G. FATIGUE STUDIES OF INCONEL AND INOR-8

W. S. Hyler and F. H. Lyon

Fatigue Studies of Inconel

This program has the objectives of obtaining basic fatigue information on Inconel and of establishing quantitative relationships among the variables of temperature, stress, strain, time, and cyclic frequency for Inconel.

The current phase of the program is concerned with the measuring and recording of strain associated with the cyclic portion of a combined load. During June the specimen-extensometer system was calibrated and checked by performing preliminary tests on a few specimens. Evaluation of results indicates that the fatigue studies of Inconel should progress as expected.

Fatigue Studies of INOR-8

A program to evaluate the fatigue properties of the alloy INOR-8 is being conducted.

Fatigue specimens have been prepared. During June preliminary experiments were performed in which specimens were cycled at 10,000 and 100 revolutions per minute.

Next month, the evaluation of the INOR-8 alloy will commence.

H-1

H. REACTOR MATERIALS AND COMPONENTS

F. A. Rough and A. W. Hare

The studies reported in this section are part of the AEC Division of Reactor Development program of research.

Tentative results of high-temperature high-pressure solid-state studies of UO_2 , BeO , La_2O_3 , and Sc_2O_3 are reported.

The programs concerned with the oxidation-resistant niobium alloys and rubbing surfaces in sodium environments have been terminated as of the end of Fiscal Year 1958.

Routine progress is reported for the basic studies of pressure bonding and the program concerned with the effect of surface preparation on the bonds obtained during pressure bonding of Zircaloy-2. Work on a fueled zirconium hydride moderator was recessed.

Valence Effects of Oxide Solutions in Uranium Dioxide

W. B. Wilson and C. M. Schwartz

This investigation is concerned with the "stabilization" of the refractory UO_2 fluorite structure by the addition of oxide additives, such that when oxidized in air at higher temperatures the transformation to U_3O_8 , as well as the sublimation of U_3O_8 , is avoided. The resulting materials are dimensionally stable and retain the same crystal structure under oxidizing and reducing atmospheres although they may be oxidized or reduced. Experimental work on the current phase of the work is essentially complete.

High-Temperature High-Pressure Solid-State Studies

W. B. Wilson and C. M. Schwartz

The use of pressure as a new degree of freedom in producing high-temperature materials for reactor components is being studied. Many aspects of the development and use of equipment evolved on this project are treated in the recent publication by H. T. Hall*. The right-circular-cylinder die equipment as depicted by Hall, with minor modifications, has been used to study reactions.

The initial studies have been concentrated on examining reactions in a few systems. Tests have been conducted at a 30,000-atm pressure or greater (due to "frictional hysteresis" upon internal heating) and at variable temperatures. The systems

*Hall, T. T., "Some High-Pressure High-Temperature Apparatus Design Considerations", Rev. Sci. Instr., 29, 267 (April, 1958).

being investigated under pressure have received preliminary study at normal pressure in relation to valence effects of oxide additives to uranium oxide. The preliminary results under pressure reported herein are tentative.

The application of simultaneous pressure and temperature in UO_2 , BeO , and La_2O_3 has not produced any detectable irreversible structural changes. Both UO_2 and U_3O_8 , mixed with BeO and Sc_2O_3 , have been studied. Under normal pressure, BeO and UO_2 are nonreactive, apparently due to the extreme difference in metal-ion sizes. However, Sc_2O_3 will react with U_3O_8 to form a stable solid solution having the fluorite structure, and is nonreactive with UO_2 . This appears to be predominantly due to the fact that the Sc^{+3} ion has a size factor favorable for reaction with U^{+6} and not with U^{+4} . Under pressure, reactions have been detected between UO_2 and Sc_2O_3 and between BeO and U_3O_8 .

When BeO was treated under pressure with UO_2 , a reaction was not detected. These early tests involving BeO and UO_2 will be reinvestigated, since various experimental failures had been encountered. Since the U^{+6} ion is about 20 per cent smaller than the U^{+4} , it was decided to mix U_3O_8 with BeO . A reaction was detected in which an unknown phase ("X" phase) was repeatedly produced. This phase, of complex crystal structure, is apparently produced at the lower temperatures, since it was first formed at 500 C. At 650 C, it was also present, together with UO_2 and BeO . At temperatures in excess of 1000 C, only the original U_3O_8 and BeO were detected.

In view of these results, and because of other suspected reactions, U_3O_8 itself was examined. In an early experiment using a nickel heater with $\text{U}_3\text{O}_8 + \text{BeO}$, the nickel was rapidly burned out and $\text{UO}_2 + \text{BeO}$ remained, indicating U_3O_8 had been reduced by the nickel heater. In view of the very rapid rate of this reaction, it was speculated that oxygen might be released under pressure by the U_3O_8 to form a cubic UO_2 -type structure saturated with oxygen, such as the mineral cleveite* or other uranium oxides of high oxygen content with the UO_2 structure.

In a recent experiment, U_3O_8 was held at greater than 30,000 atm at ambient temperature for nearly 4 hr. Subsequent examination indicated that the U_3O_8 had formed about equal amounts of the α (Zachariasen) and β modifications. The experiment is being repeated at simultaneously elevated temperature and pressure, using a platinum heater, to avoid reaction with it.

The early results seem to indicate that pressure can more easily affect the structure and composition of complex materials than of the simpler ones. It should be strongly reiterated that these results are tentative and subject to revision.

Rubbing Surfaces in Sodium Environments

J. W. Kissel and J. H. Stang

This investigation, which has been directed toward an evaluation of factors important in the friction and wear behavior of rubbing surfaces lubricated by high-temperature (above 1000 F) sodium, terminated at the end of June. During June, the

*Biltz, W., and Muller, H., Z. anorg. u. allgem. Chem., 163, 279 (1927).

H-3

activity included only further appraisals of data reported in BMI-1262 and BMI-1267. While these data are, in a sense, preliminary, they demonstrate that the friction characteristics of the principal rubbing combinations dealt with, molybdenum versus itself and WC-20 w/o cobalt binder versus itself, are sensitive to the presence of sodium at the contact surfaces, although in different ways. For example, the metallic pair exhibited lower friction coefficients with sodium present than those occurring without sodium; the cermet pair displayed the reverse effect. This divergence, in itself, is an important consideration, even though the information that is on hand is inadequate to define the exact mechanisms responsible for it. It appears that there are several factors. A detailed study of their interrelations would be required to pinpoint materials combinations and conditions required for satisfactory sodium-lubricated rubbing compatibility. Among the factors which appear important are those concerned with surface chemistry, including the formation of films which may, in the one case, act to reduce friction and, in the other, act to increase it.

