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**PROGRESS RELATING TO CIVILIAN APPLICATIONS
DURING MARCH, 1957**

by

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REPORTS RELATED TO CIVILIAN APPLICATIONS
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- BMI-1165 "Properties of Beryllium Oxide, and Carbides of Beryllium, Molybdenum, Niobium, Tantalum, and Titanium", by William P. Weber, John F. Quirk, Alexis W. Lemmon, Jr., and Robert B. Filbert, Jr.
- BMI-1166 "Hazards Summary Report for the Reflector-Control Critical-Assembly Experiments", by Francis J. Jankowski, William S. Hogan, Robert F. Redmond, Joel W. Chastain, and Sherwood L. Fawcett.
- BMI-1170 "High-Temperature Oxidation and Contamination of Niobium", by William D. Klopp, Chester T. Sims, and Robert I. Jaffee.
- BMI-1172 "Studies of Mixing in the Lower Plenum of a Quarter-Scale Flow Model of the PWR Reactor", by Herbert R. Hazard and Abraham Rotkowitz.
- BMI-1173 "Progress Relating to Civilian Applications During February, 1957", by Russell W. Dayton and Clyde R. Tipton, Jr.

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A. DEVELOPMENT OF MATERIALS FOR HANFORD REACTORS

F. R. Shober

The mechanical properties of annealed and 35 per cent cold-worked Zircaloy 3A have been determined at temperatures up to 250 C. The short-time tensile properties of the cold-worked material are superior to the annealed at temperatures included in this investigation. Creep tests on Zircalloys 2 and 3A in the 149 to 343 C range have been continued. Preparations are being made to determine the burst strength of 0.50- and 0.625-in. -diameter Zircaloy 2 tubes at 232 and 343 C.

The optimum conditions for producing a strong bond between zirconium and nickel-plated uranium appear to be less than 2 hr at 730 C and more than 6 hr at 650 C under a 10,000-psi pressure. Single crystals of aluminum are being prepared and equipment necessary for single-crystal growth of molybdenum and zirconium is being designed.

Thermal Conductivity of Uranium

H. W. Deem and C. F. Lucks

The effect of irradiation on the thermal conductivity of uranium is being studied. Exploratory work will determine the feasibility of making thermal-conductivity and electrical-resistivity measurements on uranium rods that are clad in Zircaloy 2. NaK will be the heat-transfer medium. The test specimens are being fabricated by Hanford.

Thermal-conductivity measurements will be made by the steady-heat-flow method. The apparatus has been designed and is being assembled. Some related equipment, including the thermocouple welding jig, jig holder, and constant-temperature water bath, has been assembled. All of the remaining drawings should be in the shop by April 22.

Mechanical Properties of Zirconium and Zircaloy 2

F. R. Shober, L. L. Marsh, and J. A. Van Echo

The tensile properties of Zircaloy 3A, the creep properties of Zircalloys 2 and 3A, and the burst strength of seamless zirconium and Zircaloy 2 tubes are being determined at room and elevated temperatures.

The determination of the mechanical properties at temperatures up to 482 F in air of annealed and 35 per cent cold-reduced Zircaloy 3A has been completed. The tests were performed on specimens which were taken transverse to the rolling direction, except for a few supplementary tests on specimens cut from the sheet parallel to the rolling direction. The results are shown in Table A-1. These results indicate that the mechanical properties of Zircaloy 3A are not greatly orientation dependent at the lower

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temperatures. At 482 F, some indication of preferred orientation was observed on a 35 per cent cold-worked specimen taken transverse to the rolling direction. A chevron type of fracture was observed, with the included angle equaling approximately 90 deg. Additional tests will be conducted to determine the general validity of this fracture as peculiar to cold-worked material.

TABLE A-1. MECHANICAL PROPERTIES OF ANNEALED AND 35 PER CENT COLD-REDUCED ZIRCALOY 2A

Material	Temperature, F	Elastic Modulus, 10 ⁶ psi	0.2 Per Cent Offset Yield Strength, 10 ³ psi	Ultimate Tensile Strength, 10 ³ psi	Elongation, per cent	Reduction, in Area, per cent
35 per cent cold-worked transverse	79	15.3	92.0	97.7	16.0	45.4
	212	14.5	80.2	83.5	15.5	50.5
	302	12.3	68.0	70.7	15.0	54.0
	392	12.1	61.7	64.8	16.0	60.4
	482	9.8	54.5	55.9	13.0	--
Longitudinal	392	9.85	61.9	65.0	15.5	46.5
Annealed transverse	79	14.4	56.7	68.5	32.8	48.6
	79	14.6	57.8	68.5	36.0	52.7
	212	13.6	41.7	50.7	43.0	57.7
	302	14.0	32.4	41.7	54.0	55.9
	392	13.4	24.3	33.1	56.0	60.5
	482	11.3	18.3	29.3	53.0	70.8
Longitudinal	79	14.3	54.2	69.6	50.0	48.0
	392	12.6	22.6	35.1	53.0	43.1

Creep testing of Lot 2 of Zircaloy 2 has continued to show that the creep strength of this material is less than that of Lot 1. This is shown by the total-deformation values for similar times and by the minimum creep rate. However, at the low stresses for long times (greater than 2000 hr), the creep results for the two different lots of data are similar. Data from duplicate tests of the second lot have been comparable. The results of completed tests and tests in progress are given in Table A-2.

Burst testing to determine the burst strengths of seamless zirconium and Zircaloy 2 tubes 1.5 in. in diameter has been completed. Preparations are being made to test several Zircaloy 2 tubes 0.50 and 0.625 in. in diameter at 450 and 650 F.

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

Lot	Specimen	Temperature, F	Stress, psi	Time at Test, hr	Deformation at Indicated Time, per cent							Minimum Creep Rate ^(a) , per cent per hr
					Load On	50 Hr	100 Hr	500 Hr	1000 Hr	1500 Hr	2000 Hr	
<u>Zircaloy 2</u>												
1		300	22,000	1000	0.39	0.90	0.97	1.09	1.15	--	--	0.00012
2	40-3	300	23,000	1000	0.20	2.00	2.20	2.52	2.63	--	--	0.00023
2	31-1	300	23,000	1000	0.175	1.80	1.96	2.29	2.39	--	--	0.00020
1		450	21,000	1000	0.48	0.915	0.93	0.96	0.98	--	--	0.00040
2	34-3	450	21,000	1500 ^(b)	0.59	2.45	2.465	2.49	2.49	2.495	--	<0.00001
1		450	23,000	1000	0.30	1.98	2.05	2.13	2.15	--	--	0.00003
2	40-1	450	23,000	1002.8	1.85	3.85	3.90	3.93	3.95	--	--	0.00005
1		450	24,500	22.3 ^(c)	0.30	--	--	--	--	--	--	--
2	40-4	450	24,500	1000.9	2.40	9.00	9.90	10.25	10.25	--	--	<0.00001
2	31-6	550	24,500	1002.9	7.50	15.75	16.00	16.10	16.10	--	--	<0.00001
2	31-7	550	22,500	1100 ^(b)	2.65	4.60	4.62	4.64	4.65	--	--	0.00001
1		650	9,000	3400	0.032	0.06	0.072	0.072	0.081	0.090	0.091	0.00001 ^(d)
2	3-2	650	9,000	2325 ^(b)	0.06	0.09	0.1	0.117	0.117	0.117	0.117	<0.00001 ^(d)
1		650	12,500	1000	0.17	0.42	0.428	0.448	0.46	--	--	0.000024
2	3-1	650	12,500	2000 ^(b)	0.15	0.23	0.245	0.282	0.335	0.342	0.350	0.000015 ^(d)
1		650	17,500	1000	0.54	0.655	0.675	0.711	0.742	--	--	0.00006
2	34-2	650	17,500	1500 ^(b)	1.385	1.54	1.55	1.594	1.63	1.65	--	0.000036
2	34-4	650	17,500	850 ^(b)	1.37	1.575	1.615	1.65	--	--	--	--
1		650	21,000	1000	1.50	3.87	3.89	4.06	4.36	--	--	0.0003
2	34-6	650	21,000	550 ^(b)	4.10	4.87	4.92	5.16	--	--	--	--
<u>Zircaloy 3A</u>												
	3-1	475	20,000	1000.9	0.50	2.505	2.507	(e)				
	3-2	475	22,500	1072.8	2.3	14.4	17.0	23.4	25.75	--	--	0.003

- (a) Based on creep deformation between 500 and 1000 hr.
 (b) Tests in progress.
 (c) Ruptured.
 (d) Based on creep deformation between 1000 and 2000 hr.
 (e) Extensometer became loose, test discontinued.

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Zirconium Cladding

F. R. Shoher and R. F. Dickerson

The effect of time and temperature under constant pressure on the quality of the bond formed between zirconium and nickel-plated uranium is being investigated. The bonds obtained under constant pressure at two temperatures and three time periods have been further evaluated by water quenching from 1350 F.

A section from each test sample was prepared for heat treatment. Samples were cut with care to prevent any unnecessary cracking of the bond area. Each sample was ground and polished and inspected in the bond area for cracks. The samples were then heated in a lithium-potassium carbonate salt bath to 1350 F for 20 min and water quenched. The following tabulation shows the results of the heat treatment.

Time, hr	Bonding Conditions		Appearance of Bond Before Heat Treatment	Appearance of Bond After Heat Treatment
	Temperature, F	Pressure, psi		
2	1345	10,000	No cracks	Several small cracks
4	1345	10,000	Small cracks in 50 per cent bond area	Separated area above 80 per cent bond area
6	1345	10,000	Two large cracks	Entire area cladding was separated
2	1200	10,000	No cracks	One small crack
4	1200	10,000	Cracked in three areas	Cracked areas opened up
6	1200	10,000	No cracks	No cracks

Additional heat treatments and water quenches will be made to further evaluate the strength of the bond under these conditions.

Knoop hardness measurements were made on the specimens before heat treatment across the bond area. The hardness values indicated that adjacent to the bond area, where extensive diffusion had occurred, there were zones of extreme hardness. The harder area probably associated with compound composition of nickel-uranium and nickel-zirconium were the sites where separation was initiated and propagated. The summary of results to date indicates the following:

- (1) Two and 4 hr at 1200 F are insufficient times to bond the zirconium completely to the uranium.
- (2) Four and 6 hr at 1350 F allow time for extensive diffusion and the formation of a brittle intermetallic compound.

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- (3) Six and 2 hr at 1200 and 1350 F, respectively, appear to be the better conditions, of those investigated, for producing a strong bond.

Four additional nickel-plated cores have been sealed in zirconium cans. The surfaces of two were specially prepared to obtain a smoother surface on the nickel plate. These will be pressurized at 10,000 psi at 1200 F for 2 and 4 hr, respectively. The other two will be pressurized at 1350 F for 1/2 and 1 hr. Several bare uranium cores have been prepared and sealed in zirconium cans. Times and temperatures at a 10,000-psi pressure will be investigated to determine the optimum time and temperature condition for a good bond.

Preparation of Aluminum, Molybdenum, and Zirconium
Single Crystals

E. Jablonowski, F. R. Shober, and R. F. Dickerson

Single crystals of high-purity aluminum, molybdenum, and zirconium are being prepared for irradiation studies. The single crystals are to be grown by the strain-anneal process. High-purity aluminum and molybdenum have been procured and a survey of literature on strain-anneal methods of single-crystal growth is in progress.

An aluminum ingot (99.99 w/o aluminum) was forged and rolled to 0.050-in. sheet at room temperature. Two types of tensile specimens having a 0.25-in. -wide and a 0.040-in. -wide gage section, respectively, were machined from the sheet material. The latter will yield a single crystal of approximately the size desired for irradiation studies. Irradiation specimens can be prepared from the gage section of the large tensile specimen after a single crystal has been grown. All specimens were annealed at 550 C for 6 hr and a uniform grain size was obtained. Single crystals will be grown in these specimens after plastically deforming them a critical amount.

Molybdenum bar stock has been obtained and is currently being analyzed for impurity content. For the growth of molybdenum and zirconium single crystals, modification of the strain-anneal process and annealing equipment will be necessary. An Andrade-type furnace will be designed, constructed, and used for the growth of single crystals of molybdenum and zirconium.

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B. DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS

R. J. Carlson

All of the programs aimed at developments for aluminum-clad fuel elements have been completed with the exception of work on extrusion cladding of flat plates, and topical reports will be prepared on all phases of the current work. Next month several new programs will be initiated and the progress on these programs will be included here.

Extrusion Cladding of Flat Plates

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

Two new mandrels and a new die were tested during the past month.

One mandrel was designed with a 1/16-in. radius at the corners of the core slot to minimize the possibility of the slot cracking. No cracks were observed after six tests. Both radiused and sharp-cornered cores were clad satisfactorily using this mandrel. However, excessive extrusion pressures were still encountered although an additional core guide was used to minimize cocking of the core.

The second mandrel was designed with a heavy center web that supports the tip up to the very edge of the core slot. This was done to reduce the amount of slot compression when under pressure. However, it was recognized that the heavy web would probably restrict the metal flow in the welding chamber. At a mandrel-to-die distance of 40 mils, the streams of metal issuing from the ports adjacent to the center web did not make a satisfactory seam when forming the cladding. However, with a mandrel-to-die spacing of 80 mils, a continuous cladding was produced with apparently good seams.

The new die has a shear-type orifice, but the normally flat welding chamber was tapered at a slight angle to determine if the extrusion pressure would be reduced. The results of the first test indicated that the pressure was somewhat higher than a flat-faced shear-type die.

It is planned to perform additional tests with these tools to determine their effect on metal flow under various conditions of extrusion.

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C. PLANT ASSISTANCE TO MCW

A. A. Bauer

Studies of the relationship between residual nitrogen content of UO_3 and the oxygen reactivity of UO_2 have continued and data have been re-evaluated on the basis of new nitrogen analyses. Residual nitrogen content was found to decrease with increasing temperature of denitration. The reactivity of UO_2 produced from vacuum-amorphous UO_3 varied inversely with nitrogen content; no definite relationship for UO_2 prepared by belt denitration was observed. The effect of washing on the nitrogen content of UO_3 is now being investigated.

Results of ignition analyses show that the oxygen pickup by active UO_2 occurs more rapidly and to a higher value than for inactive UO_2 .

The study of the effect of nitrogen, added in the form of powder of approximately $UN_{1.5}$ composition, on the electrical properties of uranium oxides has continued. Lowered electrical conductivity accompanies the introduction of nitrogen; however, the minimum in conductivity with increasing nitrogen content previously reported was not observed in a second set of samples prepared. No significant effect of nitrogen on the temperature dependence of conductivity has been observed. Additional measurements are being made.

The corrosion resistance of Type 304 ELC stainless steel in chloride- and fluoride-contaminated nitric acid solutions is being investigated. Corrosion rates in chloride-contaminated solutions of 18 and 30 w/o nitric acid for exposures up to 3000 hr, and in 5 w/o nitric acid solutions for exposures up to 1500 hr, have remained essentially constant. A single specimen exhibited a sudden acceleration in corrosion rate after 1906 hr of exposure; this specimen was exposed at the interface position of a 30 w/o nitric acid solution containing 0.500 w/o chloride.

Specimens from Weldon Springs stock were tested in both the welded and unwelded conditions in 18 w/o nitric acid containing additions of 0.05 w/o chloride and fluoride. Generally, better resistance was exhibited by this material than similar Battelle stock; also, the weldment seemed more resistant to corrosion than the base metal.

Aluminum additions to chloride- and fluoride-contaminated nitric acid solutions have been found to produce notable decreases in corrosion rate.

Difficulties have been encountered in the measurements of hydrogen permeability through molten magnesium fluoride slag. The zirconium employed to act as a sink for hydrogen has been found to react to an appreciable extent with one of the slag components, probably oxygen. Thus, the previously determined value for the permeability of hydrogen through the slag is in doubt. An analysis of the surface film formed on the zirconium is being performed.

Investigation of Uranium Oxides

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

The investigation of the methods of preparation of UO_3 and UO_2 is being continued. The relation between residual nitrogen content of UO_3 and oxygen reactivity of UO_2 has been re-evaluated, and new nitrogen assays have been obtained. Further studies have been made on (1) the effect of washing on the nitrogen content of UO_3 , (2) various methods of reducing UO_3 , and (3) oxygen pickup by active UO_2 at 100 C.

The nitrogen analyses reported in the last several reports had been obtained by the Devarda method and were considered to be accurate results. However, a careful study during the past month has shown that the method, as used here, is not satisfactory for UO_3 samples containing very low amounts of nitrogen. Despite recommended trapping procedures, some KOH is carried over with the ammonia and water-vapor stream. A modified Kjeldahl method has been employed, which gives good reproducibility on low-nitrogen analyses. The results of this method of nitrogen determination are shown in Table C-1. For the three methods of denitration (belt, vacuum, and pot) an increase in temperature reduces the residual nitrogen. The nitrogen contents of other preparations of Type III UO_3 will be studied next month.

