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PROGRESS RELATING TO CIVILIAN APPLICATIONS
DURING OCTOBER, 1957

by

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REPORTS RELATING TO CIVILIAN APPLICATIONS
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- BMI-1223 "Properties of Some Refractory Uranium Compounds", by M. Jack Snyder and Winston H. Duckworth.
- BMI-1224 "High-Strength Materials for Pressurized Water In-Pile Tubes", by Ward F. Simmons, Walter K. Boyd, Raeman P. Sopher, and Fred H. Lyon.
- BMI-1225 "Studies Relating to Hydrogen in Dingt Uranium", by Max J. Trzeciak and Manley W. Mallett.
- BMI-1226 "Progress Relating to Civilian Applications During September, 1957", by Russell W. Dayton and Clyde R. Tipton, Jr.

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A. DEVELOPMENTS FOR ZIRCONIUM-CLAD FUEL ELEMENTS

F. R. Shober

Equipment constructed to measure the thermal conductivity and electrical resistivity of zirconium-clad uranium specimens before and after irradiation is being calibrated using a known standard for the specimen. Calibration of the test apparatus being assembled for thermal-conductivity measurements of UO_2 will be started. Creep tests on transverse specimens of annealed Zircaloy 2 at elevated temperatures have been completed and additional tests on the 15 per cent cold-worked materials have been started. Based on early portions of the creep tests, results indicate that the cold-worked material has approximately two times the creep strength as the annealed material at comparable test temperatures.

Evaluation of data from tests to determine the bond strength of zirconium cladding bonded to electropolished uranium has indicated that metallurgical bonds containing a minimum of second-phase material are superior to those having large amounts. The formation of hydrides in zirconium during annealing has been deleterious to grain growth in an attempt to grow single crystals of zirconium by the strain-anneal method.

Thermal Conductivity of Uranium and UO_2

H. W. Deem and C. F. Lucks

An investigation directed toward determining the effect of irradiation on the thermal and electrical conductivities of uranium and on the thermal conductivity of uranium dioxide is being conducted.

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-resistivity measurements will be made on the same specimen by the voltage-drop method. The apparatus for making these measurements has been completed and is being calibrated using an Armco-iron specimen for which thermal and electrical conductivities are known. The calibration runs should be completed during November. The clad uranium specimens to be measured are being fabricated by another laboratory.

Uranium Dioxide

An apparatus for making thermal-conductivity measurements on UO_2 , both before and after irradiation, was designed and is being assembled. An absolute method is being used. Difficulties will probably be encountered in guarding and

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measuring temperatures because of the small specimen size and small heat flows involved. The apparatus was designed to be versatile and allow several approaches to temperature measurements and guarding. It is expected that calibration of the apparatus will begin during November.

Mechanical Properties of Zirconium Alloys

F. R. Shober and J. A. VanEcho

An investigation of the creep strengths of annealed and cold-worked zirconium alloys is being conducted to evaluate these alloys for elevated-temperature use. This investigation includes a study of the variation of the mechanical properties with individual lots of Zircaloy 2, determination of the creep properties of cold-worked Zircaloy 2, and the degree these properties are retained during creep at elevated temperatures.

The results from completed tests and tests in progress are shown in Table A-1. Total deformations obtained at the test temperatures and stress levels chosen have been less than 2.0 per cent. The creep rates, in many cases, have been less than 1×10^{-5} per cent per hr.

Cold-worked Zircaloy 2 is being tested over a temperature range similar to that used for the annealed material, but stress levels are generally greater by a factor of two. These tests will be 2000 hr in duration.

Zirconium Cladding

F. R. Shober and R. F. Dickerson

The metallurgical bond formed between electropolished uranium and zirconium cladding by hot hydrostatic pressing is being evaluated. The diffusion zone has been examined metallographically, and core and cladding samples have been heat treated and water quenched to test the strength of the bond. A second type of test for bond strength, consisting of welding a threaded zirconium stud to the cladding and measuring the load necessary to separate the cladding from the core, has been utilized.

Evaluation of data from tests to determine the bond strength of the cladding to the core after pressurizing has been inconclusive. Table A-2 summarizes these data. Specimens which withstood severe quenching conditions for numerous times had varying bond strengths as determined by the welded-stud tensile technique. It appears that the bond is sufficiently strong to withstand a beta heat treatment; this is a desirable condition for a uranium core material in production reactors.

One specimen is being pressurized at 10,000 psi for 6 hr at 1200 F. This specimen is to be shipped to another laboratory for examination upon completion. One other is to be pressurized at 1350 F, but selection of the time at pressure has been delayed to evaluate further the results previously obtained at this temperature.

TABLE A-1. CREEP PROPERTIES OF ANNEALED ZIRCALOY 2, ZIRCALOY 3, AND 15 PER CENT COLD-WORKED ZIRCALOY 2 AT ELEVATED TEMPERATURE

Specimen	Temperature, F	Stress, psi	Time at Test, hr	Deformation at Indicated Time, per cent							Creep Rate, per cent per hr	
				Load On	50 Hr	100 Hr	500 Hr	1000 Hr	1500 Hr	2000 Hr		Load Off
<u>Annealed Zircaloy 2, Transverse Specimens</u>												
1-7	450	20,000	2040.0	0.25	1.680	1.695	1.702	1.705	1.716	1.723	1.548	0.000014 ^(a)
15-5	550	17,000	2037.7	0.38	1.065	1.075	1.085	1.086	1.087	1.090	0.930	<0.00001 ^(a)
15-3	550	15,000	2012.5	0.16	0.470	0.475	0.480	0.480	0.490	0.490	0.386	<0.00001 ^(a)
34-7	550	14,000	2009.5	0.16	0.375	0.382	0.392	0.395	0.402	0.405	0.308	<0.00001 ^(a)
15-4	550	12,500	2033.8	0.15	0.220	0.223	0.230	0.240	0.240	0.242	0.137	<0.00001 ^(a)
15-6	650	17,850	2065.3	1.00	1.400	1.410	1.475	1.490	1.570	1.595	1.451	0.00005 ^(a)
20-5	650	14,000	1150 ^(b)	0.128	0.148	0.164	0.229	0.258	--	--	--	0.00005 ^(c)
20-4	650	12,500	2014.4	0.145	0.200	0.210	0.250	0.275	0.290	0.300	0.197	0.00002 ^(a)
<u>Annealed Zircaloy 3, Transverse Specimens</u>												
3-11	300	20,000	2011.4	0.170	1.240	1.370	1.625	1.750	1.760	1.800	1.636	0.00008 ^(a)
3-12	300	17,500	2014.0	0.140	0.750	0.840	1.035	1.100	1.110	1.113	1.008	<0.00001 ^(a)
<u>15 Per Cent Cold-Worked Zircaloy 2, Longitudinal Specimens</u>												
7-1-2	450	40,000	450 ^(b)	0.400	0.503	0.512	--	--	--	--	--	--
7-1-3	450	30,000	375 ^(b)	0.280	0.333	0.337	--	--	--	--	--	--
7-2-2	550	35,000	150 ^(b)	0.353	0.495	0.520	--	--	--	--	--	--
7-2-3	550	30,000	50 ^(b)	0.275	0.375	--	--	--	--	--	--	--
7-3-1	550	25,000	200 ^(b)	0.205	0.267	0.282	--	--	--	--	--	--
7-3-2	550	20,000	175 ^(b)	0.178	0.223	0.227	--	--	--	--	--	--
7-1-1	650	30,000	875 ^(b)	0.315	0.537	0.592	0.745	--	--	--	--	--

(a) Based on creep deformation between 1500 and 2000 hr.

(b) Test in progress.

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

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TABLE A-2. EVALUATION OF BOND FORMED BETWEEN ELECTROPOLISHED URANIUM AND ZIRCONIUM

Specimen	Pressure, psi	Temperature, F	Time, hr	Test Condition for Bond	
				Number of Water Quenches ^(a)	Bond Strength, psi
E-7	10,000	1350	1	21	1,550
E-3	10,000	1350	2	21	5,000
E-1	10,000	1350	4	21	10,350
E-6	10,000	1350	6	3 ^(b)	--
E-5	10,000	1200	2	12 ^(b)	--
E-4	10,000	1200	4	21	--
E-2	1,000	1200	6	19 ^(b)	9,370

(a) Heated in salt bath to 1345 F for 20 min, water quenched.

(b) Separation of cladding and core.

Preparation of Aluminum, Molybdenum, and Zirconium Single Crystals

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

Single crystals of molybdenum and zirconium are being prepared for irradiation studies.

The length of annealing time for molybdenum in the Andrade furnace has not yet been determined. Time consumed for fabrication and replacement of a vacuum seal on the furnace was greater than expected; consequently the furnace has not been available during a major portion of the report period for grain-growth studies. The seal has been replaced and the investigation is continuing.

Small tensile bars 0.50 and 0.125 in. in diameter are being used, in a variation of the strain-anneal method, for the growth of single crystals of zirconium. It was thought that the formation of hydrides in the zirconium bars hindered grain growth. The amount of hydride appears to have been greatly reduced by vacuum annealing at 855 C for 100 hr. Strain-annealing will continue on these specimens.

Straining of the specimen at elevated temperature to prevent the formation of deformation twins will be attempted. The regular annealing schedule will be followed to produce crystal growth.

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B. DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS

R. J. Carlson

The "right-angle" extrusion-cladding assembly has been fabricated and assembled to check for alignment and clearances between mating parts. Several small modifications are being made. Progress has been made in reducing the pressures required in the extrusion-cladding process by changing the mandrel tip-to-die spacing and by substituting M329 aluminum for 1100 aluminum in the process.

Studies of the liquidus temperature of various aluminum-uranium alloys ranging in composition from 16 to 50 w/o uranium have been completed up to 30 w/o uranium, and a study of binary alloys containing 40 to 50 w/o uranium is in progress. Data obtained to date indicate that additions of up to 1 w/o silicon and 0.5 w/o magnesium have no appreciable effect on the liquidus temperature of alloys containing up to 30 w/o uranium. The effect of these additions will be studied on 40 and 50 w/o uranium alloys.

