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PROGRESS RELATING TO CIVILIAN APPLICATIONS
DURING FEBRUARY, 1960

by

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REPORTS RELATING TO CIVILIAN APPLICATIONS
ISSUED DURING FEBRUARY, 1960

- BMI-1374 "Gas-Pressure Bonding of Zircaloy-Clad Flat-Plate Uranium Dioxide Fuel Elements", by Stan J. Paprocki, Edwin S. Hodge, Donald C. Carmichael, and Paul J. Gripshover.
- BMI-1397 "Model Studies of Flow and Mixing in the Partially Enriched Gas-Cooled Power Reactor", by Lawrence J. Flanigan, Gale R. Whitacre, and Herbert R. Hazard.
- BMI-1405 "Behavior of Rubbing Molybdenum Surfaces in Sodium Environments", by John W. Kissel, Carl W. Melton, and William A. Glaeser.
- BMI-1406 "Radiation Effects on Boron-Containing Compounds", by Donald J. Hamman and Paul Schall.
- BMI-1408 "A Method of Correlating Irradiation Effects in Dispersion Fuels", by Donald L. Keller, Lewis E. Hulbert, and Bruce W. Dunnington.
- BMI-1409 "Progress Relating to Civilian Applications During January, 1960", by Russell W. Dayton and Clyde R. Tipton, Jr.

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REACTOR MATERIALS AND COMPONENTS (AEC-DRD)

R. F. Dickerson

Structural studies of U_3O_8 - La_2O_3 solid solutions indicate that the fluorite solid solution is formed and that the lattice parameters are in general agreement with those previously obtained by an oxidation of UO_2 - La_2O_3 solid solutions of corresponding La_2O_3 content. Preliminary results from electrical studies indicate that the "valence-compensated", air-fired U_3O_8 - La_2O_3 solid solutions containing greater than equimolar amounts of La_2O_3 are poor conductors. A direct determination of the vapor pressure of uranium oxide is being undertaken for a series of UO_2 - La_2O_3 solid solutions.

Precise determinations of the oxygen content of the uranium oxides subjected to pressure has been undertaken. These analyses show that the gamma high-pressure modification of U_3O_8 has an oxygen-to-uranium ratio essentially the same as the starting U_3O_8 . In addition, reduction to lower oxides does not occur at 1300 C with applied pressures of 100,000 atm. A study of reactions under pressure of UO_2 containing Al_2O_3 and/or MgO was initiated. No reaction between UO_2 and Al_2O_3 has been detected. A sample of UO_2 containing 50 mole per cent MgO reacted to form a solid solution of fluorite structure. Since it is not certain whether this solubility was the result of excess oxygen in the UO_2 or of the pressure applied further study is in progress.

The irradiation of 11 capsules containing specimens of Type 347 stainless steel is being continued in the ETR. In addition, two capsules designed to operate at temperatures of about 600 F (gamma heating) have been inserted in the WTR. Temperature measurements of the specimens obtained during runs at 20, 25, and 30 megawatts indicate that the design temperature (600 F) will be reached when the reactor operates at 60 megawatts.

The study of high-strength niobium-base alloys for possible EBR cladding applications has been temporarily discontinued and is not reported herein. However, corrosion testing of alloys being studied for water corrosion resistance has continued. In general, alloys containing more than 45 a/o zirconium and a ternary containing 28 a/o titanium-6 a/o chromium appear most corrosion resistant. A binary alloy of niobium and 12.6 a/o vanadium possesses the optimum combination of high-temperature strength, low thermal-neutron cross section, and adequate corrosion resistance.

The fabrication of components for capsules designed for in-pile creep testing of zirconium has been initiated, and arrangements are being made for irradiations in the WTR. The decrease of internal friction in Zircaloy wire with time after straining 0.5 per cent was determined. The change noted is typical of that associated with the Köster effect and probably does not indicate strain aging. Additional tensile tests performed on Zircaloy-2 at various temperatures show the presence of a yield point after initial plastic strain which indicates a phenomenon similar to strain aging.

The studies concerned with the development of techniques for the determination of oxygen concentrations in sodium below 10 ppm have been continued. The various equipment developments and modifications needed for the various techniques are complete, and actual tests will be initiated in the near future. In previous studies of the wear and

friction of sliding surfaces in liquid sodium, ball and flat specimens, with and without sodium lubrication, were investigated. It was found that MoO_2 and Na_2MoO_4 on the surfaces reduced friction and wear. Attempts are being made to deposit coatings of molybdenum-containing compounds on metals such as stainless steel.

Valence Effects of Oxide Additions to Uranium Dioxide

W. B. Wilson and C. M. Schwartz

An investigation is being conducted on the stabilizing influence of oxide additions to uranium oxide. Previous work has been directed toward determination of the mechanism of stabilization of rare-earth oxide additions to uranium oxide exposed to an oxidizing atmosphere at elevated temperatures. Current work has been devoted to fabrication and evaluation of solid solutions of U_3O_8 containing 40, 50, and 60 mole per cent La_2O_3 . An additional sample of U_3O_8 containing 25 mole per cent Y_2O_3 and 25 mole per cent MgO was also fabricated in an attempt to lower both the amount and nuclear cross section of the additive. These compositions were sintered at 1760 C in air for periods of 1, 5, and 20 hr, which is identical to the air oxidation used previously to evaluate the stability of the solid solutions of UO_2 containing additions.

The weight losses of the above solid solutions, previously reported, were generally greater than those obtained by air oxidation of samples in which the uranium was incorporated as UO_2 . It is believed that these weight losses are the result of diffusion into the BeO setter on which the sample was heated, since the BeO was discolored following the air-oxidation treatment. Further weight-change experiments will be performed to minimize diffusion of the sample and permit a more accurate evaluation of the weight losses to be expected of these materials.

Structural studies of the U_3O_8 - La_2O_3 solid solutions indicate that they form the fluorite solid solution as previously reported by Hund. The lattice parameters, however, are considerably larger than those reported by Hund and are in general agreement with those previously obtained by air oxidation of UO_2 - La_2O_3 solid solutions of corresponding La_2O_3 content. Table A-1 gives the lattice parameters of the above solid solutions, as initially sintered at 1200 C and 1375 C each for 24 hr followed by additional air oxidation at 1760 C for 1, 5, and 20 hr. These parameters may be compared to the UO_2 - La_2O_3 solid solutions and to Hund's U_3O_8 - La_2O_3 by reference to BMI-1328.

In contrast to the behavior of the UO_2 - La_2O_3 solid solutions, the present U_3O_8 - La_2O_3 parameters given in Table A-1 show very little or no change with extensive firing at high temperature. The decrease in lattice parameter upon oxidation of the UO_2 - La_2O_3 solid solutions is associated with the oxidation of these materials.

Chemical analyses and electrical studies of these solid solutions are not, as yet, complete. A preliminary evaluation of the electrical characteristics of the U_3O_8 - La_2O_3 solid solutions indicates that the 40 mole per cent La_2O_3 was p-type while the 50 and 60 mole per cent La_2O_3 were n-type. The same change in sign of the thermoelectric power was observed for the UO_2 - La_2O_3 solid solutions previously reported. The room-temperature resistivities of the U_3O_8 -40 mole per cent La_2O_3 appear to be of the order

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TABLE A-1. LATTICE PARAMETERS OF SOLID SOLUTIONS AFTER AIR OXIDATION AT 1760 C

Sample	Intended Composition, mole per cent	Lattice Parameter After Indicated Sintering Temperature and Time, Å			
		1375 C, 24 Hr	1760 C, 1 Hr	1760 C, 5 Hr	1760 C, 20 Hr
176	U ₃ O ₈ -40 La ₂ O ₃	5.52	5.52	5.53	5.55
174	U ₃ O ₈ -50 La ₂ O ₃	5.56	5.55	5.55	5.54
175	U ₃ O ₈ -60 La ₂ O ₃	5.55 ^(a)	5.58	5.58	5.58
177	U ₃ O ₈ -25 Y ₂ O ₃ -25 MgO	5.34	5.32	5.32	5.32

(a) The low parameter was due to incomplete solid solution of the La₂O₃ at the lower 1375 C sintering temperature. The 1760 C firing was performed to homogenize the samples and evaluate their weight-loss characteristics.

of 10^2 ohm-cm, the U_3O_8 -50 mole per cent La_2O_3 near 10^4 ohm-cm, and the U_3O_8 -60 mole per cent La_2O_3 greater than 10^6 ohm-cm. Thus, the "valence-compensated" air-fired U_3O_8 - La_2O_3 solid solutions containing greater than equimolar amounts of La_2O_3 are poor conductors.

In previous experimental work the stability of the UO_2 - La_2O_3 solid solutions has been evaluated by firing compacts in air at temperatures to 1760 C and evaluating weight change. In such experiments the loss of uranium oxide by volatility or "transpiration" is qualitatively related to the weight change. A direct determination of the vapor pressure of uranium oxide is being undertaken for a series of solid solutions ranging from UO_2 to UO_2 containing 54 mole per cent La_2O_3 . The results will define the variation of the vapor pressure as a function of the La_2O_3 additive.