Basic Studies of Pressure Bonding

S. J. Paprocki, E. S. Hodge, S. D. Beck, and M. A. Gedwill

Under suitable conditions of pressure, temperature, and time, it is possible to bond metal surfaces together by the application of hydrostatic pressure. In this investigation an effort is being made to analyze the deformations of the voids which exist between the surfaces in an attempt to predict the conditions under which the surfaces will attain the sufficiently close contact needed for bonding.

Analytical Studies of Pressure Bonding

It is assumed that the deformations that occur can be analyzed in terms of a simple power law for creep. Due to the stress configuration in the pressure-bonding problem, it is necessary to consider creep behavior in a combined-stress situation. As a model with a closely related stress distribution, a thick-walled cylinder has been used. Measurements have been made of the deformations in 2S aluminum cylinders pressurized at 500 and 700 F and at various pressures and times in order to determine the creep parameters at these two temperatures. However, the observed deformations have not followed a sufficiently systematic pattern to indicate whether the assumed creep model is valid or not. As an indication of the unsatisfactory scatter, the deformations have not always behaved in a monotonic manner with increasing pressure or time. Because of the relatively small number of specimens used, the observed scatter is sufficient to cause gross errors in the values of the creep parameters. A procedure involving torsion creep tests is being considered as an alternative method of obtaining the needed creep parameters.

It is expected that the creep parameters thus attained may be applied in the case of surfaces to be pressure bonded. The voids which occur along the interface after an initial pressurizing operation can be measured and their average size taken as an initial hole diameter. This hole size can then be compared with those in specimens bonded for a longer prescribed time, in an attempt to evaluate the validity of the creep model.

Experimental Studies of Pressure Bonding

The experimental phase of the program is directed toward acquiring data which can be used to obtain creep parameters and to examine the validity of the postulated mathematical models for predicting the flow characteristics of metals during a hydrostatic pressure-bonding process. In order to obtain the experimental data, evacuated and sealed thick-walled tubular aluminum specimens are externally subjected to various sets of pressure-bonding conditions as prescribed by the analytical studies. Duplicate specimens are subjected to each set of conditions.

A series of tubular specimens has been subjected to hydrostatic pressures of 3000 and 4000 psi at 700 F for periods of 2, 4, and 6 hr. Results indicate that the extent of deformation is significantly affected by an increase in pressure at 700 F.

An attempt was made to determine if reproducible results of deformation could be achieved for two pressure-bonding operations performed under the same parameters. In order to do this, another duplicate set of tubular specimens was externally pressurized at 3000 psi and 700 F for a period of 4 hr. The results obtained for the two operations were in only fair agreement with each other.

In an effort to predict the deformation resulting between mating surfaces during a pressure-bonding process from the results obtained by the analytical studies on tubular specimens, two sets of flat-plate aluminum specimens have been machined to finishes of 250 and 750 rms, respectively. Two surfaces of the same roughness are placed in contact with their directions of machining normal to each other. Specimens consisting of each rms finish have been pressurized at 4000 psi and 700 F for the time required to bring the pressure to 4000 psi, whereupon it is immediately released. Additional flat-plate specimens with 250 and 750 rms finishes have been subjected to pressure-bonding parameters of 4000 psi at 700 F for periods of 2 and 6 hr, and are now in process of being prepared for metallographic examination.

The first attempt to obtain an initial hole size was a failure, however, since the surfaces attained virtually complete contact during the very short application of pressure. Another attempt may be made at a lower pressure and/or temperature in order to be in a range in which the deformations may be measured and compared.

Effect of Surface Preparation on Bonds Obtained During Pressure Bonding of Zircaloy-2

S. J. Paprocki, E. S. Hodge, and M. A. Gedwill

Previous experiences have indicated that the surface condition of Zircaloy-2 prior to bonding has a decisive effect on the quality of the bonds obtained during gas-pressure bonding. The objective of this investigation is intended to supplement the work that is being performed in other programs in which component bonding involving Zircaloy to Zircaloy is required. Surface preparation and bonding variables, such as the cold-worked condition of the metal, surface contamination, surface roughness, and recrystallization and grain growth, which appear to have an influence on the integrity of

H-5 and H-6

the bond obtained during pressure bonding have been isolated, and their effect on bonding will be determined. The results of this investigation will permit understanding of the relationship between surface preparation and bonding so that an optimum and economical surface-preparation process can be selected.

An evacuated and sealed Ti-Namel can containing surface-machined Zircaloy-2 specimens with 50-, 125-, 400-, and 1000-rms finishes in the hot-rolled and annealed base condition was externally subjected to pressure-bonding parameters of 10,000 psi of helium gas at 1550 F for a period of 4 hr. Preliminary metallographic examination of sections taken from these specimens indicated that complete grain growth took place across the bonding interfaces, except for the 50-rms-finished specimen, which exhibited a few minute areas of incomplete grain growth.

Specimens for the study of the effect of cold work on bonding were prepared, assembled into a multiple-specimen Ti-Namel can, and pressure bonded under the same parameters as used for the surface-machined specimens. To insure reliability of results, the can contained a Zircaloy-2 reference specimen in the annealed and belt-abraded condition. Surface layers of carbon, chromium, iron, niobium, tin, titanium, and zirconium oxide were applied to reference-conditioned Zircaloy-2 specimens, and the specimens were assembled and pressure bonded to study their effect on bonding and to observe the movement of these layers during the bonding process. In addition, annealed and belt-abraded specimens for the study of the effect of pickling Zircaloy-2 surfaces prior to bonding were also prepared, assembled, and bonded under the same pressure-bonding parameters of 10,000 psi at 1550 F for a period of 4 hr. Sections from all of these specimens were submitted for metallographic preparation. This program is being terminated with this report, but the evaluations of the bonds obtained for these specimens will be reported in other programs.

Fueled Zirconium Hydride Moderator

H. E. Bigony and H. H. Krause

No work was made during this month on the investigation of hydrides of uranium-zirconium alloys as possible fueled moderators.

I-1

I. PHYSICAL RESEARCH

F. A. Rough

The research reported below is part of the AEC Division of Research program.

This month reports of progress on a part of the projects are included. These cover studies of the properties of uranium compounds, of uranium-base ternary alloys, and of the structure of UC_2 . The balance of the active projects was last reported in BMI-1267.

The thermal-conductivity data for UBe_{13} and UC, which conclude the current phase of the uranium-compound program, were not available in time to be included in this report. Some details of preparation of the carbides are reported.

Analysis of the UC_2 structure indicates that the carbon bond is a double bond. The carbon position parameter is 0.388 ± 0.002 , giving a carbon-carbon distance of 1.34 ± 0.02 A. The carbon-carbon bond distance in the sesquicarbide is 1.35 ± 0.05 A.