The construction of the mechanical extensometer designed to study the transverse properties of uranium tubes has been completed and calibration is proceeding.

Extrusion Cladding of Flat Plates

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

Efforts directed toward the reduction of pressure required in the extrusion-cladding process were continued. With one mandrel-die combination the pressure was reduced about 15 per cent when the mandrel tip-to-die spacing was increased from 65 to 90 mils. In other tests, a 15 to 20 per cent pressure reduction was observed when M329 aluminum (99.54 w/o pure) instead of 1100 aluminum (99.0 w/o pure) billets were used. These results were taken from only a few tests, but they indicate that optimum operating conditions may be established by additional studies.

The new "right-angle" extrusion-cladding assembly is nearing completion. The tools were assembled and checked for alignment and clearances between the mating parts. Several small modifications are being made. During November, the tools will be heated to check the temperature distribution throughout the equipment, and the initial extrusion tests should be made.

Study of the Liquidus in Aluminum-Uranium Alloys

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

The aluminum-uranium system is the subject of an investigation aimed at determining the liquidus line up to 50 w/o uranium. Also being studied are the effects of additions of silicon and magnesium on the system.

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Previously reported data indicated a liquidus temperature of 885 C for a 20 w/o uranium alloy. Because this temperature is somewhat out of line with the other data, it was thought the alloy might be much higher in uranium than planned. However, chemical analyses of the nominal 17.2, 20, and 30 w/o uranium alloys revealed the actual uranium content to be 17.5, 20.4, and 30.9 w/o, respectively. A liquidus temperature of 855 C for 20 w/o uranium alloys with additions of silicon and magnesium was determined, which might be expected on the basis of finding no effects of the additions on the liquidus of the other alloys. Therefore, it might be assumed that the correct liquidus is at 855 C. Additional data are necessary to confirm this.

Binary alloys containing 40 and 50 w/o uranium have been prepared and are under investigation. Similar alloys with silicon and magnesium additions also have been prepared and will be studied once the binary work is complete. Although on the basis of results obtained it appears to date that silicon and magnesium in the amounts used for this study have no more effect on the liquidus line than the minor variations in uranium content which occur, it is felt that the 40 and 50 w/o uranium-base alloys should be examined for verification of the trend.

Preparation of Aluminum-Uranium Alloys

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

Alloys of aluminum-uranium containing low concentrations of uranium have proved satisfactory as fuel for low-temperature water-cooled reactors. However, in an effort to broaden their field of application and to enhance their economic value, it is desirable to increase the fissionable material contained in these fuels. Therefore, a program of research concerned with the melting and casting of alloys in this system containing relatively high concentrations of uranium is in progress. Since the proposed fuel-element design requires tubing clad inside and out with aluminum, it is necessary that the alloys be cast in the form of a heavy-walled cylinder suitable for coextrusion. These cylinders must be sound and homogeneous for this purpose. As a supplement to the casting program, a study of the effects of minor additions of magnesium and silicon on the casting properties of the alloys in this system containing up to 50 w/o uranium is being made.

In October, the major emphasis was on preparing the alloys necessary for the latter phase of the program. These alloys were cast in the form of 1-in. -diameter rounds 8 to 10 in. long. All of the castings made were melted in zirconia crucibles under a pressure of less than 5×10^{-3} mm of mercury and poured into heated graphite molds. The magnesium was added to the alloys in the form of a master alloy of aluminum-10 w/o magnesium. Previous to this addition, a partial pressure of helium was introduced in the melt chamber in an effort to retain the magnesium in the melt. The silicon was added both in the form of a master alloy and as pulverized elemental silicon enclosed in aluminum foil. These alloy additions were introduced into the melt approximately 2 min. before pouring. Satisfactory castings of aluminum-40 and 50 w/o uranium have been made containing these alloy additions.

Future work will be concerned primarily with centrifugal-casting studies on alloys containing up to 55 w/o uranium.

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Development of a Mechanical Extensometer

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

A mechanical extensometer with sufficient sensitivity and range to study the transverse mechanical properties of uranium tubes is being developed. Construction of this equipment is now complete and calibration tests are in progress. It is planned that this extensometer will be ready for regular test applications during December.

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C. PLANT ASSISTANCE TO MCW

A. A. Bauer

Structural and compositional variations of the oxides of uranium are continuing to receive study. A technique for the analysis of nitrogen in UO_3 has been successfully developed; however, application of this technique to UO_2 has not been successful to date. A hypothesis that increased oxygen-to-uranium ratios in the UO_2 structure is the result of an uranium-deficient lattice is being tested. UO_2 prepared for this study has been found to obey a different oxidation-rate law than does as-received MCW UO_2 .

Prolonged-exposure tests of Type 304 ELC stainless steel in fluoride- and chloride-contaminated nitric acid solutions have continued with no change in trends from those previously reported for the 5, 18, and 30 w/o acid solutions being observed. Tests have shown that in the 38 and 45 w/o solutions, the chloride must be replenished in order to keep the concentration constant. Increased corrosion rates have resulted in those solutions containing in excess of 0.10 w/o chloride. Tests in 5 w/o nitric acid solutions containing 0.10 to 0.30 w/o chloride additions indicate that an initial high corrosion rate is followed by passivity of the specimens.

Cracks have developed in the molybdenum reaction tube designed to contain the molten magnesium fluoride slag for hydrogen-permeation measurements. Consequently, a new reaction tube is to be constructed.

Preliminary viscosity data have been obtained at 1340 and 1440 C on molten magnesium fluoride slag containing up to 6 w/o magnesium oxide additions. Values on the order of 100 centipoises were obtained. Since the measurements were made in the range in which the instrument does not appear to have adequate sensitivity, modification of the viscometer will be required. MCW slag has been found to attack platinum at 1340 C. Therefore, a material other than platinum will necessarily be employed in making the viscometer spindle.

Investigation of Uranium Oxides

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

Structural and compositional variations in uranium oxides are being investigated as assistance to the production of chemically reactive uranium dioxide and in improving metal quality. The effect of the residual nitrogen content of uranium oxides upon their reactivity is being studied. A method for precise analysis of nitrogen in UO_3 has been developed. Modifications of the colorimetric and Kjeldahl methods have been made to adapt these methods for use with UO_2 , but without complete success. The main problem is in the dissolution of the UO_2 without the loss of nitrogen, and the reproducibility has been poor. Further testing will be continued in November.

The experimental work on the inactive and active UO_2 -oxygen systems has been concluded, and the results are being evaluated. In evaluating the structural and

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electrical-property data on the UO_2 -oxygen system, and, in particular, the regions of the UO_2 and U_4O_9 phases, it appears, from consideration of interstitial hole size, ion size, lattice-parameter change with oxygen content, and conductivity type, that under equilibrium conditions the oxygen-to-uranium ratio of UO_2 is increased by a loss of uranium from its lattice. If so, the formula UO_{2+x} should be written U_{1-y}O_2 . In order to verify this hypothesis, new samples with oxygen-to-uranium ratios from 2.02 to 2.32 are being prepared by oxidation at 180 C, for measurement of density in both the as-made and homogenized conditions. An interesting observation has been made during oxidation of these samples. The UO_2 to be oxidized in oxygen at 180 C was made by reduction at 1000 C to an oxygen-to-uranium ratio of 2.022. The material is light red-brown and obeys a different oxidation-rate law than does as-received MCW UO_2 . Instead of a smooth parabolic oxidation curve (time at 180 C versus oxygen-to-uranium ratio), the red-brown $\text{UO}_{2.022}$ appears to oxidize irregularly, with two or three breaks in the oxidation curve. It is planned to investigate this reaction further.

The Electrical Properties of Uranium Dioxide

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

Experimental studies are continuing at a limited rate. There are no new data to report.

The Corrosion Resistance of Selected Stainless Steels

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

The corrosion of Type 304 ELC stainless steel specimens by nitric acid solutions containing fluoride and chloride contaminations similar to concentrations possible during operation of the nitric acid reconcentrator at the uranium refinery is being studied.

Prolonged-exposure tests in 5, 18, 30, 38, and 45 w/o nitric acid, as described in Table C-2 of BMI-1220, are still in progress. The corrosion rates in 5, 18, and 30 w/o acid solutions show no changes from the trends indicated in this table. Chloride concentrations cannot be maintained unchanged in 38 and 45 w/o solutions for longer than a few days. Consequently, these tests are now run with chloride renewals being made every 2 days in 45 w/o HNO_3 and every 4 days in the 38 w/o HNO_3 solutions. Renewing the chloride concentration has had no effect on the corrosion rates in solutions containing 0.10 w/o Cl^- , or less. But in two of the 38 w/o solutions containing 0.20 and 0.25 w/o Cl^- there was an increase in corrosion rate, while a marked increase occurred in a 45 w/o solution containing 0.20 w/o Cl^- (the rate increased from 0.005 to 0.69 mil per month).

Several additional tests were started in 5.0 w/o HNO_3 solutions. Chloride contaminations of from 0.10 to 0.30 w/o are being studied. It appears from the results

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of the first 1000 hr of exposure with these concentrations that a relatively high corrosion rate is apt to occur during the first part of the exposure with 5 w/o HNO_3 solutions, after which the steel usually becomes passive.

Gas-Metal Studies

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

As part of the program of investigations related to the production of dingot uranium, a thermochemical study is being made of some of the components of the thermite reactor in which dingot uranium is produced. Presently, the study includes the permeation of hydrogen through molten magnesium fluoride slag and also measurements of the viscosity of magnesium fluoride. The permeability study is directed toward understanding the kinetics of the movement of hydrogen in the thermite reactor. These studies may suggest an effective means of lowering the hydrogen content of dingot uranium. The viscosity study includes tests of the effect of normal contaminants on the viscosity of molten magnesium fluoride.

Permeation of Hydrogen

The molybdenum diaphragm fitting reported in BMI-1226 was redesigned, fabricated, and tested to hold molten magnesium fluoride slag. During testing at molten-slag temperatures, two vertical cracks were noticed on the sides of the molybdenum reaction tube. Seepage of molten slag through the cracks occurred when an atmosphere of gas was introduced above the slag. The cracks also allowed gas to be diverted from the intended path of permeation. It is not known whether the cracks were initially present in the fabricated tube or whether they subsequently developed during experimentation as a result of thermal effects on the molybdenum or of interaction of the slag with the tube. Another tube will be constructed.