High-Pressure High-Temperature Solid-State Studies

W. B. Wilson and C. M. Schwartz

An investigation is being conducted on the effects of combined ultrahigh pressure and high temperature on the uranium-oxygen system and on the reaction of the uranium oxides with various mixed oxides. A more precise determination of the oxygen content of the uranium oxides subjected to pressure has been undertaken using microbalance techniques. The new results were obtained by reduction of the oxides to UO_2 in dry hydrogen at 900 C, followed by reoxidation to stoichiometric U_3O_8 in dry air at 750 C.

The results have shown that the original starting U_3O_8 , previously assumed to be $\text{UO}_2.67$, had gained a small amount of oxygen upon standing in air. This excess oxygen, possibly chemisorbed on the surface of the U_3O_8 powder, is incorporated into the structure of the high-pressure phases upon application of high pressure and temperature. The results of the analysis are given in Table A-2.

The results of Table A-2 show that the gamma high-pressure modification of U_3O_8 has an oxygen-to-uranium ratio of $\text{UO}_2.72$, which is essentially the same as that of the starting U_3O_8 material. The slight difference given in Table A-2 between the gamma U_3O_8 and the starting material is attributed to the loss of the chemisorbed oxygen during heating in the microbalance apparatus. In the high-pressure apparatus, this chemisorbed oxygen is incorporated into the structure of the oxide, producing slightly higher oxygen-to-uranium ratios.

The results of Table A-2 also show that reduction to the lower oxides of uranium has not occurred even at 1300 C at the higher pressure. Previous results at 50,000 atm at these temperatures showed a distinct oxygen loss upon heating to comparable temperatures.

A determination of the density and the oxygen-to-uranium ratio of the U_4O_9 material used in the present high-pressure work was undertaken. Table A-2 shows that the oxygen-to-uranium ratio corresponds to $\text{UO}_2.26$. A density determination of this material, using the helium-compression method, gave a value of 10.43 ± 0.18 g per cm^3 . A previous determination, using the vacuum-pycnometric method, yielded a value of

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TABLE A-2. MICROBALANCE ANALYSES OF THE OXYGEN-TO-URANIUM RATIO FOR URANIUM OXIDES
SUBJECTED TO PRESSURE AND TEMPERATURE

Sample	Phases	Pressure, atm	Temperature, C	Oxygen-to-Uranium Ratio
1	Initial U_3O_8	0	0	2.70
163	Gamma plus cubic ^(a)	100,000	500	2.72
164	Gamma ^(b)	100,000	700	2.73
165	Complex ^(c)	100,000	900	2.69
166	Complex	100,000	1100	2.66
167	Complex	100,000	1300	2.68
146A	Initial U_4O_9	0	0	2.26

(a) The description "cubic" here refers to the U_4O_9 cubic fluorite structure which is believed to contain considerable excess oxygen (i. e., cubic U_3O_8).

(b) Gamma here refers to the high-pressure form of U_3O_8 .

(c) The term "complex" is used to describe a structure which bears marked analogy to U_4O_9 but has lower (than cubic) symmetry.

10.33 g per cm³. The theoretical density of U₄O₉, based upon an oxygen interstitial model, requires a density of 11.31 g per cm³, while that based upon a uranium vacancy model requires a density of 10.26 g per cm³. It may be seen that the present results are in better agreement with the latter alternative. A redetermination of the helium density of the initial U₄O₉ is under way, using a larger sample, in order to obtain more precise results for correlation with the high-pressure studies of the uranium oxides.

The U₄O₉ material described above was subjected to pressures of 75,000 atm and examined by X-ray diffraction. No change or reduction to UO₂ was observed as previously experienced, indicating that the earlier sample had probably been reduced because of a crack in the heater tube in which it was contained.

The study of reactions under pressure of UO₂ containing Al₂O₃ and/or MgO was initiated. No reaction was detected between UO₂ and Al₂O₃. The UO₂-50 mole per cent MgO sample reacted to form a solid solution of fluorite structure with a lattice parameter of 5.32 Å. Excess MgO was present, indicating that complete solid solution was not obtained. It is known that excess oxygen in UO₂ increases the solubility of MgO in the UO₂ structure. Thus, it must be determined whether the solid solution formed with MgO was the result of pressure or because the sample was sufficiently oxidized to increase the MgO solid solubility. The same general reaction has been previously reported for UO₂-Sc₂O₃.

Additional calibration work was undertaken on the high-pressure apparatus to evaluate its use as a "compressible-gasket" device. The results indicated that additional sample height may be obtained through use of this technique.

Work will continue on the study of reactions of the uranium oxides with other refractory oxides.

Irradiation-Surveillance Program on Type 347 Stainless Steel

W. E. Murr, F. R. Shober, R. L. Ritzman,
and J. F. Lagedrost

The stainless steel surveillance program has as its objective the determination of mechanical-property changes occurring in AISI Type 347 stainless steel when irradiated to total fast-neutron exposures of about 0.5 to 1.5 x 10²² nvt. Information gained from capsule-irradiated subsize tensile, cyclic-strain fatigue, and impact specimens will be used to determine the type and degree of damage occurring in the operating KAPL loop and in other loops and structural members of this material operating in fast-neutron flux fields.

At the present time, 11 capsules are being irradiated in the ETR. Three capsules have been in-pile since August, 1959, and eight have been in-pile since June, 1958. The group of three will be given postirradiation anneals prior to mechanical testing, while the group of eight will be tested at room temperature. The total estimated exposures for all capsules as of January 11, 1960 (end of Cycle 25), was given previously in

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BMI-1409. ETR Cycle 26 was initiated on January 22 and ended on February 15 for an estimated total of about 3000-megawatt days; however, specific power-generation data are not available at this time.

The first two capsules designed to operate at temperatures of about 600 F by gamma heating of specimens and capsule components were inserted in Position L-7 of the Westinghouse Test Reactor on February 18. Temperature measurements obtained by thermocouples located adjacent to specimens during runs at 20, 25, and 30 megawatts indicated that the design temperature of about 600 F will be reached at 60-megawatt operation. The reactor is in the process of escalation to a 60-megawatt power level, which it should reach sometime in April. At that time, a third capsule will be inserted in the reactor.

Development of Corrosion-Resistant Niobium Alloys

D. J. Maykuth, W. D. Klopp, R. I. Jaffee,
W. E. Berry, and F. W. Fink

The mechanical-property evaluation of selected niobium alloys for possible service in pressurized-water reactors was concluded. Previous work had shown that optimum properties were obtained in a binary 12.6 a/o vanadium alloy. In the recent work, alloys of lower total alloy content were evaluated with the intent of improving fabricability and reducing the thermal-neutron-absorption cross section while still maintaining good corrosion resistance and hot strength.

Corrosion testing of alloys in 600 and 680 F water and 750 F 1500-psi steam has been continued. The results obtained to date for exposure times ranging up to 308 days are summarized in Table A-3. In general, alloys containing more than 45 a/o zirconium and a ternary containing 28 a/o titanium-6 a/o chromium appear most corrosion resistant. They possess adherent tarnish films and low weight gains. An exception is the binary 65 a/o zirconium alloy, which began to exhibit edge cracking after 168 days of exposure to 680 F water. A 12.6 a/o vanadium alloy possesses the optimum combination of high-temperature strength, low cross section, and adequate corrosion resistance.

Vanadium-zirconium and vanadium-titanium alloys containing additions of carbon and oxygen for increased strength have now been exposed 28 days to 680 F water. Weight gains for these alloys also are included in Table A-3. They are low and compare favorably with those for similar alloys which contain lower amounts of these interstitials. The results confirm earlier observations that alloys prepared from commercial-purity niobium (high in interstitials) were as corrosion resistant as those prepared from high-purity niobium (low in interstitials).

