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PROGRESS RELATING TO CIVILIAN
APPLICATIONS DURING DECEMBER 1957

By
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January 1, 1958

Battelle Memorial Institute
Columbus, Ohio

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REPORTS RELATING TO CIVILIAN APPLICATIONS
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- BMI-1228 "Vapor Deposition of Molybdenum and Niobium Coatings on Stainless Steel Tubes", by Carroll F. Powell, Danny M. Rosenbaum, Robert B. Palmer, and Ivor E. Campbell.
- BMI-1234 "A Quality-Control Corrosion Test for Uranium-Niobium Fuel Elements", by Warren E. Berry, Norman E. Daniel, Paul D. Miller, Frederick W. Fink, and Ronald F. Dickerson.
- BMI-1238 "Progress Relating to Civilian Applications During November, 1957", by Russell W. Dayton and Clyde R. Tipton, Jr.

A-1

A. DEVELOPMENTS FOR ZIRCONIUM-CLAD FUEL ELEMENTS

F. R. Shober

Several additional calibration runs have been made with test equipment designed and constructed to measure the thermal conductivity and electrical resistivity of zirconium-clad uranium before and after irradiation. Calibration of an apparatus to measure the thermal conductivity of UO_2 has shown the need for modification. Creep tests are continuing on 15 per cent cold-worked Zircaloy 2 at elevated temperatures.

Work has been initiated on an evaluation of the bond formed between nickel-plated uranium and aluminum by pressurizing at 950 F and 10,000 psi. A hot-hydrostatic-pressing technique is being used to obtain necessary bonding pressures. Growth of single crystals of molybdenum and zirconium is being attempted in an Andrade-type furnace.

Thermal Conductivity of Uranium and UO_2

H. W. Deem and C. F. Lucks

Programs directed toward determining the effect of irradiation on the thermal and electrical conductivities of uranium and on the thermal conductivity of uranium oxide are in progress.

Uranium

Exploratory work will determine the feasibility of making thermal-conductivity and electrical-resistivity measurements on pre- and postirradiated uranium, clad in Zircaloy 2, with NaK as the heat-transfer medium. A steady-state comparative method is being used to measure thermal conductivity. Electrical-resistance measurements will be made on the same specimen by the voltage-drop method. Calibration runs are in progress. The thermal-conductivity results are satisfactory, but some inconsistencies exist in the electrical-resistivity data at elevated temperatures.

Uranium Oxide

An apparatus for making thermal-conductivity measurements on UO_2 , both before and after irradiation, has been completed. A calibration run made to 450 C on a graphite specimen showed the need for a better calibration of thermocouples and/or better specimen guarding. A more elaborate guarding system is almost completed. All shop work has been completed on this project and further calibration runs will be made in January.

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Mechanical Properties of Zirconium Alloys

F. R. Shober and J. A. VanECHO

The creep strength of 15 per cent cold-worked Zircaloy 2 is being investigated at elevated temperatures to determine whether the increased strength achieved by cold working is retained for long periods under creep conditions. The investigation is being conducted at stress levels sufficient to limit total creep deformation to less than 2.0 per cent and to produce a minimum creep rate of the order of 10^{-5} per cent per hr. Eight tests are in progress.

The tests on the 15 per cent cold-worked Zircaloy 2 are in the 450 to 650 F temperature range and at stress levels ranging from 15,000 to 40,000 psi. The results obtained to this time are given in Table A-1. The test at 30,000 psi and 650 F showed a slight increase in creep rate during the last 500 hr of test. This may have been the result of the instability of the cold-worked material or the initiation of third-stage creep. A metallographic specimen will be obtained from this material to determine whether or not recrystallization has started. The remaining tests will be continued to 2000 hr.

Zirconium Cladding

F. R. Shober and R. F. Dickerson

Hot hydrostatic pressing is being investigated as a technique for bonding zirconium and aluminum to electropolished and nickel-plated uranium cores, respectively. Zirconium cladding experiments have been completed except for the preparation of one additional specimen to be evaluated at another laboratory. Pressurizing of a series of aluminum-clad, nickel-plated uranium cores will be started.

Evaluation of a specimen pressurized for 1 hr at 10,000 psi and 1350 F was made by metallographic examination and by bond-strength tests. Based on these tests and others, it appears that the best conditions for bonding at 1350 F are 2 hr at 10,000 psi. One additional specimen will be prepared for another laboratory in this manner and shipped with one prepared previously at 1200 F.

A series of 12 nickel-plated aluminum-clad specimens will be pressurized and evaluated in much the same manner as the zirconium-clad specimens. This series consists of six solid specimens prepared by fitting the cores to the cans and cold closing the ends by press forging. The other six are internally and externally cooled fuel elements, generally referred to as "I and E", having a 1/2-in. hole through the longitudinal axis of the element. These have been prepared by sizing the cans to the core and welding the ends in place. The initial test specimens will be solid-core elements pressurized at 10,000 psi and 950 F for 5 min or for 1 hr. It has been indicated that these times are the minimum and maximum times for optimum bonding conditions. The test conditions for the remaining specimens will be decided upon following evaluation of the first two.

TABLE A-1. CREEP PROPERTIES OF 15 PER CENT COLD-WORKED
ZIRCALOY 2 AT ELEVATED TEMPERATURES

Specimen	Temperature, F	Stress, psi	Time on Test ^(a) , hr	Deformation at Indicated Time, per cent								Creep Rate ^(b) , per cent per hr
				Load On	50 Hr	100 Hr	500 Hr	1000 Hr	1500 Hr	2000 Hr	Load Off	
7-1-2	450	40,000	1750	0.400	0.503	0.512	0.545	0.560	0.567	--	--	0.00003
7-1-3	450	30,000	1675	0.280	0.333	0.337	0.350	0.355	0.355	--	--	<0.00001
7-2-2	550	35,000	1450	0.353	0.495	0.520	0.595	0.625	--	--	--	0.00006
7-2-3	550	30,000	1350	0.275	0.375	0.385	0.415	0.440	--	--	--	0.00005
7-3-1	550	25,000	1550	0.205	0.267	0.282	0.305	0.323	0.33	--	--	0.000036
7-3-2	550	20,000	1515	0.178	0.223	0.227	0.245	0.250	0.265	--	--	0.00001
7-1-1	650	30,000	2015.6 ^(c)	0.315	0.537	0.592	0.745	0.875	1.025	1.183	0.882	0.00028 ^(d)
7-2-1	650	25,000	1225	0.230	0.385	0.420	0.510	0.548	--	--	--	0.000076
7-3-3	650	20,000	1230	0.180	0.300	0.325	0.380	0.397	--	--	--	0.000034
7-4-1	650	15,000	350	0.110	0.185	0.205	--	--	--	--	--	--

(a) Tests in progress.

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

(c) Test discontinued.

(d) Minimum creep rate for 2000-hr test.

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A-4

Preparation of Aluminum, Molybdenum, and Zirconium
Single Crystals

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

The preparation of single crystals of molybdenum and zirconium for irradiation studies is continuing.

A modified Andrade furnace was constructed to grow molybdenum single crystals. Several 1/4-in. -diameter molybdenum specimens were treated in this furnace for relatively short times. Some grain growth was observed. However, as a result of the large diameter of the specimens, sufficiently high temperatures necessary to grow molybdenum crystals could not be maintained. Smaller diameter 1/8-in. molybdenum rods are being prepared and will be tested soon. It is thought that a sufficiently high temperature can be obtained and maintained in the thinner rods to produce crystal growth.

Difficulties have been encountered in the growth of single crystals of zirconium. Strain-anneal methods have produced growth but the length of annealing time required to secure single crystals of any size seems to be prohibitive. Attempts to grow large beta crystals and transform them to large alpha crystals in the Andrade furnace have been unsuccessful. Several additional tests will be run to investigate cooling rates and holding times at the top temperatures. A new method is now being explored. Zirconium melting stock is being alloyed with 4.0 w/o oxygen and cast into small buttons. As this region of the phase diagram shows alpha up to the liquidus, it is thought that large crystals of alpha zirconium will be obtained upon freezing. The hexagonal-close-packed structure will be present at room temperature with slight changes in the lattice parameters due to the addition of the oxygen atoms.

B-1

B. DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS

R. J. Carlson

Work has continued on extrusion cladding flat plates utilizing the right-angle tooling. After two successful tests, the two webs of the four-porthole mandrel cracked during the third run at an extrusion pressure of about 37,000 psi. A new mandrel is being designed which will have added strength at the webs.

The study of the aluminum-uranium system is nearing completion. The liquidus temperatures have been determined for alloys containing up to 50 w/o uranium. No effects of either silicon or magnesium have been detected on the liquidus temperature; however, silicon additions of 0.5 w/o have been found to lower the peritectic temperature from 734 to about 706 C. A topical report is to be prepared.

Preliminary attempts to centrifugally cast extrusion billets of aluminum-25 w/o uranium resulted in sound ingots requiring relatively little machining for cleanup. However, a 3 to 5 w/o variation in uranium content was noted. It is felt that this segregation can be largely eliminated by merely insulating the mold from the turntable. This modification to the equipment is planned for future tests.

Extrusion Cladding of Flat Plates

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

Work was continued on the extrusion cladding of flat plates by the right-angle method.

During the third test with the right-angle extrusion-cladding tools, the two webs of the four-porthole mandrel cracked. The extrusion pressure at the time of the failure was about 37,000 psi. This failure prevented any further work on extrusion cladding with aluminum.

Although the mandrel webs were cracked, a number of steel plates were clad with red and white plasticene, for the purpose of locating and studying the seam between successive billets. The location of this seam is important in determining the bonding between successive aluminum billets.

A new mandrel is being designed which will have added strength at the webs.

B-2

Study of the Liquidus in Aluminum-Uranium Alloys

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

A study of the aluminum-uranium system is nearing completion. Binary alloys containing up to 50 w/o uranium have been investigated along with similar alloys containing silicon and magnesium additions.

From the thermal-analysis data gathered to date, liquidus temperatures appear to be 1120 C for an aluminum-41.7 w/o uranium alloy and 1190 C for an aluminum-51.2 w/o uranium alloy. These temperatures are somewhat lower than the tentative values reported in BMI-1238. No effects of either silicon or magnesium on the liquidus temperature have been detected. However, silicon additions of 0.5 w/o have been found to lower the peritectic temperature from 734 to about 706 C. Additional data are being obtained and it is expected that the investigation will be completed shortly. A topical report will be prepared.