Preparation, Fabrication, and Physical Properties
of Uranium Compounds

A. B. Tripler, Jr., M. J. Snyder, and W. H. Duckworth

Methods of ceramic fabrication and fundamental properties of some refractory uranium compounds which are potential nuclear fuels are being studied.

The potential advantage of using uranium balls in place of porcelain or alumina balls in milling uranium compounds preparatory to compacting and sintering was investigated. Arc-melted samples of UC and UC_2 were employed in the study. The monocarbide was milled for 4 hr in a rubber-lined mill with uranium balls. The average particle size of the milled material was 5.8μ compared with an average particle size of 15μ in a previous sample milled with alumina balls. The impurity content of the material milled with the uranium balls was appreciably lower, particularly in aluminum, iron, and silicon, than that of the sample milled with alumina balls. Within the limits of analysis, no significant increase in the uranium content of the monocarbide occurred.

An arc-melted sample of UC_2 first was milled for 4 hr with alumina balls. Visual examination showed the material to be relatively coarse after this period. The sample was subsequently milled with uranium balls in a rubber-lined mill. After the additional 4-hr milling, the average particle size was 3.9μ , compared with about 20μ for material milled for more than 8 hr with alumina balls. Again, impurity contents were appreciably lower than in previous samples milled to fine powders entirely with alumina balls.

Thermal-conductivity measurements are in progress on samples of UBe_{13} and UC.

A topical report is being prepared covering physical and chemical properties of UC, UC_2 , UB_2 , and UBe_{13} .

Constitution of Uranium-Base Ternary Alloys

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

An investigation of the uranium-niobium system and the effect of oxygen on the constitution of the binary system is being conducted using electrolytic uranium, ultrahigh-purity (bromide) niobium, and, in the case of alloys with oxygen, high purity-high oxygen iodide niobium. Questionable phase regions, namely, the eutectoid region and the γ_1 plus γ_2 / γ boundary, will be studied. Diffusion couples of uranium and niobium will be heat treated at temperatures above and below the peak of the γ_1 plus γ_2 loop. Electron-beam microanalysis will be used to determine composition limits of the γ phase and in calculating diffusion coefficients. Alloys of uranium with 2, 6, 20, 28.1 and 40 w/o niobium and alloys with 20, 28.1 and 40 w/o niobium plus approximately 700 ppm oxygen have been cast and are now being homogenized and hot worked. These alloys will be utilized for initial studies of the system.

Structure of UC_2

A. E. Austin and C. M. Schwartz

The positions of carbon atoms in UC_2 have been determined from powder neutron-diffraction data. The carbon position parameter is 0.388 ± 0.002 , which gives a carbon-carbon distance of 1.34 ± 0.02 Å. Thus, the carbon bond is essentially a double bond. Analysis of neutron-diffraction data for uranium sesquicarbide has indicated that the carbon-carbon bond distance is about 1.35 ± 0.05 Å.

J-1

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

C. L. Peterson, P. D. Miller, E. F. Stephan, O. M. Stewart,
J. D. Jackson, W. C. Baytos, T. E. Snoddy, and F. W. Fink

The evaluation of materials of construction for use in the various proposed processes for the recovery of spent reactor fuel elements has been continued.

The use of titanium as a material of construction for the dissolver and feed-adjustment tank in the Darex process still seems promising. Studies are under way concerning the removal of the scale that accumulates on these vessels, but serious difficulties are not anticipated.

Carpenter 20 Cb may be a possible material of construction for the dissolver vessel in the Sulfex-Thorex process if use is made of the combined cathodic protection from the dissolving stainless anodes and the inhibition from the dissolution products which accumulate in the solution.

A severe intergranular attack has been noted in the heat-affected areas of Nionel specimens exposed to the Thorex solutions. A suitable remedy for this attack will be sought. Type 329 stainless steel, Incoloy 804, the Hastelloys C and F, and crystal-bar zirconium are attacked by either the Thorex or Sulfex solutions and are not being considered for use in a dissolver in which both dissolutions are conducted.

Presently, INOR-8 appears to be a more promising material of construction than INOR-1 for the hydrofluorinator in the fluoride-volatility process using the equimolar NaF-ZrF₄ mixture.

The Darex Process

Uranium can be recovered from fuel elements containing stainless steel as a diluent or cladding by means of the Darex process. The elements are first dissolved in dilute aqua regia. The chlorides are stripped from the solution with concentrated nitric acid and, following suitable adjustment, the dissolved uranium is recovered by conventional methods of solvent extraction.

Dissolver Studies With Titanium

Operation of the flowing dissolver was terminated following 2000 hr under dissolution conditions. All the specimens from the liquid phase were covered with the usual hard, green, siliceous scale. Vigorous scrubbing did not remove this scale, but treatment with boiling 10 w/o NaOH was effective. The evaluation of these specimens is not yet complete, but preliminary examination indicated no adverse effects, corrosionwise, from this long exposure.

A new flowing-dissolver experiment is nearly ready to be started. A cleaning regimen with boiling 10 w/o NaOH will be instituted every other week in this apparatus to determine how such cleaning may affect the corrosion of the titanium specimens. During this experiment, a study will be made of the effects of injuries to the protective oxide films of titanium specimens exposed to the dissolution vapors at points where rather low nitric acid concentrations are expected.

FAT Studies With Titanium

The various types of titanium specimens exposed to boiling Initial FAT solutions were examined following 3500 hr of exposure. The specimens were covered by dark surface films but appeared to be in excellent condition.

Identification of the crystalline scale which forms on the submerged surfaces of titanium steam tubes exposed to Final FAT solutions was attempted by means of X-ray diffraction, but the results were inconclusive because of the lack of a similar pattern in the reference standards. A semiquantitative analysis by emission spectrography showed this scale to be predominantly iron and chromium with some uranium but very little nickel or silicon. It appears that some softening of this scale occurs with water and dissolution can be accomplished with boiling solutions of nitric acid, as well as other related solutions.

The Sulfex-Thorex Process

The evaluation of various metals for use as container materials for the dissolution step of a proposed Sulfex-Thorex process has continued. In this particular process, stainless-clad fuel elements of thorium or thoria would be dejacketed by dissolution in sulfuric acid. Following this, the thorium or thoria would be dissolved by a solution of 13.0 M HNO_3 , 0.05 M F^- , to give a final solution of about 8.5 M HNO_3 , 0.05 M F^- , and 1.0 M $\text{Th}(\text{NO}_3)_4$.