Elevated-temperature tensile tests were conducted on a series of binary and ternary niobium-base alloys containing 5 to 8.7 a/o vanadium. These data are summarized in Table A-4 along with similar data for the 12.6 a/o vanadium alloy for purposes of comparison. These results show that, while alloy strength decreases with total alloy content, good strengths are obtained at levels down through 5 a/o vanadium. The intentional addition of interstitial impurity elements to alloys containing 5 to 8.7 a/o vanadium

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TABLE A-3. SUMMARY OF CORROSION RESULTS OBTAINED ON NIOBIUM ALLOYS EXPOSED TO HIGH-TEMPERATURE WATER AND STEAM

Alloy Addition (Balance Niobium), a/o	600 F Water		680 F Water		750 F Steam	
	Exposure Time, days	Total Weight Change, mg per cm ²	Exposure Time, days	Total Weight Change, mg per cm ²	Exposure Time, days	Total Weight Change, mg per cm ²
<u>Commercial Niobium, Rocking-Hearth Melts</u>						
Unalloyed Nb	308	-50.5	42(a)	Disintegrated	28(a)	Disintegrated
10.5 Zr	--	--	196(a)	0.67	210	-39.7
26.1 Zr	--	--	196(a)	0.07	--	--
35.7 Zr	--	--	196(a)	0.66	--	--
45.7 Zr	--	--	196(a)	0.55	--	--
1.08 W	--	--	196(a)	-2.60	210	-64.2
4.67 W	--	--	196(a)	-29.3	--	--
9.56 W	--	--	196(a)	Cracked	--	--
2.45 Mo	--	--	196(a)	-7.10	98	Disintegrated
5.20 Mo	--	--	196(a)	-1.30	210	-184.5
7.40 Mo	--	--	196(a)	0.62	--	--
4.42 V	--	--	196(a)	0.42	210	-24.0
6.59 V	--	--	196(a)	0.73	210	1.28
8.93 V	--	--	196(a)	0.59	210	1.18
10.7 V	--	--	196(a)	0.78	--	--
13.7 V	--	--	196(a)	0.50	--	--
24.2 V	--	--	196(a)	0	--	--
4.90 Fe	--	--	196(a)	0.10	98	Disintegrated
9.41 Ti	--	--	196(a)	0.65	182	1.19
18.8 Ti	--	--	196(a)	0.48	--	--
24.3 Ti	--	--	196(a)	0.52	--	--
30.5 Ti	--	--	196(a)	0.40	--	--
33.8 Ti	--	--	196(a)	0.33	--	--
12.0 Ti-0.5 Cr	--	--	196(a)	0.66	--	--
20.2 Ti-2.1 Cr	--	--	196(a)	0.39	--	--
28.2 Ti-6.1 Cr	--	--	196(a)	0.20	--	--
12.0 Ti-4.2 Mo	--	--	196(a)	0.64	--	--
17.4 Ti-6.2 Mo	--	--	196(a)	0.54	--	--
23.1 Ti-7.8 Mo	--	--	196(a)	0.45	--	--
10.4 Ti-5.0 V	--	--	196(a)	0.56	--	--
16.1 Ti-8.4 V	--	--	196(a)	0.40	--	--
22.6 Ti-11.0 V	--	--	196(a)	0.48	--	--
<u>High-Purity Niobium, Consumable-Electrode Melts</u>						
Unalloyed Nb	308	0.70	308	-7.22	224(a)	-54.0
7.18 Mo	308	0.64	252(a)	Cracked	70(a)	Cracked

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TABLE A-3. (Continued)

Alloy Addition (Balance Niobium), a/o	600 F Water		680 F Water		750 F Steam	
	Exposure Time, days	Total Weight Change, mg per cm ²	Exposure Time, days	Total Weight Change, mg per cm ²	Exposure Time, days	Total Weight Change, mg per cm ²
<u>High-Purity Niobium, Consumable-Electrode Melts (Continued)</u>						
12.6 V	308	0.35	308	0.72	296	0.76
46.8 Zr-5.06 Ti	280	0.25	280	1.03	266	2.51
11.2 Ti-3.2 Mo	280	0.22	280	0.66	266	0.36(b)
18.8 Ti-8.7 Mo	280	0.36	280	0.66	266	0.53
9.9 Zr-9.4 V	196	0.14(b)	196	0.23(b)	182	-5.87
5.7 Zr-11.4 V	196	0.07(b)	196	0.43	182	-2.15
9.1 Ti-6.3 Cr	196	0.08(b)	196	0.51	182	-4.99
7.5 V	28	0.24	28	0.23	28	0.99
7.5 V-0.19 N	28	0.18	28	0.28	28	0.51
7.5 V-2.5 Ti	28	0.21	28	0.34	28	0.48
7.5 V-2.5 Mo	28	0.27	28	0.28	28	0.66
<u>High-Purity Niobium, Rocking-Hearth Melts</u>						
Unalloyed Nb	--	--	224	-5.73	--	--
Unalloyed Nb	--	--	84(a)	Disintegrated	--	--
Unalloyed Nb	--	--	196	-0.39	--	--
1.1 Zr	--	--	168(a)	Disintegrated	--	--
2.2 Zr	--	--	112	-17.8	--	--
5 Zr	--	--	196	-14.5	--	--
10.2 Zr	--	--	196	-0.03	--	--
40 Zr	--	--	196	0.78	--	--
65 Zr	--	--	196	-1.12, edge cracking	--	--
75 Zr	--	--	196	1.28	--	--
90 Zr	--	--	196	1.42	--	--
3.2 Ti	--	--	224	-10.3	--	--
10.5 Ti	--	--	224	0.76	--	--
25.0 Ti	--	--	224	0.48	--	--
<0.02 Cr	--	--	224	-3.87	--	--
0.5 Cr	--	--	224	-1.35	--	--
0.5 Cr	--	--	196	-4.95	--	--
<0.08 Fe	--	--	196	-1.82	--	--
0.3 Fe	--	--	112	-11.8	--	--
10 Fe	--	--	28(a)	Cracked	--	--

A-10

TABLE A-3. (Continued)

Alloy Addition (Balance Niobium), a/o	600 F Water		680 F Water		750 F Steam	
	Exposure Time, days	Total Weight Change, mg per cm ²	Exposure Time, days	Total Weight Change, mg per cm ²	Exposure Time, days	Total Weight Change, mg per cm ²
<u>High-Purity Niobium, Rocking-Hearth Melts (Continued)</u>						
10.9 Zr-5.1 Ti	--	--	196	0.60	--	--
25 Zr-5 Ti	--	--	196	-0.22	--	--
25 Zr-15 Ti	--	--	196	0.47	--	--
25 Zr-25 Ti	--	--	196	0.47	--	--
35 Zr-5 Ti	--	--	196	0.71	--	--
35 Zr-15 Ti	--	--	196	0.36	--	--
45 Zr-5 Ti	--	--	196	0.71	--	--
10 Zr-5 Mo	--	--	196	0.17	--	--
35 Zr-5 Mo	--	--	196	0.53	--	--
45 Zr-5 Mo	--	--	196	0.70	--	--
35 Zr-5 Al	--	--	196	0.26	--	--
45 Zr-5 Al	--	--	196	0.60	--	--
10 Zr-5 Cr	--	--	196	0.48	--	--
45 Zr-5 Cr	--	--	196	0.48	--	--
10 Zr-5 Fe	--	--	196	0.44	--	--
2.5 V	--	--	224	1.11	--	--
2.0 V-2.5 Ti	--	--	196	0.88	--	--
2.0 V-2.3 Mo	--	--	196	1.14	--	--
2.2 V-0.54 Fe	--	--	196	0.54	--	--
1.8 V-0.02 Cr	--	--	196	0.41	--	--
1.8 V-0.14 Al	--	--	196	0.88	--	--
2.5 V-2.5 Zr	--	--	112	-2.43	--	--
2.2 V-0.87 Ni	--	--	28 ^(a)	-1.00	--	--
4.0 V-2.3 Zr	--	--	196	0.60	--	--
5 V-25 Zr	--	--	196	-2.84	--	--
5 V-35 Zr	--	--	196	-3.52	--	--
5 V-45 Zr	--	--	196	0.60	--	--
1 Ce	--	--	56	-11.8 ^(a)	--	--
1 Y	--	--	56	-10.2	--	--
5 Y	--	--	56	-64.2	--	--
1 Ni	--	--	28	0.04 ^(b)	--	--
2.5 Ni	--	--	7	Broke, 7 days	--	--
5 Ni	--	--	7	Disintegrated	--	--
1 Pd	--	--	56	15.7 ^(a)	--	--

A-11

TABLE A-3. (Continued)

Alloy Addition (Balance Niobium), a/o	600 F Water		680 F Water		750 F Steam	
	Exposure Time, days	Total Weight Change, mg per cm ²	Exposure Time, days	Total Weight Change, mg per cm ²	Exposure Time, days	Total Weight Change, mg per cm ²
<u>High-Purity Niobium, Rocking-Hearth Melts (Continued)</u>						
5 V-2.5 Ti	--	--	56	0.51	--	--
5 V-2.5 Mo	--	--	28	0.18	--	--
5 V-2.5 Fe	--	--	28	0.21	--	--
5 V-2.5 Cr	--	--	56	0.60	--	--
5 V-2.5 Ni	--	--	28	0.42	--	--
5 V-2.5 Al	--	--	56	0.58	--	--
2.5 V-0.5 C	--	--	28	0.35	--	--
5 V-0.5 C	--	--	28	0.17	--	--
2.5 V-0.25 Ti-0.5 O	--	--	28	0.58	--	--
5 V-0.25 Ti-0.5 O	--	--	28	0.10	--	--
2.5 V-0.25 Zr-0.5 O	--	--	28	0.11	--	--
5 V-0.25 Zr-0.5 O	--	--	28	0.32	--	--
2.5 V-0.5 Ti-0.5 C	--	--	28	0.12	--	--
5 V-0.5 Ti-0.5 C	--	--	28	0.22	--	--
2.5 V-0.5 Zr-0.5 C	--	--	28	0.38	--	--
5 V-0.5 Zr-0.5 C	--	--	28	0.41	--	--
5 V	--	--	28	0.29	--	--
10 Zr	--	--	--	--	84	-4.30
10 Ti	--	--	--	--	84	0.40

(a) Off test.