Preparation of Aluminum-Uranium Alloys

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

Aluminum-uranium alloys containing high concentrations of uranium are of interest as possible fuels for low-temperature water-cooled water-moderated reactors. Certain applications require that these alloys be cast into the form of heavy-walled cylinders suitable for coextrusion. The cast shapes should be nonporous, uniform in composition, and of such quality as to require a minimum amount of cleanup. The development of casting techniques capable of yielding cast extrusion billets fulfilling these requirements is the primary concern of this program.

An investigation of the factors influencing the quality of aluminum-25 w/o uranium extrusion billets produced by centrifugal-casting techniques has been initiated. The present equipment utilizes a feed arrangement designed to permit introduction of the molten metal along the entire axis of rotation of the casting. Two types of feeders were tested. One consisted of a series of holes in the graphite feeder tube and the other a tapered slot in a graphite tube. These two feeding arrangements were used to investigate their effects on the uranium distribution in the castings. Other factors which were investigated included the speed of the mold, the pouring temperature, and the mold temperature. The melts thus produced were evaluated by sectioning. Chemical analyses were taken of selected ingots, and radiographs were obtained on selected portions of some of the castings. The analyses indicate a 3 to 5 w/o variation in uranium content.

An evaluation of the physical condition of the castings produced indicated that relatively sound ingots of this composition can be produced. Ingots currently being prepared have required relatively little cleanup.

Mold speeds between 500 and 950 rpm have been investigated, and the data indicate that the higher speeds adversely affect the interior surface of the castings. This

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adverse effect manifests itself as a number of deep channels in the last portion of the casting to solidify. These channels require excessive machining for their removal.

Additional castings utilizing mold and feeder arrangements similar to those used in the above-mentioned tests are contemplated. It is believed that the 3 to 5 w/o variations in the uranium content exhibited by these initial melts can be eliminated to a large extent by insulating the mold from the turntable.

Development of a Mechanical Extensometer

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

An evaluation of a mechanical extensometer designed and constructed to determine the transverse mechanical properties of uranium tubing is being made. Determination of the mechanical properties of irradiated uranium tubes has made the development of such an extensometer necessary. SR-4 strain gages cannot be attached directly to the uranium tubing due to the irradiation hazards present. Therefore, a mechanical extensometer that can be attached and operated remotely is a necessity.

The extensometer consists of a split ring onto which three equally spaced knife edges have been placed. Changes in diameter of the uranium tubing will activate push rods connected to each half of the split ring. Vertical motion of the push rods is transmitted to two cantilver-type strain gages and two loop-type strain gages. Readings of the strain gages are registered on a SR-4 strain-gage indicator.

The extensometer has been calibrated through the use of push-rod micrometers. Two push-rod micrometers were arranged so that they activate the extensometer push rods through the upper knife edge. The micrometers were raised in 0.0005-in. steps and the amount of corresponding deflection of the cantilevers read on the SR-4 indicators. Curves have been prepared from this information and the sensitivity of the extensometer determined.

The extensometer is now being tested by measuring the transverse properties of aluminum, yellow brass, copper, and uranium rings. All of the above rings have been machined and testing is in progress.

C-1

C. PLANT ASSISTANCE TO MCW

A. A. Bauer

Study of uranium oxide structures has continued with emphasis being placed on determining residual nitrogen contents in UO_2 . Results obtained to date have not been reproducible and additional techniques are being studied.

The corrosion resistance of Type 304 ELC stainless steel in fluoride- and chloride-contaminated nitric acid solutions is being investigated. Several tests in 5, 18, and 30 w/o nitric acid solutions have been discontinued as no change in trend of corrosion rate was observed after prolonged exposures. Tests in 38 and 45 w/o nitric acid are continuing. Only one specimen exposed to the vapor phase of 45 w/o nitric acid containing 0.2 w/o chloride has shown a sharp change in rate of corrosion, the change in rate occurring after 2000 hr on test. Aluminum has not been found to be as effective as iron additions in minimizing corrosion in fluoride- and chloride-contaminated 38 w/o nitric acid solutions.

A new reaction tube has been constructed for measurements of hydrogen permeability through molten magnesium fluoride slag. Permeability measurements were obtained during a blank run. The bottom diaphragm was found to leak during a subsequent run employing molten slag necessitating a modification of the diaphragm.

Employing a newly constructed spindle, a calibration of the viscometer designed for the measurement of magnesium fluoride slag viscosity was obtained. Subsequent measurements indicated the viscosity of molten magnesium fluoride to be less than 3 centipoises but probably greater than 1-1/2 centipoises. Based on a calculation of the effect of viscosity on the rate of uranium fall through the slag, the equipment appears to exhibit adequate sensitivity for the measurements required. The effect of additives on viscosity will now be studied.

Techniques for the measurement of surface tension are being reviewed. A bubble-pressure method appears most promising for the purpose required.

Investigation of Uranium Oxides

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

A study of uranium dioxide is being continued. During the past few months, work has been concentrated on residual nitrogen in UO_2 . The determination of the nitrogen content in the dioxide has been studied by using a modification of the procedure which was developed for the trioxide. The dioxide was first reacted with hydrogen peroxide and then dissolved in sulfuric acid. This was made alkaline with sodium hydroxide, dried, and heated to 400 C. to decompose the peroxide while maintaining alkali nitrates. The procedure was then continued, as for the trioxide, using phenol disulfonic acid to complex the nitrogen. However, the color which was developed by conversion of the acid nitration product to the alkaline salt was not reproducible. The difficulty is believed to

C-2

be due to residual peroxide or loss of nitrogen during heating. In the next month, work will be done on getting the dioxide in solution for the color test without the use of peroxide.

The Electrical Properties of Uranium Dioxide

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

This study has continued but there are no results to report.

The Corrosion Resistance of Selected Stainless Steels

C. L. Peterson, W. C. Baytos, and F. W. Fink

Investigations have been continued on the corrosion of Type 304 ELC stainless steel by chloride- and fluoride-contaminated nitric acid solutions such as might be expected to occur in the nitric acid reconcentrator at the Weldon Springs uranium refinery.

Several of the prolonged exposure tests in 5, 18, and 30 w/o nitric acid solutions have been terminated, as no change from the reported trends in corrosion rates has occurred upon continued exposure. The tests in 38 and 45 w/o acid solutions are in progress. The only sharp change in any of the corrosion rates for specimens in these tests has occurred in the case of the specimen exposed in the vapor phase of the 45 w/o nitric acid solution containing 0.2 w/o chloride. Here the rate jumped from 0.07 mil per month at 2000 hr to 1.14 mils per month after 2258 hr.

Short-term tests have been made with 38 w/o nitric acid solutions, boiling at room temperature, containing chloride and fluoride contaminations with and without large excesses of aluminum ions. These tests have shown that aluminum will not minimize the resulting corrosion in the manner that has been demonstrated for additions of excess amounts of iron.

Gas-Metal Studies

W. R. Hansen, M. J. Trzeciak, and M. W. Mallett

This is part of the program of investigations related to the production of dingot uranium. A study is being made of some of the components of the thermite reactor in which dingot uranium is produced. At present, the study includes the determination of the rate of permeation of hydrogen through molten magnesium fluoride slag and measurements of the viscosity of magnesium fluoride slag. Also, plans are being made to measure the surface tension of molten slag.

Permeation of Hydrogen Through Molten Magnesium Fluoride Slag

In order to determine the amount of hydrogen permeating the walls of the new reaction tube, a blank run was made. For this, the tube was left with a bottom section approximately five times as thick as the walls. It is believed that the amount of hydrogen permeating the bottom (at molten slag temperature) is negligible compared to that passing through the walls. Permeation of hydrogen was measured at three temperatures, 1300, 1390, and 1410 C. Permeability rates of 6.5, 12.7, and 14.6 ml (STP) per hr were obtained. Subsequent to this run, the bottom section was machined off and the reaction tube was fitted with a 3-mil molybdenum diaphragm. A run was made to measure the hydrogen permeability of slag. Molten slag seeped out at the diaphragm and the run was discontinued. Further modification of the diaphragm will be made.

Viscosity of Molten Magnesium Fluoride Slag

A molybdenum spindle 3/4 in. in diameter by 1 in. long was fabricated. Sensitivity of readings with this spindle was better than that obtained with the smaller platinum spindle (1/2 in. in diameter by 1/2 in. long) used previously. A calibration curve for the range of 4 to 12 centipoises was obtained using s-tetrabromoethane as a reference liquid. A calibration point with water at about 1 centipoise was also obtained. Measurement of the viscosity of molten magnesium fluoride slag at 1330 C indicated that it was less than 3, but probably greater than 1-1/2 centipoises.

Based on this finding, a calculation was made to determine what changes in viscosity might significantly affect the rate of fall of a uranium particle through molten slag in the thermite reactor. It was assumed that the viscosity of the molten slag was the only factor affecting the suspension of uranium particles. The calculation was made using the following modification* of Stokes' law.

$$V = \frac{2g(\sigma - \rho)a^2}{9\eta(1 + 2.4d/D)},$$

when

V = velocity

η = viscosity in poise g/(sec)(cm)

σ = density of sphere

ρ = density of fluid

a = radius of sphere

d = diameter of sphere

D = diameter of infinitely long cylinder

g = acceleration of gravity.

All factors are in cgs units.

* Merrington, A. C., Viscometry, London, Edward Arnold and Company, 1949, Chapter LV, p 46.

The calculation showed that a uranium sphere 1 mm in diameter would attain a constant velocity of about 800 cm per sec if allowed to fall in an infinitely long cylinder containing a fluid with a viscosity of 1 centipoise. For a fluid with a viscosity of 1 poise (100 centipoises) the uranium sphere would attain a constant velocity of 8 cm per sec. Thus, the velocity of a uranium particle falling through molten slag in a thermite reactor will be relatively great even if viscosity of production slag should be a hundredfold that of magnesium fluoride. Since small changes in the viscosity of the slag will have little influence on the retention of uranium particles in the slag, the present equipment has adequate sensitivity for our purposes. Measurements will now be made of the effects of additives on the viscosity of molten magnesium fluoride.