Experiments With Carpenter 20 Cb

No further cyclic studies have been conducted with specimens of Carpenter 20 Cb, nor are any more planned until more has been learned concerning the cracking that has already been observed.

As reported in BMI-1267, heat treating appeared to decrease the tendency of the specimens to crack around areas of surface deformation upon exposure to boiling sulfuric acid solutions. Minute cracks were observed to develop on heat-treated specimens in areas where no known prior surface deformation had occurred. Continued exposure of these specimens in the boiling acid solution (6 M H_2SO_4) resulted in these cracks becoming grossly enlarged. This is a serious type of attack unless further experimentation shows that it can be reliably controlled by proper heat treating or by addition of inhibitors to the solution.

J-3

Scouting experiments have indicated that cracking will not develop during a 96-hr exposure period in boiling 6 M H_2SO_4 in the presence of 5 g per liter of dissolved stainless steel, but cracking is not prevented by additions of only 1 g per liter. Further experiments continue to indicate that Carpenter 20 Cb can be afforded cathodic protection against cracking by the action of a coupled, dissolving stainless steel anode.

Experiments With Nionel

A study was carried out to see if the corrosion rates of Nionel specimens were adversely affected upon continued exposure to boiling 6 M H_2SO_4 by the presence of dissolution products. The rates were found to decrease in all cases with continued exposure and to be much lower in the presence of 5 g per liter of dissolved stainless steel than when no dissolution products were present. Additions of 35 g per liter of stainless steel resulted in a further decrease in corrosion rates. No cracking and no selective or localized attack were observed.

Welded specimens of Nionel are now being studied during cyclical exposure from Sulfex to Thorex solutions. Following five cycles, an examination showed that a serious intergranular attack is occurring in the heat-affected areas next to the weldments in specimens which have been exposed to Thorex solutions. This attack seems to be much more aggravated in specimens which have been exposed to the Initial Thorex solution. A remedy for this attack will be sought.

Scouting Experiments With Other Metals

Scouting-type experiments have been conducted with other metals in boiling Sulfex and Thorex solutions. Specimens of Type 329 stainless steel, the Hastelloys C and F, Incoloy 804, and crystal-bar zirconium were studied. None of these materials will withstand the attack of both solutions. Type 329 stainless steel and Incoloy 804 dissolve readily in the Sulfex solutions, but show good resistance to the Thorex solutions containing aluminum additions. Hastelloy F has a high rate in 6 M H_2SO_4 , while the corrosion rate of Hastelloy C is excessive in Initial Thorex solution, even in the presence of added aluminum. Zirconium looks very promising in the Sulfex solutions, but does not show good resistance to the Thorex solutions.

The Fluoride-Volatility Process

Fuel elements containing zirconium as a diluent or cladding can be recovered by a fluoride-volatility process. The first step consists of hydrofluorination of the elements in a molten bath of NaF-ZrF_4 with a stream of HF . Various materials are being evaluated for use in constructing this hydrofluorinator.

Exposure periods of 1010 hr have been completed with INOR-1 and INOR-8 specimens immersed in the molten equimolar NaF-ZrF_4 mixture at 650 C using melt containers constructed from Hastelloy B. The corrosion rates as measured both by loss in weight and by metallographic determination of the sound metal remaining are tabulated below for the specimens immersed in the molten salt at 650 C.

J-4

Specimen Material	Exposure, hr	Corrosion Rate, mils per month		Remarks
		Weight Loss	Metallographically	
INOR-8	500	1.2	--	Slight etching
	1010	1.5	1.7	Porous metal layer
INOR-1	500	2.1	--	Etching and notch
	1010	1.7	5.2	Porous metal layer

It can be seen that both materials show relatively low rates as compared with materials, such as nickel, Inconel, and Monel, evaluated earlier. Metallographic sections of these specimens showed no deep ingress of intergranular attack or selective penetration.

Specimens of INOR-8 and INOR-1 exposed for about 500 hr in the vapor above the molten salt showed penetration rates of 0.5 and 1.5 mils per month, respectively.

In general, these data show that INOR-8 has slightly better resistance to corrosion than INOR-1.

Measurements of the amount of hydrogen in the effluent gas stream indicated a much lower corrosion rate for the Hastelloy B containers than for the Inconel containers used previously.

K-1

K. DEVELOPMENTS FOR SRE, OMRE, AND OMR

F. A. Rough and J. E. Gates

Reported below are the metallurgical and postirradiation studies currently in progress.

EVALUATION OF URANIUM MONOCARBIDE
AS A REACTOR FUEL

F. A. Rough

This program is being carried out to develop uranium carbide as a fuel for the SRE.

Much of the development of techniques for preparing UC samples by casting is complete, and a major portion of the enriched specimens is available for irradiation. Progress of various phases of the program is reported below.

Casting Techniques for the Preparation of
Uranium Monocarbide

A. C. Secrest, E. L. Foster, and R. F. Dickerson

The determination of the physical properties of uranium monocarbide and the preparation of specimens for irradiation at the Materials Testing Reactor and the Battelle Research Reactor are the major goals of a program to evaluate this material as a reactor fuel.

Four UC castings have been made using uranium enriched with 8.4 w/o uranium-235. These 3/8-in.-diameter by 2-in.-long specimens will be loaded into the second and third capsules being sent to the MTR. Six identical specimens were prepared to serve as controls. Twelve enriched monocarbide castings, 1/4 in. in diameter by 1/2 in. long, have been prepared by arc melting. These castings were sealed in a capsule preparatory to being irradiated in the Battelle Research Reactor. Six similar castings were prepared as controls.

In thermal-shock tests, as-cast and machine-ground specimens of UC sealed in evacuated Vycor tubes are intact and exhibit no evidence of cracking after 100 thermal cycles (15 min at 900 C, followed by a 15-min cooling period at room temperature). During this test, the specimens were in contact with Type 304 stainless steel. No attack on the stainless was noted.

A preliminary heat-treatment study indicates that heat treating in the range of 1000 to 1500 C may prove beneficial to as-cast UC. Differences in hardness and microstructure were noted in a specimen treated at temperatures in this range. Further investigations of these changes will be made.

A single attempt at hot fabrication has been made. A specimen of as-cast UC was encased in molybdenum and reduced 50 per cent by hot rolling at 1300 C. X-ray and metallographic examination revealed that the monocarbide was severely fractured.

In order to determine the variation in composition of UC prepared by a standard melting procedure, a "proof test" has been made on eight 100-g arc-melted buttons. The castings subsequently made from these buttons are being analyzed for carbon, and metallographic examinations are being made. Results are not yet available.

Another thermal-conductivity determination is now in progress. Simultaneous measurement of the electrical resistivity of this sample is planned.