(b) Losing weight.

TABLE A-4. ELEVATED-TEMPERATURE TENSILE PROPERTIES OF NIOBIUM-BASE ALLOYS AFTER VACUUM ANNEALING AT 2190 F

Alloy	Alloy Content (Balance Niobium), a/o	Temperature, F	Tensile Properties		
			Ultimate Strength, psi	0.2 Per Cent Offset Yield Strength, psi	Elongation in 1 In., per cent
N40	100 Nb	1500	21,000	16,000	18
N81	5 V ^(a)	1500	40,000	20,000	37
N65	5 V-2.5 Ti ^(a)	1500	44,000	23,000	29
N66	5 V-2.5 Mo ^(a)	1500	43,000	24,000	9 ^(b)
N72	5 V-0.5 C ^(a)	1500	48,000	30,000	32
N78	5 V-0.5 Ti-0.5 C ^(a)	1500	47,000	26,000	42
N74	5 V-0.25 Ti-0.5 O ^(a)	1500	51,500	35,000	28
NL11	7.6 V	1200	58,000	32,000	17
		1500	53,000	34,000	34
NL12	8.7 V-0.26 N	1200	64,500	38,000	15
		1500	60,000	42,000	26
NL13	7.3 V-2.4 Ti	1200	62,000	38,000	14
		1500	53,500	34,000	23
NL14	7.6 V-2.3 Mo	1200	64,000	36,000	18
		1500	55,000	34,000	28
NL2	12.6 V	1200	77,000	49,000	18
		1500	>69,000 ^(c)	47,000	>8 ^(c)

(a) Compositions nominal.

(b) Sample fractured at pinhole in shoulder.

(c) Pin in sample shoulder ruptured.

A-13

appears as effective in improving hot strength as additions of either 2.5 a/o of titanium or molybdenum.

Creep testing of Alloys NL11 through NL14 (see Table A-4 for compositions) was completed. While these data have not yet been fully evaluated, preliminary inspection indicates that all of these alloys show good resistance to creep at 1200 F under a stress of 20,000 psi.

With completion of the mechanical-property evaluations, a topical report summarizing all of the results obtained on this research will be prepared. Corrosion testing of the alloys described in this report will be continued separately.

Investigation of the Creep Properties of Zircaloy-2
During Irradiation at Elevated Temperatures

F. R. Shober, P. B. Shurnaker, A. P. Young,
M. F. Amateau, and R. F. Dickerson

The effect of fast-neutron (>1 Mev) irradiation on the creep properties of Zircaloy-2 at elevated temperatures is to be studied by comparing total creep deformation obtained under two test conditions. Creep specimens will be stressed in the reactor and out of the reactor, and the resulting deformations will be compared. The test temperature will be 345 C. Internal-friction studies and electron microscopy of thin films of zirconium and Zircaloy-2 are being used as aids in the strain-aging studies of Zircaloy-2.

Work on an irradiation capsule to contain the creep specimens and auxiliary creep apparatus has been concerned primarily with the fabrication of components. A stress analysis of the capsule has been partially completed, and problems pertinent to the irradiation phase have been discussed with WTR personnel.

The welded metal bellows, which form a part of the stressing assembly inside the capsules, have been shipped by the manufacturer and should arrive at Battelle early in March. They will be pressure tested prior to installation in the capsules. Delivery of the electric-heater units is scheduled for mid-March; the control instrumentation for mid-April.

At the present time, it is planned to measure the deformation with a differential transformer (suitable for in-pile applications) located below and on the low-pressure side of the bellows. The transformer core will be attached to the movable ends of the loading bellows, while the transformer will be rigidly mounted to the body of the capsule. Records of the output of the transformer in both in-pile and out-of-pile tests will provide a measure of irradiation effects on the creep properties.

During March, the fabrication of components will be continued. Present plans call for the construction of a pressure mock-up and a thermal mock-up, in addition to the in-pile and out-of-pile capsules. It is probable that laboratory work with the mock-ups will begin early in April.

A-14

Internal-friction studies were continued on a second lot of Zircaloy-2 at 10 C. The effect of aging time on the internal friction of strained Zircaloy-2 was investigated during this report period. The decrease of internal friction in a Zircaloy wire with time after straining 0.5 per cent was determined. The change noted in internal friction is the typical decrease associated with the Köster effect and probably does not indicate strain aging. The specimen attained a relatively constant value of internal friction, indicating very slow recovery. Future work will consist of determining the internal friction as a function of time for strained Zircaloy-2 up to 300 C.

Tensile tests were performed at room temperature and 232, 288, and 345 C on a second lot of Zircaloy-2 annealed 1 hr at 750 C in vacuum. A definite yield-point phenomenon was evident after the specimens were plastically strained, unloaded, and reloaded at temperatures of 232, 288, and 345 C. Tensile specimens were prepared from the same material used for the internal-friction studies. Upon each subsequent loading in the tensile tests, whether loaded immediately or allowed to age at the test temperature for 30 min before reloading, a yield point was evident. The yield point in all cases was accompanied by a perceptible drop in load which was recovered by additional straining. The rapidity with which the material changes from a material which shows no yield point to a material which does implies that a rapidly diffusing interstitial could be causing the yield points.

Electron-microscopy studies have been confined to improving chemical-polishing techniques in the preparation of thin-film specimens of zirconium and Zircaloy-2. Light-microscope studies will be initiated on Zircaloy-2 which has shown the yield-point phenomenon in tensile tests. A study of the microstructure will be made with particular attention directed to the second-phase distribution and a comparison will be made with unstrained microstructures.

Determination of Oxygen in Sodium at
Concentrations Below 10 PPM

D. Ensminger, D. R. Grieser, E. H. Hall, J. W. Kissel,
J. McCallum, and W. H. Goldthwaite

The objective of this program is to prove the feasibility and demonstrate the practicality of a means of rapid oxygen determination in sodium. A sensitivity of ± 1 ppm at oxygen levels of 10 ppm and below is sought.

Novel approaches that hold promise of greater sensitivity than conventional techniques are being pursued. The initial evaluation studies are being made with "cold-trapped" sodium, 20 to 100 ppm oxygen, to establish the basic sensitivity and handling techniques.

Ellipsometry

Effort in the use of polarimetry has been concentrated on the production of a leak-tight cell containing an optically flat window. Mechanically sealed cells have been completed and are ready for receipt of sodium samples.

A-15 and A-16

Electrical Resistivity

A redesigned sodium-resistance-measuring device is virtually complete. The associated measuring equipment for the determination of resistance changes as small as 10 is under construction at present. The entire system should be completed and resistance-versus-oxygen content measurements made during the next report period.

Polarographic Studies

Recent effort has been devoted to a search for an electrolyte which is not reactive with sodium at temperatures up to about 600 F. Many potential organics have been evaluated. Thus far all of them have failed either due to sodium reactivity or through lack of conductivity. Several promising organics remain to be evaluated.

Mass Spectrometry

The sensitivity of the mass spectrometer has been increased by the incorporation of an electron multiplier. Residual gas analyses indicate an increase in sensitivity of from 500 to 700 times.

The sodium sampling device has been checked out and proven satisfactory. The reproducibility of sample weight is being determined at present. Sodium spectrum runs and oxygen determinations will be made during the next report period.

Purification-Loop Operation

Modifications to the sodium cold-trap purification loop have improved operation, and samples containing oxygen levels in the 20 to 100-ppm range can be obtained routinely.

Wear and Friction of Sliding Surfaces in Liquid Sodium

J. W. Kissel, W. Glaeser, and W. H. Goldthwaite

Friction and wear between molybdenum ball and flat specimens, with and without sodium lubrication, have been shown to vary with preparation and conditioning of the surfaces. Correlation was subsequently found between specimen conditioning techniques and the presence of surface films having specific composition. For example, MoO_2 and Na_2MoO_4 were found on surfaces which had exhibited the lowest friction and wear.

At present, vacuum-coating techniques are being developed in an effort to deposit coatings of the molybdenum-containing compounds on other base metals such as stainless steel. The thickness of such coatings will be measured, and their friction and wear properties will be determined. Friction between ball and flat steel specimens has been measured to check out the apparatus and provide baseline data.

B-1

STUDIES OF FUEL ALLOYS (AEC-DRD)

R. F. Dickerson

Niobium-base binary alloys containing 10 to 60 w/o uranium have been on test in 600 F water for 224 days. All specimens still exhibit a dull black oxide coating. All of the alloys have been exposed to 1500 F sodium for 500 hr and are currently on test for another 500 hr. The niobium-10 w/o uranium alloy was tested for 1000 hr at 1600 F under a stress of 40,000 psi without rupture. This specimen showed an average creep rate of 0.0002 per cent per hr. This same alloy has been tested for 300 hr at 2200 F under a load of 10,000 psi with no perceptible deformation after initial loading. Tests will be run at 2200 and 2500 F on the niobium-20 w/o uranium alloy. Irradiation-experiment planning and capsule design are in progress, and the long-time heat treatments necessary for the gamma-immiscibility-loop study are in progress.