Surface Tension of Molten Slag

Several techniques for the measurement of surface tension have been considered. Only two appear to be applicable to the problem. These are the ring and bubble-pressure methods*. At present it appears that the bubble-pressure method may be most satisfactory.

* Daniels, F., et al., Experimental Physical Chemistry, New York, McGraw-Hill Book Company, Inc., 1949, Chapter 4, p 62.

D-1

D. PROCESSING OF FEED MATERIALS

E. L. Foster

A continuing interest of National Lead of Ohio, in their operation of the Feed Material Production Center, is the improvement of unalloyed uranium fuel-slug material. Concrete evidence of this interest is shown in two programs relating to the processing of feed materials: the examination of factors affecting the quality of vacuum-induction melted uranium, and a theoretical study of the solidification of pure uranium ingots. In addition to determining the factors affecting the quality and solidification of uranium, the present studies should permit evaluation of melting and casting variables for future casting needs.

Solidification of Uranium

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

The work on the program concerned with the study of the solidification of uranium castings has continued. The influence of ingot configuration, mold material, pouring temperature, solidification rates, and other casting variables on the quality of high-purity uranium ingots is being investigated. To study the effects of the numerous casting variables, a mathematical model has been designed which involves a set of simultaneous equations. These equations, which permit the calculation of transient heat flow through the mold and ingot material, will describe the influence of a variable or variables as solidification proceeds. The applicability of the equations will be studied by experimental casting runs in which the variables will be closely controlled and the temperatures recorded at different critical locations in the casting.

Heat-flow equations are being programmed in an electronic computer, with specific studies being made on the effects of varying the pouring time, superheat, and environmental temperatures. Experimental tests are indicated and are being planned. These efforts will be concerned with a number of subjects, including the evaluation of thermal significance of the mold wall-ingot interface and mold wall-ingot gap formation caused by shrinkage, and the temperature dependence of some of the thermal properties of uranium and mold materials.

Examination of Factors Affecting the Quality of
of Induction-Melted Uranium

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

A study devoted to the nature of reactions and interactions of impurities which may occur with uranium during the melting cycle has for its ultimate aim a better understanding of those features of process control that are necessary for the production of

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high-quality uranium ingots. Such a study is prompted by data indicating that there may be a direct relationship between uranium fuel-slug quality and the quantity of certain nonmetallic impurities found in the metal. A better understanding of the variables involved in vacuum-melting procedures is expected to lead to improved slug quality. The experimental phase of this program centers about a laboratory vacuum-melting furnace (10-lb capacity). A second phase, a literature search dealing with the reactions that might occur during the vacuum melting of uranium, is to be executed concurrently with the experimental program.

An initial series of tests was conducted by measuring the rate and total quantity of gas evolution occurring at various controlled temperature levels in the 2000 to 2500 F temperature range. Gas samples were collected for analysis in a mass spectrometer. The main purpose of this initial series was to provide information regarding the feasibility of the test method as a means of collecting, measuring, and analyzing the gases given off during a melting cycle. The series consisted of five tests, one of which was conducted without a uranium charge at 2000 and 2500 F in order to indicate the nature and amount of background gas.

The four melting experiments made use of pure uranium dingot stock (10 to 20 ppm carbon, 1.7 to 2.5 ppm hydrogen, 10 to 40 ppm nitrogen, and 4 to 8 ppm oxygen) melted under the following conditions:

Heat	Crucible	Thermocouple Tube	Temperature Levels, F
1	MgO-coated(a) graphite	Porcelain	2000, 2200, 2300, 2400, 2500
2	Uncoated graphite	Alumina	Ditto
3	MgO-coated graphite	Ditto	Ditto
4	Ditto	Ditto	2000 and 2500

(a) Coating is a special MgO slurry formulation of National Lead of Ohio. It should be noted that in all cases where a coated crucible was used, the MgO coating flaked off the lower third of the crucible.

In all cases, a vacuum of less than 0.1μ was obtained before the furnace was isolated (exhaust valve closed) from the vacuum system. Except for the 2000 F temperature level, the vacuum system was isolated before the temperature was increased to the next level.

Pertinent observations made during the four melting tests are indicated below.

Heat 1. Bubbling of the melt occurred, and became vigorous as temperature was increased. A black sootlike material was deposited on crucible support surfaces within a well defined zone about the crucible. Definite evidence of attack of the porcelain thermocouple-protection tube was observed.

Heat 2. Throughout the test slight bubbling occurred, with little or no bubbling occurring at the 2500 F temperature level. No sooty deposits were found; however, a small quantity of brown powder was deposited on all surfaces. The alumina protection tube showed little or no attack.

D-3 and D-4

Heat 3. Vigorous bubbling occurred at a temperature of 2400 F and above. A black residue, as observed in Heat 1, was found at the conclusion of the test. A spectrographic analysis of this deposit showed that its major component was magnesium with lesser amounts of aluminum, iron, nickel, silicon, sodium, and zinc. Loss on ignition was 15 per cent.

Heat 4. Very vigorous bubbling occurred soon after melting (crucible temperature 2350 F) and continued throughout the test. The outgassing rate at 2500 F was essentially the same as that of Heat 3 for the same temperature level. As in Heats 1 and 3, a black deposit (of about the same composition of that of Heat 3) was found at the conclusion of the test. Deposition always occurred in the same well defined zone.

At the conclusion of the tests the uranium castings were sectioned and analyzed. Carbon content was appreciably increased, ranging from 1450 ppm (Heat 4) to as much as 2370 ppm (Heat 2). Oxygen content increased the most (60 to 90 ppm) in Heats 1 and 3, and the least (16 to 25 ppm) in Heat 2, for which the uncoated crucible was used. The nitrogen content did not appear to change, but the hydrogen content of the cast uranium was consistently less than the metal before melting (0.1 to 0.3 ppm) in all cases.

Although the analyses of the gas samples taken during the tests have not been completely evaluated, certain generalities are indicated. The background test indicated that moisture present in the system reacted with the graphite of the crucible to form hydrogen and carbon monoxide. At a temperature of 2500 F the reaction appeared to go to completion, yielding equal volumes of both gases. Except where gas samples were suspected to be contaminated with air, the gas samples were found to contain very large amounts of hydrogen (all greater than 90 volume per cent and several approximately 95 volume per cent hydrogen). In all instances, the CO content was less than 2 volume per cent. The remaining gases were nitrogen, H₂O, argon, CO₂, and certain unidentified hydrocarbons. It is interesting to note that nearly all CO produced by the above water-gas reaction reacts with the uranium to form uranium carbide and oxide.

Plans for the immediate future call for a more complete review of all data obtained in the present test series. Although improvements in gas-sampling techniques are indicated, the method of test appears to be reliable. As soon as the review of data has been completed and necessary sampling innovations have been completed, a second test series will be organized to include a closer study of the effect of moisture and MgO on the melting cycle.

E-1

E. GENERAL FUEL-ELEMENT DEVELOPMENT

F. A. Rough and D. C. Carmichael

Various programs of fuel-element development are reported in this section. These studies are sponsored by the AEC Division of Reactor Development.

Capsules to contain stainless steel-clad irradiation specimens having UC or UN fuel dispersed in a stainless steel matrix have been designed and are being machined. Results of tensile tests on specimens which had been cold worked 14 per cent are presented for temperatures to 1650 F. A series of fully annealed specimens is being machined for tensile testing.

An investigation is being made to determine techniques for fabricating cermet fuel elements containing high loadings of fuel compound dispersed in a metal matrix. Specimens with various loadings of UO_2 in stainless steel have been tested for strength at room temperature, and thermal-cycling tests have resulted in no flaking or internal cracking of UO_2 particles. Specimens have been prepared to determine the feasibility of cladding UO_2 -stainless steel cermets with stainless steel by a pressure-bonding technique.

A study is being conducted to investigate the feasibility of a fueled zirconium hydride moderator. Specimens are being prepared for irradiation testing, and capsule design for the tests has been completed. Phase boundaries in the zirconium-1 w/o uranium-alloy hydride system have been determined at 755 and 802 C and measurements are being made at 600 C.

Development of Dispersed UC and UN Fuel Elements

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

In addition to studies on the irradiation resistance of stainless steel-clad fuel elements which contain UC or UN dispersed in stainless steel, tensile tests are being conducted at temperatures from room temperature to 1650 F. One series of tests on 14 per cent cold-worked specimens containing approximately 24 w/o UN or UC dispersed in an 18-8 stainless steel and clad with Type 347 stainless steel gave the following results: at room temperature, an ultimate tensile strength of 110,500 psi for UN and 113,900 psi for UC specimens; at 1350 F, 38,000 psi for UC and 43,850 psi for UN; at 1500 F, 29,800 psi for UN; and at 1650 F, 10,700 psi for UC dispersions. A second series of tests to be conducted on fully annealed specimens will be initiated when machining of tensile bars is completed. Capsules for testing irradiation specimens have been designed and are being machined.

Fabrication of Cermet Fuel Elements

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

Methods of fabricating cermet fuel elements with fuel loadings of 60 to 90 volume per cent are being investigated. A bend-test procedure is being used to determine tensile strength, a water quench from 2000 F is being used to determine resistance to thermal shock, and pressure-bonding techniques are being evaluated for cladding the cermet cores.

Tensile strengths calculated from bend-test results indicate that the ultimate tensile strengths at room temperature of UO_2 -stainless steel dispersions vary from 61,800 psi for a 50 volume per cent UO_2 dispersion to 9,200 psi for an 87 volume per cent UO_2 dispersion. Examination of the data shows that the tensile strength of the specimens with 65 to 90 volume per cent UO_2 is dependent only upon the effective cross-sectional area of stainless steel. However, at lower loadings, such as the 50 volume per cent UO_2 dispersion, the tensile strength is higher than would be expected if due only to the cross-sectional area of stainless steel.

No evidence of internal cracking or flaking of UO_2 particles was noted on specimens of 74 volume per cent UO_2 dispersed in stainless steel after being subjected to 50 cycles of heating to 2000 F and quenching in water while sealed in a Vycor tube.

Specimens have been prepared for pressure-bonding studies, but experimental results are not yet available.