TABLE C-1. THE EFFECT OF DENITRATION TEMPERATURE ON NITROGEN CONTENT OF UO_3 AND ACTIVITY OF UO_2 TOWARD OXIDATION

Sample	X-Ray Type	Preparation of UO_3			Nitrogen in UO_3 , w/o	Oxygen-Uranium Ratio of UO_2 Exposed to Air
		Method	Temperature, C	Time, hr		
358	Amorphous	Belt	350	2	0.24	2.341
357	Amorphous	Belt	400	2	0.064	2.137
348	Amorphous	Belt	425	2	0.032	2.254
362	Amorphous	Belt	450	2	0.02	2.185
391	III	Vacuum	250	8	0.79	2.156
340	Amorphous	Vacuum	300	2	0.57	2.175
363	Amorphous	Vacuum	300	4	0.13	2.263
365	Amorphous	Vacuum	400	4	0.01	2.335
314C	Amorphous	Vacuum	450	2	0.002	2.357
249	III	Pot	425	2	0.075	--
181	III	Pot	MCW	--	0.043	2.052
251	U_3O_8	Pot	550	2	0.004	--

The UO_3 samples of varying nitrogen contents were hydrogen reduced to UO_2 at 600 C and then exposed to air at room temperature. The relation between the nitrogen content of the UO_3 and the activity of the UO_2 is shown in Table C-1. The oxygen-uranium ratio is indicative of the activity of the UO_2 . For UO_2 made from vacuum-amorphous UO_3 , the oxygen-uranium ratio appears to vary inversely with nitrogen. For UO_2 made from belt-amorphous UO_3 , no definite relation has been seen.

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Several samples of amorphous and Type III UO_3 , including some regular MCW production oxides, were washed in water to decrease the nitrogen content, and the effect of this treatment on the reactivity of subsequent UO_2 is being studied. The results are not complete and will be reported next month.

An active oxide with an oxygen-uranium ratio of 2.248 was made from amorphous UO_3 . It was desired to compare the oxygen-sorption rate of this material with that previously reported for MCW UO_2 . The active oxide was heated in flowing oxygen at 100 C for varying lengths of time. Results of ignition analyses are shown in Table C-2. The oxygen pickup by active UO_2 was found to reach a higher value, and in a shorter time, than did the inactive UO_2 . The X-ray patterns of samples having high oxygen-uranium ratios were diffuse, and until the powders have been annealed, the structures cannot be determined.

TABLE C-2. COMPARISON OF OXYGEN PICKUP OF ACTIVE AND INACTIVE UO_2 HEATED IN OXYGEN

Active UO_2		Inactive UO_2	
Time at 100 C, hr	Oxygen/Uranium Ratio	Time at 180 C, hr	Oxygen/Uranium Ratio
0	2.248	0	2.032
1	2.284	1	2.132
2	2.318	2	2.160
4	2.335	4	2.194
8	2.341	8	2.225
16	2.360	16	2.264

The Electrical Properties of Uranium Dioxide

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

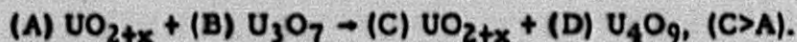
The study of the effects of nitrogen on the electrical properties of uranium oxides has been continued. Previous work has indicated that a lowered conductivity results when nitrogen is incorporated in the lattice of uranium dioxide. This finding has been verified by the preparation of additional samples of uranium oxides containing nitrogen.

"Doping" was accomplished by intimately mixing powders of $UO_{2.03}$ and $UN_{1.5}$ in predetermined ratios up to 5 mole per cent $UN_{1.5}$. The powders were hydrostatically pressed, without a binder, at 100,000 psi. The samples were then annealed at 200 C for about 120 hr to insure homogeneity.

The room-temperature conductivity of these specimens decreased, in a more or less orderly fashion, with increasing nitrogen content. However, the minimum of

conductivity versus nitrogen content apparent in the first group of specimens (see BMI-1173) was not obtained in the second group. With this exception, the magnitude and dependence of conductivity on nitrogen content of the two groups of specimens were comparable. All specimens were *p*-type.

At present the dependence of conductivity on temperature of these specimens is being measured. No significant differences between doped and undoped specimens have been noted as yet. In general, the curves consist of a straight line which changes slope between 100 and 200 C. This change of slope probably corresponds to the transition:



From room temperature to the transition temperature the slope of the curve corresponds to an activation energy of about 0.44 ev. Above the transition temperature the slope increases to about 0.50 ev. No further changes are apparent up to 500 C, the limit of the measurements. The anomalous decrease of conductivity between 400 and 500 C obtained with inactive sintered specimens is lacking in these specimens. However, measurements on only four of the eighteen specimens prepared have been completed and a thorough understanding of the effects on nitrogen on the electrical properties of uranium dioxide must await further measurements.

It is planned to complete the electrical measurements on these specimens in the near future. When the electrical measurements are completed the effect of nitrogen on the structure of the oxide will be determined by X-ray analysis. The electrical properties and structural changes will be correlated. Additional specimens containing nitrogen will be prepared for sintering studies.

The Corrosion Resistance of Selected Stainless Steels

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

Studies of the corrosion of Type 304 ELC stainless steel specimens by nitric acid solutions contaminated with chlorides and fluorides were continued during March.

A total exposure time of 3000 hr has now been reached in the prolonged-exposure tests in chloride-contaminated solutions of 18.0 and 30.0 w/o nitric acid; an exposure of 1500 hr has been reached in the case of 5.0 w/o nitric acid solutions. Table C-3 contains the data recorded during March. These data are a continuation of those given in Table C-4 of BMI-1173. All corrosion rates are remaining essentially constant or diminishing slightly with continued exposure time, with the exception of the specimen exposed at the interface position in 30.0 w/o nitric acid containing 0.500 w/o chloride. In this case, a sudden upswing in the corrosion rate has occurred after a 1906-hr exposure, as shown on page C-7.

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TABLE C-3. CORROSION OF TYPE 304 ELC STAINLESS STEEL IN CHLORIDE-CONTAMINATED NITRIC ACID

(Boiling solutions, reduced pressure)

Chloride, w/o	Specimen Position	Corrosion Rate in Exposure Time in HNO ₃ Solution Indicated, mils per month					
		1500 Hr		2000 Hr,	2500 Hr,	3000 Hr	
		5 w/o	18 w/o	30 w/o	30 w/o	18 w/o	30 w/o
0.000	Vapor	0.000			0.006	0.002	0.007
	Interface	0.002			0.012	0.003	0.013
	Liquid	0.001			0.011	0.002	0.011
0.025	Vapor	--			0.009	--	0.009
	Interface	--			0.014	--	0.015
	Liquid	--			0.011	--	0.011
0.050	Vapor	0.002			0.004	0.001	0.006
	Interface	0.002			0.013	0.004	0.013
	Liquid	0.001			0.013	0.002	0.013
0.100	Vapor	0.002			0.009	0.002	0.010
	Interface	0.071			0.014	0.002	0.014
	Liquid	0.002			0.014	0.002	0.014
0.300	Vapor	--		0.017	0.017	--	--
	Interface	--		0.024	0.023	--	--
	Liquid	--		0.016	0.016	--	--
0.500	Vapor	0.000	0.001 ^(a)		0.015	--	0.015
	Interface	1.606	0.004 ^(a)		0.820 ^(b)	--	1.381 ^(c)
	Liquid	0.002	0.003 ^(a)		0.022	--	0.023

(a) These specimens were cut from Type 304 ELC used in the Weldon Springs reconcentrator.

(b) Exposure time for this specimen was 2406 hr.

(c) Exposure time for this specimen was 2906 hr.

Note: Solutions of 5 and 18 w/o HNO₃ are boiling at 200 mm mercury, 30 w/o HNO₃ at 250 mm mercury absolute pressure.

TABLE C-4. CORROSION OF TYPE 304 ELC STAINLESS STEEL IN CONTAMINATED NITRIC ACID

(Boiling solutions, 18.0 w/o HNO₃, atmospheric pressure)

Specimen Type	Specimen Position	Chloride, w/o	Fluoride, w/o	Aluminum, w/o	Corrosion Rate in Each 48-Hr Period, mils per month							
					1st	2nd	3rd	4th	5th	6th	7th	Average
Weldon Springs Type 304 ELC (unwelded)	Vapor	.050	.050	--	1.18	1.00	0.95	0.90	0.70	0.92	0.88	0.93
	Interface				6.59	5.78	5.33	5.13	3.02	4.84	5.22	5.13
	Liquid				11.41	9.93	9.17	8.56	9.09	7.90	7.72	9.11
Weldon Springs Type 304 ELC (welded)	Vapor	.050	.050	--	1.12	1.05	0.99	0.93	0.85	0.80	0.79	0.93
	Interface				3.69	3.48	2.62	2.81	3.10	2.29	2.02	2.86
	Liquid				4.11	4.49	4.80	5.81	5.00	5.64	5.45	5.04
Battelle stock Type 304 ELC	Vapor	.050	.050	--	1.33	1.24	1.05	1.10	1.06	0.96	0.95	1.10
	Interface				11.50	12.04	8.44	9.49	8.50	10.55	8.60	9.87
	Liquid				13.63	12.50	13.96	14.18	13.16	12.35	12.99	13.25
Battelle stock Type 304 ELC	Vapor	.050	.050	.300	0.09	0.08	0.09	0.08	0.08	0.08	0.06	0.08
	Interface				0.10	0.10	0.12	0.10	0.09	0.07	0.06	0.09
	Liquid				0.10	0.09	0.10	0.09	0.09	0.08	0.09	0.09

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<u>Exposure Time,</u> <u>hr</u>	<u>Corrosion Rate,</u> <u>mils per month</u>
406	0.032
906	0.024
1406	0.022
1906	0.022
2406	0.820
2906	1.381

A similar specimen at this position exhibited severe attack during the second 48-hr exposure period of the short-period exposures preceding the prolonged tests.

Attention is called to a corrosion rate erroneously reported in Table C-4 of BMI-1173. The rate for the specimen exposed to the liquid phase of 18.0 w/o nitric acid with no chloride, after a 1000-hr exposure, should read 0.002 mil per month, rather than 0.011 as printed.

Many of the test solutions have been analyzed for chlorides following the exposure periods. So far, there has been no indication that chlorides are lost during prolonged boiling. This is true even in the case of the 30.0 w/o nitric acid solutions containing 0.500 w/o chloride which have boiled for 500 hr at 250 mm mercury absolute pressure.

Specimens cut from the material which is being used in the fabrication of the Weldon Springs reconcentrator were exposed in both the welded and unwelded condition to 18.0 w/o nitric acid solutions containing 0.050 w/o chloride and 0.050 w/o fluoride. The results with this steel are generally better than those obtained with Battelle stock Type 304 ELC. From the rates recorded in Table C-4, it would appear that the welded specimens were somewhat more resistant than the unwelded. The weldment does seem more resistant than the base metal; however, this lower corrosion rate may be due partly to the uncertainty in measuring the surface area of these specimens.

The weldments were produced by the shielded metal-arc process, using electrodes of Type 308 ELC stainless steel. In cutting up into samples, the weldments were so positioned as to leave an area of the specimen which was not heat affected. Microscopic examination of the weldments showed no severe corrosion resulting from the welding process. There was no evidence of the so-called knife-line attack in the heat-affected areas.

During this same series of tests, a study was made of the effect of aluminum additions on the corrosivity of chloride- and fluoride-contaminated nitric acid solutions. Specimens cut from Battelle stock Type 304 ELC stainless steel were exposed to 18.0 w/o nitric acid solutions containing 0.050 w/o chloride, 0.050 w/o fluoride, and 0.300 w/o aluminum additions, boiling at atmospheric pressure. Table C-4 gives the corrosion rates measured during seven 48-hr exposure periods. The effect of the aluminum additions on the corrosion rates is very pronounced. The rate for the specimen exposed in the liquid phase has been decreased over hundredfold by the addition of aluminum. The reduction in corrosion rates for the other two specimen positions is not quite so high, but, in all cases, a satisfactory resistance to attack has been imparted by the aluminum additions.

The survey of chloride retention by nitric acid concentrations of greater than 30.0 w/o was delayed by other studies, which include finding a suitable method for following the fluoride concentration during the exposure test periods. Present plans call for completing both of these studies during April, in addition to continuing with the prolonged-exposure tests already under way.

Gas-Metal Studies

M. J. Trzeciak and M. W. Mallett

As part of Mallinckrodt Chemical Works' investigation of their thermite reaction to produce uranium metal, a study of relationships between hydrogen and the materials of reaction is being made. The objective is to determine the movement of hydrogen within the reactor during reaction. In turn, this will lead to an understanding of the mechanism by which hydrogen enters the uranium metal produced. Presently, the permeation of hydrogen through molten magnesium fluoride slag is being measured.

A tentative value for the permeation of hydrogen through magnesium fluoride slag was reported in BMI-1173. A close examination of the experimental system was made to evaluate the correctness of the reported value. It was found metallographically that the zirconium being used as a sink for hydrogen was reacting to an appreciable extent with some component of the slag, possibly oxygen. As a result of this reaction, a hard film was formed on the surface of the zirconium. This film presumably could inhibit reaction of hydrogen with zirconium. This would influence the magnitude of the value obtained for permeability of hydrogen through slag. An analysis of the film is being made. In addition, a compatibility study involving zirconium with slag and cp magnesium fluoride is being conducted.

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D. PROCESSING OF FEED MATERIALS

E. L. Foster, Jr.

Two studies now in progress are of direct interest in the processing of feed materials. One, concerned with the development of salt baths for the heat treatment of uranium, has been in effect for some time. A new study, whose objective is the elimination of galling in dies used for the compacting of uranium tetrafluoride-magnesium mixtures, has just been initiated.

The pickup of hydrogen by uranium has been noted in the salt baths used for its heat treatment at the Feed Metals Processing Center (FMPC). In laboratory studies conducted previously, it was determined that uranium heat treated in two different salt systems contained less than the expected quantities of hydrogen. It is the purpose of a program now in progress to continue and confirm on a pilot-plant scale these laboratory-scale results. The two salt baths selected for further studies were 50 w/o sodium chloride-50 w/o potassium chloride and 50 w/o potassium chloride-50 w/o sodium carbonate.

These systems plus additions to the expected contamination level from baths now in use have been tested for periods up to 3 weeks. Samples of uranium heat treated in them have been analyzed for hydrogen, and the weight loss during heat treatment noted. In addition, qualitative data on the relative attack by the salt baths on the electrodes and containers used have been gathered.

Data from the incomplete studies indicate that uranium specimens heat treated in all the baths picked up on an average less than 1 ppm of hydrogen. Further, within this range of contamination, the all-chloride baths were more effective in keeping the hydrogen pickup lower than were the chloride-carbonate baths. Uranium-slug weight losses continue low, on the order of 0.2 w/o for specimens immersed for 13.5 min.

The compacting of uranium tetrafluoride (UF_4)-magnesium mixtures has been envisioned as a means of increasing the efficiency of the bomb reduction charges. In doing this, the charges are mixed and then briquetted in a closed die. In briquetting, after repeated operation, galling occurs between the ram and die wall. If no cleanup step is incorporated in the compacting operation, the ram sticks and further compacting is made impossible.

A program has been initiated to study methods of alleviating this galling. It is felt that the magnesium in the compacts is responsible for the present difficulties; the planned program not only includes the use of die lubricants found successful for magnesium but also a scrutiny of die-tolerance and -design considerations.

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Salt-Bath Development

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

Hydrogen pickup by uranium rods heat treated in pilot-plant scale salt baths is being studied. Two salt mixes which resulted in low hydrogen pickup in previous laboratory work (50 w/o sodium chloride-50 w/o potassium chloride, and 50 w/o potassium chloride-50 w/o sodium carbonate) are being used in the present program. The planned experimental program, consisting of three 3-week runs with the NaCl-KCl mix and two 3-week runs with the KCl-Na₂CO₃ mix, is nearly complete. Hydrogen analyses (conducted by NLO at Fernald) have been obtained for the uranium specimens treated in the first three runs (one with the NaCl-KCl mix and two with the KCl-Na₂CO₃ mix). These results show that average hydrogen pickup was less than 1 ppm, and, as indicated by the previous work, the hydrogen pickup in the all-chloride salt bath was lower than that in the chloride-carbonate salt baths.

Testing of Salt Bath 4, consisting of 93.5 w/o Nu-Sal (50 w/o NaCl-50 w/o KCl) plus 2.9 w/o Li₂CO₃ and 3.6 w/o K₂CO₃ has been completed. The mild steel pot used for the previous Nu-Sal test (Salt Bath 1) was used for this salt-bath test. New mild steel electrodes were used, similar to those used for tests of Salt Baths 2 and 3. These electrodes were made from 2.5 in. cold-rolled bar stock planed down to 2-in. bars to remove rolling cracks. Four groups of two specimens each were treated in this salt bath: one group when the bath had aged 1 week, one group when the bath had aged 2 weeks, and two groups when the bath had aged 3 weeks. The first two groups were treated by the modified procedure, which consists of immersing a pair of uranium specimens in the salt bath at 1350 F for 13.5 min, quenching in water for 2 min, rinsing for 10 sec in a second tank of water, and finally rinsing in a 140-ml sample of deionized water to be used for chloride analysis. The rinse sample for chloride determination was omitted for the two groups treated when the bath had aged 3 weeks. One of the groups treated in the 3-week-old bath was treated for 6.75 min instead of 13.5 min.

Work was begun with Salt Bath 5, consisting of 93.5 w/o Nu-Sal, and 1.6 w/o Li₂CO₃, and 4.9 w/o K₂CO₃. The same mild steel pot and mild steel electrodes used for the previous Nu-Sal test (Salt Bath 4) were retained for this salt-bath test. When the bath had aged 1 week, one group of 2 specimens was treated by the original procedure, which omits separate rinse samples for chloride determination.