Melting studies designed to develop techniques for the preparation of thorium-uranium alloys containing finely divided, homogeneously disposed uranium particles are continuing. Thermodynamic studies indicate that the equilibrium nitrogen pressure for the formation of ThN at 3600 K is about 2 atm. Thorium was arc melted once under a pressure of 2-1/2 atm of nitrogen. The resultant compound was analyzed to be ThN_{0.86}. Additional attempts will be made to prepare ThN by melting the thorium a number of times under 2-1/2 atm of nitrogen. Ternary additions of 1.5 w/o molybdenum or 2 w/o niobium decrease the 600 C secondary-creep rate of the thorium-10 w/o uranium alloy by a factor of about 2, while the ternary addition of 10 w/o zirconium increases the creep rate of the binary thorium-uranium alloy.

Development of Niobium-Uranium Alloys

J. A. De Mastry, S. G. Epstein, A. A. Bauer, and R. F. Dickerson

Niobium-rich niobium-uranium alloys have potential as high-temperature metallic fuels. For this reason, a program to investigate the fabrication characteristics, mechanical and physical properties, corrosion behavior in various media, and irradiation behavior of these alloys has been undertaken.

Alloys containing from 10 to 60 w/o uranium were prepared from three grades of niobium, grades variously containing from 0.03 to 0.07 w/o oxygen and 0.02 to 0.74 w/o zirconium. No effect due to the oxygen and zirconium variations was observed on the fabrication characteristics or the tensile properties of the niobium-uranium alloys.

The testing of niobium-uranium alloys in 600 F water has been in progress for 224 days; the data obtained are shown in Table B-1. While the majority of the alloys being tested show total weight gains, the weight gains are now decreasing. All specimens being tested have a dull black oxide coating.

Specimens of each alloy composition have been examined after exposure to sodium at 1500 F for 500 hr. All specimens exhibit a dull black oxide. Table B-2 shows weight

B-2

TABLE B-1. CORROSION DATA FOR NIOBIUM-URANIUM ALLOYS^(a) IN 600 F WATER

Nominal Alloy Content (Balance Niobium), w/o	Impurity Content		Specimen Condition	Total Weight Change in 600 F Water, mg per cm ²	
	Oxygen ^(b) , ppm	Zirconium ^(c) , w/o		In 210 Days	In 224 Days
10 U	680	0.74	Fabricated	0.60	0.63
	1190	0.17	Fabricated	-0.56	-0.76
	3170	0.02	Fabricated	-3.08	-3.51
20 U	458	0.74	Fabricated	0.86	0.95
	523	0.17	Fabricated	1.85	1.93
	198	0.02	Fabricated	0.71	0.76
30 U	586	0.74	As cast	0.15	0.10
	669	0.17	Fabricated	-2.27	-2.32
	165	0.02	As cast	-0.43	-0.59
40 U	661	0.74	As cast	0.40	0.43
	579	0.17	As cast	0.17	0.20
	261	0.02	As cast	0.54	0.62
50 U	375	0.74	As cast	0.27	0.15
	334	0.17	As cast	0.34	0.37
	271	0.02	As cast	-0.38	-0.54
60 U	471	0.74	As cast	-5.32	-6.02
	273	0.17	As cast	-4.73	-5.44
	192	0.02	As cast	-1.52	-1.67

(a) Average of duplicate specimens.

(b) Analyzed value.

(c) Nominal composition.

TABLE B-2. CORROSION DATA FOR NIOBIUM-URANIUM ALLOYS^(a) AFTER 500 HR IN SODIUM AT 1500 F

Nominal Alloy Content (Balance Niobium), w/o	Impurity Content		Specimen Condition	Total Weight Change After 500 Hr in 1500 F Sodium, mg per cm ²
	Oxygen ^(b) , ppm	Zirconium ^(c) , w/o		
10 U	680	0.74	Fabricated	0.03
	1190	0.17	Fabricated	0.05
	3170	0.02	Fabricated	-0.10
20 U	458	0.74	Fabricated	0.15
	523	0.17	Fabricated	0.17
	198	0.02	Fabricated	-0.04
30 U	165	0.02	As cast	0.18
40 U	261	0.02	As cast	0.55
50 U	271	0.02	As cast	0.33
60 U	192	0.02	As cast	0.49

(a) Average of duplicate specimens.

(b) Analyzed oxygen content.

(c) Nominal composition.

B-3

changes after exposure. These specimens are being re-encapsulated and will be tested under the same conditions for an additional 500 hr.

Stress-rupture tests of a niobium-10 w/o uranium alloy at 1600 F under a stress of 40,000 psi were discontinued, since the specimen did not rupture after 1000 hr. This specimen showed an average creep rate of 0.0002 per cent per hr; this rate includes the initial deformation on loading. A second specimen of this alloy has been loaded under a stress of 10,000 psi at 2200 F. This test has progressed 300 hr with no perceptible deformation after initial loading. Upon completion of this test a specimen of the same alloy will be tested at 2500 F. Specimens of the niobium-20 w/o uranium alloy are to be tested at 2200 F and 25,000 psi, and at 2500 F at an as yet undetermined stress. Specimens are being machined for this purpose.

Capsules are being designed for irradiation studies of niobium-10, -20, and -30 w/o uranium alloys and two niobium-uranium-zirconium alloys. It is planned to irradiate specimens of these to approximately 1 a/o burnup at 1600 and 1800 F.

Wire bars of binary niobium-uranium alloys are being heat treated in order to investigate the effects of oxygen on the composition limits of the gamma immiscibility loop which occurs in this system. The alloys, ranging in composition from 10 to 58 w/o niobium, are being heat treated at temperatures of 650 to 940 C for times of 1 to 2 months. Evaluation of the phase composition limits will be made by metallographic examination. Wire bars of ternary uranium-niobium-zirconium alloys containing 50 and 60 w/o uranium have been homogenized and are being given extended heat treatments at temperatures of 500, 650, and 750 C. The effects of the zirconium additions on the niobium-uranium immiscibility gap and on the stabilization of the body-centered-cubic solid solution will be investigated in this study.

Development of Thorium-Uranium Alloys

M. S. Farkas, R. W. Endebrock, A. A. Bauer, E. L. Foster,
and R. F. Dickerson

Thorium-uranium and thorium-uranium-base alloys are being investigated with the objective of improving their irradiation stability and corrosion resistance. The effect of thorium purity, casting methods, and fabrication on the size and distribution of uranium-rich particles is being investigated. Thorium-5 to 25 w/o uranium-base ternary alloys that have been and are now being studied contain either 5 to 25 w/o zirconium or small additions of molybdenum or niobium; quaternary alloys containing niobium and zirconium additions are also being studied. The preparation and properties of thorium and thorium-uranium carbides and nitrides are also being investigated.

Melting studies of relatively large scale melts of thorium-uranium alloys will commence when materials become available. Meanwhile, a search of the literature has continued for methods applicable to the preparation of a finely divided, homogeneously dispersed alloy of uranium in thorium. Initially the effect of various melting variables on the micro and macro homogeneity of the thorium-10 w/o uranium composition will be studied. Later, the effect of small additions of niobium and molybdenum will be investigated.

B-4

The thermodynamics of thorium-nitrogen reaction have been studied. Although only estimates of the free energy of formation of ThN are available, the equilibrium nitrogen pressure for the formation of ThN at 3600 K is estimated to be about 2 atm. In a preliminary test of the preparation of ThN it was found that ThN_{0.86} could be obtained by melting thorium once under a pressure of 2-1/2 atm of nitrogen. X-ray analysis confirmed that the major phase present was ThN, while metallographic examination shows that the second phase present was thorium. Additional attempts to prepare ThN will be made by melting thorium several times under about 2-1/2 atm of nitrogen.

Tensile and creep tests at 600 and 700 C are being performed on thorium-5, -10, and -20 w/o uranium alloys and on thorium-10 w/o uranium alloys containing additions of 1.5 w/o molybdenum, 2 w/o niobium, 10 w/o zirconium, or 10 w/o zirconium-2 w/o niobium. Data obtained thus far are presented in Tables B-3 and B-4. Examination of these data show that the addition of 1.5 w/o molybdenum or 2 w/o niobium decreases the 600 C secondary-creep rate of the thorium-10 w/o uranium alloy by a factor of about 2, while the addition of 10 w/o zirconium increases the creep rate. However, the 600 C tensile data show that the thorium-10 w/o uranium-10 w/o zirconium alloy is superior to the base binary alloy. Failure of this alloy to exhibit low creep rates is attributed to the fact that phase transformations occur near the test temperature.