Fueled Zirconium Hydride Moderator

H. E. Bigony and J. B. Vetrano

The capsule design for the irradiation of the hydride of a zirconium-2 w/o enriched uranium alloy has been completed and the specimens are being prepared. The capsule is double walled. The inner chamber contains the specimens and is filled with hydrogen, while the outer chamber contains NaK as a heat-transfer fluid. The specimens have a diameter of 3/16-in. and are hydrided to a hydrogen-to-zirconium atomic ratio of about 1.6.

The 755 and 802 C dissociation-pressure isotherms of a zirconium-1 w/o uranium alloy hydride have been run. They show considerable deviation from Sieverts law in the single-phase regions. Rather diffuse inflections are apparent at points corresponding to phase changes in the binary system zirconium-hydrogen. This diffuseness makes it impossible to determine the lower concentration limit of the beta phase. The other phase boundaries as determined from the curves are shown in Table E-1.

Data for a zirconium-1 w/o uranium alloy hydride at 700 C and for a zirconium-50 w/o uranium alloy hydride at 600 C were not usable due to a leak in the system which permitted oxidation of the specimen. Measurements are currently being made at 600 C for the zirconium-1 w/o uranium hydride.

E-3 and E-4

TABLE E-1. PHASE BOUNDARIES IN THE ZIRCONIUM-1 w/o URANIUM ALLOY HYDRIDE SYSTEM

Phase-Boundary Designation	755 C		802 C	
	Hydrogen Sorption, cm ³ per g	Pressure, cm	Hydrogen Sorption, cm ³ per g	Pressure, cm
$\alpha (\alpha + \beta)$	5.48	0.076	2.22	0.060
$\beta (\beta + \delta)$	113.0	4.10	114.9	13.0
$(\beta + \delta) \delta$	176.7	4.70	177.2	15.2

F-1

F. STUDIES OF URANIUM AND URANIUM-ALLOY FUELS

F. A. Rough and A. W. Hare

This section reports the research and development work on various uranium-alloy fuels which is being sponsored by the Reactor Development Division of the AEC.

Irradiation of test Capsule BMI-8-5 is continuing at the MTR.

For the program designed to develop metastable gamma-phase uranium alloys, corrosion data are reported on various uranium ternary and quaternary alloys.

Routine progress is reported on the program concerned with the preparation of uranium-niobium alloys.

In the research on the mechanism of uranium corrosion, work is continuing to determine the exact oxygen content of uranium which has been corroded in steam at 200 C.

Radiation Stability of Uranium-Zirconium Alloys

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

The literature survey to summarize the present technology on this system is continuing. All other work is recessed pending receipt of capsules (including BMI-8-5) now at the MTR for irradiation.

Development of Gamma-Phase Uranium Alloys

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

A program designed to develop a metastable gamma-phase alloy by ternary and quaternary alloying is being conducted. Such an alloy is expected to have good corrosion resistance, improved resistance to irradiation damage, and fairly good hot strength. Alloying additions being investigated are chromium, molybdenum, niobium, ruthenium, vanadium, and zirconium.

As part of the investigation, all the alloys are being corrosion tested in 500 F water in the as-quenched condition. The following tabulation lists the alloys which are exhibiting the best corrosion resistance after 504 hr of exposure.

F-2

Alloy	Weight Loss, mg per cm ²
U-20 w/o Zr-5 w/o Mo	2.98
U-20 w/o Zr-2 w/o Ru	6.03
U-40 w/o Zr-3 w/o Mo	1.46
U-40 w/o Zr-5 w/o Mo	2.12
U-10 w/o Nb	1.23
U-10 w/o Nb-1 w/o Zr	1.32
U-10 w/o Nb-3 w/o Zr	0.62
U-10 w/o Nb-10 w/o Zr	0.58
U-10 w/o Nb-1 w/o Mo	1.57
U-10 w/o Nb-3 w/o Mo	1.55
U-10 w/o Nb-5 w/o Mo	1.55
U-10 w/o Nb-2 w/o V	0.25
U-10 w/o Nb-0.5 Cr	0
U-20 w/o Nb-3 w/o Mo	0.12(gain)
U-7.5 w/o Mo-3 w/o Nb	6.51
U-7.5 w/o Mo-5 w/o Nb	5.43
U-20 w/o Zr-3 w/o Nb-3 w/o Mo	3.16
U-10 w/o Zr-5 w/o Nb-3 w/o Mo	1.79
U-7.5 w/o Mo-5 w/o Zr-3 w/o Nb	4.42

All the above alloys will also be corrosion tested in 680 F water. The only alloys to show relatively poor corrosion resistance as a group are the uranium-7.5 w/o molybdenum-zirconium alloys, which suffered high weight losses, and these will not be tested in 680 water.

Thermal-analysis and hot-hardness measurements are being made on all alloys. However, insufficient data have been collected to date to draw any conclusions. A detailed study of the transformation kinetics of these alloys is being planned.

The uranium-zirconium-niobium alloys, prepared for a previous project aimed at developing a corrosion-resistant uranium-zirconium ternary alloy, are continuing to corrode at approximately the same rates after 1848 hr in 680 F water. A uranium-23.8 w/o zirconium-6.1 w/o niobium specimen, water quenched from 900 C and aged 24 hr at 500 C, is corroding at a rate of 0.04 mg/(cm²)(hr), while uranium-23.2 w/o zirconium-4.84 w/o niobium specimens in furnace-cooled, air-cooled, and quenched-and-aged conditions are corroding at rates between 0.095 and 0.25 mg/(cm²)(hr). A uranium-27 w/o zirconium-1.9 w/o niobium alloy, air cooled from 800 C and aged for 24 hr at 500 C, is corroding at a rate of 0.12 mg/(cm²)(hr) after 672 hr in 680 F water. By comparison, a uranium-50 w/o zirconium alloy corrodes at a rate of approximately 0.2 mg/(cm²)(hr).

Among Zircaloy 2-clad specimens being tested with simulated pinhole defects, a uranium-23.2 w/o zirconium-4.84 w/o niobium alloy in the water-quenched condition is still on test after 1848 hr in 680 F water. A similar clad alloy, water quenched and aged, is in good condition after 672 hr on test. Also in good condition after 672 hr in 680 water is a clad uranium-27 w/o zirconium-1.9 w/o niobium alloy which was air cooled from 800 C and aged for 24 hr at 500 C.

Preparation of Uranium-Niobium Alloys

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

Uranium-niobium alloys have been the subject of a broad program of study in reference to their use as reactor fuels. The first part of the program has been concerned principally with the review of literature pertaining to these alloys. Specifically of interest has been information on alloy preparation, constitutional relationships, kinetics, heat-treatment effects, physical and mechanical properties, corrosion behavior, and the effects of radiation. The review of literature is nearly complete. Further and more recent information on these subjects is being gathered by direct contact with laboratories presently engaged in work relating to this alloy system.

A part of the over-all program is concerned with the development of methods for the preparation of high-quality alloy material. In this phase of the program, the need for material of known composition and quality has been recognized. Since further work will be required to fill in or resolve conflicting reports on the technology of the alloy, it is evident that alloys of high integrity will be needed. The literature review has indicated that impurities, in addition to their effect upon the fabricability of high-niobium-content alloys, may have an important effect upon transformation relationships of low-niobium-content alloys.

The preparation of alloy material is presently concerned with three facets of the problem: (1) evaluation of melting methods, (2) fabrication of alloys of varied composition, and (3) appraisal of the availability of high-quality niobium.

Recent efforts have been concerned with the evaluation of alloy material prepared by consumable-electrode arc-melting techniques, and fabricated by cladding in titanium. Further evaluations of materials prepared by the skull arc-melting procedure is in progress. Fabrication of specimens prepared by both of these procedures after cladding in molybdenum is planned. Additional efforts are in progress to adapt existing equipment so that larger scale, skull-type arc melting can be conducted.

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

Work is continuing on the identification of the aqueous corrosion products of uranium as an aid to understanding the corrosion process. Although the product has a UO_2 -type structure, a material balance between weight loss and hydrogen evolution during corrosion has not been obtained when the composition of the corrosion product was assumed to be stoichiometric UO_2 . Since it is difficult to analyze the corrosion product for oxygen-to-uranium ratio, other tests are being made to determine the oxidation state of the uranium in the reaction product.

During December, the study of the optical properties of uranium, UO_2 , and UO_3 were continued. No new data are available. A study also was made of the L_{III} X-ray absorption edge for UO_2 , $\text{UO}_2 \cdot 4$, and UO_3 . The breadth of the absorption edge for UO_2

F-4

was found to be 0.0012 ± 0.0005 Å, while for UO_3 the absorption edge measured 0.0022 ± 0.0005 Å wide. In the case of $\text{UO}_{2.4}$, an irregular absorption edge suggests a more complex electronic structure than that of UO_2 or UO_3 . The absorption edges in the oxides UO_2 and UO_3 were nearly straight lines but of different slopes. During the next period, an attempt will be made to improve the reproducibility of absorption-edge measurement.

Corrosion studies were conducted on uranium in steam at 200 C. Calculations, based on the amount of metal consumed and the amount of hydrogen evolved in the reaction, indicate an oxygen content in the corrosion product ranging from $\text{UO}_{1.93}$ to $\text{UO}_{2.03}$. Ignition analyses continue to reveal $\text{UO}_{2.40}$ in the corrosion product. However, there are strong indications that the corrosion product oxidizes further when the reaction vessel is opened and air reaches the test samples. Present equipment is being modified to permit the analysis of the uranium oxide without removing it from the corrosion-test vessel.

G-1 and G-2

G. FATIGUE STUDIES OF INCONEL

W. S. Hyler and R. J. Favor

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

Conditions to be investigated are (1) temperatures of 1200, 1400, and 1600 F, (2) load ratios of $A = 0.25$, 0.7 , 2.0 , and ∞ , and (3) cyclic frequencies of 1 and 10 cps.