Hydrogen-analyses results were received for two specimens treated in Salt Bath 1, six specimens treated in Salt Bath 2, eight specimens treated in Salt Bath 3, and two specimens treated in Salt Bath 4. These results and the four hydrogen analyses reported last month are recorded in Table D-1. The hydrogen contained in individual specimens treated for 13.5 min ranged from 0.21 to 1.10 ppm. Average hydrogen contents for all specimens treated for 13.5 min in each salt bath were 0.41, 0.87, and 0.68 ppm for Salt Baths 1, 2, and 3. The low average hydrogen content was obtained with an all-chloride bath (Salt Bath 1). The greater Li₂CO₃ content of Salt Bath 2 compared with that of Salt Bath 3 (3.1 w/o versus 1.6 w/o) may explain the greater average hydrogen content of specimens treated in Salt Bath 2 (0.87 compared with 0.68 ppm). However, humidity data which have not been computed and correlated at this point may explain some of the difference in hydrogen pickup between Salt Baths 2 and 3. The two specimens treated for 135 min in Salt Bath 3 had an average hydrogen content of

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**TABLE D-1. HYDROGEN CONTENT OF URANIUM SPECIMENS TREATED IN SALT BATHS^(a)
1, 2, 3, AND THE FIRST WEEK IN SALT BATH 4**

Specimen	Hydrogen Content, ppm		
	Individual Results	Average for Each Group	Average for Each Salt-Bath Test
1-1-a	0.51	0.36	
1-1-b	0.21		
1-2-a	0.40	0.39	0.41
1-2-b	0.37		
1-3-a	0.40	0.49	
1-3-b	0.57		
2-1-a	0.73	0.91	
2-1-b	1.09		
2-2-a	0.95	1.03	0.87
2-2-b	1.10		
2-3-a	0.83	0.66	
2-3-b	0.49		
3-1-a	0.97	0.80	
3-1-b	0.62		
3-2-a	0.61	0.74	0.68
3-2-b	0.87		
3-3-a	0.51	0.50	
3-3-b	0.48		
3-3-c ^(b)	1.15	1.13	
3-3-d ^(b)	1.11		
4-1-a	0.52	0.50	
4-1-b	0.47		

(a) Salt Bath	Composition
1	93.5 w/o Nu-Sal (50 w/o NaCl-50 w/o KCl) and 6.5 w/o K-2 (50 w/o KCl-50 w/o Na ₂ CO ₃)
2	93.0 w/o K-2, 3.1 w/o Li ₂ CO ₃ , and 3.9 w/o K ₂ CO ₃
3	93.5 w/o K-2, 1.6 w/o Li ₂ CO ₃ , and 4.9 w/o K ₂ CO ₃
4	93.5 w/o Nu-Sal, 2.9 w/o Li ₂ CO ₃ , and 3.6 w/o K ₂ CO ₃

(b) These specimens were treated for 135 min instead of 13.5 min.

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1.13 ppm compared with an average hydrogen content of 0.50 ppm for the two specimens treated at the same time for 13.5 min. The large differences obtained with some duplicate samples are greater than would be expected from the hydrogen-content test. NLO at Fernald will analyze ten untreated uranium specimens to determine the hydrogen-content variation that exists before heat treating in the experimental salt baths.

All of the uranium specimens treated in Salt Bath 4 and the two specimens treated so far in Salt Bath 5 showed no visible salt coating after quenching. Analysis of rinse samples obtained from the uranium specimens treated when Salt Bath 4 was 1 and 2 weeks old showed from 8.35×10^{-6} to less than 5.95×10^{-6} g per cm^2 of specimen surface. In view of such low levels of chloride pickup on the surface of the uranium specimens, these analyses have been discontinued. However, it has been suggested that NLO at Fernald determine the chloride content of the cuttings they obtain from the surface of the uranium specimens for a more positive test of chloride adherence to uranium. Visible surface corrosion of the uranium specimens was negligible in all cases. Weight losses were between 0.2 and 0.3 per cent for the specimens treated the standard 13.5 min. Weight losses of the two specimens treated for 6.75 min were slightly lower, 0.18 per cent. The actual weight losses are recorded in Tables D-2 and D-3. Metallographic examinations of the corrosion specimens suspended in the salt baths are in progress.

The planned work consists of completing the treatment now in progress of uranium specimens in Salt Bath 5. Work will be begun on correlation of the data obtained from the five salt-bath tests.

TABLE D-2. WEIGHT LOSS OF URANIUM SPECIMENS TREATED THE FIRST, SECOND, AND THIRD WEEKS IN SALT BATH 4

89.5 w/o Nu-Sal, 2.9 w/o Li_2CO_3 , and 3.6 w/o K_2CO_3 .

Specimen	Weight Loss ^(a)	
	G	Per Cent
4-1-a	2.1	0.22
4-1-b	1.8	0.20
4-2-a	2.1	0.22
4-2-b	2.5	0.26
4-3-a	2.2	0.23
4-3-b	2.0	0.21
4-3-c ^(b)	1.7	0.18
4-3-d ^(b)	1.7	0.18

(a) Incurred during the operations of heating, quenching, rinsing, and drying the specimens.

(b) These specimens were treated for 6.75 min instead of 13.5 min.

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TABLE D-3, WEIGHT LOSS OF URANIUM SPECIMENS TREATED THE FIRST WEEK IN SALT BATH 5

93.5 w/o Nu-Sal, 1.6 w/o Li_2CO_3 , and 4.9 w/o K_2CO_3 .

Specimen	Weight Loss ^(a)	
	G	Per Cent
5-1-a	2.1	0.23
5-1-b	2.2	0.21

(a) Incurred during the operations of heating, quenching, rinsing, and drying the specimens.

Galling Problems in Compacting of Magnesium-Green Salt Blends

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

At the request of NLO, a program has been initiated designed to eliminate galling of die walls while briquetting uranium tetrafluoride (UF_4) and magnesium blends preparatory to bomb reduction. It is believed that this galling is caused when magnesium particles become wedged between the ram and die wall and are extruded along the length of the wall as the ram is operated. After several operational cycles the amount of magnesium that builds up on the die wall becomes sufficient to cause the ram to stick. Three factors are being considered in efforts to eliminate this galling problem. These are the design of the die, die lubrication, and the die material.

With regard to die design, several inserts with various die clearances will be made and used between the ram and compact to determine the effect of die-ram clearance on the galling problem. A die is being made with a bore diameter of 2-3/4 in. and having a slight taper (10 min) in the section where the final compacting of the biscuit is done.

In addition to the new die design, several lubricants will be used on the die and ram surfaces to facilitate ram movement during compacting. The lubricants will be chosen for the most part from those used in the fabrication of magnesium. In using these lubricants, no regard will be paid as to their effect in contaminating the pressed compacts until some are found to be suitable. Then the characteristics of the lubricants such as composition and volatility will be considered.

The possibility of using a different die material will not be considered until after the die-design and lubrication studies have been completed.

The die has been designed and is being constructed. Compacting will begin when it is completed. A literature survey has been conducted on the lubricants used in the forming of magnesium shapes. It has been found that dispersions of graphite, such as Aquadag suspended in various carrier media, have been used for the most part in the

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extrusion of magnesium shapes. Graphite dispersions have also been used to prevent galling in hot pressing uranium and magnesium blends. In addition, several of the stearates have also been used with success in fabricating magnesium.

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E. GENERAL FUEL-ELEMENT DEVELOPMENT

F. A. Rough

In the development of refractory fuel elements, emphasis is continuing on fission-product losses and uranium losses from graphite-UO₂ specimens.

The sinterability of BeO is improved considerably by the combined addition of MgO and CaO, but is unaffected by the separate additions of either MgO or CaO.

Studies intended to evaluate the radiation performance of various uranium alloys and compartmentalized specimens are in varying stages of progress. Being studied are alloys of uranium-niobium, uranium-zirconium, and uranium-chromium-molybdenum, and specimens of stainless steel with dispersed particles of UC or UN.

Water-corrosion studies of uranium give results which are readily explained, if more trivalent uranium is produced in the early stages of corrosion.

CERAMICS

Refractory Fuel Elements

G. B. Engle, M. J. Snyder, J. F. Quirk,
D. N. Sunderman, and M. Pobereskin

Research is being performed on problems associated with fuel elements for high-temperature gas-cooled reactors. Current studies are on fabrication and design of graphite fuel elements and the diffusion of uranium in dense BeO.

BeO

Studies to determine the penetration of uranium into a dense BeO body were continued. Previously, uranium was detected at a depth of 0.001 in. below the treated surface of a UO₂-BeO diffusion couple which had been heated at 2300 F for 20 hr in vacuo. This month, no uranium was detected after an additional cut of 0.001 in., or a total of 0.002 in. below the treated surface of the couple, indicating relatively slow diffusion of uranium into the BeO under these conditions.

Additional UO₂-BeO diffusion couples are being prepared, using dense BeO tubes and wedge-shaped slabs to obtain more precise information on the diffusion. Several slabs were pressed from readily sinterable BeO powder and are being sintered in hydrogen. Extruded tubes of essentially theoretical density were bisected and are being examined with a microscope. Efforts will be made to make UO₂ adhere to the surfaces of these specimens, and the couples, so prepared, will be heated for various periods in the temperature range 2000 to 3000 F.

Graphite

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

Previously, several lots of rod-shaped graphite elements, containing UO_2 rods at their axes and differing in the fabrication methods used in their preparation, were irradiated at room temperature. The fractions of the total fission gases produced during irradiation which diffused from the specimen during subsequent heat treatments in the range 1650 to 2200 F are being measured and the products identified. Initial data have been obtained and are being evaluated.

In studies to determine uranium losses from a UO_2 -graphite element, a run was made at 2000 F for 28 hr using high-purity nitrogen (dew point, -100 F). No significant damage occurred to the UO_2 -graphite element (0.45 per cent weight loss). Uranium vapor was collected in a water-cooled trap immediately below the furnace tube. The trap was acid washed and the wash solution submitted for uranium analysis.

After a new supply of high-purity nitrogen has been procured, the same sample will be reheated for a longer period. Uranium will be collected and analyses made at intermediate times.

In planned work, studies of uranium diffusion in BeO will be continued. Fabricating techniques of UO_2 -graphite elements will be evaluated with respect to their ability to hold fission gases. A large number of specimens will be fabricated, using the best techniques, and irradiated for further evaluation of fission-gas retention. Uranium-loss tests on UO_2 -graphite elements will be extended to a longer time at 2000 F.

Characterization of Sinterable Oxide Powders

J. F. Quirk and N. Mosley

Relations between sinterability and the basic properties of oxide powders are being investigated. From previous work on BeO prepared by thermal decomposition of $\text{Be}(\text{OH})_2$, impurity content of the hydroxide appeared to be an important factor affecting sinterability of the oxide powder.

Current work is aimed at finding the effect on sinterability of specific impurities.

Experiments last month showed that certain sinterable BeO powders had appreciable amounts of surface-concentrated impurities, principally MgO and CaO , whereas a pure nonsinterable powder did not.

During March, experiments were made to determine the effect of specific contaminants, MgO , CaO , and Al_2O_3 , on the sinterability of pure BeO prepared from ammonia-precipitated $\text{Be}(\text{OH})_2$. Gaseous NH_3 was used to precipitate $\text{Be}(\text{OH})_2$ from BeSO_4 solutions that were deliberately adulterated with aluminum, calcium, or magnesium ions in amounts equivalent to about 1 of w/o oxide. The hydroxides were separated and dried

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without washing, and were calcined 2 hr at 1470 F to form BeO. The BeO powders were compacted hydrostatically at 50,000 psi and sintered for 1 hr at 2600 F in argon-diluted hydrogen. Sinterability of the contaminated powders is indicated by the bulk-density values given in Table E-1. Results for a sample of uncontaminated oxide and for a readily sinterable impure BeO designated Sample C-5659 are shown for comparison.

TABLE E-1. EFFECT OF CONTAMINANTS ON SINTERABILITY OF BeO POWDERS

BeO ^(a) Sample	Contaminant ^(b)	Bulk Density of Sintered BeO Compacts	
		G per Cm ³	Per Cent of Theoretical
LP-30	None	2.77	91.6
LP-31	MgO	2.76	91.3
LP-32	CaO	2.09	69.1
LP-33	Al ₂ O ₃	2.79	92.2
LP-34	2/3 MgO, 1/3 CaO	2.93	96.9
C-5659	Impure	2.94	97.2

(a) All BeO samples except C-5659 were made from NH₃-precipitated hydroxides.

(b) Added to the starting BeSO₄ solutions in amounts equivalent to 1 w/o total oxide contaminant.

The results indicate that contamination with MgO or CaO alone did not improve the sinterability of the pure BeO, but sinterability was improved considerably by addition of these oxides together in the ratio of two parts MgO to one part CaO. Contamination with Al₂O₃ did not improve sinterability, and the CaO addition appeared to inhibit sintering. Spectrographic examination of the powders will be made in order to correlate these results with previous experiments.

For the production of reactor-grade BeO, the ammonia precipitation of Be(OH)₂ has a considerable advantage over the commercial process employing NaOH as the precipitant. Undesirable impurities, such as sodium and the contaminants usually present in NaOH, are avoided by use of gaseous ammonia and, in addition, the process is simpler, requiring no heating, dissolving, or washing of the precipitate. In view of these advantages, some of this month's experiments on the effect of MgO and CaO contaminants will be repeated to assess the reproducibility of the process.

METALLURGY

Uranium Alloys for High-Temperature Application

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

The elevated-temperature properties of binary uranium alloys are being obtained as part of an evaluation of these alloys for reactor-fuel application. At present, alloys of uranium-7, 10, 15, and 20 w/o niobium are being studied.

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Tables E-2 and E-3 list linear-thermal-expansion data from room temperature to 950 C obtained upon specimens of uranium-7 and 10 w/o niobium alloys water quenched from 790 C. The measurements were made in a recording quartz-tube dilatometer, and include data from two heating and two cooling curves. A portion of the variation in expansion coefficients between data from the two heating and cooling curves is a result of the sluggishness of transformation in these alloys and the continued process of transformation during heating and cooling. The first heating curve, in particular, was obtained on material which was gamma quenched, while, subsequently, the alloys all had transformed to some degree during the slow temperature cycling.

An additional phase of this program is concerned with irradiation and subsequent inspection of alloys of uranium-10 and 20 w/o niobium and uranium-15 w/o zirconium. The first of three temperature-controlled (725 C) capsules has been loaded with one specimen of each of these alloys, and has been shipped to the MTR. The capsules will be irradiated to produce approximately 1 a/o burnup of the fuel and will be returned to Battelle for examination.

Preparation and Properties of Low-Melting Alloys

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

The experimental development of low-melting castable uranium alloys for possible use as a power-reactor fuel is in progress. A uranium-5 w/o chromium-1 w/o molybdenum alloy is undergoing investigation at this time. Small pin-type specimens of the as-cast alloy will be irradiated at temperatures of 600 C and lower to a burnup of approximately 1 a/o.

Small pin-type specimens of the as-cast alloy have been prepared. The pins are to be evaluated by conducting postirradiation measurements of dimensions, density, linear thermal expansion, electrical resistivity, and thermal conductivity and by comparing these results with those obtained from the pins prior to irradiation. The same measurement will be made following irradiation. The results will be compared with information available from previous studies made on the binary uranium-chromium eutectic alloy.

Compartmentalized Fuel Elements

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

The phase of this program concerned with the preparation of irradiation specimens is now in progress. The two types of specimens to be irradiated are 24.2 w/o UN and 24.3 w/o UC dispersed in Type 347 stainless steel. All irradiation specimens will be clad with Type 347 stainless and will contain fully enriched uranium compounds.

A series of dummy specimens containing 24.3 w/o natural UC dispersed in stainless has been roll clad. A 6-to-1 reduction at 1800 F was used to give a final core

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TABLE E-2. LINEAR-THERMAL-EXPANSION COEFFICIENTS FOR URANIUM-7 w/o NIOBIUM

Temperature, C	Linear-Thermal-Expansion Coefficient, 10^{-6} per deg C			
	First Heating	First Cooling	Second Heating	Second Cooling
20-100	12.6	13.5	13.1	13.2
200	12.5	13.5	13.0	13.3
300	12.7	14.1	13.4	13.8
400	13.1	14.5	14.0	14.3
450	13.0	--	--	--
460	12.5	--	--	--
475	11.5	--	--	--
500	10.7	15.2	14.4	15.0
525	10.1	--	--	15.3
550	9.9	16.3	--	16.3
570	--	19.9	--	--
575	--	20.2	--	20.2
600	10.3	20.2	14.7	20.2
650	11.5	--	15.3	--
665	--	--	15.0	--
675	15.7	--	20.1	--
700	15.9	20.1	20.4	20.1
800	16.1	20.1	19.9	20.0
900	16.5	20.0	20.1	20.0
950	16.5	20.0	20.1	20.0

TABLE E-3. LINEAR-THERMAL-EXPANSION COEFFICIENTS FOR URANIUM-10 w/o NIOBIUM

Temperature, C	Linear-Thermal-Expansion Coefficient, 10^{-6} per deg C			
	First Heating	First Cooling	Second Heating	Second Cooling
20-100	13.3	13.2	15.9	13.7
200	13.3	13.3	15.8	13.6
300	13.5	13.9	16.0	14.1
400	14.0	14.3	16.6	14.5
425	--	--	16.6	--
450	--	14.6	16.4	14.7
460	--	--	--	--
475	--	15.0	--	--
500	14.2	15.9	15.7	16.0
515	--	--	--	16.6
525	13.8	16.6	15.3	16.8
535	--	17.0	--	17.0
550	12.8	17.4	14.8	17.2
575	11.5	--	14.5	--
585	11.2	--	--	--
600	11.1	17.4	14.5	17.2
625	11.2	--	--	--
650	11.6	--	14.9	--
660	--	--	15.3	--
665	--	--	--	--
675	14.8	--	17.6	--
685	15.1	--	18.8	--
700	15.1	17.6	19.1	17.4
800	14.8	17.8	18.9	17.6
900	14.8	17.9	18.9	17.7
950	14.9	18.1	18.9	17.8

thickness of 50 mils and a cladding thickness of 5 mils. Metallographic specimens are being prepared.