TABLE B-3. TENSILE PROPERTIES OF THORIUM-URANIUM AND THORIUM-URANIUM-BASE ALLOYS AT 600 AND 700 C

Alloy Composition (Balance Thorium), w/o	Test Temperature, C	0.2 Per Cent Offset Yield Strength, psi	Ultimate Tensile Strength, psi	Elongation, per cent
5 U	600	11,600	15,950	44
10 U	600	13,800	17,970	41
20 U	600	11,200	15,160	50
10 U-1.5 Mo	600	11,220	17,710	43
10 U-2 Nb	600	12,550	18,000	37
10 U-10 Zr	600	19,600	28,400	44
10 U-10 Zr-2 Nb	600	19,200	25,640	48
5 U	700	6,590	11,700	51
10 U	700	8,800	10,300	44
20 U	700	6,710	8,170	58
10 U-2 Nb	700	8,450	10,800	66
10 U-10 Zr-2 Nb	700	12,100	13,910	77

TABLE B-4. CREEP PROPERTIES OF THORIUM-URANIUM AND THORIUM-URANIUM-BASE ALLOYS AT 600 C

Alloy Composition (Balance Thorium), w/o	Stress, psi	Minimum Creep Rate, per cent per hr	Elongation, per cent
5 U	3500	0.0007	--
10 U	1500	0.0028	--
10 U	3500	0.0100	3.2
10 U	4000	0.0135	--
20 U	1500	0.0014	--
20 U	3500	0.0126	--
10 U-1.5 Mo	3500	0.0013	--
10 U-1.5 Mo	5500	0.0085	2.6
10 U-2 Nb	3500	0.00174	1.0
10 U-2 Nb	7000	0.0625	12.63
10 U-10 Zr	3500	0.037	7.2

C-1

FISSION-GAS RELEASE FROM REFRACTORY FUELS (AEC-DRD)

J. B. Melehan, D. A. Vaughan, R. H. Barnes,
H. Sheets, S. D. Beck, and F. A. Rough

Various aspects of a basic program of study of release of fission products from UO_2 are reported in this section. The objective is to understand releases from sintered UO_2 ; thus, in-pile study of sintered material is being initiated, and supporting studies of diffusion and of characterization are in progress.

Characterization of Sintered UO_2 and Model of Gas Release

The pore size, pore distribution, and grain size are being evaluated for specific UO_2 bodies to aid in interpreting the fission-gas release rates during irradiation and postirradiation heat treatment. Two specimens from a batch of sintered UO_2 have been examined for pore size and distribution in the as-polished condition. The sintering treatments were a 15-hr ball milling, a 40,000-psi forming pressure, and a 1-hr sinter at 2800 F in hydrogen. The density was measured pycnometrically to be 95.8 per cent of theoretical. The surface area of this lot of specimens, as determined by nitrogen adsorption, was found to be 410 cm^2 per g, or 4480 cm^2 per cm^3 . This large surface area for 95 per cent dense bodies was surprising. Recent microporosity analysis has indicated a large error in the density as measured pycnometrically. For the two bodies examined, pore fractions of 0.21 and 0.18 were obtained by lineal analysis which included only those pores larger than 3 μ . Thus, it would appear that the density of this lot is approximately 80 per cent of theoretical rather than 95.8. Based upon this pore measurement, the surface area, due to pores, was computed to be $\approx 30 \text{ cm}^2$ per cm^3 , where the average pore diameter is 9 μ and the mean free path between pores is 30 μ . It would appear from this analysis, that the high surface area, as obtained by the BET method, for this case cannot be attributed to the large pore volume of these specimens. Smaller pores in these specimens are being analyzed by microscopic methods using higher magnification. Also, during the next period, attempts will be made to separate grain-boundary pores from intragranular pores. In this latter investigation, the grain-boundary surface area will be determined, since it is possible that the high surface area might be due to grain-boundary surface adsorption of nitrogen.

Also, during this period, five groups of specimens were prepared for study that are expected to exhibit wide differences in total surface area, as determined by the BET method. This was accomplished by selectively altering fabrication conditions.

Fabrication conditions, densities, porosities, and oxygen-to-uranium ratios of the five groups of specimens, are shown in Table C-1. Note that porosity measurements were made by xylene adsorption. It would be expected that some of the porosity that is closed to xylene will be open to nitrogen. Examination of polished sections of representative specimens is planned.

Future work also will include an investigation of the effect of sintering conditions (time, temperature, and atmosphere) on the density, oxygen-to-uranium ratio, and

microstructure of UO_2 pellets made from oxidized UO_2 . Additional specimens will be made for surface-area measurements.

A preirradiation examination of four preparations of UO_2 (two sintered bodies, a fused UO_2 , and a single crystal) has been completed on a second series of specimens. This series is being irradiated in the as-polished condition to determine the effects of irradiation upon surfaces. Light and electron metallographic studies will be made after irradiation, and the results will be compared with the results of the preirradiation examination of these specimens and with those of the previous study of surface changes during irradiation.

Diffusion in UO_2

The experimental study of fission-gas release from uranium dioxide during postirradiation heat treatment is continuing.

During February, measurements of fission-gas release from a 1.57-g flat plate of single-crystal UO_2 were made at 1205 C. The plate was irradiated for 16 hr in an estimated effective thermal-neutron flux of 1×10^{13} nv. The results are being analyzed.

Measurements of the release rates of fission gases from additional plates of single-crystal UO_2 in the range of 1200 to 1500 C are scheduled to continue.

Preparation for In-Pile Study

The apparatus for in-pile study of fission-product release from UO_2 has been reinserted into the BRR, but difficulties with the air-monitoring system developed; these difficulties are being corrected. It is anticipated that actual operation at temperature will begin soon.

For these in-pile studies, diffusion-type releases of fission gas can best be measured and understood when an equilibrium is established for a nuclide at a given temperature. Ideally, at equilibrium, the concentration pattern reaches a steady state, and the rate of release is a constant fraction of the rate of production. For spherical geometry, this fraction is a function of the dimensionless parameter μ only, assuming a constant rate of production of the nuclide and assuming that no diffusion of the precursor occurs:

where

$$\mu = a^2 \lambda / D$$

a = equivalent sphere radius, cm

λ = decay constant, sec^{-1}

D = diffusion coefficient, cm^2 per sec.

C-3 and C-4

Equations have also been derived for spherical geometry describing the ratio of the rate of release to the rate of production for the case where the precursor diffuses. The results are expressed as a function of μ_1 and μ_2 , which are the dimensionless parameters (defined above) for the parent and daughter nuclides, respectively.

TABLE C-1. FABRICATION CONDITIONS, DENSITIES, POROSITIES, AND OXYGEN-TO-URANIUM RATIO OF UO_2 SPECIMENS

Specimen	Fabrication Conditions				Density and Porosity(a)				
	Ball-Milling Time, hr	Preoxidation Temperature, F	Forming Pressure, psi	Sintering Temperature, F	Bulk Density		Porosity, per cent		Oxygen-to-Uranium Ratio
					G per Cm ³	Per Cent of Theoretical	Open Pores	Closed Pores	
79-26	2	--	40,000	2600	10.17	92.6	0	8.4	2.00
79-28	2	--	40,000	2800	10.38	94.5	0	5.5	2.00
79-30	2	--	40,000	3000	10.38	94.5	0	5.5	2.00
80	15	--	20,000	3000	10.58	96.3	0	3.7	2.01
81	15	1000	20,000	3000	10.58	96.3	0	3.7	2.00

(a) Densities and porosities are the average of measurements on three pellets, and were measured by xylene immersion. Theoretical density of UO_2 was taken as 10.98 g per cm^3 .

D-1

GENERAL FUEL-ELEMENT DEVELOPMENT (AEC-DRD)

S. J. Paprocki

Techniques are being developed for the fabrication of dense cermet-type fuel materials consisting of a mixture of 80 volume per cent UO_2 and 20 volume per cent metal. Stainless steel, chromium, molybdenum, and niobium metal matrices are being investigated. The properties of the cermet fuels are being characterized by evaluation of structure and determination of physical and mechanical properties.

A study is being conducted concerned with the solid-phase bonding of molybdenum and niobium. Techniques have been developed for the self-bonding of niobium, and the process is being applied to the fabrication of niobium-clad compartmented UO_2 fuel plates and assemblies.

The kinetics involved in a solid-phase pressure-bonding process are being investigated. A study is being made to determine the effects of metal structure, surface roughness, and bonding parameters of time, temperature, and pressure on the subsequent area of contact and bond characteristics.

Fabrication of Cermet Fuel Elements

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

Currently pressure bonding is being used to fabricate cermets for thermal-conductivity, mean linear thermal-expansion, electrical-resistivity, modulus-of-rupture, and thermal-shock-resistance measurements. Matrix materials include stainless steel, chromium, molybdenum, and niobium.

Cermet rods of 80 volume per cent UO_2 dispersed in the various matrix materials have been pressure bonded to 90 per cent of theoretical density or greater. Caps of iron, silver, and solder have been placed on the ends of the rods in preparation for thermal-conductivity measurements. Measurements have been made to 800 C on two of the rods; however, an analysis of the data has not been made at this time.