During December, evaluation at 1200 F and a load ratio of $A = 2.0$ was initiated. Early tests indicated that deformation in the first few cycles was of an appreciable magnitude as compared with negligible amounts in all previous tests at 1600 F and at $A = \infty$ at 1200 F. This behavior made it advisable to make some supplemental studies. These involved tensile tests at 1200 and 1600 F and metallographic examinations of fatigue specimens. The tensile data are summarized below:

Specimen	Temperature, F	0.2 Per Cent Offset Yield Strength, psi	Tensile Strength, psi	Elongation in 1 in., per cent	Reduction in Area, per cent
5-23	1200	17,200	58,500	47	52
3-25(a)	1200	26,000	57,000	50	55
13-46	1600	14,800	17,400	99	95

(a) Specimen 3-25 fatigue tested under completely reversed stress prior to tensile testing.

The strain rate was 0.003 in. per in. per min and the specimens were heated 1/4 hr at temperature before testing. Specimen 3-25 was previously tested under completely reversed stress at 1200 F and a maximum stress of 26,000 psi.

Metallographic work is summarized below:

Specimen	Type of Section	ASTM Grain Size	Evidence of Deformation	Vickers Hardness, DPH
8-4	Longitudinal	2	Twins	168
11-29	Longitudinal	>1	None	203
9-25	Longitudinal	2	None	170

The results of these studies indicate that some strengthening of the material possibly due to strain hardening may have occurred, and that the apparent fatigue strength (under fully reversed loading) is of the order of twice the nominal yield strength. To obtain data comparable with results previously reported, testing will have to be confined to stresses not much greater than the yield strength. Creep will necessarily be small.

The 1200 F study will be continued and a limited study begun at 1400 F at a load ratio of $A = \infty$ (completely reversed stress) at 1 cps.

H-1

H. REACTOR MATERIALS AND COMPONENTS

F. A. Rough and A. W. Hare

Various research programs concerned with reactor materials and components and supported by the Reactor Development Division of the AEC are reported in this section.

Routine progress on the oxidation-resistant niobium alloy program is reported.

The fabrication data and oxidation and electrical data on UO_2 and La_2O_3 solid solutions are discussed for the program concerned with the valence effects of oxide solutions in uranium oxide. Electrical-measurement data are also reported on UO_2 samples containing CaO and ThO_2 in various amounts.

Modification of the first die for high-temperature high-pressure solid-state studies is still in progress. This change will permit more efficient screening studies at lower pressures in a smaller press.

It is reported that the apparatus necessary to run experiments to obtain information about friction-behavior of sliding surfaces immersed in sodium has been completed and tests of the equipment have been initiated.

The basic studies of pressure-bonding phenomena are reported in this section. Routine progress of both the analytical and experimental phases of this effort are discussed.

Oxidation-Resistant Niobium Alloys

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

The program on oxidation behavior of niobium and niobium alloys is continuing, with the objective of applying principles derived from the study of binary alloys to development of improved ternary alloys, and of further study of the principles of oxidation as applied to niobium alloys.

No experimental work was conducted during December. A technical paper describing the results of continuous weighing tests on binary alloys and correlating the oxidation behavior in terms of atomic size, valence, and ability to form new, stable oxides is being prepared.

Compositions for ternary alloys are now being selected. These alloys will be processed into strip and their oxidation behavior studied by the continuous weighing method at 1000 and 1200 C.

Valence Effects of Oxide Solutions in Uranium Dioxide

J. Moody, A. Gerds, W. M. Albrecht, R. E. Bennett,
W. B. Wilson, and C. M. Schwartz

Several experiments were performed to attempt to produce dense and homogeneous bodies of UO_2 and La_2O_3 . The La_2O_3 powder was vacuum dried to remove adsorbed and combined water prior to mixing with the UO_2 . The powders were weighed and mixed in an argon-filled dry box and compacted hydrostatically at 100,000 psi using a binder composed of 1 w/o beeswax in carbon tetrachloride. The compacts were placed on high-fired UO_2 blocks and sintered in vacuum.

Three compositions were studied: UO_2 -60, 50, and 40 w/o La_2O_3 . Compacts were sintered at 1700 to 1800 C for times of 2 to 4 hr. Slight melting appeared to occur at 1800 C. At 1700 C, no evidence of melting was noted. The densest body was produced with the 40 w/o La_2O_3 -60 w/o UO_2 mixture by sintering at 1700 C for 4 hr, re-crushing in a dry box through 325-mesh screen, and then repressing and resintering at 1700 C for 4 hr followed by slow cooling. This body appeared to shrink very little during the second sintering and was relatively free of cracks and voids. Other La_2O_3 - UO_2 compositions will be treated in a similar manner. Attempts also will be made to produce dense bodies of mixtures of Sc_2O_3 and UO_2 .

A study is being made of the stability, in air at high temperatures, of solutions of various oxides in UO_2 . In these experiments, the rate of weight change will be determined using a microbalance. Currently, the microbalance is being modified so that specimens can be heated to temperatures of about 2500 to 3000 F. A platinum resistance furnace was constructed inside the reaction tube of the microbalance. Tests are being made to determine the techniques of operation of the apparatus.

Room-temperature electrical measurements have been completed on a series of sintered specimens composed of UO_2 and varying amounts of a second metal oxide. The results of these measurements are collected in Table H-1. The resistivity values listed in Table H-1 have not been corrected for density variations among the various specimens. Common practice is to refer the measured resistivity of a sintered specimen to a calculated theoretical density. This will be done for the above systems when sufficient information concerning exact composition, structure and lattice constants is obtained. However, with the possible exception of those samples containing La_2O_3 , it is not expected that the correction will affect the relative values of resistivity listed in Table H-1. Unfortunately, the specimens containing La_2O_3 contained cracks and voids which may have contributed to the measured resistivities.

It is obvious from Table H-1 that the presence of metal ions of valence or size different from that of U^{+4} has a pronounced effect on the resistivity of UO_2 . Although there is some doubt about the measurements, it appears that the p -type conductivity of UO_2 decreases with increasing La_2O_3 content. The same appears to be true for additions of ThO_2 . The reasons for this effect are not clear at present and an understanding must await a detailed examination of the dependence of electrical properties on temperature.

H-3

TABLE H-1. ELECTRICAL PROPERTIES OF UO_2 -METAL OXIDE SYSTEMS

Specimen	Composition, w/o	Resistivity, ohm-cm	Type of Conductivity
13662-9-1	100 UO_2	3.84×10^2	P
13662-9-2	95 UO_2 -5CaO	1.30×10^2	P
13662-9-3	90 UO_2 -10CaO	8.25×10	P
13662-9-5	50 UO_2 -50 ThO_2	2.07×10^4	P
13662-9-6	45 UO_2 -55 La_2O_3	1.4×10^4	P
--	50 UO_2 -50 La_2O_3	1.1×10^5	P
--	60 UO_2 -40 La_2O_3	2.5×10^3	P

An opposite effect is observed with additions of CaO. That is, the p-type conductivity of UO_2 increases with CaO content. From the extrinsic portion of the curve of conductivity as a function of temperature (measured from -78 to 800 C), an activation energy of 0.36 ev was calculated as the energy necessary to free a positive charge carrier in samples containing CaO. This value is comparable to the activation energy found for positive carriers in UO_2 containing excess oxygen. These results suggest the calcium ions are substituting for uranium ions in the UO_2 lattice. Since the calcium atom does not have a sufficient number of electrons to share fully in the fluorite arrangement, "holes" are introduced in the valence band of the lattice and the p-type conductivity of UO_2 is increased.

Pure, sintered specimens of UO_2 (with low excess-oxygen content) usually became intrinsic electrical conductors at about 600 C. The intrinsic region of the specimens containing CaO was not reached even though the resistivity measurements were extended to 800 C. These measurements were not extended further because the parts of the holder became soft and conductive at temperatures somewhat above 800 C. The effect of additions of CaO on the energy gap of UO_2 (about 3.0 ev) would probably not become apparent below 1000 C.

Measurements of the temperature and time dependence of the resistivity of samples containing La_2O_3 reveal a surprising aging effect. The measurements listed in Table H-1 were obtained shortly after the specimens were prepared. After several days' aging at room temperature the resistivity of these samples decreased by at least an order of magnitude. Reheating the specimens to above 500 C tends to return them to their original condition, indicating the reversibility of the aging reaction. This behavior is to be compared with the resistivity of pure UO_2 which increases when specimens are quenched from elevated temperatures.

The extrinsic behavior of the resistivity of aged UO_2 - La_2O_3 specimens is thus a function of the history of the specimens. The behavior of preheated samples, on the other hand, appears to be reproducible. The activation energy necessary to free a positive carrier calculated from these curves is about 0.60 ev.

Again it was impossible to measure the intrinsic properties of these samples since they were still extrinsic at 800 C.

H-4

It is now planned to further investigate the factors determining the reproducibility of the electrical properties of $\text{UO}_2\text{-La}_2\text{O}_3$ specimens. It is also planned to adapt the present equipment to extend the electrical measurements to temperatures above 1000 C.

High-Temperature High-Pressure Solid-State Studies

W. B. Wilson and C. M. Schwartz

Final construction, modification and assembly of the first die design has been completed. The unit was heated in air to 1000 C, specifically to test the insulation, as a preliminary to testing and calibration in the press. Pressure calibration will be accomplished initially, by using the pressure transition points of Cs, Ba, Tl, and Bi.

A simplifying modification of this initial die design, utilizing existing parts, has been designed and construction has been started on auxiliary components. This modification will permit screening studies at lower pressures to be carried out more efficiently in a smaller press.

The second or heated geometrical-advantage-type die design has been completed and construction awaits testing of the first unit. It is expected that testing and preliminary calibration of the first unit will be accomplished in January, 1958.

Rubbing Surfaces in Sodium Environments

J. W. Kissel and J. H. Stang

During December, the inert-atmosphere apparatus for experiments to obtain information about friction behavior of sliding surfaces (sphere against a flat plate moving at about 1 cm per sec) immersed in molten sodium (up to 1500 F) was assembled and vacuum-tightness checks were initiated. Plans were made for preliminary runs to check the operation of the sodium-loading and temperature-control devices and for friction experiments using a conventional lubricant such as mineral oil. The latter will be conducted principally to examine the sensitivity and reproducibility of the system provided to measure the static and kinetic frictional forces.

According to present plans, test materials for the oil-lubrication experiments will be confined to fairly conventional steels. For the liquid-metal tests, however, materials such as molybdenum, Inconel, and a limited number of TiC- and WC-base cermets will be employed because of their high-temperature-strength characteristics and resistance to attack by sodium.