Dummy specimens containing UN dispersed in stainless are being prepared and will be used to establish exact fabrication techniques for preparation of UN-bearing irradiation specimens.

CORROSION

Mechanism of Uranium Corrosion

J. B. Schroeder, C. M. Schwartz, H. D. Hannan,
P. D. Miller, R. S. Peoples, J. W. Moody, and H. Goering

The mechanism of aqueous uranium corrosion is being studied to develop corrosion-resistant reactor fuel elements.

The investigation of the oxide produced by corrosion in water at 100 C has been continued. The oxygen-to-uranium ratios of additional samples have been determined in a microbalance as described in BMI-1173. The ratios resulting from corrosion-produced oxide are: 2.19^a, 2.26^a, and 2.19. Commercial UO₂ which was placed in boiling water for 24 hr before ignition in the microbalance gave a ratio of 2.07. A sample of corrosion product which was boiled an additional 24 hr after the metal had been removed from the test also gave an oxygen-to-uranium ratio of 2.07.

These results, and the hydrogen deficit reported earlier, can be readily explained, if some trivalent uranium is produced in the early states of corrosion. An analytical technique is being developed to test this hypothesis.

Further corrosion tests on dingot material at 200 C in a wet flowing-argon atmosphere have been made. The inconsistency of the results cannot be explained and a modification of the test apparatus is in progress. This consists mainly of a thermostatically controlled oil bath in which the reaction vessel can be immersed and maintained at any desired temperature.

Work was continued on procedures for purifying uranium by floating-zone techniques. Spectrographic analyses on specimens from Ingots 12169-11 and 12169-12 have been obtained and are given in Table E-4.

^aReported in BMI-1173 and included here for completeness.

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TABLE E-4. SPECTROGRAPHIC ANALYSIS OF FLOATING ZONE MELTED URANIUM

Specimen	Impurity, (ppm)									
	Al	Ca	Co	Cu	Fe	Mg	Mn	Mo	Ni	Si
1	10	10	15	10	20	10	5	4	4	20
12169-11A	10	8	15	10	20	2	5	4	4	40
-11B	10	10	15	10	30	6	8	5	5	20
12169-12A	10	8	10	3	20	2	6	6	5	15
-12B	10	8	20	3	20	4	6	6	6	15

It will be recalled that Ingot 12169-11 was given five passes at 1 in. per hr and Ingot 12169-12 was also given five passes at 1 in. per hr with a direct current of about 7 amp being applied to the ingot during zone melting. Specimen 1 in Table E-4 refers to the original material before zone melting. Specimens 12169-11A and 12169-12A were from the first parts of the ingots to freeze and Specimens 12169-11B and 12169-12B were from the rear.

The spread of values shown in Table E-4 are of the order of the accuracy of the analyses. Therefore, conclusions about the effect of the electric current cannot be drawn. In either case, purification of the ingots was slight.

At present an ingot is being given five passes at 1/2 in. per hr. When this experiment is finished it is planned to give an ingot a large number of "heat passes", i.e., a hot zone somewhat below the melting point will be passed along the ingot.

F-1

F. STUDIES OF ZIRCONIUM-URANIUM ALLOYS

F. A. Rough

Studies of zirconium-uranium alloys have been in progress for some time. Some of the programs have been completed, but research is continuing in order to give a further understanding of mechanical properties, corrosion behavior, and irradiation performance.

Preliminary results on high-temperature water corrosion at additional temperatures have been obtained. Activation energies are given for corrosion of alloys containing 30 to 60 and 7 to 15 w/o uranium.

Preliminary radiations of the 22 w/o uranium alloy confirm its excellent stability and indicate that it is not sensitive to wide variations in microstructure at burnups near 1 a/o. Greater burnups will be required to establish the significant effects attributable to microstructure.

Heat Treatment and Mechanical Properties
of Zirconium-Uranium Alloys

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

The effect of oxygen on the decomposition of the high-temperature solid solution of uranium in zirconium is being studied. The kinetics of the transformation and the resultant type of structure obtained are being determined by interrupted quenching of samples and subsequent metallographic examination. Mechanical properties are being evaluated as a function of heat treatment and structure.

It appears that 15 w/o uranium alloys containing 0.29 w/o oxygen precipitate alpha zirconium much more rapidly than alloys containing only 0.094 w/o oxygen. The alpha precipitation at 400 and 450 C in the 0.29 w/o oxygen alloys resulted in beta enrichment and formation of epsilon prime (supersaturated epsilon) within 10 min at 450 C and within 1 min at 400 C. Alpha appeared as discrete particles as well as the "basket-weave" Widmanstätten structure. Epsilon prime was observed as small grains within the prior beta grains. Proeutectoid alpha formed at temperatures between 500 and 580 C within the period of 10 to 100 min for the 0.29 w/o alloy. No epsilon prime was observed, although the presence of the phase might possibly be detected by X-ray diffraction. Other temperatures for the 0.094 w/o oxygen alloys have not been analyzed as yet.

Proeutectoid alpha zirconium precipitated in the 10 w/o uranium-0.29 w/o oxygen alloy within 1 min at 400 C, within 10 min at 450 C, and within 100 min at 500 C. The 10 w/o uranium-0.094 w/o oxygen alloy showed no alpha within 10 min at either 450 or 500 C. It appears that a "knee" exists below the eutectoid temperature for these alloys. It is not possible to tell from the microstructures if epsilon prime formed in these alloys. A phase which may be epsilon prime was masked by large amounts of acicular alpha which formed during the final quench.

Additional alloys and temperatures will be analyzed as the photomicrographs become available. Meanwhile, specimens for tensile-property tests are being prepared.

Corrosion of Zirconium-Uranium Alloys

W. Chubb, R. F. Dickerson, R. S. Peoples, and W. E. Berry

Evaluation of Clad Samples

Some zirconium-uranium alloys are being clad with Zircaloy 2 for an examination of the corrosion behavior of such units. The cores will be sponge zirconium containing 22, 50, and 70 w/o uranium. The clad samples will be heat treated in a variety of ways, defected, and corrosion tested in 680 F water.

The components necessary for roll cladding have been collected, machined, and assembled. The roll-cladding operation will be performed as soon as the rolling equipment becomes available. It is hoped that specimens will be available for corrosion testing in about 3 weeks.

Evaluation of Bare Samples

The corrosion behavior of zirconium-uranium alloys is being evaluated in high-temperature water as part of the over-all program on the study of the properties of zirconium-uranium alloys. Alloys being studied contain 7 through 70 w/o uranium additions to crystal bar, sponge, and high-oxygen sponge zirconium.

Corrosion rates have been established in 600 and 680 F high-purity water. Long-time tests are being continued in 500 F water in an attempt to establish corrosion rates for alloys containing less than 30 w/o uranium. Corrosion tests are now under way in 550 and 640 F water and corrosion rates have been calculated after 1000 hr of exposure. Data from the 500 F water tests do not fall on the temperature-dependency plot of the linear corrosion rates at the other four temperatures, indicating a possible difference in reaction kinetics at this temperature. At a given alloy level, corrosion rates increase with increasing oxygen content in the zirconium melting stock, although the slope of the temperature-dependency curve is generally the same for the three different melting stocks. Based on preliminary results, an activation energy of approximately minus 22,000 cal per mol was determined for 7 through 15 w/o uranium alloys and approximately minus 18,500 cal per mol was determined for 30 through 60 w/o uranium alloys.

Development of Corrosion-Resistant 70 w/o Uranium Alloys

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

An investigation aimed at improving the corrosion resistance of the zirconium-70 w/o uranium alloy either by heat treatment or by ternary additions to the base alloy is being conducted. Ternary additions are being made either to tie up the free alpha

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uranium found in the 70 w/o alloy in the form of a compound or to stabilize the gamma phase. Additions of molybdenum, niobium, silicon, and titanium are being studied for this purpose.

Corrosion tests of base 70 w/o alloy specimens and of ternary alloy specimens, bare, in 680 F water, have been completed. The results of these tests indicated that additions of molybdenum, silicon, and titanium did not improve the corrosion behavior of the alloy (see BMI-1173). Alloys containing niobium are being prepared in order that the effect of this addition can be determined.

In addition, alloys similar to those already tested have been prepared by duplex melting and have been fabricated to flat plates. Specimens are to be clad with Zircaloy 2 and corrosion tested employing a pinhole defect. Preparations for cladding are now under way.

Radiation Stability of Zirconium-Uranium Alloys

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

The investigation of the effects of irradiation on zirconium-uranium alloys is being continued.

Initial studies to determine the effect of heat treatment and microstructure on the stability of the zirconium-22 w/o uranium alloy have been completed. The results and the various heat treatments employed are shown in Table F-1. The heat treatments employed are identified by letters and consisted of the following:

Heat Treatment C.

36 hr at 1550 F, furnace cooled 1/2 F per min from 1550 to 1200 F, and furnace cooled to room temperature.

Heat Treatment E.

36 hr at 1550 F, water quenched; annealed 1/2 hr at 1020 F, water quenched; annealed 10 hr at 1400 F and furnace cooled.

Heat Treatment F.

36 hr at 1550 F, water quenched; annealed for 22 days at 1020 F and furnace cooled.

Heat Treatment C produced microstructures leaving a dispersion of coarse alpha-zirconium plates in an epsilon matrix; Heat Treatment E resulted in the precipitation of alpha zirconium in the prior beta grain boundaries surrounding a fine dispersion of alpha zirconium in epsilon; and Heat Treatment F produced a fine dispersion of alpha zirconium and epsilon.

TABLE F-1. PRE- AND POSTIRRADIATION DATA ON ZIRCONIUM-02 w/o URANIUM ALLOY SPECIMENS

Identification		Preirradiation Heat Treatment(a)	Temperature During Irradiation, C	Total Calculated Krypton Released, % of theoretical	Per Cent Change			Remarks	
Capsule	Specimen				Density	Length	Diameter		
BME 2-1	EN-19	C	-400	0.95	0.4	-4.06	2.3	1.60	No appreciable bow or warpage is evident
BME 2-1	EN-27	E	-400	0.85	--	-3.28	1.4	-0.20	Small amount of surface roughening, no bow or warpage is evident
BME 2-1	EN-4A	F	-400	0.85	0.4	-3.41	2.0	0.00	Machining marks are quite evident -- slightly warped
BME 2-2	EN-00	C	-400	0.74	--	-3.41	6.4	2.19	Warped in center section of the specimen
BME 2-2	EN-05	E	-400	0.76	4.5	-2.38	10.7	-1.89	Very slight bow is evident
BME 2-2	EN-5A	F	-400	0.88	2.5	-3.41	2.2	4.09	Slightly warped
BME 2-3	EN-02	C	-400	0.92	0.4	-2.88	4.9	2.89	No bow or warpage is evident
BME 2-3	EN-08	E	-400	0.98	0.2	-3.14	5.2	0.10	No bow or warpage is evident
BME 2-3	EN-11	F	-400	0.76	0.4	-2.75	2.3	2.29	Slightly bowed
BME 2-4	EN-03	C	400	0.34	1.2	-2.73	1.4	0.90	Very smooth surface -- no bow or warpage is evident
BME 2-4	EN-09	E	400	0.34	0.2	-2.33	-4.1	0.40	Very smooth surface -- no bow or warpage is evident -- one end of the specimen was broken in the capsule
BME 2-4	EN-12A	F	400	0.13	0.4	-1.97	0.5	0.40	Very smooth surface -- no bow or warpage is evident

(a) The letter denotes the following type of heat treatment:

C - 24 hr at 1550 F, furnace cooled 1/2 F per min from 1550 to 1200 F and furnace cooled to room temperature.

E - 24 hr at 1550 F, water quenched; annealed 1/2 hr at 1020 F, water quenched; annealed 24 hr at 1400 F and furnace cooled.

F - 24 hr at 1550 F, water quenched; anneal for 22 days at 1020 F and furnace cooled.

(b) These calculations are based on densimeter readings.

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F-5 and F-6

Three sets of specimens of each heat treatment were irradiated without temperature control to an intended burnup of 2 a/o (based on total atoms). Burnups achieved are given in Table F-1. The calculated maximum temperature was about 400 C. Specimens in a temperature-controlled capsule were irradiated at 650 C to a calculated burnup of about 0.15 a/o total. These particular specimens received such a low burnup because after only one cycle in the MTR the capsule leads were inadvertently cut and the capsule had to be removed from the reactor.

The specimens irradiated at about 400 C had an average of 4.05 per cent volume expansion per 1 a/o burnup. Although there is no significant effect of the various heat treatments upon the volume expansion or upon the amount of fission-gas release, the specimens receiving Heat Treatment E appear to show a slightly greater resistance to growth and warpage. Higher burnups will be required to evaluate the effects of heat treatment in the 22 w/o uranium alloy.

The specimens irradiated at 650 C were in the alpha zirconium-plus-beta field during irradiation. These specimens experienced about four times as much volume expansion per a/o burnup as did the specimens irradiated at epsilon-phase temperatures (400 C). This may be the effect of the decreased strength of the beta phase. However, there was no significant difference in the percentage of fission gas released.

Chemical burnup analyses will be made on selected specimens to verify the dosimeter burnups.

Zirconium-50 w/o uranium specimens are now being encapsulated for irradiation to 2 a/o burnup in the MTR.

G-1

G. CORROSION STUDIES OF ZIRCONIUM

R. S. Peoples

Research described in this section pertains to a study of the long-term corrosion properties of zirconium-base cladding materials in high-temperature water and steam.

Long-term corrosion tests have been continued for samples of Zircaloy 2 and modified Zircaloy 2 compositions (lower tin with iron and/or nickel) exposed to 600 and 680 F degassed water and 750 F (1500 psi) steam.

Corrosion Studies of Zirconium and Zirconium Alloys

W. K. Boyd and R. S. Peoples

Samples from seven different melts of Zircaloy 2 continue to evince excellent resistance to 600 F degassed water. No indication of breakaway or transition in corrosion rate has been observed after exposure periods approaching 3-1/2 years. Total weight gains are slightly in excess of 50 mg per dm².

In other studies, intentionally contaminated samples of Zircaloy 2 are being evaluated in 680 F degassed water. Results based on up to 21 months of exposure may be summarized as follows:

- (1) Nitrogen contents in excess of 200 ppm destroy corrosion properties of Zircaloy 2.
- (2) In the range 500 to 1500 ppm, hydrogen has no significant effect on corrosion behavior of samples exposed to 680 F water.
- (3) A substantial reduction in corrosion resistance occurs with oxygen levels in excess of 6000 ppm.

Zirconium Corrosion Committee Program

As a part of the Zirconium Corrosion Committee program to develop zirconium-base alloys superior to Zircaloy 2, extended-exposure data are being obtained in 680 F degassed water and 750 F steam. Alloys under study contain 0.25 to 0.50 w/o tin plus 0.0 to 0.50 w/o iron and/or nickel with and without additions of nitrogen and aluminum. Weight-gain data based on up to 546 days of exposure are summarized in Tables G-1 through G-3. Results continue to indicate that:

- (1) Heat treatments consisting of either a slow cool from 775 C or a water quench from 900 C do not significantly alter the corrosion behavior of tin-iron or tin-nickel alloys.