Viscosity of Molten Slag

Viscosity measurements of ACS reagent-grade magnesium fluoride with and without additions of magnesium oxide have been made using a Brookfield viscometer. Initial data indicate that the absolute viscosity of molten magnesium fluoride is of the order of 100 centipoises at both 1340 and 1440 C and with magnesium oxide concentrations up to 6 w/o. Since the viscosities of most elements and compounds change rapidly with temperature, there is some question as to the accuracy of the data. The present measurements were made in the range of viscosity in which the viscometer does not appear to have adequate sensitivity. It is planned to modify the viscometer to overcome this deficiency and to recheck the data.

A compatibility study between platinum (used in the spindle of the viscometer for viscosity measurements) and the as-received slag was made. Platinum showed attack after a 15-min immersion in the slag at 1340 C. The attack is believed due to the

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metallic-uranium contaminations in the slag. Hence it will be necessary to use a spindle of a different material, probably molybdenum, for measurements on the as-received slag.

D-1

D. PROCESSING OF FEED MATERIALS

E. L. Foster

Two studies in progress at Battelle are concerned with subjects of interest in the processing of feed materials. These are: the examination of factors affecting the quality of vacuum induction-melted uranium, and a theoretical study of the solidification of pure uranium ingots. Both of these are related to the vacuum melting and casting of uranium and reflect the interest of National Lead of Ohio (NLO) in the improvement of uranium for fuel slugs. However, of more importance in the solidification study is the evaluation of factors such as ingot shape and mold material for future casting needs.

Solidification of Uranium

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

A theoretical study concerned with the solidification of uranium is in progress. This program, of interest to NLO, has as its primary objective a more comprehensive understanding of the problems inherent in casting large masses of uranium. Such a study should yield information valuable in determining the effects of mold materials, ingot shapes, pouring temperatures, and other casting variables on the quality of the ingots produced. The general nature of the program is such that this information, when properly interpreted, will enable those concerned to evaluate the effects of ingot shapes and mold material on the quality of castings that may be produced in the future.

Since its inception, the program has concentrated largely on the mathematical treatment of heat flow in the casting and mold. The initial analytical information has consisted of a set of simultaneous equations which would describe the transient heat flow through the materials. Programming of the initial formulation of the relationships for determining the heat flow during the solidification of a cylindrical casting has been completed. A pie slice of the casting and mold material was used for this study because the heat flow is expected to be axially symmetric. This pie slice, divided into 20 separate cells, permits computation of the heat flow on the basis of a "lumped constant" approximation to the real casting. This "lumped constant" concept implies that each cell is characterized by its central temperature which is determined by the heat flow between it and the adjacent cells. The equations for heat flow were programmed for solution by the IBM 650 digital computer, and trial computations were made. The results of these trial computations were obtained in the form of temperature-time data for each of the 20 cells considered.

The time-temperature data obtained showed the expected decay of temperatures. This is considered evidence of the validity of the method of analysis employed. However, it should be noted that these initial results should not necessarily be expected to represent the real situation until certain additional refinements are incorporated. The work now being performed is to further develop the mathematical model to include the following effects:

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- (1) The temperature dependence of the heat flow constants
- (2) The release of the heat of fusion
- (3) The separation of the casting from the mold wall during the solidification process.

Also, a finer subdivision of the casting and mold material will be used to increase the number of points at which the temperatures will be determined.

This initial analysis has been useful in verifying the methods of analyses used, and it has also indicated that physical constants and experimental data must be obtained to permit a more refined analysis. These refinements should assist in bringing the mathematical solution into agreement with the results noted in production practice.

Future work will be concerned with a continuation of the mathematical analysis as discussed above and with completion of the literature survey. This is desirable as an aid to the evaluation of the metallurgical considerations of the problem and to the compilation of the factors and constants associated with the solidification of castings.

Examination of Factors Affecting the Quality of
Induction-Melted Uranium

E. L. Foster and R. F. Dickerson

Uranium fuel slugs may be produced by a series of operations that include the vacuum-induction melting and casting of large ingots. In these operations, large quantities of materials are handled. For economic reasons, it is necessary to recycle material from the various fabrication operations. Unfortunately, the type and quantity of impurities in the material charged for melting varies to a great extent, and existing data indicate that the quality of uranium-fuel slugs may be related to the quantity and type of nonmetallic impurities present in the uranium metal. As a result, specifications have been written to relate this data to the acceptance of fuel slugs for reactor use. Further, there is little doubt that ultimate metal quality is very closely associated with the type of charge material and the procedures followed in the melting operation.

National Lead of Ohio, in connection with its operation of the Feed Materials Processing Center, has an active interest in the resolution of the factors involved in the melting of uranium. In the past, it has conducted several studies concerned with the effect of charge materials on ingot chemistry and has integrated its findings into production runs. However, the resolution of all the factors involved is difficult. Some of the difficulties are caused by the following: (1) the large castings that are produced makes detailed evaluation costly and cumbersome; (2) the charged materials vary in composition (the charge may be composed of as-reduced derbies, compacted machine chips, ingot cropping, fabrication scrap, or mixtures of these materials); (3) the portions of charges differ for different materials and in some instances for the same material; and (4) the chemical reactions that may occur during melting are unknown and, therefore, no controls have been set up.

D-3 and D-4

As a result of these difficulties, it has been requested that a study be initiated to determine the basic variables involved in the vacuum melting of uranium metal. This study will consist of two phases. The initial phase is concerned with a literature survey to gather known data on the kinetics and thermodynamics of the reactions that may occur. The second phase is experimental in nature and will deal with studies of reactions which take place during vacuum melting. Charges of known compositions will be subjected to analysis after melting. This analysis will include samples from ingots, crucible skulls, slag, and condensables. In addition, particular note will be made of outgassing products by the use of a mass spectrometer.

Efforts to date have been preparatory in nature in that materials have been gathered and equipment adaptation has begun.

E-1

E. GENERAL FUEL-ELEMENT DEVELOPMENT

F. A. Rough and D. C. Carmichael

Fuel-element-development programs supported by the AEC Division of Reactor Development are reported in this section. Stainless steel-clad specimens containing UN or UC fuel dispersed in a stainless steel matrix are being machined for tensile tests.

Methods of preparing cermet fuel elements with high loadings of fuel in a metal matrix are being investigated. Procedures have been determined for hot pressing cermet cores to a high density, and electrical-resistivity measurements have indicated that the resistivity of UO_2 is decreased by a factor of 10^6 when a small amount of metal matrix is present.

A study is being made of the feasibility of a fueled zirconium hydride moderator. Construction and calibration of an apparatus for measuring hydrogen-sorption isotherms to determine phase relations in the hydrogen-zirconium-uranium system have been completed.

Development of Dispersed UC and UN Fuel Elements

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

A limited number of tensile tests are to be conducted on UN and UC dispersions. Specimens containing 24.2 w/o UN or 24.3 w/o UC dispersed in stainless steel and clad with Type 347 stainless steel have been fabricated for this purpose, and are now being machined to size.

Fabrication of Cermet Fuel Elements

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

Methods of fabricating cermets with high loadings of uranium are being investigated. Systems being studied are 60 to 90 volume per cent loadings of UO_2 , UN, or UC in a stainless steel matrix and 60 to 90 volume per cent of UO_2 in molybdenum.

A hot-pressing technique has proven to be the most feasible method of compacting cermet cores to a high density (greater than 90 per cent of theoretical). The method consists of welding the cores in a picture-frame pack and press forging at 1800 to 2100 F to a 35 per cent reduction in thickness. Higher density compacts are obtained when stainless steel is used as a picture frame than when mild steel is used. The optimum temperature for pressing appears to be 1900 F. At higher temperatures the pack is so ductile that little reduction of the core occurs, and at lower temperatures the entire assembly is so resistant to deformation that high pressures are required.

E-2

The electrical resistivity of 74 volume per cent UO_2 in stainless steel has been measured on compacts of 82 to 92 per cent of theoretical density. In general, as the density increases, the resistivity decreases. However, it should be noted that even specimens with densities of 82 per cent of theoretical have values of approximately 1.5×10^{-3} ohm-cm as compared to an approximate value of 1×10^{-3} ohm-cm for pure UO_2 . Electrical resistivity was also measured on specimens of 80 volume per cent UN in stainless steel which had a density of 94 per cent of theoretical. Values of approximately 1.5×10^{-4} ohm-cm were obtained. Thus it appears quite feasible to lower resistivity values by a factor of 10^6 with small additions of stainless steel or other metals.

Tests to determine elongation and bend strength are now in progress on these specimens but have not been completed.

Fueled Zirconium Hydride Moderator

H. E. Bigony and J. B. Vetrano

During October, the system for measuring hydrogen-sorption isotherms was completed, the volume of the sorption system was determined from room temperature to 800 C, and the Autovac vacuum gage was calibrated against a McLeod gage for dry hydrogen gas.

Measurements of hydrogen-sorption isotherms are being started on a sample of zirconium-50 w/o uranium alloy, as planned originally and reported in BMI-1220, in order to check results obtained against those of Gulbransen. These measurements will cover a temperature range of 600 to 650 C and will be reported next month.

After completing the limited study on the zirconium-50 w/o uranium alloy, the investigation will be concentrated on measuring hydrogen-sorption isotherms on the zirconium-1 w/o uranium alloy. Samples in the vicinity of phase boundaries will also be prepared for high-temperature X-ray diffraction investigation and electron-fluorescence examination. As reported previously, these techniques should be of value in defining phase boundaries more accurately, as well as in indicating whether there has been any segregation of the alloy components.

F-1

F. STUDIES OF URANIUM AND URANIUM-ALLOY FUELS

F. A. Rough and A. W. Hare

Uranium and uranium-alloy fuel studies sponsored by the Division of Reactor Development are reported in this section.

Irradiation of zirconium-uranium specimens is continuing. Capsules of both 22 and 50 w/o uranium specimens are at the MTR.