H-5

Basic Studies of Pressure Bonding

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

An investigation is being conducted to determine the fundamental principles associated with hydrostatic pressure bonding. The greatest emphasis has been concerned with the prediction of the bonding parameters (time, temperature and pressure) necessary to insure that the surfaces obtain sufficiently close contact to bond.

Analytic Studies

Experiments have been conducted using hollow circular cylinders of prescribed dimensions subjected to external pressure. Using the theory of ideal plasticity and the von Mises yield condition, a relationship among the pressure, the yield point of the material, the dimensions of the specimen, and the final diameter of the hole was found. The results of the experiments described below showed that the amount of deformation was dependent upon time during the early part of the pressure-bonding process. Since time is not considered in the von Mises theory, this variable needs to be studied using a different theoretical basis.

It is recognized that material does not behave ideally. Thus it is postulated that an effective yield point, dependent on time and temperature, needs to be utilized. The yield point for the material in simple tension as determined for a given temperature is being utilized with the experimental data obtained from these experiments to establish an empirical relationship between the effective yield point and time.

An analytic examination under certain simplifying assumptions, of the pressures required to bring almost flat surfaces into contact, indicates that the pressure depends only on the yield point of the material and is not affected by the geometry of the surfaces. It is anticipated that the yield point to be used must be the effective yield point discussed above. The knowledge of the effective yield point over a suitable range of temperatures and times can lead to the optimum choice of the bonding parameters.

Experimental Studies

In the experimental phase of the program, specimens are designed and prepared to obtain data which can be compared with calculated values in order to determine the validity of using postulated mathematical models to predict the flow characteristics of metal during pressure bonding. Thick-walled aluminum tubes with both ends sealed have been externally pressurized at different temperatures to determine if the amount of deformation produced can be previously predicted using the von Mises shear-strain-energy criterion for yield.

A series of specimens prepared with various conditions of temperature and pressure were metallographically measured to determine if the amount of deformation obtained was identical to that predicted by the von Mises theory. For these specimens the amount of deformation obtained at 400 and 700 F at 4,000 psi revealed a pronounced

H-6

temperature dependence that can be predicted from the theory. Specimens pressurized at 700 F using 4,000 and 5,000 psi exhibit no appreciable difference in deformation with the increase in pressure. Additional specimens are being subjected to a constant temperature of 700 F at significantly higher and lower pressures to study further the effect of pressure on the flow of metal during the primary stages of the pressure-bonding operation. To investigate further the various phenomena involved in the pressure bonding process, additional specimens will be prepared as prescribed by the analytical studies.

I-1

I. PHYSICAL RESEARCH

F. A. Rough and B. W. Dunnington

In this section are reported various physical studies in the field of metallurgy and ceramics. Progress is reported on studies of uranium-base ternary alloys, of properties of uranium compounds, of solid-state reactions in the UNC system, and of the structure of UC_2 . Routine progress was made in other projects including studies of bonding fundamentals, of niobium-gas reactions, and of hydrogen migration in zirconium hydride, but no report sections are included for the latter studies.

In the study of constitution of uranium-base ternary alloys, efforts are directed toward locating a ternary eutectoid in the system uranium-zirconium-molybdenum. Results are also reported for temperatures from 550 to 750 C in the uranium-rich corner of the system uranium-zirconium-silicon.

Studies of properties of uranium compounds are continuing with emphasis on UC, UC_2 , and UBe_{13} . Specimens for thermal-expansion and thermal-conductivity measurements are being ground from sintered compacts of uranium monocarbide. Specimens of UC_2 and UBe_{13} are also available for physical measurements.

The solid-state study of the system UNC by high-temperature X-ray diffraction to determine the extent of solid solution of uranium carbides with uranium nitrides, the existence of ternary phases and the stability of the various materials under vacuum, argon, or nitrogen atmospheres is continuing. It is yet too early to obtain a coordinated picture of this system.

The structure of UC_2 is being reinvestigated to determine the validity of the assumed tetragonal structure and to determine the carbon positions. Preliminary X-ray diffraction studies have been made and attempts to extract single crystals of UC_2 by etching techniques are in progress.

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 investigated.

In the preparation of sintered compacts of uranium monocarbide, bulk densities of 98 per cent of theoretical or greater have been achieved by increasing the sintering time to 2-1/2 hr or more. No difficulty was encountered in grinding these high-density compacts to regular cylindrical shapes with a dimensional tolerance of ± 0.001 in. Specimens for thermal-expansion and thermal-conductivity measurements will be prepared.

Sintered uranium monocarbide specimens corrode very rapidly in boiling water at 1-atm pressure. In approximately 1 hr the sintered specimens had disintegrated. The corrosion products were powdery and olive green in color. Calculations showed that the weight of corrosion product corresponded to complete conversion of the uranium in the samples to U_3O_8 .

Small sintered compacts of UBe_{13} appeared to be more stable in boiling water at atmospheric pressure than did the UC. Although the sintered specimens disintegrated after several hours, the particles retained their metallic luster after a 10-hr exposure.

Sintered compacts of uranium nitride, having a bulk density of 90.3 per cent of theoretical, have been prepared. Previously, a maximum density of 88.5 per cent of theoretical was obtained.

Hydrostatically compacted specimens of UBe_{13} are being sintered in an argon atmosphere. These specimens and sintered compacts of UC and UC_2 prepared this month will be used for physical and chemical measurements.

Constitution of Uranium-Base Ternary Alloys

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

A study of phase relationships existing between the intermetallic phases of the uranium-zirconium and uranium-molybdenum systems is in progress. The tentative ternary cut, as presented in BMI-1220, has not been changed appreciably.

Efforts are now being directed toward determining the ternary eutectoid composition of the uranium-zirconium-molybdenum system. The method employed is as follows: an alloy believed to be near the eutectoid in composition was heat treated at 900 C and water quenched. Another specimen of the same alloy was heat treated at 550 C and water quenched. In both cases, the gamma phase was retained. An electron-probe analyzer was used to determine the chemical composition of the gamma phase in both specimens. By noting the change in composition of the gamma phase on going from the high to the low temperature, it was possible to estimate the location of the eutectoid. New samples are being prepared for further study of the eutectoid composition based on this technique.

The phase relationships existing in the high-uranium corner of the uranium-zirconium-silicon system are also being studied. Phase equilibria at lower temperatures are essentially the same as reported in BMI-1201 for the high temperatures, except as altered by the eutectoid transformation occurring in the uranium-zirconium system. At 705 C the following phase fields, extending from the uranium-zirconium binary, exist: beta plus Zr_5Si_3 plus Zr_2Si ; beta plus gamma plus Zr_2Si ; $gamma_1$ plus Zr_2Si ; $gamma_1$ plus $gamma_2$ plus Zr_2Si ; $gamma_2$ plus Zr_2Si . No evidence of Zr_4Si has been found. Similarly, at 675 C the following phase fields exist: alpha-plus beta-plus Zr_2Si ; alpha-plus $gamma_2$ -plus Zr_2Si ; and $gamma_2$ plus Zr_2Si . At 550 C, delta uranium-zirconium forms and is in equilibrium with Zr_2Si ; also the three-phase field alpha plus delta plus Zr_2Si is present.

Experimental work is near completion and a final report will be prepared.

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

A. E. Austin, A. F. Gerds, and C. M. Schwartz

This ternary system is being studied to determine the extent of solid solution of uranium carbides with uranium nitrides, the existence of ternary phases and their stability under vacuum, argon, and nitrogen atmospheres. The various compositions have been prepared by solid-solid reaction and solid-nitrogen (gas) reaction. BMI-1232 reported results on formation of $U(N, C)$ solid solution by reaction of equimolar amounts of UN with UC, U_2N_3 with UC and with UC_2 .

Reactions at 1000 C of equimolar ratios of UN with carbon and UC_2 with UN have been investigated. The reaction of UN with carbon produced the solid solution $U(N, C)$ with a minor amount of UC_2 . The reaction UC_2 with UN gave the solid solution $U(N, C)$ with a trace of UC_2 . The lattice constants of these solid solutions indicated a deviation from Vegard's law. Table I-1 gives product compositions and lattice constants. Additional reactions between varying amounts of UN and UC were made, in order to obtain solid-solution samples to check the variation of the lattice constants with the ratio of carbon to nitrogen. A composition of 7 parts UN plus 3 parts UC yielded a solid solution with lattice constant essentially linear between those of UN and UC.

TABLE I-1. COMPOSITIONS AND PHASES IN PRODUCTS OF REACTIONS UN + CARBON AND UC_2 + UN

Starting Mixture	Product Composition, at%			Phases in Product
	Uranium	Carbon	Nitrogen	
UN + Carbon	45.2	42.3	12.6	$U(N, C) a_0 = 4.9474 \pm 0.0003 + UC_2$
UC_2 + UN	49.4	38.4	11.8	$U(N, C) a_0 = 4.9487 \pm 0.0003 + \text{trace } UC_2$
7UN + 3UC	52.5	12.5	35.2	$U(N, C) a_0 = 4.9088 \pm 0.0005$

Reactions with excess ratios of UC and UC_2 to U_2N_3 were carried out for other compositions to seek the limits of the $U(N, C)$ solid-solution field. Solid solutions close to the UC composition were formed. The product of reaction with excess UC_2 contained, in addition to $U(N, C)$, the phase UC_2 apparently unchanged in lattice constant, indicating very little nitrogen solid solution in UC_2 .

When UC is heated in 1 atm of nitrogen at 1000 C, its unit cell is expanded from 5.01 to 5.35 Å at temperature indicating extensive gas-solid reaction. The crushed powder examined at room temperature showed a doubled cubic cell similar to that of U_2N_3 . Bulk samples will be prepared for analysis to determine possible solid solution of carbon in this phase. Additional reactions with UC_2 are planned.

Structure of UC₂

A. E. Austin and C. M. Schwartz

The structure of UC₂ is being reinvestigated to determine the validity of the assumed tetragonal structure and to find the carbon positions. The structure data are needed to aid in interpretation of the thermal behavior and uranium-carbon bonding of UC₂. BMI-1232 indicated that the X-ray diffraction intensities for UC₂ powder did not agree with those calculated for the tetragonal structure but suggested that the monoclinic ThC₂-type structure should be considered.