In addition to the preparation of additional irradiation specimens for the MTR, future plans include heat-treatment studies, thermal-shock tests at 1100 C, and investigation of the effect of varying compositions on the physical properties of cast UC.

Irradiation-Capsule Design for Uranium Carbide

R. B. Price, R. H. Barnes, and W. H. Goldthwaite

The efforts on preirradiation, encapsulation, irradiation, and postirradiation phases of the Atomic International UC radiation-effects program are being continued. Irradiations in both the MTR and BRR are planned.

The first MTR capsule, BMI-23-1, has been loaded and shipped to the MTR. All requirements except the final IDO approval have been satisfied in preparation for the tentative insertion in Position A-28-NE during the Cycle 107 shutdown. A three-cycle irradiation at 1×10^{14} nv is planned. This can be reduced to one or two cycles if desired. The "Plutonium Run" now tentatively scheduled for Cycle 107, may be an important factor in the irradiation of the first and possibly succeeding capsules. All six MTR capsules are thermocoupled capsules.

The capsule bodies and some of the specimen-basket components for the remaining five MTR capsules have been fabricated. Some of the specimens for the next two MTR capsules have been received and are being radiographed as a first step in the preinspection examination.

The BRR capsule (no thermocouples) has been loaded and is scheduled for insertion in Core Position 56. A 30 to 60-day irradiation at 0.5 to 1.0×10^{13} nv is planned. Flux measurements have been made; also, aluminum-0.58 w/o cobalt dosimeter is attached to the outer capsule wall.

The BRR capsule-specimen postirradiation program consists essentially of heat treating the irradiated UC specimens at various temperatures, removing fission-gas

K-3

samples at various time intervals by freezing, and determining the krypton-85 and/or xenon-133 activity, using a gamma spectrometer.

The following burnup figures are given as a rough guide to irradiation planning:

Reactor	Exposure		Equivalent Burnup of Uranium-235, a/o	Over-All Perturbation Factor
	Time, days	Flux (Perturbed), nv		
MTR	15	2×10^{13}	0.14(a)	-0.22
BRR	60	5×10^{12}	0.14(a)	-0.46

(a) Burnup of 0.14 a/o $\approx$ 1000 MWD/T of uranium.

The long-term NaK compatibility studies at 1100 F are continuing.

POSTIRRADIATION STUDIES OF SRE, OMRE,
AND OMR FUEL MATERIALS

J. E. Gates

Postirradiation examination of thorium-11 w/o uranium specimens irradiated in two six-specimen capsule trains is nearing completion. These trains, NAA-15-6 and NAA-15-7, are part of the SRE fuel-evaluation program.

Two irradiated capsules of UO_2 specimens, also part of the SRE program, are expected to arrive at Battelle during July. An examination of these specimens should begin soon after their arrival.

The dummy element containing specimens of structural materials for use in the OMR will arrive during July. Effects of corrosion induced by an organic reactor coolant and radiation on the physical properties of these materials will be studied.

An irradiated OMRE fuel element originally scheduled to be shipped during June is now scheduled to arrive during July. Planning for handling the evaluation of this element is under way, with the actual evaluation to begin as soon as possible after the element arrives.

SRE Fuel Material

G. E. Lamale, J. H. Saling, W. J. Braun,
and J. E. Whitney

Thorium-Uranium Specimens

The postirradiation evaluation of specimens recovered from the NAA-15-7 capsule train is nearing completion. Density measurements on Specimens B, D, E, and F indicated decreases of 2.8 to 4.8 per cent. Since Specimen A had swelled appreciably at one end, density measurements will be confined to the unswelled thermocouple end. Diameter measurements of Specimens B, E, and F indicate increases of 1.02 to 2.07 per cent, and length measurements of the same specimens indicate increases of 1.18 to 1.75 per cent. Two diameter measurements of Specimen D were made even though it appeared friable. These measurements were not sufficiently great in number for a detailed comparison with the preirradiation data.

Gas samples were taken from Specimens A, C, and F of Train NAA-15-7. An amount of fission gas equivalent to 0.02 per cent of the theoretical amount formed based on 1 a/o burnup was recovered from Specimen C. No fission gas was recovered from Specimens A and F. Apparently there was a significant air leak at the capsule-punch seal and it is doubtful that the data on Specimen C are significant.

The second capsule train, NAA-15-6, containing the same type of specimens as NAA-15-7, was received at the hot-cell facility during June. This train has been separated into capsules. Four capsules were sampled for fission gas, and all capsules were opened. The specimens were numbered 1 through 6, starting at the bottom of the train.

The visual examinations of the specimens of NAA-15-6 reveal them to be in good condition. They had a high metallic luster. All specimens were removed from their thermal shields without undue difficulty. Specimens 4, 5, and 6 were somewhat bulged at the ends away from the thermocouples. After the end with the thermocouple was cut off, the remaining 7/8-in. cylinder was used for the measurement of density.

Specimens 1, 2, and 3 showed increases of diameter of 1.29 to 1.64 per cent and increases of length of 1.09 to 1.38 per cent. The range of density decreases for the same specimens was from 2.58 to 4.15 per cent. Densities on Specimens 4, 5, and 6 will be made on the thermocouple ends.

The capsule-puncturing apparatus was modified for Train NAA-15-6. Preliminary results indicate that gas has been recovered from three capsules in this train. The data are being evaluated and will be reported next month.

The evaluation of the specimens in both trains will continue. The planned program includes cutting burnup disks and making metallographic cross sections. In addition, the thermal shield of Specimen D will be sectioned adjacent to the bad area. Specimens B (NNA-15-7) and 2 (NAA-15-6) will be placed in the dilatometer for heat treatment up to 1800 F at a rise of 3 C per min. Evaluation of cesium-137 in NaK will be studied for Specimens A (NAA-15-7) and 1 (NAA-15-6).

K-5 and K-6

Uranium-Dioxide Specimens

Two capsules of uranium-dioxide specimens, NAA-28-1 and NAA-29-1, will arrive during July. The postirradiation evaluation will include a detailed visual examination of the fuel jacket, a detailed examination of the pellets, including weight, dimensions and densities, and stereomacrographs will be made. Metallography will be performed on selected specimens and replicas taken on three specimens.

OMRE Fuel Elements

R. J. Burian, D. K. Dieterly, D. N. Sunderman,
and J. E. Whitney

The first irradiated OMRE fuel element is to be shipped to Battelle in July. Design and assembly of remote equipment is continuing. A dummy run in the hot cells will be made before the actual element is handled. It is planned to initiate the examination of this element soon after its arrival.