In the studies of gamma-phase uranium alloys, routine progress is reported on new studies. Studies begun last year of 70 w/o uranium alloys containing various amounts of zirconium and niobium reveal that the samples continue to look good in water at 680 F after 840 hr.

The program on preparation of uranium-niobium alloys is concerned with evaluation of methods suitable for 20 and 80 w/o niobium.

Studies of uranium corrosion are proceeding routinely. Methods of analysis of the chemical valence state by X-ray absorption spectra are being calibrated prior to evaluation of uranium valence in the corrosion product.

Radiation Stability of Uranium-Zirconium Alloys

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

Irradiation-effects studies on uranium-zirconium alloys are continuing. The temperature-controlled Capsule BMI-8-5, which contains two uranium-50 w/o zirconium specimens, is operating satisfactorily in the MTR. The specimens are being irradiated at a temperature of 675 C in an estimated thermal-neutron flux of from 0.5 to 1.0×10^{-14} nvt.

A noncontrolled capsule, which contains three zirconium-22 w/o uranium specimens of three different heat treatments, is at the MTR awaiting irradiation. Indications are that this capsule will not be inserted into the MTR until December, 1957.

The literature survey which had been initiated to summarize the technology of uranium-zirconium alloys is being continued.

Development of Gamma-Phase Uranium Alloys

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

The development of a corrosion-resistant gamma-phase uranium alloy through ternary or quaternary alloying with chromium, molybdenum, niobium, ruthenium, vanadium, and zirconium is the object of this investigation.

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As part of a preliminary screening investigation of these alloys, a rough study of the transformation kinetics is being made employing hardness measurements. To date, uranium-niobium-base alloys with ternary additions of zirconium have exhibited the greatest thermal stability. However, the uranium-molybdenum-base alloys have yet to be studied. Also, all alloys are to be corrosion tested in 500 F water in the quenched condition.

Additional data have been obtained on uranium-zirconium alloys prepared for a previous program aimed at developing a corrosion-resistant 70 w/o uranium ternary alloy. A base uranium-23.8 w/o zirconium-6.1 w/o niobium alloy, water quenched from 900 C and annealed for 24 hr at 500 C, continues to exhibit a corrosion rate of approximately $0.04 \text{ mg}/(\text{cm}^2)(\text{hour})$ after 840 hr in 680 F water. Uranium-23.2 w/o zirconium-4.84 w/o niobium alloys in the quenched and 500 C annealed condition along with air-cooled and a furnace-cooled specimen are still on test after 840 hr, although their corrosion rates are somewhat higher, ranging from 0.10 to $0.36 \text{ mg}/(\text{cm}^2)(\text{hour})$. The same alloy air cooled and annealed at 500 C was removed from test after 24 hr because of a high weight loss.

Similar alloys, clad and artificially defected, and tested in the as-clad and water-quenched conditions were removed from test after 672 hr because of the failure of the Zircaloy 2-to-Zircaloy 2 bond. Actual corrosion damage to the core was confined to a blister at the defect.

Additional tests of these alloys are being conducted.

Preparation of Uranium-Niobium Alloys

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

Attention, in varying degrees, has been directed to alloys of the uranium-niobium system. Specifically, the uranium-10 w/o niobium alloy has been studied in connection with its application as a blanket material. Other alloys containing niobium in greater quantities have received only limited attention; however, favorable properties as a high-strength high-temperature fuel-element material have been indicated. Because of these favorable properties, alloys of the entire system are deserving of a more intense study.

The program was initially concerned with a literature survey of past work on alloys of this system. The survey and evaluation of past work include information on alloy-preparation procedures, constitution relationships, kinetics, heat-treatment effects, physical and mechanical properties, corrosion behavior, and the effects of irradiation on the alloys. Several portions of the survey have been completed.

Experimentally, the program is mainly concerned with the development of procedures for preparing high-integrity alloy material for study. In this vein, ingots containing 20 and 80 w/o niobium have been arc melted, sectioned for examination, and remelted. The specific alloys chosen for study were representative of the

F-3 and F-4

phase-diagram relationships in the areas of interest. Further, very little information on preparation procedures at these alloy contents was available in the literature.

Radiographic examination made after the first melt showed extensive banding-type segregation. This type of segregation has been noted in the arc melting of uranium-10 w/o niobium alloys. It has been found to be caused by a combination of density- and heat-fluctuation factors that are present during solidification of some arc-melted alloys. Evaluation of the persistence and degree of segregation after remelting is progressing.

The study of preparatory techniques for the alloys in question will be continued. Since the indications to date are that all alloys across the diagram tend to "band" it is essential that a homogeneous material be prepared and that the alloy system be studied with this material. Consumable-electrode arc-melting studies will continue and the use of skull-melting techniques will be investigated. The literature survey will be completed and evaluated.

Mechanism of Aqueous Corrosion

J. B. Schroeder, C. M. Schwartz, D. A. Vaughan, W. E. Berry,
P. D. Miller, and F. W. Fink

The mechanism of aqueous uranium corrosion is being studied to aid in the development of more corrosion-resistant reactor fuel elements. Previous studies on the composition of the 100 C corrosion products have failed to account for all of the metal consumed. The presence of some divalent or trivalent metal in the corrosion product might account for the lack of balance in proposed reactions.

During October, the effect of the chemical state of uranium on its X-ray absorption spectrum was investigated by methods described by Cabannes*. The results on tetravalent and hexavalent uranium compounds are being compared with the published work in order to calibrate the techniques before applying the method to the study of corrosion products.

In the program for studying corrosion at temperatures above 100 C, uranium samples have been exposed 15 min in water at 200 C. One group of samples was covered with a loose-fitting aluminum jacket to contain the oxide formed. The second group was tested bare. Metallographic examination of samples from both groups revealed no hydride formation. X-ray diffraction patterns of several oxide layers on samples from both groups revealed diffuse patterns of the UO_2 fluorite-type structure. These results compare with those obtained in steam at the same temperature.

Further tests are planned to establish the role of hydrogen in the corrosion process.

*Cabannes, Jean, Compt. rend., 225, 537 (1947).

G-1 and G-2

G. FATIGUE STUDIES OF INCONEL

W. S. Hyler and R. J. Favor

This program was initiated with the objectives of obtaining basic fatigue information on Inconel and also to establish a quantitative relationship between the variables of temperature, stress, strain, time, and cyclic frequency for Inconel subjected to repeated stressing.

Test conditions to be investigated are (1) test 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 October, collection of data at the 1600 F level at all load ratios and both cyclic frequencies has been concluded.

In the next phase of the program the evaluation will be made at the 1200 F level and at the four load ratios and two cyclic frequencies utilized at 1600 F. After the program has been completed at 1200 F, the data collected will be compared with the 1600 F data and preliminarily analyzed.

H-1

H. REACTOR MATERIALS AND COMPONENTS

F. A. Rough and A. W. Hare

In this section are reports of research on reactor materials and components supported by the AEC Division of Reactor Development.

Current study of oxidation-resistant niobium includes testing of oxygen for niobium and further evaluation of binary alloys.

Routine progress is reported on the work on the valence effects of oxide solutions in uranium dioxide.

The construction of dies is in progress for high-temperature high-pressure studies. The objective is the production of new phases having desirable high-temperature properties.

Equipment design and assembly is continuing for the study of rubbing surfaces in sodium environments.

Basic studies of pressure bonding are continuing. Techniques have been developed for sealing specimens, and data are being obtained on bonding at various conditions for correlation with postulated mathematical models.

Oxidation-Resistant Niobium Alloys

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

This program is an investigation of the oxidation behavior of niobium at elevated temperatures and the effects of alloying on oxidation behavior. Current work includes oxidation testing of oxygen-free niobium at 600 to 1200 C and testing of niobium binary alloys at 1000 and 1200 C.

Seven pure niobium samples, which had been degassed at 2200 C, were reheated in vacuum to 2300 C to insure removal of oxygen. Three of these samples have been subsequently saturated with oxygen prior to oxidation testing, while four were tested in the oxygen-free condition. Data on these four tests are given in Table H-1, along with data for nine alloy samples. Tests at 600 and 1000 C gave linear oxidation rates after short initial parabolic deviations. At 1200 C, the rate decreased with time, a result of decreasing sample area caused by rapid attack at the edges. However, at 800 C, a black, adherent oxide was formed and the parabolic rate law was followed. The reason for this behavior is not known.

Tests on niobium-15 and -17.5 a/o vanadium at 1000 C indicated that the rates transform from parabolic to linear after 3.5 and 0.5 hr, respectively. These tests fix the most oxidation-resistant composition in this system at 12.5 a/o vanadium. This alloy was previously shown to oxidize parabolically at a low rate. Niobium-1 and -10 a/o tungsten oxidized slightly faster than pure niobium. The reactions are believed

TABLE H-1. OXIDATION DATA FOR NIOBIUM AND NIOBIUM ALLOYS IN DRY AIR

Sample	Composition, a/o	Test Temperature, C	Parabolic Rate Constant, $\text{mg}^2/(\text{cm}^2)(\text{hr})$	Transition to Linear Rate, hr	Linear Rate Constant, $\text{mg}/(\text{cm}^2)(\text{hr})$	Weight Gain, mg per cm^2 , in Indicated Time					
						1 Hr	2 Hr	3 Hr	4 Hr	5 Hr	6 Hr
0-79	100 Nb(a)	600	3.8	0.2	2.79	3.7	6.7	9.3	11.8	14.3	16.6
0-80	100 Nb(a)	800	590	--	--	24.3	33.9	40.3	--	--	--
0-71	100 Nb(a)	1000	65.6	0.1	23.3	23.5	46.0	68.4	93.0	117	140
0-77	100 Nb(a)	1200	--	0	93(b)	107.7	--	--	--	--	--
0-72	Nb-15V	1000	89.2	3.5	2.6	9.0	13.2	16.3	19.0	21.7	24.2
0-73	Nb-17.5V	1000	71.3	0.5	11.2(c)	8.9	13.6	18.7	24.6	33.0	44.0
0-70	Nb-1W	1000	--	0	45(b)	46	76.5	101	120	136	152
0-74	Nb-10W	1000	--	0	32.1(b)	32.4	57.5	76.5	92.0	106	115.5
0-75	Nb-24Cr(d)	1000	--	0	149.7(e)	66	179	--	--	--	--
0-76	Nb-10Ti	1200	--	0	42.3(b)	54.0	90.5	--	--	--	--
0-81	Nb-20Ti	1200	602	0.3	23.4	29.8	53.0	74.6	--	--	--
0-82	Nb-25Ti	1200	193	0.45	10.6	15.8	26.8	37.4	47.4	57.0	67.0
0-78	Nb-24Cr(d)	1200	--	0	70.8	73.0	139	--	--	--	--

(a) Degassed by vacuum annealing 1 hr at 2300 C.