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TABLE G-1. CORROSION OF ARC-MELTED (ROCKING-HEARTH METHOD)

Alloys Melted

Heat	Alloy Composition (Balance Zirconium), w/o	750 F Steam			680 F Water	
		Exposure Time, days	Weight Gain, mg per dm ²	Remarks ^(*)	Exposure Time, days	Weight Gain, mg per dm ²
Heated 1 Hr at 775 C in Vacuum and Furnace Cooled						
						<u>Tin and</u>
462	Unalloyed	1-1/2	768	W	210	211
463	Unalloyed	1-1/2	839	W	210	210
464	0.225Sn-0.28Fe	104	108	M	546	97
465	0.235Sn-0.30Fe	104	105	M	546	98
466	0.225Sn-0.38Fe	134	100	M	546	93
467	0.225Sn-0.39Fe	134	100	M	546	87
469	0.255Sn-0.27Fe	111	102	M	546	84
468	0.255Sn-0.28Fe	111	105	M	546	85
470	0.255Sn-0.33Fe	136	102	M	546	83
471	0.255Sn-0.34Fe	111	107	M	546	82
473	0.275Sn-0.26Fe	106	105	M	546	73
472	0.295Sn-0.28Fe	106	115	M	546	81
475	0.275Sn-0.28Fe	148	109	M	546	78
474	0.285Sn-0.34Fe	106	104	M	546	74
477	0.475Sn-0.44Fe	112	112	M	546	76
478	0.515Sn-0.47Fe	142	109	M	546	76
481	0.525Sn-0.47Fe	136	106	M	546	80
479	0.575Sn-0.50Fe	136	95	M	546	70
476	0.485Sn-0.51Fe	124	100	M	546	80
480	0.515Sn-0.53Fe	124	102	M	546	77
						<u>Tin and</u>
488	Unalloyed	1-1/2	680	W	120	138
489	Unalloyed	1-1/2	570	W	154	111
491	0.175Sn-0.03Ni	249	114	M	434	109
490	0.185Sn-0.04Ni	231	103	M	434	102
492	0.135Sn-0.08Ni	249	109	M	546	85
493	0.185Sn-0.08Ni	189	112	M	546	94
495	0.175Sn-0.19Ni	249	107	M	546	94
494	0.195Sn-0.19Ni	249	115	M	546	107
496	0.215Sn-0.04Ni	207	78	W, F	546	118
497	0.225Sn-0.04Ni	249	117	M	482	102
498	0.225Sn-0.07Ni	213	102	M	546	90
499	0.225Sn-0.08Ni	207	118	M	546	89
501	0.215Sn-0.20Ni	213	100	M	546	94
500	0.225Sn-0.41Ni	249	115	M	546	90
502	0.275Sn-0.03Ni	213	108	M	482	102
503	0.275Sn-0.04Ni	213	108	M	482	108
505	0.275Sn-0.08Ni	213	100	M	546	103
504	0.285Sn-0.08Ni	213	100	M	546	90
506	0.265Sn-0.19Ni	213	103	M	546	107

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SPONGE ZIRCONIUM IN 150 F STEAM AND 680 F DEGASSED WATER

at Battelle

Remarks ^(*)	150 F Steam		Remarks ^(*)	680 F Water		Remarks ^(*)
	Exposure Time, days	Weight Gain, mg per dm ²		Exposure Time, days	Weight Gain, mg per dm ²	

Heated 1 Hr at 900 C in Vacuum and Water Quenched

Iron

M, F	10	1680	W	28	Lost weight	W, F
M, F	10	1325	W	28	66	W, F
M	136	109	M	560	95	M
M	136	109	M	560	80	M
M	136	104	M	560	95	M
G	136	100	M	560	80	M
G	184	105	M	560	90	M
G	136	100	M	560	69	G
G	147	100	M	560	84	M
G, I	136	105	M	560	94	M
G	184	108	M	560	82	M
G	147	95	M	560	81	M
G	147	102	M	560	84	M
G	136	100	M	560	88	M
G	147	111	M	560	103	M
G	136	113	M	560	71	M
G	136	105	M	560	96	M
G	136	110	M	560	109	M
G	147	106	M	560	101	M
G	136	104	M	560	106	M

Nickel

M	26	Lost weight	W, F	42	261	W
W	26	Lost weight	W, F	42	231	W
M	206	105	M	532	134	W, S
M	242	117	M	564	150	W, S
M	168	Lost weight	S, F	532	94	M
M	242	113	M	532	101	M
M	242	116	M	532	101	M
M	206	102	M	532	95	M
M	261	Lost weight	W, F	532	104	M
M	261	Lost weight	W, F	532	101	M
M	168	Lost weight	S, F	532	125	M
M	242	106	M	532	101	M
M	201	111	M, S	532	87	M, S
M, S	206	107	M	532	87	M, S
M	140	Lost weight	W, F	532	100	M
M	206	Lost weight	S, F	532	102	M
M	261	Lost weight	W, F	532	111	M, S
M	242	110	M	532	104	M
M	206	103	M	532	100	M

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TABLE G-1.

Heat	Alloy Composition (Balance Zirconium), w/o	750 F Steam			680 F Water		Tin and (Con-
		Exposure Time, days	Weight Gain, mg per dm ²	Remarks ^(a)	Exposure Time, days	Weight Gain, mg per dm ²	
507	0.255zr-0.10Ni	207	148	M	546	105	
508	0.565zr-0.04Ni	249	106	M	434	102	
509	0.565zr-0.04Ni	213	102	M	546	105	
510	0.545zr-0.07Ni	213	103	M	546	106	
511	0.565zr-0.07Ni	207	117	M	546	98	
513	0.515zr-0.18Ni	207	120	M	462	101	
512	0.545zr-0.20Ni	207	118	M	546	112	
							Tin, ppm,
483	0.515zr-0.14Fe-0.090Ni	124	104	M	490	110	
482	0.485zr-0.23Fe-0.097Ni	148	110	M	546	85	
485	0.475zr-0.27Fe-0.13Ni	136	102	M	546	128	
484	0.485zr-0.31Fe-0.13Ni	136	102	M	546	94	
486	0.485zr-0.38Fe-0.18Ni	148	106	M	546	93	
487	0.485zr-0.44Fe-0.18Ni	148	116	M	546	104	

(a) Key to remarks: G - black tarnish film (good material)

M - milky

W - white oxide

F - flaking (heavy oxide)

S - segregated.

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(Continued)

Remarks ^(*)	150 F Steam			Remarks ^(*)	250 F Water		Remarks ^(*)
	Exposure Time, days	Weight Gain, mg per dm ²			Exposure Time, days	Weight Gain, mg per dm ²	
<u>Nickel</u> (continued)							
M	204	117		M	532	110	M
M	206	108		M	532	115	M
M	206	139		M	532	112	M
M	206	131		M	448	109	M
M	206	141		M	532	110	M
M	201	126		M	532	115	M
M	206	133		M	532	117	M
<u>and Nickel</u>							
M	136	148		M	430	154	M
M	136	105		M	560	100	M
M	136	107		M	504	113	M
M	136	104		M	560	100	M
M	130	107		M	560	103	M
M	130	107		M	560	121	M

TABLE G-2. CORROSION OF ZIRCONIUM-BASE ALLOYS MELTED

Heat	Alloy Content					700 F Steam			600 F Water	
	Sn, w/o	Fe, w/o	Ni, w/o	N, ppm	Al, ppm	Exposure Time, days	Weight Gain, mg per dm ²	Remarks(*)	Exposure Time, days	Weight Gain, mg per dm ²
						Vacuum Annealed 2 Hr at 775 C and Furnace Cooled				
986	0.24	0.25	--	44	58	200	103	M, WP	518	116
974	0.25	0.25	--	48	63	200	102	M, WP	406	178
997	0.26	0.25	--	33	46	186	98	M, WP	434	105
984	0.26	0.25	--	47	68	212	109	M, WP	406	106
1001	0.26	0.27	--	55	62	181	145	M, WP	378	21
1003	0.48	0.41	--	219	43	135	120	M, WP	294	125
975	0.49	0.36	--	57	45	186	115	M, WP	434	102
1000	0.49	0.38	--	43	50	186	130	M, WP	434	109
983	0.50	0.39	--	53	70	194	118	M, WP	406	102
998	0.51	0.38	--	48	47	186	111	M, WP	434	106
990	0.48	0.25	0.23	47	56	186	111	M, WP	462	105
985	0.49	0.20	--	47	64	194	109	M, WP	462	101
1007	0.50	0.21	0.19	60	51	135	110	M, WP	294	160
999	0.53	0.20	0.18	64	46	186	123	M, WP	406	101
995	0.65	0.22	0.19	38	57	186	116	M, WP	434	103
1024	0.25	0.25	--	63	68	181	115	M, WP	378	109
1026	0.25	0.25	--	63	67	186	121	M, WP	406	106
1030	0.50	0.44	--	68	47	186	123	M, WP	618	93
1028	0.49	0.28	--	71	38	181	116	M, WP	406	100
1014	0.25	0.25	--	60	145	147	112	M, WP	322	116
1012	0.25	0.25	--	50	179	147	107	M, WP	322	Lost weight
1013	0.26	0.26	--	54	275	135	170	M, WP, S	182	118
1010	0.51	0.39	--	46	58	135	105	M, WP	266	110
1009	0.48	0.38	--	57	148	147	118	M, WP	294	170
1011	0.50	0.39	--	62	275	135	123	M, WP	266	97
1025	0.50	0.21	0.19	65	62	186	114	M, WP	378	97
1023	0.53	0.22	0.20	63	60	181	119	M, WP	378	104
1017	0.49	0.21	0.19	53	135	181	137	M, WP	378	121
1016	0.51	0.21	0.18	48	154	135	107	M, WP	322	114
1015	0.50	0.21	0.19	51	265	147	148	M, WP	266	125

(*) Key to remarks: M, WP - milky with a white oxide pattern
 W - white oxide
 F - flaking (heavy oxide)
 S - segregated.

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AT THE BUREAU OF MINES FOR ZIRCALOY 3 EVALUATION

Remarks(*)	150 F Steam		Remarks(*)	680 F Water		Remarks(*)
	Exposure Time, days	Weight Gain, mg per dm ²		Exposure Time, days	Weight Gain, mg per dm ²	
	Heated 2 Hr at 900 C in V. cum and Water Quenched					
M, WP	170	118	M, WP	504	76	M, WP
M, WP	170	106	M, WP	504	81	M, WP
M, WP	182	111	M, WP	504	77	M, WP
M, WP	170	102	M, WP	504	83	M, WP
M, WP, F	182	111	M, WP	504	88	M, WP
M, WP	135	136	M, WP, S	448	100	M, WP
M, WP	170	110	M, WP	504	81	M, WP
M, WP	170	135	M, WP	504	96	M, WP
M, WP	170	107	M, WP	504	85	M, WP
M, WP	170	132	M, WP	504	81	M, WP
M, WP	170	111	M, WP	504	94	M, WP
M, WP	170	107	M, WP	504	90	M, WP
M, WP	135	113	M, WP	364	106	M, WP
M, WP	170	134	M, WP	504	109	M, WP
M, WP	170	106	M, WP	504	91	M, WP
M, WP	158	134	M, WP, S	504	115	M, WP
M, WP	170	127	M, WP, S	504	91	M, WP
M, WP	158	127	M, WP	504	103	M, WP
M, WP	158	128	M, WP	504	103	M, WP
M, WP	158	128	M, WP	476	100	M, WP
W, F	123	112	M, WP	448	105	M, WP
M, WP	105	108	M, WP	364	122	M, WP
M, WP	105	107	M, WP	448	118	M, WP
M, WP	105	112	M, WP	448	105	M, WP
M, WP	93	115	M, WP	364	121	M, WP
M, WP	158	132	M, WP	504	106	M, WP
M, WP	158	142	M, WP	448	100	M, WP
M, WP	105	105	M, WP	448	105	M, WP
M, WP	93	98	M, WP	420	121	M, WP
M, WP	93	108	M, WP	364	141	M, WP

TABLE G-3. CORROSION OF SPONGE-BASE ZIRCONIUM ALLOYS IN 750 F STEAM AND 680 F DEGASSED WATER

Alloys Vacuum Annealed 1 Hr at 775 C and Furnace Cooled

Heat	Alloy Composition (Balance Zirconium), w/o	750 F Steam			680 F Water		
		Exposure Time, days	Weight Gain, mg per dm ²	Remarks ^(a)	Exposure Time, days	Weight Gain, mg per dm ²	Remarks ^(a)
556	0, 05Fe-0, 002Ni-0, 007N	105	Lost weight	W, F	333	105	M
555	0, 05Fe-0, 002Ni-0, 007N	105	Lost weight	W, F	305	193	W, F
557	0, 05Fe-0, 15Ni-0, 007N	207	99	M	417	88	LM
558	0, 05Fe-0, 15Ni-0, 015N	172	111	M	305	107	M
559	0, 05Fe-0, 15Ni-0, 0225N	159	114	M	277	114	M
560	0, 05Fe-0, 15Ni-0, 030N	118	137	M	221	140	M
563	0, 05Fe-0, 15Ni-0, 007N-0, 55Sn	166	104	M	417	94	M
564	0, 05Fe-0, 15Ni-0, 015N-0, 55Sn	159	103	M	361	103	M
565	0, 05Fe-0, 15Ni-0, 0225N-0, 55Sn	159	113	M	305	103	M
566	0, 05Fe-0, 15Ni-0, 030N-0, 55Sn	136	119	M	305	130	M
567	0, 25Fe-0, 25Sn-0, 007N	207	89	M	417	74	G
568	0, 25Fe-0, 25Sn-0, 015N	172	109	M	417	109	M
569	0, 25Fe-0, 25Sn-0, 0225N	172	127	M	305	101	M
570	0, 25Fe-0, 25Sn-0, 030N	159	114	M	277	115	M
573	0, 40Fe-0, 50Sn-0, 007N	172	110	M	417	88	M
574	0, 40Fe-0, 50Sn-0, 015N	172	119	M	417	90	M
575	0, 40Fe-0, 50Sn-0, 0225N	136	104	M	361	105	M
576	0, 40Fe-0, 50Sn-0, 030N	124	116	M	305	109	M
579	0, 20Fe-0, 20Ni-0, 50Sn-0, 007N	159	130	M	417	96	M
580	0, 20Fe-0, 20Ni-0, 50Sn-0, 015N	159	132	M	417	115	M
581	0, 20Fe-0, 20Ni-0, 50Sn-0, 0225N	118	120	M	305	119	M
582	0, 20Fe-0, 20Ni-0, 50Sn-0, 030N	118	108	M	305	104	M
581	0, 05Fe-0, 15Ni-0, 007N-0, 02Al	159	132	M	221	126	M
571	0, 25Fe-0, 25Sn-0, 007N-0, 02Al	136	112	M	249	121	M
577	0, 40Fe-0, 50Sn-0, 007N-0, 02Al	124	114	M	305	119	M
583	0, 20Fe-0, 20Ni-0, 50Sn-0, 007N-0, 02Al	136	112	M	417	113	M
582	0, 05Fe-0, 15Ni-0, 007N-0, 01Ti	159	105	M	305	115	M
572	0, 25Fe-0, 25Sn-0, 007N-0, 01Ti	118	115	M	277	112	M
578	0, 40Fe-0, 50Sn-0, 007N-0, 01Ti	159	119	M	417	87	M
584	0, 20Fe-0, 20Ni-0, 50Sn-0, 007N-0, 01Ti	136	108	M	417	102	M

(a) Key to remarks: G - black tarnish film (good material)
M - milky
W - white oxide
F - flaking (heavy oxide)
LM - light milky.

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- (2) Sponge-base alloys containing 0.2 to 0.5 w/o tin plus 0.03 to 0.5 w/o nickel are more resistant to 750 F steam than are alloys containing similar combinations of tin and iron. The reverse, however, is true for 680 F water exposure.
- (3) As the nitrogen content is increased from 70 to 300 ppm, there is a progressive increase in the corrosion rate of 0.25 to 0.5 w/o tin-0.25 to 0.50 w/o iron alloys.
- (4) The modified Zircaloy 2 alloys show a decrease in corrosion resistance with aluminum contents of 200 ppm.

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H. REACTOR-MATERIALS DEVELOPMENT

F. A. Rough

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

Enameling of uranium is being continued to eliminate the crazing generally obtained thus far.

Numerous data are reported on air-oxidation tests of niobium in dry air and also reported is the progress in preparation of additional niobium alloys for evaluation in oxidizing conditions.

Preliminary data indicate that the gamma eutectoid in uranium-rich uranium-titanium alloys is near 3 a/o titanium.

In studies of vapor deposition of molybdenum, a new apparatus for the deposition of molybdenum on the inside of small-bore stainless steel tubes is being constructed.

Enameling of Special Metals

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

The possibility of enameling to increase the usefulness of metals of interest to the AEC is being investigated. In the development of enamel coatings for uranium, the major problem to date has been the elimination of enamel crazing during the cooling period of the firing cycle. Previous work has reduced the degree of crazing, but this defect has never been entirely eliminated.

In a continuation of last month's work, the elimination of crazing was sought by compositional variation of a phosphate enamel containing lead oxide, iron oxide, and sodium oxide. These compositions maintained the calculated P_2O_5 content at 55 mole per cent while varying the other components. None of the enamels was totally free of crazing.

Next month, a new technique of enamel application will be tried in an attempt to achieve craze-free coatings using enamel compositions already developed. First, a thin enamel coating will be fired on the uranium. Then, a thin metallic layer will be deposited on the enamel by vacuum metallizing methods. Finally, a cover coat of enamel will be fired on the metallic layer. It is anticipated that the ductile intermediate-metallic layer might decidedly reduce or eliminate crazing by relieving localized stresses in the coating.

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Oxidation-Resistant Niobium Alloys

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

This program is an investigation of the oxidation and contamination behavior of niobium and niobium alloys. Present work is concerned with continuous-weighting air-oxidation tests on pure niobium; niobium alloys are being fabricated into strip for similar tests.

Continuous-weighting oxidation tests in dry air have been conducted on eight samples of pure niobium. The weight-gain data, which do not follow a parabolic or linear law, were reported for previous tests as constants for the general oxidation equation

$$W = Kt^n .$$

However, additional tests indicate that oxidation proceeds almost linearly after various incubation periods. Therefore, the data obtained for the latest tests are reported as linear rates (given in Table H-1).

Two duplicate samples of 20-mil annealed niobium sheet, 0-25 and 0-26, were oxidized at 800 C under identical conditions. Appreciable differences in both weight gains and linear rates were observed between the two samples, suggesting that considerable scatter exists in oxidation data at 800 C. One oxidized sample, 0-26, was vacuum heated 10 hr at 1000 C to investigate the stability of the oxide. This treatment blackened the scale and caused severe embrittlement of the metal core by intergranular oxide. The scale is being examined by X-ray diffraction.

Six samples of 95-mil niobium sheet, 0-27 through 0-32, were oxidized in dry air to determine if difference in oxidation behavior could be detected between wrought and annealed niobium. At 600 C, the wrought sample oxidized faster than the annealed sample after 160 min. At 800 C, the wrought sample consistently oxidized about 15 per cent faster than the annealed samples, while at 1000 C, the wrought sample oxidized about 15 per cent slower than the annealed sample. With the exception of that at 600 C, the differences in behavior between wrought and annealed samples can be explained by data scatter. For example, the 1-hr weight gains for annealed samples in dry air range from 2.35 to 2.80 at 600 C (six samples), 18.3 to 37.0 at 800 C (eight samples), and 30.3 to 45.5 mg per cm² at 1000 C (four samples).