OMR Fuel and Structural Materials

R. J. Burian, W. J. Braun, and J. E. Whitney

Two dummy elements containing specimens of various structural materials of interest to the OMR program will be examined. The specimens were designed for tensile, impact, corrosion, and bend tests, and will provide data on the effects of radiation and corrosion by the organic coolant used in the reactor. The first dummy element is scheduled for shipment to Battelle early in July.

L-1

L. STUDIES OF SODIUM-TANTALUM COMPATIBILITY
AT ELEVATED TEMPERATURES

J. H. Stang

The studies reported here are concerned with various phases of the technology of tantalum and are part of the Los Alamos Scientific Laboratory's LAMPRE development program. Included are investigations of creep in flowing sodium and in static helium and weldability. In the creep studies, direct evidence shows that the strength of tantalum is about the same in 1200 F sodium as it is in 1200 F helium.

In the welding studies, emphasis has been placed on correlating the carbon content in tantalum with weld soundness. In recent experiments, sound welds were made in several experimental tantalum-carbon alloys which were prepared from unweldable tantalum. Investigation of the effect of ultrasonic energy applied to tantalum during and subsequent to welding to reduce grain size has been terminated. It has been shown that maximum refinement can be effected by applying ultrasonic energy during welding; however, the coupling problem may limit this process to relatively simple structures.

Tantalum-Sodium Compatibility Studies

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

During June, activity in this program, which is concerned with determining the creep of tantalum specimens immersed in 1200 F sodium circulating in polythermal loop systems, was as follows:

- (1) Five standard creep experiments, started in April, were concluded.
- (2) One special creep experiment in which the creep specimen was exposed for 455 hr to flowing sodium prior to application of load was continued.
- (3) One control experiment with helium as the specimen environment was started and concluded.

The test material in these runs was commercial arc-cast tantalum from a single lot which, as discussed in BMI-1267, did not reach a sufficient temperature during the original annealing treatment to provide a full anneal.

Table L-1 presents creep data from these various runs. Among the principal points indicated by these data are the following:

- (1) At 26,000 psi, the minimum creep rates for the standard exposures and the helium control exposure are comparable. This result leads to the general conclusion that effects from sodium exposure are not particularly important in the tantalum-creep situation at the 1200 F level. Posttest inspections of various specimens confirm this point since unusual corrosion effects have not been found.

L-2

TABLE L-1. CREEP DATA FOR PARTIALLY ANNEALED ARC-CAST TANTALUM SPECIMENS AT 1200 F

Stress, psi	Time in Progress, hr	Time to Reach Indicated Percentage Deformation, hr				Total Deformation, per cent	Minimum Creep Rate, per cent per hr
		0.1	0.2	0.5	1.0		
<u>Standard In-Sodium Exposures^(a)</u>							
20,000 ^(b)	258	2	5	--	--	0.40	0.0001
22,000	1100	4	240	--	--	0.43	0.0001
26,000	1090	<1	3	12	264	1.74	0.0007
26,000	934	<1	1.8	7	18	2.00	0.0004
26,000 ^(b)	998	<1	<1	4	130	4.36 ^(c)	0.0012
<u>Special In-Sodium Exposure^(d)</u>							
26,000	712	<1	<1	<1	7	2.23	0.0019
<u>Control Exposure in Helium</u>							
26,000	600	<1	14	300	--	0.64	0.0003

(a) Load applied at start of exposure.

(b) The fluid temperature cycle in these experiments was 160 F. In other sodium tests, it was 80 F.

(c) Part of this unusually high total deformation was incurred during startup periods following power-failure shutdowns.

(d) Load applied after 455 hr to reduce the oxygen content of the test piece prior to initial loading.

L-3

- (2) While the first-stage creep (and, consequently, total final deformation) of specimens in the sodium runs is consistently quite high, this does not reflect specimen behavior but rather is traceable to a short-coming of the creep-monitoring technique used in conjunction with the flow-loop systems. This technique is based on measuring the total deformation of the specimen rather than gage-section deformation; while, as explained in previous reports, the specimens are strengthened at the grips to avoid excessive distortion in the vicinity of the pinholes, enough still occurs to be measurable. However, it seems to be confined to first-stage creep, presumably because the pinhole quickly deforms to accommodate the pin and, consequently, local stresses are lowered to a level that does not foster continued distortion.
- (3) In the special in-sodium exposure, the current minimum creep rate appears to be somewhat higher than those for the standard exposures. It seems likely that the reduced strength indicated stems from the fact that the oxygen content of the tantalum specimen was reduced by the prior exposure to sodium. (Corrosion exposures of tantalum to high-purity sodium demonstrated conclusively that there is a migration of oxygen into the sodium.) However, it must be remembered that commercial tantalum is not a uniform product and, hence, that a relatively small variation in creep behavior may denote only a variation in material.

Further sodium experiments are not planned in this program. The only remaining experiment contemplated is a duplicate control run in helium with the partially annealed material. It might be mentioned that in this test specimen creep will be monitored both by an internal extensometer and an external dial gage in an attempt to obtain more information about the pinhole-deformation problem described above.

High-Temperature Mechanical Properties of Tantalum

D. C. Drennen, M. E. Langston,
C. J. Slunder, and J. G. Dunleavy

Analyses of thermally degassed fine-grained tantalum produced for inert-atmosphere creep testing by thermal degassing, cold rolling, and vacuum annealing are listed below along with analyses of the parent as-received sintered bar.

	<u>As-Received</u> <u>Sintered Bar</u>	<u>Treated</u> <u>Material</u>
Analysis, ppm		
Oxygen	68	10-16
Nitrogen	40	20
Carbon	40	20-30
Hydrogen	0.2	0.7
Hardness, VHN	84	73

L-4

Metallographic examination showed that the average grain diameter was reduced from about 0.100 mm for the original material to about 0.030 mm after cold rolling (80 per cent reduction) and annealing the thermally degassed material.

Creep tests at 1200 F in helium are in progress on 30-mil strip specimens of the specially prepared material. Results will be compared with those from 1000-hr tests previously run on annealed arc-cast, annealed sintered, and thermally degassed coarse-grain sintered tantalum.