Extraction of single crystals of UC₂ has been tried. By etching in a solution of sodium dichromate in phosphoric acid, fine crystals of UC₂ were obtained; however, these were too small for study. An ingot of cast UC₂ is being heat treated in the cubic-phase field and slow cooled through the transformation in an attempt to obtain larger crystals.

J-1

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

C. L. Peterson, P. D. Miller, E. F. Stephan,
T. E. Snoddy, W. D. Beasley, 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 Zircex Process

In the Zircex process, use is made of the volatility of ZrCl_4 to separate uranium from zirconium by hydrochlorinating the fuel element at elevated temperatures. Both a liquid-phase and a gas-phase hydrochlorination have been under study.

The screening study of materials exposed to boiling $\text{AlCl}_3 \cdot \text{NH}_4\text{Cl}$ in the presence of gaseous HCl has been completed. Table J-1 summarizes the data obtained. Of the five materials which were exposed for 7 days, molybdenum, nickel, and Hastelloy X have corrosion rates sufficiently low to be worthy of further consideration. Platinum, gold, and tungsten also showed little attack but are probably less practical as construction materials. The nickel specimens and the specimen of Hastelloy X exposed to the liquid phase showed slight intergranular penetration at the surface. Molybdenum, Hastelloy C, and Nichrome V appear to be structurally sound.

The results from the gas-phase hydrochlorinator cyclic tests were summarized in BMI-1238.

No further work in either of the above programs is contemplated in the immediate future.

The Darex Process

A new series of evaluations has been started in the flowing dissolver apparatus which has been described previously. Twenty-six titanium specimens are being exposed to the liquid and vapor in the flask and to the vapor and condensate in the side arm leading to the condenser. The specimens include examples of butt welds, lap welds with two beads, and lap welds with one bead in Ti-55A alloy. Stressed specimens of Ti-75A material are also included in this series.

Plans are under way for starting an evaluation program for titanium in simulated feed-adjustment tank (FAT) solutions.

TABLE J-1. MATERIALS EXPOSED TO BOILING $\text{AlCl}_3 \cdot \text{NH}_4\text{Cl}$ PLUS HCl GAS AT 425 C

Metal	Corrosion Rate, mils per month, for Indicated Exposure			
	24-Hr Exposure		1-Week Exposure	
	Liquid	Vapor	Liquid	Vapor
Platinum	0.00	0.00		
Gold	0.01	0.01		
Tungsten	0.1	0.3		
Molybdenum	0.6	0.4	0.04	0.02
Nickel	6.4	0.2	1.2	0.08
Hastelloy X	8.2	0.5	2.7	0.07
Nichrome V	--	--	6.2	0.13
Hastelloy C	12.3	1.6	8.4	1.6
Hastelloy C stabilized	10.2	1.0		
Inconel X	10.4	0.2		
Illium R	10.5	0.7		
Hastelloy B	11.4	2.0		
Monel	11.9	0.9		
Type S-590	13.6	0.7		
Inconel	14.0	0.1		
Hastelloy F	16.1	0.6		
Type S-816	16.5	0.2		
Carpenter 20 stabilized	17.6	0.7		
Hastelloy W	18.2	2.4		
Haynes 36	40.2	4.2		
Haynes 25	43.9	0.9		
Haynes 30	50.6	0.2		
Haynes 23	66.5	2.4		
Haynes 21	82.9	3.8		

J-3

Fluoride-Volatility Process

Fuel elements containing zirconium as a diluent or cladding can also be reclaimed by a fluoride-volatility process. The first step in this process consists of hydrofluorinating the elements in a molten bath of $\text{ZrF}_4\text{-NaF}$ with a stream of HF . A material of construction for this hydrofluorinator is being sought.

Results with Inconel and Hastelloy W were reported in BMI-1238. Tests have now been completed with INOR-8, "A" Nickel, and deoxidized copper. Table J-2 summarizes the data obtained to date. Corrosion rates are given from calculations based on weight loss (specimens cleaned in ammonium oxalate) and metallographic sectioning. It can be seen that the corrosion rate for INOR-8 was only 0.09 mil per month, while for Hastelloy W, a slight weight gain of 80 mg was noted. The durability of deoxidized copper was not established by the test reported because a white metallic coating was plated onto the specimen during exposure. The outer portion of this layer was soluble in nitric acid and gave a confirmatory test for nickel. No confirmation for chromium was obtained on a portion soluble in hot hydrochloric acid. Metallographic sections showed that this metallic layer was approximately 1/2 mil thick. As would be expected, grain growth occurred in the copper specimen during exposure.

TABLE J-2. PRELIMINARY RESULTS FROM FLUORIDE-VOLATILITY STUDIES

Test	Exposure Time, hr	Specimen Material	Corrosion Rate, mils per month	
			Determined by Weight Loss	Determined by Metallography
1	36	Inconel	13.5	120.
2	96	Inconel	22.2	85.
3	2	Inconel	2.0	1410.
4	24	Inconel	38.6	--
5	250	Hastelloy W	(a)	--
6	250	INOR-8	0.09	--
7	192	"A" Nickel	8.9	--
9	8	Deoxidized copper	(b)	--

(a) Weight gain of 80 mg was noted.

(b) Corrosion rate not determined; white metallic coating plated on specimen during exposure.

J-4

Photomicrographs of exposed INOR-8 specimens showed some selective attack and penetration. This was confined to the area of the tube at which gas impingement took place. The other portions appeared unattacked. Longer exposures are planned for this material.

The photomicrographs show some intergranular attack on the exposed nickel specimens. This occurred over the entire surface of the specimen.

Two modified experimental assemblies are now being evaluated for use with longer term exposures. Monel and INOR-1 specimens are under test in these units.

K-1 and K-2

K. EVALUATION OF A REFLECTOR-CONTROLLED
HETEROGENEOUS BOILING REACTOR

R. G. Wilson, D. A. Dingee, J. W. Chastain

An attempt is being made to determine the effects of voids which run completely through the reactor such as those caused by (1) the control-rod channels and (2) the method of introducing boiling voids in earlier reflector-control studies. It is believed that these voids may serve as streaming paths for neutrons.

During December, experiments were performed with the core (a 24-in. cube) containing 32 voids, each 1-1/2 by 1-1/2 by 24 in., distributed in a uniform array throughout the core. Two situations were examined: one with fuel strips located in the voids and the other with no strips in the voids. The data obtained in the present experiments include critical mass, rod worth, and detailed flux plots through the core and in the neighborhood of fuel and voids. Results of these experiments will be compared with earlier tests during which the reactor geometry and total moderator content were the same but with no axial through-voids present in the core.

During the remainder of the program, two more core conditions will be examined. These will have larger numbers of voids of smaller cross section but constant moderator content in the core. Comparable data will be obtained.

L-1

L. STUDIES OF SODIUM-TANTALUM COMPATIBILITY
AT ELEVATED TEMPERATURES

J. H. Stang

Research is being conducted to investigate the tantalum-sodium combination which is to be present in LAMPRE I, a fast reactor being developed at Los Alamos Scientific Laboratory. Tantalum will be present in this reactor as the container material for the molten plutonium-alloy fuel while sodium will serve as the heat-transfer fluid.

During December, one flow-loop system was made ready for experiments to determine the strength and corrosion characteristics of tantalum specimens being exposed to 1200 F peak-temperature sodium. Assembly of four other similar systems is in progress..

Recent recrystallization and annealing experiments to obtain fine-grained low-impurity tantalum for inert-atmosphere creep testing have indicated that satisfactory grain refinement (starting with a coarse-grained material) can be achieved by cold reduction to 80 per cent and over followed by annealing. Best results were obtained with unidirectional rolling.

Studies using ultrasonic energy subsequent to welding have indicated that the higher the temperature of tantalum at the time of the application of energy the finer the grain size in the weld zone. Resistance-welding studies indicated that two tube-to-header joints under investigation are promising from the viewpoint of mechanical strength. However, it has not been possible to obtain satisfactory fusion bonds by resistance welding since electrode sticking has accompanied melting in the weld zone.

Tantalum-Sodium Compatibility Studies

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

This research is directed toward an evaluation of the creep and corrosion behavior of stressed tantalum specimens during exposure to 1200 F flowing sodium, which has an oxygen level in the <10-ppm range. The basic design of the polythermal flow loops to be employed in this investigation was described in some detail in BMI-1238. Briefly, the loops are fabricated of Type 316 stainless steel and are equipped with facilities for stressing sodium-immersed tantalum specimens and for monitoring resulting specimen creep rates.

During December, the assembly of the first creep-corrosion loop assembly was completed and, late in the month, the system was charged with prepurified sodium for decontamination. The sodium used here was purified by prior cold trapping and gettering in an auxiliary flow system; all sodium introduced into the test loop systems, whether initial cleanup or for actual testing, will receive a similar prior purification treatment. During test runs, additional purification will take place continuously since the sodium circulated through the loop will pass through a zirconium-foil-filled hot trap.

L-2

The first creep test of tantalum in a sodium environment will start in early January; also during January the installation of four additional loop systems will be nearly completed. The initial test will feature arc-cast tantalum sheet stressed at about 17,500 psi; according to present plans, the run will be of about 1000 hr in duration. Data now on hand from inert-atmosphere creep tests of tantalum (covered in the next subsection of this report) indicate that the normal amount of deformation for such material at 1200 F is 1.5-2.0 per cent per 1000 hr with the above load.

High-Temperature Mechanical Properties of Tantalum

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

As mentioned in BMI-1238, emphasis is being placed on obtaining fine-grained degassed tantalum for 1200 F inert-atmosphere creep testing. The treatment involves solid-state degassing at about 4800 F, followed by work hardening with cold reduction and recrystallization annealing. Studies were made during December of the recrystallization behavior of large-grained thermally degassed arc-cast tantalum sheet after cold rolling to various deformations. Annealing temperatures of 2350 F and 2650 F, an annealing time of 10 min, prior deformations of 57 through 95 per cent reduction, and unidirectional rolling versus alternate cross rolling were investigated. Fairly satisfactory grain refinement occurred with recrystallization after cold reductions of about 80 per cent and over, when the rolling was largely in one direction. Additional tests are planned to verify these results. The mean grain size area obtained at 80 per cent reduction was about 0.002 mm^2 , as compared with 0.001 mm^2 for annealed commercial 0.030-in. sheet from arc-cast tantalum used in previous tests.