Depth of contamination has been measured on a sample of pure niobium previously reacted with pure dry oxygen at 500 C for 150 min. The depth for a hardness increase of 50 Knoop points was 0.0049 cm. The diffusivity of oxygen in niobium at 500 C was calculated from the hardness traverse to be $10.13 \pm 3.07 \times 10^{-10}$ cm² per sec, slightly higher than the value of 5.68×10^{-10} cm² per sec obtained by extrapolation of higher temperature diffusivities (BMI-1170).

Melting and fabrication of niobium alloys into strip for oxidation testing is continuing on niobium-5 and 25 a/o vanadium and niobium-2.5, 5, 7.5, 10, and 25 a/o molybdenum alloys. As shown in Table H-2, niobium-2.5 a/o molybdenum and niobium-5 a/o vanadium were easily cold fabricable into 50-mil strip; but niobium-5,

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TABLE H-1. AIR-OXIDATION RATES FOR PURE NIOBIUM IN DRY AIR

Sample	Condition	Oxidation Temperature, C	Duration of Test, min	Time to Become Linear, min	Linear Rate, mg/(cm ²)(hr)	Weight Gain, mg per cm ² , for Indicated Exposure					
						1 Hr	2 Hr	3 Hr	4 Hr	5 Hr	6 Hr
0-25	Annealed ^(a)	800	120	6	28.3	36.9	61.0	--	--	--	--
0-26	Annealed	800	120	55	18.3	30.9	49.1	--	--	--	--
0-31	Annealed	600	360	130	1.76	2.8	4.4	6.0	7.8	9.5	11.5
0-32	Wrought ^(b)	600	295	20	1.71 ^(c)	2.8	4.5	6.6	9.1	12.4 ^(d)	--
0-29	Annealed	800	360	45	12.1 ^(e)	17.4	30.2	43.9	60.0	77.0	94.2
0-28	Wrought	800	264	110	15.2	24.7	39.3	54.6	70.1	--	--
0-27	Annealed	1000	117	45	30.8	40.1	71.0 ^(d)	--	--	--	--
0-30	Wrought	1000	360	5	30.0 ^(f)	30.0	61.3	89.8	114.2	136.9	158.6

(a) Annealed 1 hr at 1500 C.

(b) Cold rolled from 0.615 in. in diameter to 0.099-in. strip, 84 per cent reduction.

(c) Rate began increasing after 160 min; during final 20 min of test, rate was 3.66 mg/(cm²)(hr).

(d) Extrapolated value.

(e) Rate was 12.1 mg/(cm²)(hr) from 45 to 150 min, and 17.0 mg/(cm²)(hr) from 210 to 360 min.

(f) Rate was 30.0 mg/(cm²)(hr) from 5 to 160 min, and 22.1 mg/(cm²)(hr) from 210 to 360 min.

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TABLE H-2. COLD FABRICATION OF NIOBIUM-BASE ALLOYS

Alloy Composition, a/o	Cast Hardness, VHN	Initial Reduction, per cent	Results	Annealing Temperature, C	Final Reduction, per cent	Results
Nb-5V	152	71.9	Good	1500	50.0	Good
Nb-2.5Mo	128	75.2	Good	1500	50.0	Good
Nb-5Mo	154	24.1	Cracked			
Nb-7.5Mo	175	24.2	Cracked			
Nb-10Mo	199	7.8	Cracked			
Nb-10V	196	22.5	Good	1500		Good
Nb-12.5V	209	23.3	Good	1500 See		Fair
Nb-5Mo	154	22.7	Good	1500 text		Good
Nb-7.5Mo	175	23.0	Good	1500		Fair
Nb-10Mo	199	14.0	Cracked			
Nb-12r	176	86.4	Good, VHN 227			
Nb-5Zr	167	13.3	Cracked			
Nb-10Zr	209	12.5	Cracked			
Nb-25Zr	272	11.0	Cracked			
Nb-35Zr	289	39.4	Cracked			
Nb-1W	127	83.3	Good, VHN 164			
Nb-5W	165	79.4	Good, VHN 229			
Nb-10W	196	34.5	Cracked			
Nb-25W	299	18.9	Cracked			

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7.5, and 10 a/o molybdenum were unfabricable. After remelting, these three ingots plus niobium-10 and 12.5 a/o vanadium were cold rolled in partially opened, grooved rolls which provided side restraint, annealed, and flat rolled into 100-mil strips with several intermediate anneals. However, niobium-10 a/o molybdenum was still unfabricable, and will be encapsulated and hot rolled with niobium-25 a/o vanadium and niobium-25 a/o molybdenum.

Exploratory fabrication tests were performed on small pieces of niobium-zirconium and niobium-tungsten alloys prepared previously. As seen in Table H-2, niobium-1 a/o zirconium and niobium-1 and 5 a/o tungsten were fabricable. The hardness data indicate that neither element increases the rate of work hardening up to the limit of cold fabricability.

Plans for future work include:

- (1) Six-hr continuous-weighting tests on wrought and annealed niobium at 1200 C and on annealed niobium alloys at 1000 C in wet and dry air
- (2) Fabrication of niobium alloys containing chromium, molybdenum, tungsten, vanadium, and zirconium for oxidation testing
- (3) Investigation of the niobium-oxygen reaction at 400 and 550 C.

Uranium-Titanium Alloy Studies

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

The determination of the phase equilibria of the uranium-rich end of the uranium-titanium system is being conducted. The martensitic transformation in this end of the system is being studied in conjunction with its effect on the mechanical properties of uranium-titanium alloys.

The hot hardness and the length change of samples as a function of temperature are being used to determine the phase boundaries. These methods are being supplemented by metallographic examination of alloys equilibrated at various temperatures and quenched to room temperature. Equilibrations have been performed at 680, 700, 730, 750, and 770 C. Alloys which were either wholly or partially gamma at temperature show a martensitic structure in the quenched samples. No martensite has been observed in 0.6 or 1.1 a/o titanium alloys quenched from temperatures up to 750 C. The beta/beta plus gamma transus, as determined by inflections in the hardness-temperature curves is slightly above 750 C for the 1.1 a/o alloy. A 2.1 a/o alloy quenched from 730 C consisted of some martensite and transformed beta. On the basis of work to date, it appears that the eutectoid composition is close to 3 a/o and not 4 a/o as reported by Knapton. Dilatometric work is still in progress for alloys containing from 3 to 6 a/o titanium.

Alloys containing up to 6 a/o titanium have been solution treated 1 hr at 800 C and water quenched. Two samples of each alloy will be tested in tension to determine the

mechanical properties. Additional samples have been tempered for 1 hr at 300, 400, and 500 C from which mechanical properties will be determined.

Vapor Deposition of Molybdenum Coatings
on Stainless Steel Tubing

C. F. Powell and I. E. Campbell

Current research includes the development of processes for vapor depositing molybdenum on the inside and outside of small-bore Type 304 stainless steel tubes.

In prior work, adherent molybdenum coatings were applied to the inside and outside of 20-in. lengths of the small tubing by hydrogen reduction of MoCl_5 vapor at reduced pressure. Uniform coatings were obtained over the outside of these lengths of tubing, but only nonuniform coatings, covering only 2 to 3 in. of tubing, were obtained on the inside, in spite of using several different techniques for applying the internal coatings.

Work for the past month was concerned with the study of a new coating technique and the construction of a coating apparatus using this principle.

This technique is expected to be particularly suited to the application of coatings to the inside of long lengths of small-diameter tubing. It consists of passing one of the constituents of the coating atmosphere, such as a mixture of argon and MoCl_5 vapor, through the uniformly heated tube for a brief interval. The other constituent of the coating atmosphere, the hydrogen, is then passed through the tube in the reverse direction for a similar brief interval. These cycles are repeated several times per second. Metal deposition will occur only where the two bodies of gas mix. For gas flows within the laminar region, mixing should occur only by diffusion in a narrow zone between the two bodies of gas. Metal should be deposited uniformly throughout the tube as this diffusion zone oscillates from one end of the tube to the other. The study of the operation of this principle was made with liquid flow in small-diameter glass tubes at Reynolds numbers of 1 to 10 (the range generally used in previous deposition work). It showed that little or no mixing of the two separate fluid bodies occurred so long as the zone of separation was kept within the small-bore tubing, and that this zone of separation developed the theoretical shape as it moved through the tube. Estimates of the diffusion rates existing in the gaseous systems involved, made from the results of past deposition work, showed that high deposition rates should be obtainable by this technique for gas flows within the laminar region (Reynolds numbers below 2000).

A coating apparatus using this principle of operation has been constructed but has not been tested because of breakage of part of the vacuum-pumping system. Tests of the moving-temperature-gradient coater, mentioned in BMI-1173, were held up for the same reason.

Tests of the two pieces of coating equipment mentioned above will be made during the coming month.

I-1

I. PHYSICAL METALLURGY

F. A. Rough

Various research programs sponsored by the AEC Research Division are reported in this section.

In the study of uranium compounds, the fabrication studies of uranium silicides and of UAl_2 are completed. Thermal-expansion coefficients are reported for USi , USi_2 , USi_3 , and UAl_2 specimens up to 950 C.

Further data on the effects of varying grain size and strain rate on the tensile transition in alpha uranium are discussed.

Interpretation of a portion of the ternary system uranium-molybdenum-zirconium has been made. Further data are being obtained. Studies this month of the uranium-carbon system are confined largely to the alteration of the high-temperature X-ray diffraction camera.

At present, equipment is being set up to measure contact areas continuously in bonding tests involving single-asperity bonds.

Additional data are reported for the niobium-hydrogen equilibrium. Solubilities of hydrogen in niobium appear to be a maximum of 0.1 to 0.15 (hydrogen-to-niobium atomic ratio).

PROPERTIES OF URANIUM COMPOUNDS

The fundamental properties of some refractory binary compounds of uranium, other than oxides, are being investigated to provide bases for their consideration as nuclear fuels.

Preparation, Fabrication, and Physical Properties

W. D. McKee and M. J. Snyder

In nonextensive fabrication experiments, sintered-powder compacts as noted next have been prepared.

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Compound	Sintering Temperature, F	Bulk Density	
		G Per Cm ³	Per Cent of Theoretical
U ₃ Si ₂	2820 (1550 C)	12.0	98.0
USi	2860 (1620 C)	9.8	94.0
USi ₂	2840 (1560 C)	9.0	97.5
USi ₃	2730 (1500 C)	7.6	93.0
UAl ₂	2460 (1350 C)	7.15	88.0

Ceramic-fabrication investigations on these compounds are now believed sufficiently thorough for the purposes of this program, and future effort on these ceramics will be restricted to property investigations.

Thermal-expansion coefficients, measured in a vertical quartz-tube dilatometer under a pressure of 5×10^{-5} mm of mercury are given in Table I-1.

TABLE I-1. MEAN LINEAR EXPANSION COEFFICIENT

Compound Temperature Range, C	Linear Thermal Expansion Coefficient, 10^{-6} per deg C							
	USi		USi ₂		USi ₃		UAl ₂	
	Heating	Cooling	Heating	Cooling	Heating	Cooling	Heating	Cooling
20-100	20.0	18.8	15.9	15.1	15.5	15.5	14.9	15.2
20-200	18.8	18.1	15.6	14.9	15.3	15.6	14.8	15.2
20-300	17.8	18.2	15.5	15.8	15.4	16.3	14.7	16.0
20-400	17.4	17.9	15.6	15.8	15.7	16.7	15.0	16.1
20-500	16.9	17.7	15.5	15.9	15.8	17.1	15.1	16.4
20-600	16.4	17.7	15.5	16.3	15.6	17.8	15.2	16.7
20-700	16.1	17.6	15.7	16.6	15.3	18.4	15.4	17.0
20-800	15.9	17.6	16.2	16.7	15.1	18.8	15.6	17.3
20-900	15.8	17.4	16.8	16.7	15.0	18.9	16.1	17.1
20-950	15.7	17.3	16.9	16.6	14.9	19.1	16.2	17.1

Thermal-diffusivity and electrical-conductivity measurements on these ceramics are in progress. Also, progress was made in preparing ceramics of UB₂, UB₄, and UAl₃. An intensive effort is being made to obtain outside sources of the uranium compounds of interest to expedite the collection of data on these compounds.

Reaction With Gases

W. M. Albrecht and M. W. Mallett

A study is being made of the reactions of certain uranium compounds with several of the common gases. Oxygen was reacted with irregular polyhedral specimens of U₃Si₂, about 3/16 in. across and roughly equiaxed. U₃Si₂ at 400 C and 1 atm of

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pressure burned nonisothermally. At 300 C, the reaction followed an approximately parabolic law. Additional rate data are being obtained between 360 and 400 C to determine where nonisothermal burning of the material begins. Data also will be obtained on the reaction of oxygen and nitrogen with USi , USi_2 , and USi_3 .

Heat-of-Formation Measurements

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

As previously described, the heats of formation of various refractory uranium compounds are to be determined. Thus far in the program, emphasis has been placed on determining this quantity for USi_2 by comparing its heat of combustion with the heat of combustion of uranium and silicon. The oxygen-bomb and the sodium peroxide fusion techniques are both being investigated with the hope that at least one will prove successful. The effort during the past month has been concentrated on the peroxide fusion method.

The fusion bomb was modified so that the fusion cup now is surrounded by air during the fusion. During fusion, the temperature in the cup rises to above 350 C. After about 2 min, the temperature falls, drawing water into contact with the outside of the fusion cup, thus hastening temperature equilibrium. Pulverized samples of USi_2 and silicon reacted well with the peroxide fusion mixture in the presence of a combustion aid such as sulfur or carbon. The oxidation was not complete, however, probably because a small amount of sample was thrown against the cold wall and top of the fusion cup. Methods of recovering the unreacted sample are being investigated.

Uranium chips did not react satisfactorily with the peroxide fusion mixture in the experiments made to date. However, it seems likely that an adequate technique can be established using UO_2 powder as the reacting material. This approach will be investigated in the future.

A platinum sample holder, electrodes and fuse wire have been obtained and installed in the oxygen bomb. Emphasis during the coming month will be on finding a suitable combustion aid for the oxygen-bomb combustions and toward characterizing the products of the reactions.

INVESTIGATION OF THE TENSILE TRANSITION
IN ALPHA URANIUM

L. L. Marsh and G. K. Manning

A study of the mechanical properties and the metallography of plastic deformation in uranium is being conducted over the range of temperatures defining the tensile transition. Other variables being studied include grain size and strain rate; the objective is to determine the effect of grain size and strain rate on deformation behavior.

Evidence based on an incomplete evaluation of data suggests that the tensile-transition curve is displaced in temperature approximately 100 F by an increase in grain size from ASTM No. (3) to ASTM No. (-2). Similarly, increasing the strain rate from approximately 0.01 to 1.0 per sec displaces the transition phenomenon up in temperature by approximately 50 F. However, the elongation at fracture-versus-temperature curves for the two quoted strain rates are not homologous; marked deviation occurs in the fast strain-rate series at higher temperatures. Thus, there seems to be some relaxation process associated with deformation which is being affected by the strain-rate increase. The nature of this change in relaxation behavior is not known at present. Perhaps as more evidence is obtained on the metallography of plastic deformation in the temperature range above 150 F, the reason will become clear.

CONSTITUTION OF URANIUM ALLOYS

Uranium-Base Ternary Alloys

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

An investigation of delta-phase relationships in the uranium-zirconium-molybdenum system is proceeding. The studies are being carried out on specimens with compositions that lie on a line joining the delta phases of the uranium-zirconium and the uranium-molybdenum systems (uranium-31.5 at/o molybdenum and uranium-74 at/o zirconium). Additional specimens with compositions near the ends of the line joining the delta phases were prepared to study the extent of the terminal solid solutions.

The specimens were arc-cast of biscuit uranium, high-purity molybdenum sheet, and crystal-bar zirconium. Thermal-analysis specimens and metallographic specimens were prepared and heat treated at appropriate temperatures. Phase relationships are being established on the basis of differential thermal analysis, metallography, and X-ray diffraction.

A tentative ternary section in Figure I-1 is presented on the basis of data thus far obtained. Information on the molybdenum-rich end of the diagram is incomplete, as is shown by the dashed lines. High-temperature X-ray-diffraction data are being obtained to further establish these phase relationships.

The temperature of the gamma-to-delta transformation of the uranium-zirconium system is lowered by the addition of molybdenum to a temperature of 454 C, this temperature being well established by thermal-analysis results. The gamma-to-delta transformation of the uranium-molybdenum system appears to be lowered by the addition of zirconium on the basis of X-ray diffraction data; however, this reduction is not supported by thermal data and additional studies on this region are planned. The isothermal reaction occurring between γ_1 , γ_2 , and $ZrMo_2$ and γ_1 , alpha uranium, and $ZrMo_2$ is established at 604 C, by thermal-analysis data. It is evident that $ZrMo_2$ is stable throughout most of the system. The $ZrMo_2$ is in equilibrium with alpha uranium below 604 C, thus preventing equilibrium between the delta phases.