(b) Reaction rates of these samples decreased with time due to decreasing surface area caused by excessive edge attack.

(c) An increasing reaction rate with time was noted after 0.5 hr, becoming linear after a 5-hr oxidation.

(d) Composition corrected for heavy melting losses.

(e) An accelerating reaction was observed for 1 hr, after which the rate became linear.

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linear, although the rates decreased with time because of decreasing surface areas. Niobium-24 a/o chromium oxidized rapidly at 1000 C. The appearance of the oxidized sample suggested that the scale was partially liquid during oxidation.

At 1200 C, niobium-10, -20, and -25 a/o titanium oxidized about five times faster than the same alloys at 1000 C, but the rate for niobium-25 a/o titanium still represented a ninefold improvement over pure niobium. The times for transition from parabolic to linear rates were much shorter than at 1000 C. Niobium-24 a/o chromium oxidized linearly at a slightly lower rate than at 1000 C, but the true rate may be masked by evaporation of the low-melting complex oxide.

Future work plans include:

- (1) Continuation of oxidation tests on binary alloys at 1200 C.
- (2) Oxidation testing of oxygen-saturated niobium for comparison with degassed niobium.
- (3) Electrical-resistivity measurements on selected alloy scales to assist in establishing the mechanism of oxidation.
- (4) Chemical analyses on selected alloys and scales.

Valence Effects of Oxide Solutions in Uranium Dioxide

W. B. Wilson and C. M. Schwartz

The purpose of this program is to study the mechanism of stabilization of the fluorite structure of UO_2 , under oxidizing conditions, by additions of lower valence oxides to form solid solutions. Stabilization appears to be effected by a process of compensation for the valence change of the uranium ion during oxidation.

Work was continued during the month on evaluating methods of producing samples which will be most suitable for electrical measurements. Both vacuum and hydrogen sintering of $\text{La}_2\text{O}_3 \cdot \text{UO}_2$ solid solutions have been performed. These materials are currently being examined to determine their electrical characteristics.

Thermal-microbalance studies have been initiated to evaluate the oxidation stability of materials prepared to date, for correlation with the electrical and structural studies. Chemical analyses and density measurements are also being made to determine the nature of possible solid-state defects present.

High-Temperature High-Pressure Solid-State Studies

W. B. Wilson and C. M. Schwartz

The purpose of this research is to investigate the ability of pressure to produce new phases having desirable high-temperature properties. From the phase rule, pressure represents a degree of freedom which is essentially ignored in most phase diagrams. By varying pressure, new phases may be formed, some of which may be metastably retained (i. e. diamond and borazon).

The chief advantage of high pressure is that it tends to produce phases having high cohesive energy density and high bond symmetry, which are very desirable physical attributes for high-temperature materials. The ability to produce new phases is a result of the effect of pressure on the electronic structure of the solid. The effect is of such magnitude that some insulators may become metallic if adequate pressure is supplied to decrease the energy gap between valence and conduction band. In the case of metals, the band overlap is probably responsible for the inability to permanently retain transition phases produced by pressure. The role of temperature, aside from its usual effect as a degree of freedom, is to promote reactions kinetically.

Very little is known of the effect of pressure to alter, reversibly or irreversibly, the extent of solid solutions. Likewise, the physical conditions of ion size, valence, temperature, etc., affecting the production and retention of new phases are not understood.

In the present program, work will be undertaken to determine some of the effects of pressure relative to crystal physics, and simultaneously be directed toward producing materials for reactor use. As a preliminary step, however, it is necessary first to develop suitable means of supplying heat and pressure to the specimen to be studied. Final construction of the first die design is being completed, and it is expected that experimental work using this unit will be undertaken in November. The second die design, of a "geometrical-advantage" type, will be started after the first is finished. The second unit will be aimed at achieving the ultimate pressure and temperature compatible with the properties of existing materials. It is anticipated that pressures of the order of 1 to 3×10^6 psi and temperatures in excess of 2000 C will be attained.

Rubbing Surfaces in Sodium Environments

J. W. Kissel and R. W. Endebrock

A study of factors which contribute to the friction behavior of surfaces sliding in sodium maintained at temperatures up to 1500 F is under way. The objective is to provide information which can be employed in the design of sodium-flow-system components featuring sliding parts.

During October, the design of an apparatus for experiments to evaluate the low-sliding-speed friction characteristics of sodium-immersed spherical specimen-flat specimen combinations was nearly completed. The more important components

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incorporated into the apparatus to permit such evaluations are described briefly below; these components will be integrated into a small chamber which can be evacuated and filled with clean inert gas prior to friction experiments.

- (1) Flat-specimen assembly. The function of this assembly will be to provide a sodium-immersed flat specimen ($7/8$ by $1-1/4$ by $1/4$ in.) which can move in a lateral direction at a constant rate of speed while contacting a spherical specimen mounted above it. To accomplish this, the flat specimen will be inserted in a holder attached to a fairly stiff arm. This arm will be supported by an axial ball bushing mounted on the housing and will be coupled to an external variable-speed gear-head motor drive. The seal at the housing will be of the bellows variety.

The quantities of sodium used will be small, the only criterion being that the surface of the flat specimen be adequately covered. A sodium dispenser will be incorporated into the system to obtain the small controlled quantities required.

- (2) Spherical-specimen assembly. This assembly will provide for the mounting of a small ($1/2$ -in. maximum diameter) spherical specimen at one end of a $3/8$ -in. -OD tube about 24 in. long; as indicated above, this specimen will ride on a flat specimen suspended beneath it. The other end of this tube will serve as the core of a linear differential transformer, a feature which will provide a means for measuring the lateral movement of the spherical specimen caused by frictional drag. This lateral movement will be opposed by a calibrated vertical restraining spring attached to the $3/8$ -in. tube at a point about 4 in. from the transformer section. The actual drag forces involved will be determined from the deflections of this spring.
- (3) Specimen-heating assembly. The specimens will be heated during an experiment by conventional heating elements mounted within the housing. A temperature-control thermocouple will be embedded in the holder provided for the flat specimen.
- (4) Housing. The housing will be a stainless steel cylindrical shell which, as indicated above, will serve essentially as a controlled-atmosphere dry box. Sight glasses will be mounted at appropriate locations to allow visual observations of specimen positions and movements.

The combinations of materials to be evaluated in this program will be chosen on the basis of corrosion resistance and behavior in sodium established in prior compatibility screening studies. While specific combinations have not yet been chosen, it is likely that some of the studies will be concerned with molybdenum, metal-bonded TiC and WC, and a few of the attack-resistant high-temperature alloys.

Basic Studies of Pressure Bonding

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

The phenomenon of pressure bonding is being investigated in an effort to establish criteria for optimum bonding conditions. Particular interest is devoted to the case where two bodies at elevated temperatures are pressed together by pressure which is applied hydrostatically to the external surfaces.

Analytical Studies of Pressure Bonding

The study of the pressures needed to establish intimate contact between two microscopically rough surfaces hydrostatically pressed together has continued. The following analysis based on Hertz's theory of contact was used: Let two almost flat surfaces be brought into contact. It is assumed that contact is made at a point, or along a straight line, in some influenced region. Depending on whether the contact is a point or a line, this region is assumed to be a circle of some radius a or an infinitely long strip of width $2a$; more precisely it is assumed to be an almost flat surface element of a sphere or cylinder. The distance between the two bodies at a is small compared to a . Due to the external hydrostatic pressure, p , acting in a direction normal to the contacting surfaces, a force or force per unit length will be exerted across the contacting surface from each body to the other. Based on Hertz's theory and treating the system as elastic, the size of the area in actual contact and the distribution of stresses on the surface can be computed. The area of contact occupies only a small portion of the influenced region. Compressive stresses equal to the hydrostatic stress are superposed on the stresses parallel to the plane of the surface. The value of p is then determined for which the elastic-stress configuration at a reaches the yield condition. This value of p is found to be equal to the yield stress in simple tension in the case of the cylinder, and somewhat under that, according to the value of Poisson's ratio, in the spherical case.

It should be emphasized that the above analysis gives only a rough estimate of the pressure for which the surface becomes plastic. The chief source of error lies in considering an elastic-plastic body as fully elastic. It is felt, however, that such a simplifying assumption is justifiable in order to estimate the pressure required for complete yielding of the surface, and presumably then, for complete contact.

Experimental Studies of Pressure Bonding

In the experimental phase of the program, specimens are designed, fabricated, and pressure bonded under various conditions in order to obtain experimental results which can be correlated with values calculated on the basis of postulated mathematical models. Thick-walled aluminum tube specimens are being pressurized to determine the applicability of the von Mises shear-strain-energy criterion for yield to the pressure-bonding process.

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Techniques have been developed for sealing the aluminum specimens. A specimen pressurized at 4000 psi and heat treated for 3 hr at 500 F deformed in accordance with the value predicted from the mathematical model. In order to determine the effect of time on the amount of deformation achieved during pressure bonding, specimens have been treated at 500 F and 4000 psi for 1 hr. These specimens will be evaluated to determine if the extent of deformation can be correlated to the 3-hr treatment.

Additional specimens have been prepared and will be pressurized under various conditions to be prescribed by the analytical studies. To insure reliability of results, duplicate specimens are being bonded for each set of pressure-bonding parameters.

I-1

I. PHYSICAL RESEARCH

F. A. Rough and B. W. Dunnington

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

In the study of high-density UB_2 specimens, three phases were found by metallographic examination and the microhardness of each phase was measured. Grain-boundary porosity was observed in a sintered specimen of UN.