Weldability of Tantalum for High-Temperature Systems

S. M. Silverstein and R. P. Sopher

The objective of one of the phases in this program is to determine underlying reasons why arc welds in some commercial lots of tantalum are porous while consistently sound welds can be made in other lots purchased from the same source. Studies conducted to date have indicated the possibility of a relationship between carbon content and the extent of porosity. In attempts to determine the specific correlation, samples were prepared by diffusing carbon into tantalum while it was heated to elevated temperatures. However, as discussed in BMI-1267, examination of the treated specimens showed a nonhomogeneous carbon distribution which made it difficult to study the weld-metal porosity problem. During June, various amounts of carbon were added to samples of tantalum while in the molten state during arc melting. Four samples were prepared in which 20, 50, 100, and 200 ppm of carbon were added to an unweldable lot of tantalum (containing 10 ppm carbon). The samples were melted in a tungsten-electrode arc furnace under approximately 1/3 atm of helium. A water-cooled copper crucible was used for the experiments. Subsequently, the arc-melted buttons were fabricated to 0.020-in. sheet. Welds made on these four materials were radiographically and metallographically free of porosity. In addition to obtaining complete analyses of the test materials, work is now in progress to observe the effects of arc melting alone on the weldability of the unweldable base material that has been used.

The study to determine the effect on grain size of applying ultrasonic energy to weldments at approximately 1100 C has been terminated. Fragmentation or mechanical breaking up occurred rather than grain refinement. As a result, a contiguous grain structure existed which probably would have both low strength and ductility.

The work on the effects of applying ultrasonic energy during welding also has been terminated. This study has shown that grains which were one-third the size of unexcited tantalum could be produced in a weld zone. However, the difficulty involved in coupling an ultrasonic source to a specimen limited all work to simple bead-on-plate welds. This coupling problem would tend to limit the application of ultrasonic energy during welding to relatively simple structures.

M-1

M. DEVELOPMENTAL STUDIES FOR THE PWR

R. W. Dayton

The present phase of the program for studying improved mixing in a PWR air-flow model is being concluded with the preparation of a report on the results of the study.

The use of color film in the panoramic camera for examining plate-type sub-assemblies has been found to improve the ability of the camera to discriminate between various types of defects.

The major developments in the study of pressure-bonded oxide fuel plates have been the preparation of a considerable number of plates by the best process, the observation of brittleness in the Zircaloy-2 when Ti-Namel, but not stainless steel, jackets are used, the successful bonding of plates without jackets and, also, of plates using receptacle plates assembled from Zircaloy-2 strips.

Reactor-Flow Studies

L. J. Flanigan and H. R. Hazard

Air-flow studies in a quarter-scale model of the PWR are being conducted to determine the effects of lower-plenum geometry on mixing and flow distribution in Core 2. Previously reported work includes studies of mixing in the lower plenum with eleven different lower-plenum configurations.

In June, the swirl-vane configuration, with vanes positioned at an angle of 30 deg from a radius and with a shield coolant-flow rate of 19.6 per cent of model flow, was selected as a best compromise between mixing and pressure drop. With this configuration, studies were made to determine core-flow distribution, shield velocities and directions, flow-baffle pressure loss, and mixing for two-, three-, and four-loop operation. The studies were made with the basic 7.5-ft core design and with the basic 7.5-ft core design with swirl vanes.

A report covering all the studies made with the 7.5-ft core design up to June 30 is being prepared. The studies made previously with the 9-ft core design are included in the report. This completes the present phase of this study.

Improved Techniques for Corrosion Detection
in Subassemblies and Clusters

G. G. Cocks and C. M. Schwartz

It is the purpose of this work to investigate means of interpreting the photographs produced by the panoramic camera, and to improve the optical system in order to

facilitate interpretation and recognition of the various surface features, such as corrosion, acid stain, and topography.

During the past month, photographs of water-channel walls were taken on color film and on black-and-white film, using both plane and cylindrical probe mirrors. The surfaces of the water channels had corrosion marks, dents made by hammering, and large shallow depressions (also resulting from hammering). These surface defects are recognizable when color film is used with either the flat or the cylindrical probe mirror. Apparently, the use of color film will make it possible to distinguish corrosion spots from these surface defects.

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

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

A flat-plate Zircaloy-2-clad fuel element containing compartmented uranium dioxide fuel is being considered for PWR Core 2. An investigation is being conducted to study the preparation of these elements by gas-pressure bonding. In this process, the components are fabricated to final size, cleaned, and assembled into a Ti-Namel envelope which is then evacuated and sealed. The assembly is subjected to helium-gas pressure at elevated temperatures to obtain bonding of the components. After removal from the envelope, the element is heat treated in a barium chloride salt bath at a temperature in the beta-phase region for Zircaloy-2. Studies have shown that a bond possessing incomplete grain growth across the interface can be improved by a beta heat treatment. The transformation is accompanied by an increase in grain size and sufficient driving force to induce additional grain growth across the interface. A large number of fuel-element samples have been bonded and are being subjected to numerous evaluation tests to attempt to determine the optimum procedures for preparing these elements. Three sets of specimens have been bonded for irradiation testing. All of these elements were bonded at temperatures from 1450 to 1550 F for 4 hr at a pressure of 10,000 psi and were then heat treated at 1850 F for 5 min and rapidly cooled.

A series of 75 specimens approximately 1/8 by 2-3/8 by 11-1/4 in. has been pressure bonded for a comprehensive evaluation of fabrication procedures. The tests employed for these elements include intercompartment leak tests, deflection and bursting of compartments, corrosion of purposely defected compartments, waterlogging tests, metallographic examination of bond quality and fuel-cladding reaction, bend and peel tests, and corrosion of the Zircaloy-2 cladding. The cladding components for these specimens had belt-abraded bonding surfaces. The uranium dioxide cores were coated with graphite to prevent fuel-cladding reaction; the Ti-Namel spacers placed between the envelope and the cladding were chromium plated to attempt to avoid diffusion of iron into the Zircaloy-2. The specimens have been placed on test. Results of peel tests and intercompartment leak tests have indicated good bond integrity.

The cladding of many of these specimens became embrittled during the pressure-bonding process. This embrittlement resulted from a pickup of hydrogen and diffusion of iron from the chromium-plated Ti-Namel spacers. To avoid hydrogen contamination,

M-3 and M-4

the spacers were outgassed at a temperature of 350 F. This eliminated most of the hydrogen pickup. The diffusion of iron is apparently governed by the type and thickness of chromium plate. Although the same plating technique was utilized for all of the spacers, the results were inconsistent. Some of the bonded specimens exhibited extensive iron diffusion, whereas others showed no evidence of iron diffusion and embrittlement. Specimens are now being prepared with a thin stainless steel barrier layer between the Ti-Namel spacers and the cladding. The stainless spacers prevent iron diffusion; however, they do not flow as readily during pressure bonding as the chromium-plated Ti-Namel spacers.

Most of the pressure-bonded specimens exhibited an edge effect. This is manifested by poor flow of metal at the edge of the specimens with the result that poor bonding is usually obtained a distance of about 1/8 in. from the edge toward the center along the entire edge of the specimens. An increase in the thickness of the pack by use of heavier spacers promotes flow at the edges and eliminates this effect. It may, however, prove to be less costly to trim the edges of the specimens after bonding.