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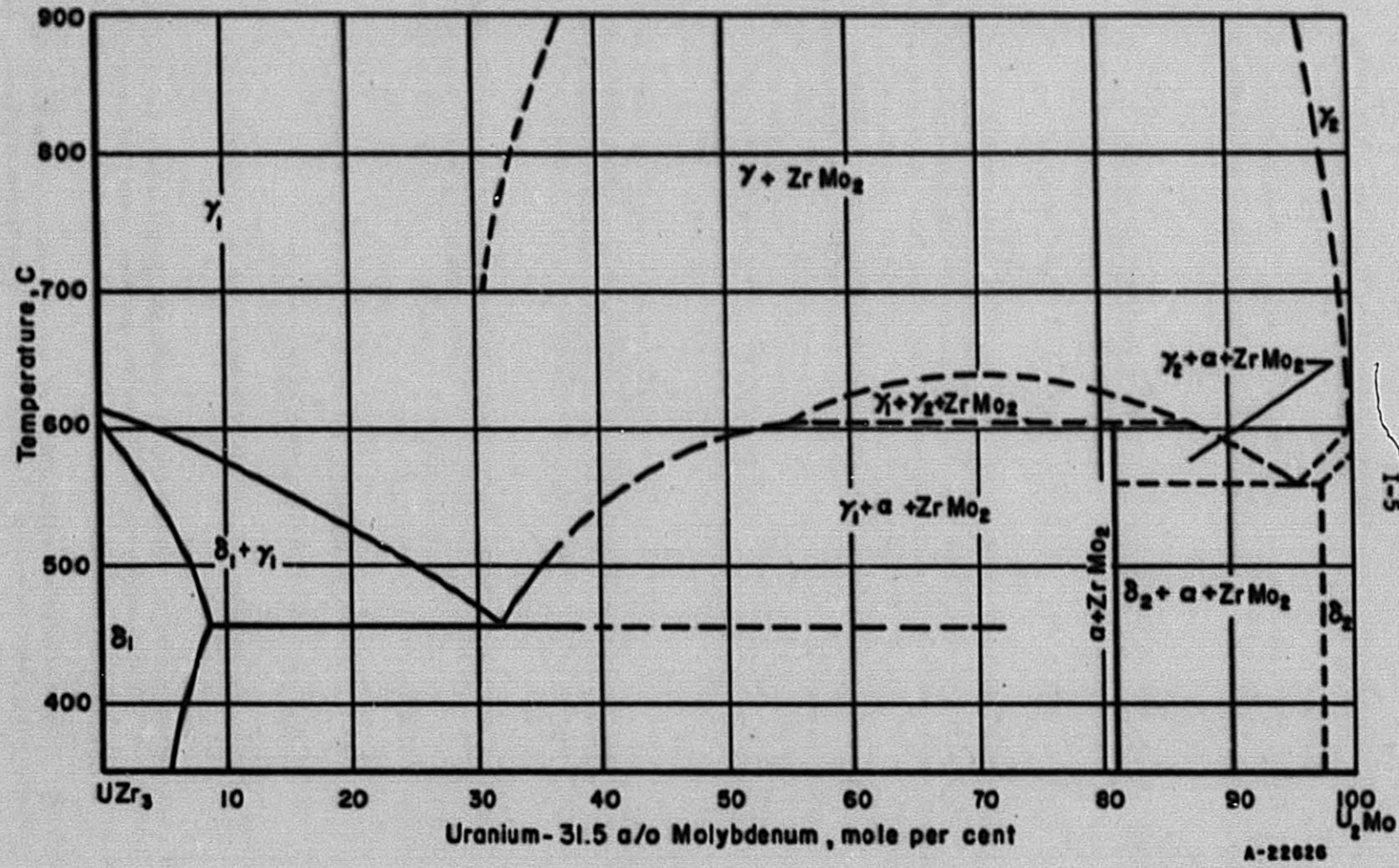


FIGURE I-1. TENTATIVE DIAGRAM SHOWING PHASE RELATIONSHIPS BETWEEN INTERMEDIATE DELTA PHASES OF THE URANIUM-ZIRCONIUM AND URANIUM-MOLYBDENUM SYSTEMS

Investigation of terminal-solid-solubility specimens is progressing by X-ray diffraction studies.

Investigation of the uranium-zirconium-silicon system is proceeding with a metallographic examination of specimens whose compositions lie in the uranium-rich corner of the ternary system.

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

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

Experimental work on the uranium-carbon system was somewhat curtailed during March to permit minor alterations of the high-temperature diffraction camera. Tungsten heating elements were substituted for tantalum, and work was started to achieve the highest possible temperatures from the instrument. Several modifications were found necessary. At a 1200-w input, temperatures near 2100 C were achieved when a vacuum failure occurred. This failure was due to radiation melting of the seals around the beryllium windows. Additional baffling was obviously needed; therefore, 0.5-mil aluminum foil was placed in front of the beryllium windows. A successful exposure was obtained on UC_2 at 2000 C with a 750-w input at a vacuum of 4×10^{-6} mm of mercury. However, vaporization of the aluminum baffle made it necessary to substitute a metal of lower vapor pressure. Nickel foil, 0.5 mil thick, is being tried.

The beryllium windows themselves were resealed with intimate contact to the water-cooled brass support, in order to keep the windows cool. While initial difficulties were experienced in obtaining a suitable vacuum, it now appears that the instrument may be modified satisfactorily to operate in excess of 2200 C in the 10^{-5} mm of mercury range.

The improvement offered by the use of tungsten as a heating element will permit direct examination of the uranium-carbon system at higher temperatures than previously attained. Work will continue to define the immiscibility region of UC + UC_2 and to study phase relationships at elevated temperatures.

STUDY OF BONDING FUNDAMENTALS

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

This research program is being conducted to study the solid-state bonding process and to determine the effects of experimental conditions and material properties on the formation and growth of bonded areas.

The experimental problem involves the study of microscopic-scale bond formation at the single-asperity junction between a needle point and a plane surface. To

I-7

date, the breaking strength of the junction has been the only criterion available for study of bond formation and growth under various conditions. From strength values, bond-growth rates and such rate-controlling factors as time, temperature, and pressure were studied. Each individual test could, however, supply only a single item of information - the breaking strength of a junction under a single combination of experimental conditions. Recently, efforts have been made to devise a direct method for measurement of the contact area. Several techniques were considered, but the most promising one involves measurement of electrical properties of the bond area. Holm*, in a treatment of electrical-contact theory, relates contact area to measurable properties by the expression

$$\text{Contact area} = \pi \left(\frac{PI}{2E} \right)^2,$$

in which P = electrical resistivity of the contact material

I = current through the contact

E = potential drop across the contact.

This expression may require modification for this particular application.

The necessary electrical circuit and measuring instruments have been set up to provide a continuous record of contact potential and current from which a similar record of contact area can be derived. Heretofore, it was impossible to follow the change in contact area for any individual test.

Several preliminary experiments have been conducted utilizing the potential-recording equipment. The graphical data reveal an increase of contact area with time. No definite conclusions can be made at this point. Future work will include more extensive use of the potential-measuring technique to follow bond formation and growth.

NIOBIUM-GAS REACTIONS

M. W. Mallett, W. M. Albrecht, and A. Levy

The kinetics and mechanisms of the reactions of niobium with hydrogen and nitrogen are being investigated. Currently work is being done on the niobium-hydrogen system. Investigation of the temperature-pressure-composition equilibrium diagram of the niobium-hydrogen system is being continued.

The work at 10 to 1000 mm of mercury has been extended to include temperatures from 100 to 900 C and compositions up to atomic ratios of hydrogen to niobium of 0.85. Based on the data obtained thus far, tentative phase boundaries have been defined. In these ranges the maximum solubility of hydrogen in the niobium phase is of the order of atomic ratios of 0.1 to 0.15. A hydride phase was found to exist at temperatures below

*Holm, R., Electric Contacts, Stockholm, H. Geber (1946).

500 C and high atomic ratios; for example, a hydride phase with an atomic ratio of about 0.5 formed at 400 C and a hydrogen pressure of about 120 mm of mercury. Additional equilibrium data are being obtained.

Work is being initiated to determine the rates of diffusion of hydrogen in niobium. Hydrogen concentration gradients have been prepared in niobium cylinders at 600 C. Chemical analyses of the gradients are incomplete.

J-1

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

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

Investigations of the corrosion problems associated with the recovery of spent reactor fuel elements have been continued during the month of March as part of a program of assistance to ORNL.

The Zircex process utilizes the differential volatility of the chlorides to separate zirconium from uranium. Current research is concerned with evaluation of materials to be used in construction of vessels in which Zircex processing may be performed. During March, elimination tests have continued; Haynes 21 and Vitallium exhibit comparable corrosion rates; Haynes 23 and 36 are not corrosion resistant.

The Darex process uses a dilute mixture of HCl and HNO₃ to dissolve stainless steel and uranium prior to separation. Materials for the construction of processing vessels are being studied. Zirconium has been found unacceptable as a dissolver-vessel material. Type 347 stainless steel shows good corrosion resistance to extraction solutions.

The Zircex Process

Hydrochlorinator - Dissolver Studies

Elimination Tests. Eleven materials have been exposed to boiling solutions of 5.0 M HNO₃, 0.4 M UO₂(NO₃)₂ containing 500 ppm chloride. Ten of these materials, tantalum, titanium, Vitallium, Haynes 25, Type S-816, Type S-590, Carpenter 20, Carpenter 20 stabilized, Type 304 ELC stainless steel, and Type 347 stainless steel, all appear to be satisfactorily resistant with corrosion rates of less than 1 mil per month after 6 weeks of exposure. The highest corrosion rates were exhibited by Haynes 25, with Type S-590 and Type S-816 showing rates nearly as high. It was necessary to eliminate Hastelloy C from this series of tests after a 329-hr exposure because of the high corrosion rates encountered with this metal. The exposure of the other materials will be continued for at least 8 weeks.

Specimens cut from cast Haynes 21, 23, and 36 were compared with wrought specimens of Vitallium, exposing both types of specimens to the 15.0 M HNO₃, 0.4 M UO₂(NO₃)₂ solutions through which HCl gas was bubbled. Haynes 23 and 36 specimens were eliminated after 1 week because of high corrosion rates. Both Vitallium and Haynes 21 specimens survived the first week without too severe an attack but had to be discontinued from test at the end of the second week because of the high corrosion rates which were occurring. In general, the rates measured for Haynes 21 specimens were somewhat higher than those for Vitallium. These same tests will be repeated using wrought specimens of the Haynes alloys.

A titanium specimen failed due to severe attack during one of the first series of elimination tests made in 15.0 M HNO_3 , 0.4 M $\text{UO}_2(\text{NO}_3)_2$, with HCl gas bubbled through. This specimen was exposed in the vapors of this solution and was not attacked until some time during the fourth week of exposure. In an attempt to repeat this phenomenon, three similar tests are under way. In addition, a fourth test is being made under similar conditions, except that a weaker acid solution (11.0 M HNO_3) is being used. Four weeks of exposure have already been completed with no evidence of the catastrophic type of attack. It has been observed that titanium specimens exposed to the return drip of the condensate are attacked to a greater extent than are specimens which are so positioned in the vapor that condensate return seldom contacts them. These tests will be continued until the failure occurs or until it is reasonably certain that continued exposure will not lead to failure.

A new set of elimination tests has been started preliminary to the cyclic experiment. In this set, specimens of Type S-816, Haynes 21, Carpenter 20, Type 304 ELC, and Type 316 are being exposed to aqueous solutions containing 0.4 M UCl_3 . These studies have just started and the results will be reported next month.

Cyclic Tests. Equipment is ready for exposing specimens of Type S-816, Haynes 21, Carpenter 20, Type 304 ELC, and Type 316 to cyclic conditions consisting of a hot cycle of about 14 hr at 600 C in HCl, hydrogen, and ZrCl_4 , followed by a short (2 hr) cycle in boiling solutions of from 0.0 to 15.0 M HNO_3 containing uranium and chloride additions. In addition to the specimens which will be made to undergo the exposure to the cyclic conditions, tensile specimens of the materials will be left in the furnace and evaluated at various time intervals after exposure to the hot cycle only.

Chloride-Stripper Studies

Both tantalum and titanium specimens are being exposed to boiling solutions which are considered to be somewhat representative of those which might occur during the stripping of chloride from the dissolver solution. The solutions are 9.0, 11.0, and 14.0 M HNO_3 containing 0.4 M $\text{UO}_2(\text{NO}_3)_2$ with HCl gas being bubbled through each one. After 5 weeks of exposure, there has been no attack on any of the tantalum specimens. There has been a slight amount of corrosion of the titanium specimens exposed in the 9.0 M HNO_3 solution. The corrosion rate for the specimen exposed to the vapor phase of this solution has risen from no measurable rate to 0.10 mil per month over a 4-week period, while the specimen exposed in the liquid phase shows a corrosion rate which started at 0.44 mil per month after a 1-week exposure and has dropped to 0.11 mil per month after 4 weeks.

The Darex Process

Dissolver Studies

Eight weeks of exposure have been completed for specimens of tantalum, titanium, and Type S-816 in both the beginning and midpoint dissolver concentrations of the Darex process. The data for the first 4 weeks of exposure in these solutions have already

J-3

been reported in Table N-3, BMI-1173 (page 60), and Table N-4, BMI-1161. The corrosion rates have either remained essentially constant or decreased very slightly during the additional 4 weeks.

Four weeks of exposure have been completed with specimens of tantalum, titanium, titanium alloy, zirconium, Zircaloy 2, and Type S-816 in the final dissolver concentration of the Darex process. The data for these tests are included in Table J-1. None of these materials is seriously attacked by these solutions.

Specimens of zirconium are being exposed to the beginning and midpoint solution concentrations, and wrought specimens of Haynes 21 are being exposed in all three of the Darex dissolver solutions.

Dissolution Studies

A dissolver has been assembled in which a Type 304 stainless steel rod is being dissolved in contact with titanium in a continuous flow of 2 M HCl-5 M HNO₃. Corrosion specimens of several materials are being exposed to the off gases from this dissolution both at the liquid line and high in the condenser. It has already been shown that zirconium is severely attacked by the off gases.

Another experiment is under way in which a dissolver is operated on a batchwise basis. The beginning acid concentration of 2 M HCl-5 M HNO₃ is used to dissolve Type 304 stainless steel rod until no more will dissolve in the 3, 5 liters of boiling acid. Corrosion specimens of several materials are also being exposed in this experiment. No results are available at the present time.

Galvanic Effects

Half-cell and open-cell potentials have been measured in 2 M HCl-5 M HNO₃ solutions at boiling for uranium, stainless steel, and titanium electrodes. The half-cell potentials of the uncoupled electrodes with respect to the saturated calomel electrode were: titanium, +0.50 v (noble); Type 304 stainless steel, -0.28 v (active); and uranium, -0.71 v (active). When coupled to Type 304 stainless steel, the titanium electrode polarized to +0.03 v; coupled to uranium, it polarized to -0.07 v. In either case, a gas, which was thought to be hydrogen, could be seen evolving from the titanium electrode. About one hundred times the amount of current flows when titanium and uranium electrodes are connected than flows when the connection is made between tantalum and uranium.

Two experiments have been assembled in which either a titanium or a tantalum electrode is brought by an external emf to the potential in boiling 2 M HCl-5 HNO₃ that they would have if uranium were dissolving in contact with them. The tantalum electrode has been exposed a little more than 5 weeks, the titanium more than 2. Small sections have been removed from the tantalum and titanium weekly. The tantalum is slowly increasing in hydrogen content, as shown by vacuum-fusion analysis. No evidence of hydride formation has been obtained as yet by metallographic examination. The titanium is covered with a bluish-gray scale. X-ray analysis of this has shown it

TABLE J-1. DAREX BOILING FINAL-DISSOLVER-SOLUTION^(a) TESTS

Specimen	Specimen Position	Corrosion Rate ^(b) , mils per month							
		166 Hr		327 Hr		500 Hr		661 Hr	
		A	B	A	B	A	B	A	B
Tantalum	Vapor	g ^(c)	g	0.00	g	g	g	g	g
	Interface	g	g	0.00	g	g	g	0.00	g
	Liquid	g	g	0.00	g	g	g	0.00	g
Titanium	Vapor	0.00	g	0.01	g	0.00	0.00	0.01	0.01
	Interface	g	g	0.00	0.00	g	g	0.00	0.00
	Liquid	0.02	g	0.01	g	0.01	g	0.01	g
Titanium alloy (6 w/o Al, 4 w/o V)	Vapor	0.01	0.04	0.01	0.08	0.02	0.06	0.04	0.08
	Interface	g	g	g	g	g	g	0.00	0.00
	Liquid	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Zirconium	Vapor	g	g	g	g	0.00	g	g	g
	Interface	g	0.01	0.00	0.00	0.00	0.00	0.00	0.00
	Liquid	0.01	0.01	0.00	0.00	0.00	0.00	0.00	0.00
Zircaloy 2	Vapor	g	0.00	g	g	0.00	0.01	0.00	0.00
	Interface	0.01	0.01	0.00	0.00	0.00	0.00	0.00	0.00
	Liquid	0.01	0.01	0.00	0.00	0.00	0.00	0.00	0.00
Type S-816	Vapor	0.08	0.21	0.07	0.15	0.06	0.12	0.05	0.09
	Interface	0.89	1.11	0.88	0.99	0.77	0.89	0.79	0.86
	Liquid	0.93	1.03	0.90	0.97	0.90	0.94	0.90	0.94

(a) These solutions are: 1.00 M H⁺, 1.78 M Cl⁻, 4.46 M NO₃⁻, 1.10 M Fe³⁺, 0.30 M Cr³⁺, 0.12 M Ni⁺⁺, 0.40 M U⁶⁺.
The specific gravity is about 1.38 at 25 C.