In the uranium-zirconium-silicon system, evidence has been shown for complete solubility between U_3Si_2 and Zr_3Si_2 .

X-ray examination of mixtures of uranium-nitrogen and uranium-carbon compounds indicate additivity of solid solutions between UN and UC with some evidence of expansion of the U(N, C) solid-solution-phase field.

Structure factors calculated for UC_2 from new X-ray data are in disagreement with previous structure factors, indicating that the assumed tetragonal structure may be somewhat in error.

An upper limit was found for the diffusion rate of hydrogen in beta zirconium at 600 to 700 C. A preliminary value for the diffusion rate of hydrogen in delta zirconium hydride is in reasonable agreement with extrapolations from previous low-temperature data.

Preparation, Fabrication, and Physical
Properties of Uranium Compounds

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

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

A sample of UBe_{13} , which was largely single phase and had a bulk density slightly over 100 per cent of the theoretical value, is being subjected to gas kinetic studies. Reactions of UBe_{13} with oxygen, nitrogen, and water vapor are being investigated. The results will be reported next month when the experiments have been concluded.

A successful method for preparing dense specimens of UB_2 was described in BMI-1226. Essentially, it consisted of making UB_2 by a solid-solid reaction in vacuum between UH_3 and boron powders, grinding the material, compacting it hydrostatically, sintering it in vacuum, and finally arc melting the sintered compact. Two phases were observed in the specimen made by using stoichiometric quantities of the two elements. The second phase occurred along the grain boundaries and was believed to be a uranium-rich phase. Re-examination of the metallographic specimen after 2 weeks of storage revealed that reaction with air had resulted in an "etch" that had brought to light a third

phase. The latter appeared as tan areas in the principal phase, which was thought to be UB_2 . Knoop hardness measurements (200-g load), made on the three phases were as follows:

Principal phase	
White area	1508
Tan area	870
Phase at grain boundary	679

Because it was believed that the phase along the grain boundaries was rich in uranium, a new specimen was prepared using 2 w/o excess boron. Metallographic examination of the final specimen showed it to have much less of the grain-boundary phase. The bulk density of this specimen was 96.5 per cent of theoretical as compared with 99.5 per cent of theoretical for the specimen made with stoichiometric amounts. The average Knoop hardness of the second preparation was 1077 (200-g load). No hardness measurements were made of the grain boundary-material because there was so little of it. This specimen will be "etched" in air and Knoop hardness measurements will be made on any separate phases that become evident. The specimens will be examined by X-ray diffraction. A portion of the more pure specimen will be used for gas kinetic studies.

A compact of uranium nitride powder sintered in vacuum at 1850 C for 1 hr had a bulk density 82.5 per cent of theoretical. An additional 3 hr at 1850 C increased the bulk density to 84 per cent of theoretical. Metallographic examination showed that there was good contact between the grains but that some porosity existed at the grain boundaries. The average Knoop hardness of the specimen was 544 (100-g load). The UN powder used in the above work had an average particle size of $66\ \mu$. A portion of the powder has been milled to an average particle size of $7\ \mu$. This material will be sintered and the sintered compact will be examined.

Arc-melted UC which had been previously ball milled to an average particle size of $15\ \mu$ was further reduced to $9\ \mu$. Sintering experiments will begin shortly.

Sintered specimens of UC_2 that are large enough for thermal-conductivity measurements are being made.

Constitution of Uranium-Base Ternary Alloys

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

A study of the phase relationships which occur between the delta phases of the uranium-zirconium and uranium-molybdenum systems is nearly complete. An attempt is being made to obtain equilibrium structures in samples heat treated at low temperatures and thus to verify the present tentative diagram. Also, an attempt is being made to pinpoint the ternary eutectoid composition.

The uranium-zirconium-silicon system is being studied by means of metallography, X-ray diffraction, and thermal analysis of uranium-rich samples. Tentative

I-3

isotherm data have been collected and will be reported after more study has verified them. The existence of Zr_4Si has not been definitely established, however, a compound high in zirconium is thought to exist and is arbitrarily called Zr_4Si . Zr_3Si_2 and U_3Si_2 are both tetragonal and have similar lattice parameters, thus complete solid solution is expected. Experimental evidence to support this are the lattice-parameter measurements of a $(UZr)_3Si_2$ pattern. Below is a tabulation showing the comparative lattice constants of U_3Si_2 , Zr_3Si_2 , and $(UZr)_3Si_2$.

Material	Lattice Constant, A	
	a_0	c_0
U_3Si_2	7.3299	3.9004
$(UZr)_3Si_2$	~7.15	~3.73
Zr_3Si_2	7.081	3.701

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

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

This ternary system is being investigated to determine the thermal stability of uranium carbides, solid solutions of uranium carbides with uranium nitrides, and ternary phases, and their stability with respect to a nitrogen atmosphere. Solid-solid and solid-nitrogen reactions are being studied.

The binary phases UN, U_2N_{3+x} , UC, and UC_2 have been prepared. Initial solid-solid reactions were carried out using equimolar ratios of nitrides and carbides. Compacts of minus 200-mesh powder were mixed and compressed under a protective atmosphere of argon. The first reactions were carried out in BeO crucibles under vacuum at 1700 to 1800 C. The uranium nitride reacted with the beryllium oxide to form UO_2 and Be_3N_2 , as well as with the uranium carbide. Subsequent reactions have been carried out at 1800 C in graphite crucibles under argon, with the compacts supported on mixed nitride and carbide powder. Data obtained on the latter reactions are given in Table I-1.

TABLE I-1. SOLID-SOLID REACTIONS OF URANIUM NITRIDES AND CARBIDES AT 1800 C

Starting Mixture	Product Composition, a/o			Product Phases By (a) Indicated Determination Method	
	N	C	U	X-Ray Diffraction	Metallography
UN + UC				Fcc $a_0 = 4.9254 \pm 0.0006$ A	UN-like
U_2N_3 + UC	32.3	16.75	51.0	Major Fcc $a_0 = 4.9068 \pm 0.0006$ A	UN-like
				Minor Fcc $a_0 = 4.941 \pm 0.001$ A	UC-like
				Minor of UO_2	UO_2
U_2N_3 + UC_2	13.3	37.8	49.0	Fcc $a_0 = 4.9480 \pm 0.0003$ A	UN-like

(a) For comparison, the constant of UC was 4.9598 ± 0.0005 A and that of UN was 4.8897 ± 0.0003 A at room temperature (25 C).

The reaction of the equimolar mixture of UN plus UC yielded a solid solution with a lattice constant linear with UN and UC, within the errors of measurement. This indicates additivity of the lattice constant according to Vegard's law for compositions on the pseudobinary diagram line between UN and UC. The reaction of U_2N_3 with UC_2 produced essentially a single face-centered-cubic phase which is slightly rich in nitrogen plus carbon. Its lattice constant showed a marked deviation from that expected, on the basis of its carbon-to-nitrogen ratio and the linear relation observed for UC-UN compositions. This is further indication that this sample is not an additive solid solution between UN and UC, and, therefore, suggests an expansion of the $U(N, C)$ solid-solution phase field.

Further work is in progress to verify the additivity of solid solutions on the join between UN and UC, and to study the reaction of UN plus carbon and UC_2 . Subsequently it is planned to examine reactions of the carbides and solid solutions with nitrogen gas.

Structure of UC_2

A. E. Austin and C. M. Schwartz

The structure of UC_2 has been reported as tetragonal, of the calcium carbide type. Litz* discussed two possible configurations of carbon atoms in the body-centered tetragonal structure. Recent data have disclosed that UC_2 transforms around 1800 C to a face-centered-cubic structure. Furthermore, for the assumed tetragonal structure, there is a marked anisotropic thermal expansion in which the c_0 spacing goes through a maximum value. The present study was undertaken to reinvestigate the structure of UC_2 and determine the positions of carbon atoms in it, to contribute to an understanding of uranium-carbon bonding and to interpret the thermal behavior.

The amplitude of the structure factors for the assumed tetragonal structure were calculated from X-ray diffraction intensities from UC_2 powder. Intensity data from pure tungsten powder mixed with the UC_2 were used to place the intensities on an absolute basis. The observed intensities were corrected for the Lorentz-polarization and multiplicity factors. Absorption correction was eliminated by use of the tungsten mixture. The structure factors computed are in marked disagreement with those calculated for the tetragonal structure proposed by Litz. The data appear more in accord with those of monoclinic ThC_2 , after allowance for changes in multiplicity. The possibility of a ThC_2 type of structure will be studied. Additional X-ray diffraction data will be obtained. It is planned also to obtain neutron-diffraction data in the near future.

Migration of Hydrogen in Zirconium Hydride

J. B. Vetrano and W. M. Albrecht

As reported in BMI-1213, this program is concerned with a study of the phenomenon of hydrogen migration in zirconium hydride. An attempt will be made to apply thermodynamics of the steady state using data on patterns of migration as a function of

*Litz, L. M., Garrett, A. R., Croxton, F. C., "Structure of Carbides of Uranium", J. Am. Chem. Soc., 70, 1718 (1948).

I-5 and I-6

time, temperature gradient, and original composition, and data on the diffusion coefficients of hydrogen in beta zirconium and in the delta hydride.

During October, the experimental work was concentrated on the determination of the rates of diffusion of hydrogen in zirconium hydride. Initial experiments with beta zirconium were made using the concentration-gradient technique. Attempts were made to produce hydrogen-concentration gradients in beta-phase zirconium cylinders by dissolving thin hydride films for 20 and 30 min at 600 and 700 C, respectively. Analyses of layers machined radially showed that the hydrogen was distributed uniformly throughout the specimen indicating that heating times were too long. However, it was estimated from these results that the diffusion rate for hydrogen in beta zirconium is of the order of 10^{-4} cm² per sec at 600 to 700 C. Thus, only about 1 min would be required to produce a suitable concentration gradient. Such time is not experimentally feasible since the time required to cool the specimens from the experimental temperatures is greater than this. Therefore, another experimental technique, based on the rate of uptake of hydrogen by the zirconium cylinder is being tried.