The possibility of eliminating the Ti-Namel envelope by fusion edge welding the fuel-element components before pressure bonding appears promising. Five elements with belt-abraded Zircaloy-2 components were edge welded and pressure bonded. Preliminary tests indicate that good bonding was attained. The cladding is ductile and does not appear to be contaminated. This method of preparing specimens would reduce costs and eliminate the problems associated with spacer reactions and edge effects.

Specimens have also been prepared using strip components to form the receptacle plate of the element, which may have an economic advantage over the one-piece punched receptacle plate. Elements have been bonded using three different frame configurations. These specimens are being evaluated for bond integrity between compartments. In addition, two specimens approximately 3 ft long have been prepared for pressure bonding to determine the feasibility of bonding larger elements. One of the specimens has a one-piece punched receptacle plate, and the other has a receptacle plate assembled from small strips.

Three sets of irradiation specimens have been fabricated using belt-abraded components and graphite-coated cores. One series of specimens exhibited good bonding, but extensive iron contamination of the Zircaloy-2 cladding occurred through the chromium plating of the Ti-Namel spacers.

N-1 and N-2

N. DEVELOPMENTS FOR THE MGCR

W. H. Goldthwaite

This research is part of the MGCR program. A design concept which is of considerable interest features carbon dioxide as the coolant and graphite as the moderator. In this concept, coolant temperatures up to 1500 F, at a pressure of 2000 psi are anticipated, and the graphite must be clad with a material resistant to CO₂ at this temperature.

The effect of radiation on the design materials is being studied at Battelle.

Investigation of the Effect of Irradiation on Clad
Graphite Specimens in a CO₂ Environment

J. C. Smith, R. H. Barnes, and W. S. Diethorn

A program has been initiated to investigate the effects of radiation on clad graphite specimens in a 1500 F 1000-psi CO₂ environment. A one-capsule experiment is being conducted in which six AGOT graphite specimens clad with Type 310 stainless, Type 446 stainless, or Inconel will be irradiated for 5 weeks in the Battelle Research Reactor. Two specimens of each will be irradiated. The results of postirradiation examinations, including metallography, will be compared with results from duplicate out-of-pile exposures conducted elsewhere for evidence of the effect of radiation on corrosion by the gaseous environment and reactions at the graphite-cladding interface.

The capsule has been designed, fabricated, and loaded. Out-of-pile proof tests are under way. The irradiation will start in early July; the results are to be available by early September.

Design and Operation of the Irradiation Capsule

The irradiation capsule, about 18 in. long by 1-3/4 in. in diameter, features three 1000-w electric heaters brazed to a 7/8-in.-OD furnace tube within which the specimens can be heated to 1500 F. The furnace tube is open to the outer pressure shell, which will be cooled by the reactor pool water to about 160 F. Four thermocouples will be arranged to provide information on the axial and radial temperature gradients at the specimen location. All materials of construction are Type 304 stainless steel except for the specimen basket fabricated from Type 310 stainless 1/16-in.-diameter rods.

During irradiation, the capsule will be connected to a CO₂ tank and gas-sampling equipment. Gas samples will be taken at the beginning of the exposure and at approximately 6, 24, and 72 hr later. Analyses of these initial gas samples will indicate how frequently the gas should be changed during the 5-week irradiation in order to simulate some aspects of a dynamic gas environment with a static test.

O-1

O. DEVELOPMENTS FOR NMSR

A. W. Hare and R. F. Dickerson

These studies are part of the Nuclear Merchant Ship Reactor program. Research currently in progress is concerned with the fabrication of UO_2 fuel pellets and loop-test fuel pins and the design and construction of irradiation-test capsules.

Fabrication of Urania Fuel Pellets for Loop-Test Studies

H. D. Sheets, C. Hyde, and A. G. Allison

This program is aimed at the preparation of UO_2 specimens containing 3 to 7-1/2 per cent U^{235}O_2 for the NMSR program. Specimens having bulk densities of about 91 per cent of theoretical are desired.

Previously, processing conditions for fabricating pellets having the required densities were determined and fifty 1/4-in. -diameter 1/4-in. -long and two hundred 1/2-in. -diameter 1/2-in. -long pellets suitable for in-pile testing were fabricated.

During June, an additional 140 satisfactory pellets 1/4 in. in diameter and 1/4 in. long, and 160 satisfactory pellets 1/2 in. in diameter and 1/2 in. long were prepared. Spectrochemical analyses of representative specimens of three of the four lots of UO_2 are given in Table O-1.

Additional UO_2 powder containing 3 per cent and 6 per cent of U^{235}O_2 will be required to complete fabrication of the necessary pellets.

TABLE O-1. SPECTROGRAPHIC ANALYSES OF UO_2 SPECIMENS

Enrichment, per cent U^{235}O_2	Impurity Content, ppm													
	Fe	Be	B	Si	Mn	Mg	Pb	Ni	Mo	Cu	Co	Ca	Al	Cr
2.83	60	<0.5	3	40	2	40	<2	500	50	15	10	250	50	30
2.83	80	0.5	1	140	4	150	<5	400	160	35	50	200	900	15
4.06	60	<0.5	3	50	4	50	<2	500	40	50	20	250	100	60
4.06	60	<0.5	3	50	3	40	<2	500	40	25	20	250	15	60
6.05	60	<0.5	2	15	2	15	<2	40	25	10	20	50	100	10
6.05	40	<0.5	1	10	1	10	<2	30	20	5	5	40	20	5

Fabrication of Loop-Test Fuel Pins

S. Alfant, A. W. Hare, F. A. Rough, and R. F. Dickerson

The fabrication of fuel pins for the test loop is still in progress. The test pins for the first in-pile loading have been completed, however, the fabrication of the necessary out-of-pile and control specimens for the first loading and the in-pile, out-of-pile, and control specimens for the second loop loading have been temporarily delayed. These in-pile specimens share the loop irradiation facility with other irradiation specimens which are currently being fabricated. As soon as these specimens are fabricated they will be loaded into fuel-pin holders and shipped to the MTR for irradiation tests.

Preirradiation Measurements

R. B. Price, R. H. Barnes, J. C. Smith,
D. B. Hamilton, and W. H. Goldthwaite

In-Pile Loop Program

Work continued at a low rate of effort as no fuel pins or pin assemblies were available for identification and preirradiation measurements.

Fuel Capsule-Irradiation Program

The capsule-irradiation program to evaluate the effects of nuclear radiation on the proposed fuel elements for the NMSR is continuing.

The capsule design has not progressed past the conceptual stage as some of the details of the program have not been definitely decided upon.

CRT:RWD/all