(b) Specimens A and B are duplicate specimens.

(c) g indicates a small weight gain of not more than 0.8 mg for tantalum, 0.6 mg for titanium, 15.5 mg for titanium alloy, 1.3 mg for zirconium, and 1.6 mg for Zircaloy 2.

J-5 and J-6

to be titanium dioxide. No hydrides have been found in the titanium either, although no vacuum-fusion analyses have been made with the titanium specimens.

Chloride-Stripper Studies

As in the Zircex process, specimens of tantalum and titanium are being exposed to solutions which are considered somewhat representative of some which might occur during the operation of stripping chloride from the dissolver product. In this case, the solutions are 7.0, 9.0, and 10.6 M HNO₃ containing uranium and the various ions, which would arise from the dissolution of stainless steel, in the form of nitrates. In addition, HCl gas is bubbled through these solutions while they are boiling. The tantalum specimens have been exposed for 5 weeks and the titanium specimens for 4 weeks. So far, there has been no evidence of corrosive attack of either material in any of the solutions.

Purex Acid Extraction Studies

Unstressed specimens of Type 347 stainless steel have been exposed to Purex acid solutions, both high and low in uranium and ions from the stainless steel content, as might arise from the Darex process. These solutions have been contaminated with up to 1.00 w/o chloride. After over 9 weeks of exposure, there has been only a slight weight loss, with no tendency toward stress corrosion as yet.

Specimens of Type 347 stainless steel, stressed to 90 per cent of their yield strength, and other specimens, bent around a mandrel until stressed beyond their yield strength, have been exposed to similar solutions for over 3 weeks. No evidence of stress corrosion has been observed to date.

K-1

**K. EVALUATION OF A REFLECTOR-CONTROLLED
HETEROGENEOUS BOILING REACTOR**

J. W. Chastain

This work is directed toward developing a system of control for an unmanned heterogeneous boiling reactor. This system of control is operated by power demand. A greater power demand causes a decrease in steam pressure above the reactor core. The lowered pressure permits water to flow from a surge tank into the annular reflector tank surrounding the reactor core. This reduces the neutron leakage and causes the reactor to operate at a higher stable power level.

The hydraulic mock-up was shut down during most of the month awaiting new inserts for the control valve.

The critical-assembly experiments for the cold-water case are complete and work has started on the hot-non-boiling condition.

Critical Experiment

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

The purpose of this experiment is the evaluation of the effectiveness of a reflector system of control and an assessment of the feasibility of this system. An evaluation of the long- and short-term transient behavior of the reactor system will be obtained by using the results from the critical assembly as input data in the reactor simulator.

The "cold" case, that is the case where the reflector and moderator were unvoided and the core had an aluminum-to-hydrogen atomic ratio of 0.358, was completed during March.

Expanded polyethylene having a density of 0.65 g per cm³ has been ordered for the radial reflector to simulate water at 417 F and 300 psi. This plastic is expected to arrive early in April. Meanwhile, experiments are being conducted with the radial reflector unvoided and the moderator and axial reflector voided to have nuclear properties equivalent to water at 417 F and 300 psi. No results for this case are available at the present time.

The results given in Tables K-1 and K-2 below are the final results of the cold case.

It should be noted in Tables K-1 and K-2 that the errors given for the critical mass are not probable errors but an estimated error. This error is due to the difficulty in maintaining a geometrically homogeneous loading while adjusting the final critical mass. Experience has shown this error to be of the magnitude of plus or minus 1 per cent.

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K-2

TABLE K-1. REACTIVITY AND CRITICAL MASS FOR POISONED^(a) CASE

Reflector Position From the Bottom of the Core, in.	Critical Mass, g	Reflector Worth, reactivity per in.
24	4060 ± 51	6.63 ± 0.34 × 10 ⁻⁴
18	5172 ± 52	25.46 ± 2.25 × 10 ⁻⁴
14	5272 ± 53	40.67 ± 1.51 × 10 ⁻⁴
12	5383 ± 54	39.31 ± 1.49 × 10 ⁻⁴
10	5465 ± 55	39.81 ± 0.95 × 10 ⁻⁴
6	5671 ± 57	26.97 ± 0.83 × 10 ⁻⁴
0	5964 ± 60	6.63 ± 0.44 × 10 ⁻⁴

(a) The poison consists of a 1/8-in. -thick sheet of polyethylene impregnated with 0.119 g per cm³ of boron (based on B₄C content), forming an annular ring between the inner and outer reflector.

TABLE K-2. REACTIVITY AND CRITICAL MASS FOR UNPOISONED CASE

Reflector Position From the Bottom of the Core, in.	Critical Mass, g	Reflector Worth, reactivity per in.
24	5062 ± 51	5.61 ± 0.14 × 10 ⁻⁴
0	5584 ± 56	8.98 ± 0.65 × 10 ⁻⁴

Figures K-1, K-2, and K-3 are graphical representations of the data given in Table K-2. Figure K-1 is a plot of the differential reflector worth versus reflector height. By performing a numerical integration on the area beneath this curve data were obtained which yielded the curve shown in Figure K-2 (integral reflector worth versus reflector height). The total worth of the reflector in the cold case was found to be 6.14 ± 0.29 per cent reactivity.

Figure K-3 is a graph of the critical mass versus reflector position.

Simulator Studies

B. B. Gordon and J. J. Stone

No work was done on the simulator studies during the month. The inserts for the control valves should arrive shortly and work will resume.

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K-3 and K-4

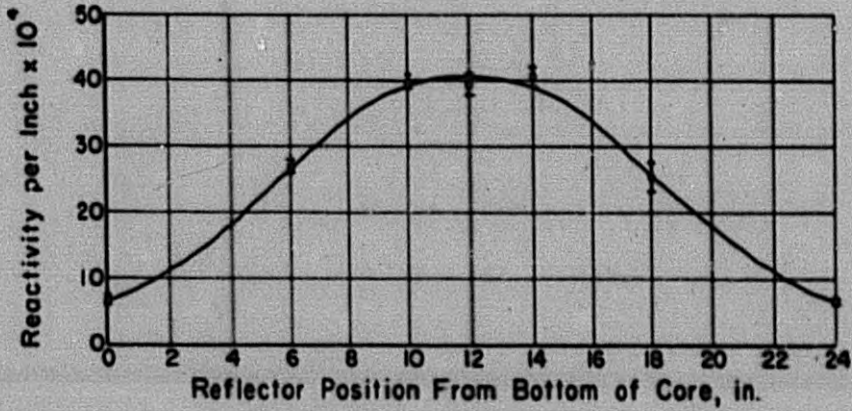


FIGURE K-1. DIFFERENTIAL REFLECTOR WORTH VERSUS REFLECTOR HEIGHT

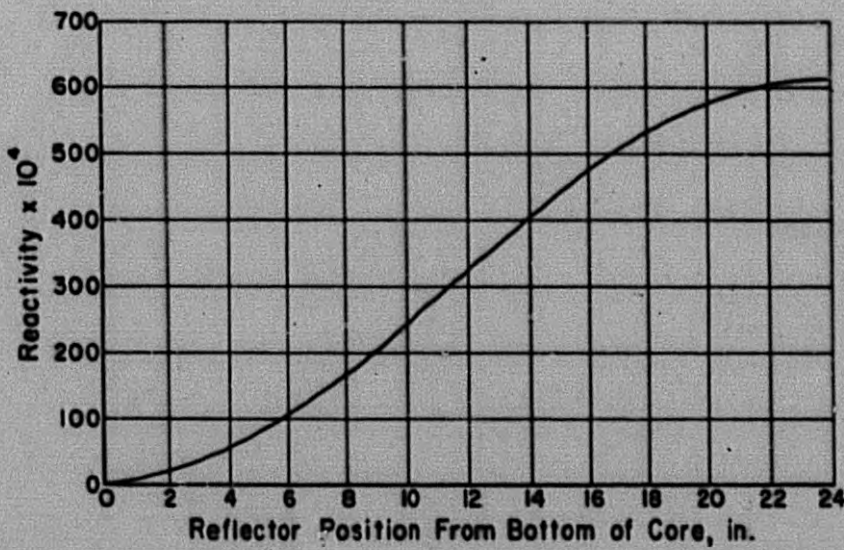


FIGURE K-2. INTEGRAL REFLECTOR WORTH VERSUS REFLECTOR HEIGHT

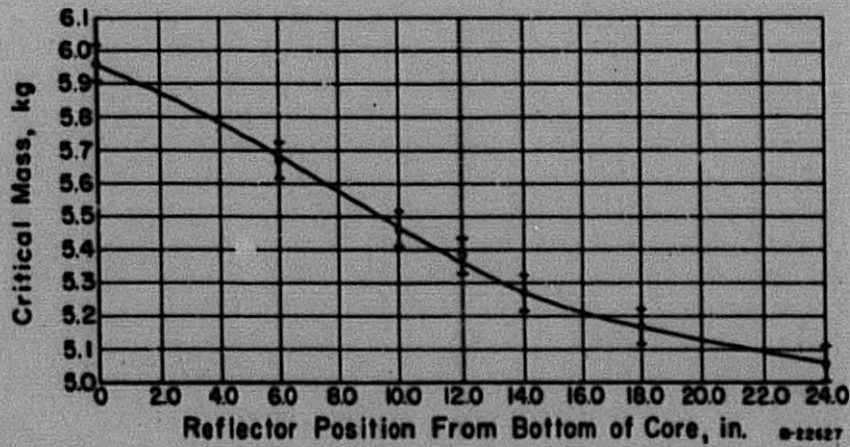


FIGURE K-3. CRITICAL MASS VERSUS REFLECTOR HEIGHT

L-1

L. STUDIES OF SODIUM-TANTALUM COMPATIBILITY AT
ELEVATED TEMPERATURES

J. H. Stang

Research is being conducted on several problems posed by the containment of high-temperature sodium by tantalum. This over-all program is complementing the effort under way at Los Alamos Scientific Laboratory to develop LAMPRE I, a fast reactor system.

During March, several modifications were incorporated into the first polythermal flow-loop system of the type in which tantalum specimens will be exposed to 1500 F peak-temperature sodium. It is expected that these changes will provide the reliability in the system necessary for long-duration runs. This system, along with three others, is scheduled to be placed in full operation during April.

Creep-rupture testing of tantalum has been delayed because of the necessity to repair leaks in the newly constructed special-furnace test rig. In the most informative experiment to date, a specimen of degassed sintered tantalum failed on loading with 20,000 psi at 1200 F. Several improvements in the vacuum-degassing apparatus were made and a 5-hr-long degassing exposure of several tantalum samples at a temperature level in the neighborhood of 4800 F was successful.

Welding studies made on reclaimed arc-cast tantalum sheet have shown the existence of weld porosity similar to that found in a majority of the welds made in sintered tantalum. However, some very recent work on virgin arc-cast material has been somewhat more encouraging. Investigations to determine the best distance of separation of small tantalum tubes welded to a thin tantalum header have shown that a separation of 1/16-in. gave good fillets in the weld joints.

The gettering crucible assembly, to be employed in the experiments to develop an analytical method for oxygen in sodium in the 0.0001 w/o range, was altered somewhat during March to permit the addition of an auxiliary fluid-charging system. The amalgamation apparatus was readied for service analyses of oxygen in sodium; recent runs with this apparatus have yielded reproducible results in the 100-ppm range.

Tantalum-Sodium Compatibility Studies

G. E. Raines, C. V. Weaver, and W. H. Goldthwaite

An appraisal of the behavior of tantalum in 1500 F peak-temperature sodium environments is the main objective of this study. This is to be accomplished by operating Type 316 stainless steel forced-convection flow loops with tantalum inserts in the hot legs. Each loop will be provided with special trapping features which will permit close control of the amount of oxygen in the sodium as a primary variable. Contemplated oxygen limits range from less than 10 ppm to 200 ppm.

L-2

During March, various experiments to obtain operational experience with the first loop system continued. The need for several revisions, in addition to those found during February and discussed in BMI-1173, became apparent. The principal difficulty encountered recently was failure of the main heat-input furnace. This unit has been redesigned to incorporate quartz infrared lamps which should provide both reliability and ease of maintenance. Final modifications were completed at the end of the month and it is now expected that formal experimental work with the revised rig can start in early April. Five more loops were assembled during March; at least three of these are scheduled to be in operation by April 30.

Although completely reliable operation of the first loop system has not been achieved as yet, its general performance thus far is encouraging. For example, in one run the charge of sodium was purified by cold trapping to less than 10 ppm oxygen, as indicated by plugging-indicator measurements. Following this, the charge was successfully circulated through the zirconium-foil-filled gettering trap for 22 hr at 1000 F and for 5 hr at 1200 F. This additional purification step is provided in the system to remove oxygen in the 0-10 ppm range. The actual efficiency of this operation cannot be determined at present. However, when the analytical technique for determining oxygen in sodium below 10 ppm is established (discussion of this phase of the work is included in the subsequent subsection entitled "Determination of Oxygen in Sodium"), it will be used to advantage in obtaining information such as this.

The High-Temperature Mechanical Properties
of Tantalum

J. C. Stewart, C. J. Slunder, J. G. Dunleavy,
and A. M. Hall

The effect of oxygen content, up to about 2000 ppm, on the creep and stress-rupture properties of tantalum in the temperature range 1000 to 2000 F is being studied. True evaluation of the data obtained will depend on the control of factors such as grain size and carbon and nitrogen content.

In March, emphasis was placed on the initial operation of the first inert-gas-blanketed stress-rupture furnace. Delays were experienced while system leaks were traced and repaired. In a preliminary test a degassed tantalum specimen (fabricated of sintered stock) failed quickly upon loading to 20,000 psi at 1200 F. The test will be repeated using annealed specimens.

Recent improvements in the induction degassing apparatus have made it possible to degas tantalum test bars at temperatures of 4500 to 4800 F. Eight bars have been degassed in this apparatus thus far; their hardness, after an exposure lasting 5 hr, was reduced from 169 to 90-100 VHN (10-kg load).

In the immediate future the remaining (total of four) creep-test units will be leak tested, and put into operating condition. Creep tests will be started at 1200 F using the annealed sintered tantalum now on hand. However, a batch of arc-cast material from a single lot of reclaimed powder has been ordered and is expected to be delivered to

L-3

Battelle in early April. This batch is to provide a standard material for the bulk of the stress-rupture experiments now planned.

Weldability of Tantalum for High-Temperature Systems

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

This program is directly concerned with the weldability of the small all-tantalum tube-to-header assembly which will be used as the core of LAMPRE I. Past welding studies have indicated the welds (made by the tungsten-electrode arc process) in 30-mil sintered tantalum sheet taken from one batch of stock have consistently been sound while welds in 40-mil sintered sheet from another batch have been porous. At present, further chemical analyses of these materials are being made to ferret out elements which might account for the marked differences in behavior. As discussed in BMI-1173, carbon, hydrogen, nitrogen, and oxygen analyses failed to shed light on the issue. During March, welding studies using the 30-mil sheet continued. Specimens taken from it were joined to thin-wall sintered tantalum tubes (3/16-in. ID by 0.015-in. wall) from a new batch of stock. Although, as before, welds in the sheet were sound, gross porosity was detected in the weld deposit joining this sheet to the tubing. This porosity was similar to that found in the tube-to header joints made with thin-walled sintered tubes from the batch of tubing first employed in this work.

Weldability studies were also made during March on two thicknesses of reclaimed arc-cast tantalum. Metallographic examinations of sections made through welds in this material indicated porosity similar to that encountered in the case of welds in 40-mil sintered sheet reported previously. In addition to this work with reclaimed arc-cast material, initial work started late in the month to investigate the weldability of 20- and 30-mil sheet from a recently procured supply of virgin arc-cast material. Studies thus far made with this sheet have indicated that sound porosity-free welds can be produced with it. Additional work is scheduled during April to check out these current findings.

A number of simulated tube-to-header assemblies were made with a welding fixture which was designed to aid in the welding of tantalum tubes to a tantalum header. These assemblies were made to determine the most desirable spacing of the tantalum tubes in the header to achieve complete weld penetration of the back side of the tube-to-header joints. Complete weld penetration would eliminate the sharp notch formed by the tube-and-header plate. The results of this study made on two thicknesses of header sheet (30-mil-thick tantalum and 19-mil-thick tantalum) and using three different tube separations (1/32 in., 3/64 in., and 1/16 in.) indicated that a 1/16-in. separation gave the best fillet welds with the least chance of weld burn-through.

Determination of Oxygen in Sodium

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A method based on the extraction of oxygen from sodium by a "getter" metal which in turn is analyzed by a vacuum-fusion procedure is being appraised as a technique for

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determining oxygen levels in sodium to a sensitivity of 0.0001 w/o. During March, the construction of a stainless steel apparatus which can deliver a metered amount of filtered sodium directly into the main crucible of the gettering assembly was completed. In this apparatus liquid sodium is forced into a calibrated volume (about 100 cm³) by gas pressure (prepurified nitrogen) and then forced into the gettering bomb. Compression-type tube fittings have been used in the construction to facilitate changing the metering volume should this become necessary. Necessary alterations were made to the bomb for the addition of the metering attachment. The leak tightness of the assembled system has now been established and gettering experiments will start shortly.

Several experiments were completed during the past month to determine operational techniques with the amalgamation apparatus. Oxygen contents of about 100 ppm were reproduced to ± 10 ppm. This apparatus appears to be ready for service analyses.

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