Other diffusion experiments using the moving-boundary technique were made with delta zirconium hydride at 200 and 400 C. In this technique the rate of movement of the hydride-metal (delta-alpha) boundary into the hydrogen-saturated alpha core is measured. At 200 C the rate of movement of the boundary is slow and long times (greater than 6 hr) are required to obtain a measurable movement of the boundary. The initial experiment at 200 C indicated that the diffusion coefficient of hydrogen in delta zirconium hydride is of the order of 10^{-9} cm² per sec. Additional experimental work is necessary to establish the diffusion coefficient. At 400 C a coefficient of 1.1×10^{-7} cm² per sec was obtained. This value is in good agreement with 2.1×10^{-7} cm² per sec obtained by an extrapolation of the low-temperature data presented by Gulbransen and Andrew.* Further experiments are being made at 300 and 500 C.

*Gulbransen, E. A., and Andrew, K. F., J. Electrochem. Soc., 101, 560-566 (November, 1954).

J-1

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

C. L. Peterson, P. D. Miller, E. F. Stephan, E. L. White,
T. E. Snoddy, W. D. Beasley, and F. W. Fink

The evaluation of materials of construction for various vessels in the proposed fuel-element recovery processes has been continued.

Platinum, gold, tungsten, molybdenum, and nickel all seem worth further investigation for use for the liquid-phase Zircex hydrochlorinator.

The results of metallographic sectioning of specimens of Haynes 25 and of Type S-816 alloys after exposure in the Zircex cyclic test indicate that the use of Type S-816 as a container material for the vapor-phase hydrochlorinator dissolver can no longer be recommended and the value of Haynes 25 in this application would be highly questionable.

No deleterious effects upon titanium weldments or crevices exposed in simulated Darex dissolver and stripper solutions were revealed by metallographic examination.

Progress is being made in running the fluoride-volatility corrosion-testing apparatus for extended periods, and tests with Hastelloy W and INOR-8 specimens are, or soon will be, under way.

The Zircex Process

Uranium may be separated from fuel elements which contain zirconium as a diluent or cladding by means of the Zircex process. Hydrochlorination of the fuel element at elevated temperatures with subsequent evolution of the volatile ZrCl_4 is followed by dissolution of the uranium ash in nitric acid. Uranium is recovered by normal methods of solvent extraction, once the nitric acid solution has been stripped of chlorides and properly adjusted. Two methods (liquid and vapor phase) are presently under consideration for the hydrochlorination step.

Several more materials have been exposed in molten $\text{AlCl}_3 \cdot \text{NH}_4\text{Cl}$ in the presence of HCl gas in screening experiments to evaluate possible container materials for a liquid-phase Zircex hydrochlorinator. Along with platinum and gold, as reported in BMI-1226, tungsten was found to have good resistance. Nickel and Hastelloy X have corrosion rates of less than 10 mils per month. Hastelloy C stabilized, Inconel X, Illium R, Monel, Type S-590, Inconel, Hastelloy F, and Carpenter 20 stabilized, in order of decreasing resistance, are in the 10 to 20 mils-per-month range, while Haynes 23 and Haynes 21 have rates of 67 and 83 mils per month, respectively.

Some 7-day tests have been made with nickel and Hastelloy C, and such a test with Hastelloy X is now in progress. A lower corrosion rate of 1.2 mils per month was measured for the nickel specimen whose rate had been 6.4 mils per month during the

J-2

24-hr test. The rate for a particular Hastelloy C specimen dropped from 12.2 to 8.4 mils per month upon the longer exposure.

The Zircex cyclic tests are designed to evaluate materials of construction for a one-vessel vapor-phase hydrochlorinator dissolver. The corrosion rates for specimens of Haynes 25 and Type S-816, cycled from a hot HCl-H₂ atmosphere to 15 M HNO₃ solutions, have risen to values which indicate that the use of these materials in such a system would be impractical.

Both Haynes 25 and Type S-816 specimens have shown low corrosion rates when cycled into either 3 M or 5 M HNO₃ solutions, as measured by weight losses of the specimens. However, metallographic sectioning of the specimens has revealed that intergranular attack and severe scaling are occurring with both materials, and the apparent corrosion rate calculated by weight-loss data is considerably lower than the actual rate obtained by measuring the amount of good metal remaining.

Accurate determinations of corrosion rates by sectioning have not been completed, but some general observations have been made. The intergranular attack seems to have penetrated to about twice the depth in specimens of Type S-816 that it has in specimens of Haynes 25 exposed in comparable environments. Also the attack seems to be manifested to the greatest extent on specimens which have been submerged in the boiling liquid. There may also be a direct relationship between the amount of attack and the concentration of chlorides in the boiling solutions, although this has not been definitely established.

At this point, the use of Type S-816 as a container material in the Zircex process cannot be recommended and the use of Haynes 25 would appear to be highly questionable.

The Darex Process

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

The only investigation connected with the Darex process, which was under way last month, was a metallographic examination of the fusion and spot weldments and crevices in titanium specimens exposed in the continuous and batch dissolvers and in the chloride-stripper experiments. No stress cracking or other deleterious effects could be detected in these specimens.

Fluoride-Volatility Process

Fuel elements containing zirconium as a diluent or cladding can also be reclaimed by the fluoride-volatility process. The first step of this process consists of hydrofluorinating the elements in a molten bath of ZrF₄-NaF with a stream of HF. A material of construction is being sought for this hydrofluorinator.

J-3 and J-4

Much of the month was spent in modifying the exposure equipment to enable runs of longer than a few hours to be made without plugging of the lines. At present, the situation seems to be in hand and a 250-hr-exposure test with Hastelloy W at 650 C is under way. Following completion of this run, a similar run with INOR-8 is contemplated.

The longest exposure of Inconel was 96 hr. Other runs with Inconel, made before the equipment was modified to its present form, were considerably shorter but did indicate that the passage of HF gas is necessary for attack, as little attack occurred on Inconel when submerged in the melt with only nitrogen passing through. Examination of the surfaces of Inconel specimens which have been attacked shows that a selective leaching has occurred but, as yet, little is known concerning the mechanism of this attack or which component is leached.

K-1 and K-2

K. EVALUATION OF A REFLECTOR-CONTROLLED
HETEROGENEOUS BOILING REACTOR

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

A new phase of this work has been started. The object of this portion of the research program is to attempt to provide basic information on neutron streaming from a critical-assembly experiment made up of strips of material. The voids between the strips provide streaming paths for neutrons which may affect significantly the results obtained from the reflector-control experimentation.

During October, the first core design was finalized and work on setting it up was begun. The method of construction is identical to the original core; however, the new core geometry is a 24-in. cube rather than a right cylinder, and is unreflected. The fuel cans have been loaded and are being stored in the vault until modification of the core structure to accept the new geometry is completed.

The initial core loading will be approximately 7.5 kg based on preliminary estimates obtained from simple two-group calculations. Initial criticality with the modified geometry will be made early in the next month.

L-1

L. STUDIES OF SODIUM-TANTALUM COMPATIBILITY
AT ELEVATED TEMPERATURES

J. H. Stang

The reports in this section deal with materials investigations which furnish information to the LAMPRE I development program at Los Alamos Scientific Laboratory. The investigations are concerned specifically with establishing creep data for various grades of tantalum in environments of 1200 F peak-temperature sodium and 1200 F inert gas and with developing techniques for making welds in tantalum which do not exhibit porosity or extensive grain growth. Recent exploratory work to evaluate resistance welding as a method of eliminating the latter troublesome effects has indicated that resistance welds of good metallurgical structure can be obtained in tantalum.

Tantalum-Sodium Compatibility Studies

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

During October, construction and assembly of various components of a series of newly designed Type 316 stainless steel flow loops started. These loop systems will be used to evaluate the creep behavior of stressed tantalum inserts during exposure to an environment of polythermal (peak temperature of 1200 F) flowing sodium having an oxygen content of <10 ppm. The effects that will be carefully scrutinized are (1) any reduction of strength owing to the slight amounts of corrosion which prior loop experiments with the tantalum-sodium combination have pinpointed and (2) any accelerated corrosion processes that can be attributed to stress alone.

According to the present schedule, the first group of experiments in this research will begin around the first of next year.

High-Temperature Mechanical Properties
of Tantalum

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

The creep properties of tantalum at 1200 F in a helium atmosphere are being investigated. During October, tests were continued on three types of material: low-oxygen-content tantalum, annealed arc-cast tantalum, and annealed sintered tantalum. Table L-1 presents data now on hand from these recent experiments; prior observations that the strength of sodium-exposed (reduced oxygen) sintered material compares favorably with that of the original annealed sheet are substantiated.

L-2

TABLE L-1. RECENT CREEP DATA FOR TANTALUM SHEET^(a) TESTED AT 1200 F IN HELIUM

Stress, psi	Time in Progress, hr	Time, hr. to Reach Indicated Percentage Deformation						Total Deformation, per cent	Minimum Creep Rate, per cent per hr
		0.1	0.2	0.5	1.0	2	5		
<u>Annealed Sheet From Sintered Tantalum</u>									
12,500	500 ^(b)	--	1	4	7.5	--	--	--	--
<u>Annealed Sheet From Arc-Cast Tantalum</u>									
15,000	170 ^(b)	15	35	--	--	--	--	--	--
<u>Annealed Sheet From Sintered Tantalum Exposed to Sodium at 1200 F for 33 Days</u>									
15,000	1655	0.4	1.0	8.0	16	130	1450	2.25	0.0015
12,500	500 ^(b)	20	40	--	--	--	--	--	--
10,000	1563	400	850	4500 (est.)	--	--	--	0.25	0.00008

(a) Analyses of the test materials are as follows:

Material	Contaminant Level, ppm				Hardness (5-Kg Load), VHN
	Oxygen	Nitrogen	Hydrogen	Carbon	
Sintered sheet	270	150	0.2	20	170
Arc-cast sheet	214	150	0.3	10	185
Sodium-exposed sheet	15-43	130; 160; 270	5	20	130

(b) Test still in progress.

Weldability of Tantalum for High-Temperature Systems

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

Tantalum welding studies now under way are concerned with (1) uncovering basic reasons for porosity in welds in some lots of tantalum and (2) evaluating techniques to minimize or prevent grain growth such as is present in welds made by the standard argon-shielded tungsten-arc process. Techniques now scheduled for evaluation include the use of ultrasonics to reduce grain size in welds made by normal welding processes and resistance welding. The latter process, if successful, should also eliminate weld porosity.