As a result of the large reductions needed to provide satisfactory grain size, thermal degassing will be investigated on tantalum of greater thickness (about 0.150 in.) than that used to date. This, however, will be delayed temporarily since the water-jacketed Vycor furnace chamber failed during recent degassing runs. A new modified chamber is now under construction.

Inert-atmosphere creep tests at 1200 F on annealed arc-cast tantalum and on tantalum after exposure to sodium (for reduction of oxygen level) were continued during December. The creep data obtained were generally in accord with data previously established. In addition to these continued tests, a creep test on a tantalum weldment was also started during the month.

Weldability of Tantalum for High-Temperature Systems

R. L. Koppenhofer, J. J. Vagi, S. M. Silverstein, and R. P. Sopher

Research is being conducted to appraise the usefulness of ultrasonic energy in preventing excessive grain growth in tantalum welds. Work during December with bead-on-plate welds has indicated that relatively small grain sizes can be obtained in the weld if the plate is ultrasonically excited immediately after arc welding. It appears, however, from examinations of various weld-zone sections along the length of such welds, that the

L-3 and L-4

temperature of the solidified material at the time of application of energy influences final grain size. Sections taken from areas near the start of the weld, which had the greatest opportunity for cooling, displayed larger grains than sections taken from areas near the end of the pass; the latter were at relatively high temperatures during the period of energy application. While application of ultrasonic energy to tantalum in the molten state is of considerable interest, experiments to date have indicated that shaking out of molten metal is a leading problem. As pointed out in BMI-1238, it may be that a reduced depth of penetration will be helpful in resolving this situation.

Resistance-welding studies conducted on two additional tube-to-header joint designs have proved promising, and as a result further testing is planned for each design. Concurrent with these tests, the evaluation of electrode materials has been continued. The best electrode material found to date is tungsten which is internally and externally cooled during welding. However, it has, in general, been found that electrode sticking occurs if enough heat is generated to cause melting in the weld nugget.

Another technique that has been used to evaluate resistance welds is the peel test. The findings indicated that good strength properties had been obtained in welds in which only a solid-phase bond existed. As yet, bonds formed by a melting process have not been evaluated. Once electrode materials satisfactory from the standpoint of electrode sticking have been found, fusion-type welds will be made for subsequent strength determinations.

M-1

M. DEVELOPMENTAL STUDIES FOR THE PWR

R. W. Dayton

Flow studies in the PWR model were made to determine the effect of flow deflection during two-, three-, and four-loop operation. This completes the present phase of the work.

Preliminary tests have been made with the X-ray photometer being constructed for the examination of fuel elements. The changes found to be required are being made. Sensitivity tests will be made soon.

In studies of pressure bonding fuel plates containing uranium dioxide cores, good bonds have been obtained consistently when using annealed material finished by belt abrasion. Cold-rolled Zircaloy 2 components, similarly finished, do not grow grains at all places across the interface between components, though bonds of this type may be as strong and corrosion resistant as any. Reaction between Zircaloy 2 and uranium dioxide can be avoided completely by a metal coating on the oxide. Radiation samples are being prepared.

Reactor Flow Studies

L. J. Flanigan and H. R. Hazard

Air-flow studies in a quarter-scale model of the PWR reactor are being conducted to determine the effects of lower-plenum geometry on mixing and flow distribution in Core 2 with a 9-ft core length. Previously reported work includes core-flow distribution studies and mixing studies involving various geometric changes in the lower plenum, selection of one semicircular flow-deflector design as a best compromise between mixing and pressure drop, and an in-place calibration of the control orifice and measuring orifice in each fuel assembly over a range of flows.

Core-flow distribution, shield velocities and directions, flow-baffle pressure loss, and mixing for two-, three-, and four-loop operation were determined during December. The studies were made with the basic 9-ft core design and with the basic 9-ft core design with flow deflectors.

The addition of flow deflectors around each inlet improved mixing for all loop configurations with very little effect on flow distribution in the core or thermal shields. Flow deflectors increased the static-pressure drop between the inlet pipe and the core inlet, downstream of the flow baffle, by a factor of 1.95 over that obtained with the basic 9-ft core design.

An interim report covering all the studies made with the 9-ft core design is being prepared. This completes the work to be done under the present contract.

X-Ray Photometric Examination of Fuel Elements

J. B. Schroeder and C. M. Schwartz

An X-ray photometer, utilizing a collimated X-ray source and scintillation detector, is being constructed.

Preliminary tests of the X-ray tube mounted on the lathe showed that the intensity of scattered X-radiation from the vicinity of the tube head was high. Additional lead shielding has been provided to reduce this to a safe level. Minor additions were made to the electronic circuitry to permit proper coupling. A fast-count-rate meter has been ordered, which may make it possible to eliminate the interpolation scales circuit and improve the counting statistics.

It is planned to test the sensitivity by using 0.135-in. -thick filler specimens into which steps have been milled.

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

S. J. Paprocki, E. S. Hodge, D. C. Carmichael,
P. J. Gripshover, and H. D. Hanes

An investigation is being conducted to determine the feasibility of preparing a plate-type fuel element containing compartmented uranium dioxide fuel by pressure bonding; this process utilizes gas pressure at elevated temperatures to achieve bonding. Investigations are being conducted to determine suitable conditions for obtaining Zircaloy-to-Zircaloy bonds and to develop a suitable barrier layer to prevent fuel-cladding reaction. Irradiation specimens are also being prepared for irradiation testing.

A study of various methods of surface preparation has revealed that annealed or hot-rolled Zircaloy 2 that had been machined or belt abraded can be satisfactorily pressure bonded consistently. Cold-rolled material prepared in a similar manner does not yield comparable bonds. Methods of barrel finishing, pickling, hot peening, and vapor blasting which have been investigated do not produce surfaces that are sufficiently clean for pressure bonding, as reported previously.

The use of an annealing heat treatment before or after bonding is being investigated to determine if there is a resulting improvement in bonding. Sections of specimens that had been previously bonded were placed in an argon-filled Vycor bulb and annealed for 1-1/2 hr at 1750 F. These specimens showed substantially increased grain growth across the bond interface. Similar results have been obtained with a 4-hr heat treatment at 1650 F in a vacuum furnace. It has also been found that any heat treatment in the beta region will produce 100 per cent grain growth across the interface regardless of slight bond contamination. Examination of a specimen with a nickel barrier layer indicated no core-cladding reaction even after a beta anneal. Specimens are also being prepared from material that has been beta treated before bonding.

M-3

Large flat-plate specimens (11 by 2.25 by 0.08 in.) have been pressure bonded under various conditions, using both simulated ceramic fuel and uranium dioxide cores. Specimens that were bonded with as-received surfaces which had been annealed, vapor blasted, and pickled, revealed poor bonding due to contamination on the pickled surface. Grain growth across the bond interface was increased to 50 per cent by a 4-hr anneal at 1650 F and to 100 per cent by a 15-min beta anneal. Several specimens which were bonded when using components which had several mils of material machined from the surface resulted in 100 per cent grain growth across the interface. Similar specimens that were belt abraded exhibited 100 per cent grain growth across the bond interface at the solid ends but only 80 per cent at the ribs, where slight contamination was visible. Since specimens prepared from solid plates that had been belt abraded exhibit 100 per cent grain growth, it appears that the excessive belt pressure on the ribs during abrading of the punched receptacle plate causes contamination which prevents complete grain growth. This difficulty could be corrected by punching the receptacle plates from belt-abraded stock.

An investigation has revealed that if the reaction of the Zircaloy cladding with the Ti-Namel spacer during pressure bonding is found to be detrimental, it can be eliminated through the use of very thin stainless steel barrier shims between the spacers and the cladding. A thin chromium plate on the spacers will also provide adequate protection; this is also being considered.

To reduce production costs, however, it may be desirable to eliminate the can entirely. Specimens with the cladding fusion welded to the frame around the edges were pressure bonded and examined metallographically. Encouraging results were obtained with this technique; however, the contamination introduced during the welding operation was too severe to permit obtaining a completely satisfactory bond. Welding techniques will be investigated further to determine whether omitting of the protective can for pressure bonding results in considerable cost reduction.

Results of the study of the reaction occurring between the uranium dioxide cores and the Zircaloy 2 cladding during pressure bonding indicate the amount of reaction is not as severe as originally predicted from reaction studies and that a barrier layer may not be necessary if pressure bonding times of 4 to 5 hr at 1550 F are used. If a barrier layer is necessary, electroplated or chemical-displacement coatings of chromium, nickel, or iron appear suitable. Specimens rendered defective and sectioned specimens containing a 0.001-in. barrier layer of iron exhibited no growth or attack after 6 weeks in 750 F steam. Specimens containing 0.0005-in. electroplated coatings of chromium evidence no attack after 2 weeks of testing. Specimens intentionally made defective and containing nickel foil, Kanigen nickel, and electroplated nickel barriers have grown only a small amount after 6 weeks in 750 F steam.

X-ray analysis of cores exposed to 750 F steam indicated that some of the uranium dioxide was oxidized to U_3O_8 by oxygen contained in the corrosion media. A specimen made defective and with no barrier layer between the Zircaloy clad and the Al_2O_3 - UO_2 core has exhibited 0.011-in. growth after 6 weeks as a result of the formation of U_3O_8 . Additional corrosion tests in 680 F deoxidized water have been initiated. The specimens on test, which contain intentional defects, are Zircaloy-clad Al_2O_3 -21 w/o UO_2 cores and 100 w/o UO_2 cores with various barriers and heat treatments. Specimens identical to those run in the 750 F steam are being run in the 680 F deoxidized water.

M-4

A series of irradiation specimens, which were pressure bonded before the deleterious effect of cold work was determined, had cold-rolled cladding components and consequently did not exhibit 100 per cent grain growth across the bond interface. Additional specimens of the same type are being prepared from hot-rolled material, and stainless steel barriers are being used to prevent the Ti-Namel-Zircaloy reaction. Components for additional sets of irradiation specimens have been machined.