Five cores each of 80 volume per cent UO_2 -chromium and -niobium have been pressure bonded for 3 hr at 10,000 psi, at temperatures ranging from 1900 to 2200 F. Machining of the cores into specimens for modulus-of-rupture measurements was impossible due to the low density achieved by pressure bonding at these temperatures. Additional cores are being prepared for pressure bonding at higher temperatures in an attempt to obtain cermet cores of 90 per cent of theoretical density or greater. In addition, 1 by 1 by 0.100-in. 80 volume per cent UO_2 -molybdenum and -stainless steel cermet cores have been prepared for pressure bonding for 3 hr at 10,000 psi at temperatures ranging from 2250 to 2350 F. After pressure bonding, specimens 0.085 by 0.150 in. will be machined from the cores for modulus-of-rupture measurements.

Gas-Pressure Bonding of Molybdenum- and Niobium-Clad Fuel Elements

S. J. Paprocki, E. S. Hodge, and P. J. Gripshover

Fabrication of molybdenum- and niobium-clad fuel elements containing ceramic and cermet fuels is being studied. Investigations are directed toward determination of suitable bonding conditions to produce strong metallurgical bonds in fuel elements of various configurations. Evaluation of specimens is based on the quality of bond obtained and the properties of the cladding material after bonding.

The studies during the past report period have been primarily concerned with the development of niobium-clad fuel elements. Specimens have been prepared and evaluated to determine the relative merit of various materials as spacer materials that can later be dissolved out of assemblies to form coolant channels. These included graphite-coated molybdenum, graphite-coated Ti-Namel, magnesium oxide, molybdenum, uranium dioxide, Vycor, and window glass. Chemical analyses were made to determine if any increase in impurity level of the niobium was introduced by the barrier material. Results of these tests indicated that uranium dioxide was the only material which did not change the chemical composition of the niobium during bonding. Magnesia, Vycor, and window glass raised the oxygen content to an unacceptable level, while the molybdenum alloyed with the niobium specimen, causing severe embrittlement. Results with the graphite-coated specimens indicated the carbon content was increased approximately 100 ppm during bonding; furthermore, they showed that the coating must be much thicker on the Ti-Namel because of the rapid diffusion of carbon into iron at these temperatures. The introduction of the carbide phase into the microstructure is not desirable, however, and carbon does not appear to be satisfactory as a barrier material unless the bonding time can be shortened to minimize carbon diffusion.

In an effort to determine the strength of the niobium-to-niobium bonds formed at optimum bonding parameters, two flat-plate tensile specimens have been prepared for evaluation. One specimen, a control, consisted of a single plate 0.100 in. thick by 0.5 in. wide by 8 in. long; the second specimen consisted of two plates 0.1002 by 0.5 by 4 in. which were butt bonded to form a single 8-in.-long plate. Both specimens were subjected to bonding conditions of 2100 F and 10,000 psi for 3 hr. After bonding, a metallographic examination was made of the butt bond. It was noted that some lack of grain growth had occurred in some areas, which is not characteristic of the bonds normally achieved. Both specimens were pulled in tension until fracture occurred. The control specimen fractured at a load of 1990 lb with considerable necking before fracture. The butt-bonded specimen fractured in the bond area under a load of 1850 lb with some necking. Some separation occurred at the edge of the tensile specimen under lighter loads. It is significant to note, however, that necking did occur in the center of the bond area, indicating that area was well bonded. Tensile tests are being repeated using rod-type specimens which may give more reliable data.

D-3

Factors Affecting Pressure Bonding

J. W. Spretnak and G. W. Cunningham

The mechanism and kinetics of solid-phase bonding of metals under application of heat and pressure are being studied. One phase of the program involves an investigation of the relationship between hot hardness and bonding pressure. Specimens are prepared for evaluation by hot pressing a 500-rms surface of OFHC copper in contact with an 8-rms OFHC copper surface. A total time of less than 1 min is required to apply and release the desired load. This short time is used so that the bonding time will be comparable with that used in obtaining hot-hardness values.

Specimens have been prepared by applying pressures of 2000 to 24,000 psi at temperatures of 200, 400, 600, and 700 F. Comparison of pressure used with values of Vickers hardness expressed in psi indicates that apparent contact between the surface of the two metals is reached at pressures of approximately 5 to 10 per cent of the hot hardness. Apparent contact is used to describe the condition such that after polishing and etching very lightly that a bond line can be detected, but no voids are visible under the light microscope.

Metallographic examinations show that the bond lines produced by low pressures do not have uniform etching characteristics. The bond line produced at the points of initial contact etches more rapidly but does not greatly differ in appearance from that of an ordinary grain boundary. Along the areas which were the last to be pressed into contact and thus the areas which were pressed at the lowest pressure, etching is very rapid, and, after a prolonged time, wide, deep holes which follow the contour of the bond interface are produced. As bonding pressures are increased, these areas decrease, and etching becomes more uniform along the bond line. Pressures of approximately 75 per cent of the hot hardness are required to provide a uniform bond line.

This etching behavior suggests that, in areas where low bonding pressures are obtained, very few atoms on the two surfaces are close enough to be involved in bonding, and thus etching occurs almost as rapidly as on two free surfaces. Annealing studies also support this opinion. Sections of specimens bonded at 700 F and pressures of 4,000, 12,000, and 24,000 psi were annealed 15 hr at 1000 C. After polishing and light etching, large bond-line areas of the 4,000-psi specimen were found to be not in contact. More of the area was in contact in the 12,000-psi specimen, and nearly all the area was in contact in the 24,000-psi specimen. No grain growth across the interface could be observed in the 4,000- and 12,000-psi specimens, but grain growth across 10 to 20 per cent of the bond interface could be detected on the 24,000-psi specimen. The separation of the bond line in particular areas during annealing suggests that weak bonds were formed during the initial application of pressure, but that these bonds were broken during high-temperature annealing in an effort to minimize surface energy.

Sections of the 700 F series have also been used to prepare diffusion couples with a copper-4 w/o silver alloy. The diffusion of silver along the bond line and grain boundaries will be studied to obtain information on the nature of the bond line as related to applied pressure.

D-4

Specimens are being prepared to study the effect of pressure at higher temperatures (1000, 1200, and 1400 F). The effect of grain size will also be studied at 400, 700, and 1000 F.

E-1

GAS-PRESSURE BONDING OF CERAMIC, CERMET,
AND DISPERSION FUEL ELEMENTS
(AEC-FUEL CYCLE)

S. J. Paprocki, S. W. Porembka, D. L. Keller, E. S. Hodge,
C. B. Boyer, and J. B. Fox

This study is concerned with the development of a fuel-element fabrication process which will maintain or improve quality while reducing manufacturing costs. The gas-pressure-bonding method is being investigated since it appears to be a likely method for achieving these objectives. The fuel systems being evaluated in this program include the ceramic, cermet, and dispersion fuels with a major emphasis on the ceramic fuel system. It is anticipated that such fuel systems will offer the greatest promise of high burnups over a range of operating temperatures. The program is directed toward the simultaneous densification of powder fuels and cladding with stainless steel.

Uranium Dioxide Compaction Studies

Compaction studies of the uranium dioxide powders are based on the development of high-density green compacts which will subsequently achieve a desired range of densities during pressure bonding. Previous studies of UO_2 compaction involved the cold-pressing characteristics and tap densities of seven commercial powder types and several powder mixtures, centrifugal packing, ultrasonic compacting, presintering, binder removal, and the effect of stoichiometry. At the present, investigations on the effects of particle-size range and the effects of mixtures of UO_2 on the attainable pressure-bonded density are in progress.

The effect of adding ceramic grade uranium dioxide to both the fused and special dense oxides was investigated. The object of this approach was to achieve the major part of densification during pressure bonding with the ceramic grade material. The results, which are given in Table E-1, do not show a definite density trend with the amount of ceramic grade addition, although the highest density was attained with a 25 w/o ceramic grade addition to the fused uranium dioxide. In general, higher pressure-bonded densities were achieved than those previously attained with the as-received fused and special dense powders.

Flash roasting to attain off-stoichiometry uranium dioxide powders is being studied as a possible means of activating the powders prior to pressure bonding. Previous tests have shown that the off-stoichiometry powders return to stoichiometry during the pressure-bonding operation. The results of initial flash-roasting tests with the special dense oxide are given in Table E-2. In addition to flash roasting of the fused oxides, further work will involve the compacting and pressure-bonding characteristics of such powders.

TABLE E-1. EFFECT OF ADDITIONS OF CERAMIC GRADE UO_2 TO FUSED OR SPECIAL DENSE UO_2 POWDERS ON PRESSURE-BONDED DENSITY

Type of UO_2 Compact	Green Density		Postbonding ^(a) Density	
	Average, g per cm^3	Per Cent of Theory	Average, g per cm^3	Per Cent of Theory
90 w/o fused (as received)-10 w/o ceramic (minus 325 mesh)	8.63	78.7	10.1	92.2
85 w/o fused (as received)-15 w/o ceramic (minus 325 mesh)	8.46	77.2	9.78	89.2
80 w/o fused (as received)-20 w/o ceramic (minus 325 mesh)	8.35	76.2	10.3	94.0
90 w/o MCW special dense (as received)-10 w/o ceramic (minus 325 mesh)	8.36	76.3	10.1	92.2
85 w/o MCW special dense (as received)-15 w/o ceramic (minus 325 mesh)	8.18	74.6	10.3	94.0
80 w/o special dense (as received)-20 w/o ceramic (minus 325 mesh)	8.06	73.5	10.1	92.2
75 w/o fused (as received)-25 w/o ceramic (as received)	8.32	75.9	10.5	95.8

(a) Bonding conditions for all compacts: 3 hr at 2100 F and 10,000 psi.