L-3 and L-4

During October, equipment was assembled which will permit coupling of ultrasonic energy to areas of arc welds. Emphasis, however, was placed on making various exploratory studies pertinent to resistance-welding problems. These included the following.

- (1) Initial spot-welding tests on 0.020-in. -thick arc-cast tantalum sheet using a 50-kva precision spot-welding machine. The purpose was to obtain information about bonding characteristics and to determine any effect of forging pressures on grain size in weld nuggets. These pressures were applied at various times during and after the weld time in the welding cycle. Metallographic examinations of specimens indicated that sound metallurgical bonds were obtained and that grain size was much smaller than in arc welds. Forging-pressure effects on grain size were not discernible.
- (2) Examining methods for cleaning tantalum prior to resistance welding. While a successful pickling solution for cleaning has not, as yet, been devised, initial welding tests have indicated that a cleaning method which involves abrading with crocus cloth, followed by degreasing in acetone, might be satisfactory.
- (3) Experiments to check the limit to which sintered tantalum tubing, 0.160 in. in ID and 0.189 in. in OD, can be expanded without rupture. These tests, which were carried out by compressing a rubber cylinder inside lengths of tubing, were in connection with a proposed joint design in which a tube would be inserted in a close-fitting hole in a header and would be expanded to obtain firm contact between the parts for resistance welding. It was found that expansions of about 0.0015 in. (on the outside diameter) occurred prior to rupture if an outside restraint was not present. However, when a 0.030-in. -thick restraining sleeve was present, the prerule expansion increased to about 0.003 in.

The phase of the program concerned with the problem of porosity in arc welds is being delayed pending delivery of an additional supply of tantalum. This material, which is expected in about 4 weeks, will consist of unweldable grades that can be used in studies wherein constituents which are responsible for the unweldability are identified.

M-1

M. DEVELOPMENTAL STUDIES FOR THE PWR

R. W. Dayton

Further flow tests have been made of devices for improving mixing in the PWR flow model. Improved mixing has been obtained, though at high pressure loss, and studies are continuing to obtain good mixing with lower pressure drops.

Construction of the apparatus for X-ray photometric examination of fuel elements has been started.

Studies of the preparation of Zircaloy-clad UO_2 fuel plates are continuing. The nature of the bond produced depends on the surface finish of the Zircaloy; machined or abraded surfaces give good bonds, and pickled, grit-blasted, or shot-blasted surfaces give poor bonds, presumably because of contamination. Removal of such contamination has not yet been successfully accomplished. Barrier layers of iron, nickel, and chromium are proving successful at preventing reaction between Zircaloy and UO_2 , and effort is being concentrated on these materials.

Reactor Flow Studies

L. J. Flanigan and H. R. Hazard

Studies in a quarter-scale air-flow model of the PWR reactor are being conducted to determine the effects of lower-plenum geometry on mixing and flow distribution in Core 2. Previously reported work includes preparation of the model for the present studies, core-flow distribution studies, and mixing studies involving various geometric changes in the lower plenum. Mixing was improved by two previous changes.

In October, the semicircular flow deflectors installed around the inlets were positioned 25 deg above the horizontal to direct the flow immediately above adjacent flow deflectors. Mixing was the same as was obtained with the deflectors positioned horizontally. The maximum amount of fluid from the traced inlet found in any fuel assembly was 51 per cent, which is the best mixing which has been achieved. The static pressure drop between the inlet pipe and downstream of the flow baffle was 56.5 in. of water as compared with a drop of 50.4 in. of water corrected to the same flow rate with the flow deflected tangentially. The static pressure drop between these two points with Core 1 configuration was 3.8 in. of water; for the basic configuration for Core 2 with a 1/2-in. conical extension on the flow baffle, the static pressure drop was 5.4 in. of water corrected to the same flow rates.

It appeared that this high pressure loss resulted from flow restriction through the flow deflectors, so semicircular deflectors which provided more flow area were installed 3/4 in. from the edge of each inlet to direct the flow tangentially around the bottom dome. The corrected static pressure drop was reduced to 11.3 in. of water, but mixing was also reduced. The maximum amount of fluid from the traced inlet found in any fuel assembly was 70 per cent.

M-2

In November, several additional mixing devices will be studied in an attempt to obtain better mixing with low pressure loss.

X-Ray Photometric Examination of Fuel Elements

J. B. Schroeder and C. M. Schwartz

An X-ray photometer is being built to detect weld porosity and uranium segregation in alloy fuel elements.

The shop work necessary to mount the X-ray tube, detector, and sample on the lathe bed has been started. When completed, it will be possible to scan a fuel plate in either the longitudinal or transverse direction.

The counting circuit will consist of a scintillation detector, preamplifier, linear amplifier, two fast decade scaling units, and a count-rate meter. These components, with the exception of the count-rate meter, are on hand or are expected to be delivered shortly.

On the basis of the results of the earlier feasibility study and the requirements for rapid and economical scanning of specimens, it is anticipated that counting rates of the order of 10^5 per sec, or higher, must be measured. Conventional count-rate meters do not attain such counting rates, and thus it was necessary to provide some means, such as the fast decade scaling units, to scale down the rate.

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

The use of compartmentalized Zircaloy-clad UO_2 fuel elements is being considered for the PWR Core 2. An investigation is being conducted to determine the feasibility of preparing these fuel elements by pressure bonding, a process which utilizes gas pressure at elevated temperatures to achieve bonding. Major emphasis is being placed on the development of satisfactory methods for preparing Zircaloy 2 surfaces for bonding and the investigation of various barrier layers to prevent the Zircaloy 2- UO_2 reaction during pressure bonding.

A series of specimens is being pressure bonded to determine surface-preparation methods for Zircaloy 2 which will result in good metallurgical bonds as shown by grain growth across the original interface and no visible contamination at the bond. Zircaloy plates have been prepared by abrasive-belt finishing, pickling, vapor blasting, barrel finishing, and shot peening, and have been bonded at 1550 F and 10,000 psi for 4 hr. Excellent bonding has been obtained when an abrasive belt has been used to remove 1 to 3 mils from the surface of hot-rolled Zircaloy 2.

M-3

Specimens will be prepared from additional hot-rolled and cold-rolled Zircaloy 2 plates which are being belt abraded to determine the most satisfactory abrasive, grit size, coolant, and rate and amount of metal removal. Shot-peened, vapor-blasted, and pickled specimens have exhibited poor bonding due to contamination along the bond interface. A specimen which was shot peened with hardened steel shot and one which was treated with hydrochloric acid after shot peening also showed contamination along the bond interface.

Specimens fabricated from Zircaloy which was vapor blasted with alumina or zirconia were not well bonded and demonstrated poor corrosion resistance because abrasive was embedded along the bond interface. This contamination was not removed from alumina-blasted plates which were boiled for 5 min in sodium hydroxide before bonding, but a specimen similarly treated for a longer period of time has not yet been examined. Various single and multiple pickling treatments for Zircaloy 2 have not produced satisfactory bonds, and electron-diffraction studies have revealed the presence of tin, tin oxide, or zirconium oxide on all such surfaces. No combination of pickling baths has been found to eliminate all contaminants from the surface. A specimen with vapor-deposited tin on the surface is being prepared in order to determine if tin is preventing bonding, and an additional specimen is being fabricated from pickled sponge zirconium to determine if the alloying elements in Zircaloy 2 are the source of contamination.

Barrel-finished Zircaloy plates have been prepared using alumina slurries, and specimens from this material have been pressure bonded and are to be examined. The effect of cold work and surface roughness is being investigated. A specimen consisting of Zircaloy 2 plates having metallographically polished surfaces, which should be smooth, cold-worked, and free of contamination, will be pressure bonded and examined. Wire brushing and ultrasonic cleaning of surfaces are also being investigated.

An investigation of the fuel-to-clad reaction is being conducted to determine the feasibility of using various barrier layers to eliminate the reaction between the UO_2 cores and the Zircaloy 2 cladding during pressure bonding. A standard specimen without a barrier layer between the Al_2O_3 -21 w/o UO_2 core and the Zircaloy 2 clad was pressure bonded at 1550 F and 10,000 psi for 5 hr, and metallographic examination revealed a 0.0002-in. zone of uranium-zirconium reaction products and 0.003 in. of oxygen diffusion into the Zircaloy. A portion of this specimen was made defective with a 0.040-in. hole and has been corrosion tested in 750 F steam for 3 days without any detectable growth of the sample. However, the corrosion resistance of the Zircaloy 2 was decreased, apparently due to reaction with the core. Samples with iron, nickel, niobium, molybdenum, and palladium foils between the core and clad have been bonded to determine the feasibility of using metallic barrier layers. The palladium, molybdenum, and niobium layers reacted with the Zircaloy and exhibited poor corrosion behavior.

Metallographic examination indicated very slight reaction between the iron barrier layer and the clad, and no visible reaction between the fuel and the barrier. Corrosion attack of the iron layer was apparent, but there was no growth evidenced in the sectioned and defective specimens. A large diffusion zone was evident between the nickel foil and the Zircaloy 2 clad; however, a sectioned corrosion specimen shows only slight attack in this reaction zone, and a defective specimen indicated no growth after 72 hr. Specimens with Kanigan nickel coatings have been pressure bonded and are on test, but

M-4

results are not yet available. A 0.5μ vapor-deposited chromium coating diminished the fuel-clad reaction; thicker chromium coatings are being investigated.

Nonmetallic coatings of ZrO_2 , CaO , ThO_2 , and MgO have been applied to oxide cores, and specimens containing these cores are being bonded; however, the work on oxide coatings is being terminated because of dusting and flaking of such layers. A study has indicated that it is not feasible to electrodeposit nickel over a graphite coating to insure against dusting of the graphite. An attempt is being made to vapor deposit a protective layer of chromium over the graphite. Methods of electroplating UO_2 cores with chromium, iron, and nickel have been developed, and specimens are to be bonded using cores plated with various thicknesses of these metals.