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E-3 and E-4

TABLE E-2. EFFECT OF FLASH ROASTING ON UO_2 STOICHIOMETRY

Sample ^(a)	Heat-Treating Data		Oxygen-to-Uranium Ratio
	Time, min	Temperature, F	
SD-3	5	400	2.02
SD-6	5	450	2.01
SD-9	5	500	2.02
SD-12	5	550	2.04
SD-15	5	600	2.08
SD-18	5	650	2.09
SD-20	2	700	2.05
SD-21	5	700	2.22

(a) All samples were special dense UO_2 with an oxygen-to-uranium ratio of 2.00.

Gas-Pressure Bonding

Included in the gas-pressure-bonding studies is the determination of the densification, permeability, and thermal-conductivity characteristics of various types and combinations of UO_2 powders in the form of both tapped powders and cold-pressed pellets. Investigations are being conducted to evaluate various fuel-element shapes for the simultaneous densification and cladding of the UO_2 powders. Also under study are various fuel-element shapes containing high-density UO_2 which lend themselves to pressure bonding and which cannot be produced by other conventional fabrication methods.

Several tubes of both cold-pressed pellets and tap-packed powders of various types of UO_2 were pressure bonded. These were prepared to evaluate different pressure-bonding cycles with identical specimens, establish the effect of select particle-size mixtures upon densification, and produce permeability and thermal-conductivity test specimens. These specimens are in process of being evaluated.

In a process using powdered stainless steel for a spacer material and for end plugs, it was demonstrated that the stainless steel can be densified to 97.3 per cent of theoretical during pressure bonding. Further work will be aimed at matching the consolidation of the stainless steel powder components to that of the uranium dioxide fuel.

F-1

DEVELOPMENT OF URANIUM CARBIDE (AEC-FUEL CYCLE)

F. A. Rough and W. Chubb

As part of the USAEC Fuel-Cycle Development Program, research on the preparation and properties of uranium carbides is in progress. This program includes investigations of the preparation and forming of uranium carbides by powder metallurgical and by melting and casting techniques. The mechanical, physical, and chemical corrosion and compatibility characteristics of uranium carbides and their alloys are being determined. A study of diffusion rates in the uranium carbides has been undertaken, and an effort is being made to understand the nature of irradiation-damage processes in carbides.

Data reported during the first 10 months of research have included information on the variation of density and resistivity with composition, information on the compatibility of carbides with common metals, strength data, and diffusivity data. In addition, the arc-casting process has been scaled up to produce 5-kg castings, and alloys of uranium monocarbide have been found which show increased strength and corrosion resistance.

Work during the last month resulted in a compact pressed and sintered to 89 per cent of theoretical density at a composition of approximately 6 w/o carbon. A series of castings 3/4 in. in diameter and 8 in. long has been prepared by skull casting and has been found to contain between 4.78 and 5.26 w/o carbon. This indicates considerable progress in compositional control in the skull-casting process.

Four compression tests on uranium-4.8 w/o carbon alloys yielded an average compressive strength of 54,100 psi. This is in excellent agreement with the previously reported value of 54,500 psi. Two compression tests on uranium-7.0 w/o carbon alloys gave an average compressive strength of 78,300 psi. This increased strength at this composition is in agreement with the trend indicated by hardness data.

Data on the rate of self-diffusion of uranium in uranium monocarbide indicate that the rates of self-diffusion of uranium in uranium monocarbide are over 100 times lower than the rates of interdiffusion of uranium and carbon in the carbide. By implication, carbon must diffuse much faster than uranium in these carbides.

Alternate Fabrication Methods for UC

S. J. Paprocki, D. L. Keller, D. E. Kizer, and J. M. Fackelmann

Various techniques are being studied for producing UC powder and dense UC compacts by methods other than arc melting. The reaction of fine uranium powder with methane or propane has been accomplished at 700 and 540 C, respectively, to produce fine UC powder of near stoichiometric composition. The methane reaction

yields a product containing UC_2 and a small quantity of an unknown as determined by X-ray diffraction. The propane reaction product has a very strong but diffuse pattern of UC, showing no UC_2 nor alpha uranium, and a small quantity of an unknown. Recently, the reaction of these alkanes with uranium has been studied in an apparatus employing a stagnant partial pressure of alkane gas and hydrogen. The partial pressure of hydrogen in the system has been varied from 40 mm of mercury to 0.7×10^{-3} mm of mercury with no marked effect upon the rate of reaction with methane or propane. Also, increasing the reaction temperature by 100 C has had no marked effect upon the rate of reaction with methane and propane. The apparatus is being modified to pass the alkane gas over the uranium in an attempt to increase the reaction rate.

Uranium carbide powders produced by the uranium-methane reaction were sintered 1 hr in vacuo at a temperature of 1950 C. The average density increased from 7.62 to 11.17 g per cm^3 . This powder, containing 5.98 w/o total carbon, was described in BMI-1409. As may be seen by comparing these data with previously reported data, raising the sintering temperature from 1900 to 1950 C has little effect on densification.

Compacts of UC mixed with 15 volume per cent of metallic chromium have been sintered at 1800 and 1900 C to help understand the effect of a liquid phase on the densification of these compacts. No bulk-density measurements could be made because the specimens fractured during the heat treatment; however, these specimens will be examined metallographically and chemically in an effort to obtain useful information.

In addition, samples of uranium hydride plus 5 w/o carbon are being vacuum hot pressed in an attempt to achieve a high green density. These compacts will then be reacted at higher temperatures to form UC.

Melting and Casting Techniques for Uranium-Carbon Alloys

B. C. Boesser, W. M. Phillips, E. L. Foster, and R. F. Dickerson

Reliable techniques for the production of high-quality cast shapes of uranium carbide are being developed. Large cast shapes of uranium carbide currently are being prepared by skull-type arc melting.

As reported previously, it has been determined that the major difficulty in attaining compositional control is segregation in the carbide skull in which the melts are made. Improvements in charging practices and melting techniques have decreased this segregation, although samples taken from several spots in the skull showed carbon compositions from 4.5 to 5.6 w/o. The improvement in skull homogeneity is demonstrated by the analyses of castings produced this month. Several 2-in. -diameter by 8-in. -long castings were cast into cold molds and found to be homogeneous and sound and to possess carbon contents ranging from 4.3 to 5.3 w/o. Even better compositional control was attained in a number of 3/4-in. -diameter by 8-in. -long ingots that incidentally were cast in molds heated to 2200 F. These castings were sound and homogeneous and possessed very good surfaces. These ingots ranged in composition from 4.78 to 5.26 w/o carbon.

F-3

The remaining apparent lack of composition control may be attributable to skull inhomogeneities and to variable pickup of carbon from the electrode tip.

Research during the next month will involve the production of 3/4-in. -diameter by 8-in. -long cylindrical shapes of uranium carbide with a nominal content of 5 w/o carbon. The realization of compositional control will be sought by homogenization of the skull, by standardization of melting times, and by study of the effect of atmospheres on electrode-tip erosion and the gas content of the cast shapes.

Metallurgical and Engineering Properties of Uranium Monocarbide

W. M. Phillips, W. L. Foster, and R. F. Dickerson

Uranium carbide is being studied in terms of such properties as density, resistivity, thermal conductivity, strength, and corrosion resistance. To both define and improve the characteristics of this material, the variables of impurity content, carbon content, alloy content, and heat treatment are being evaluated in relation to the above properties.

Previous work has indicated that additions of molybdenum, niobium, and tantalum to uranium monocarbide result in a considerable increase in strength, while additions of the carbides of molybdenum, titanium, and vanadium increase the corrosion resistance of uranium monocarbide in Santowax. Specimens of both the monocarbide and sesquicarbide containing 1, 5, 10, or 20 w/o additions of metal and their carbides are currently undergoing physical, mechanical, and corrosion testing.

As shown by Table F-1, the average transverse rupture strengths of the uranium-carbon binary alloys are considerably less than the compressive rupture strengths. It has been observed that the transverse rupture strength tends to decrease as the time between machining and testing is increased. This observation suggests that the transverse rupture strengths shown in Table F-1 may be highly dependent upon the surface condition of the specimens.

It is hoped that current tests on uranium carbide containing ternary metal additions will show whether the apparent strength increase in transverse rupture testing is a true strength increase or is a phenomenon resulting from a reduction or elimination of surface microcracking. As part of the above program, determinations of the modulus of elasticity and strain at rupture are being made.

Uranium Monocarbide Diffusion Studies

W. Chubb, R. W. Getz, and F. A. Rough

The rate of self-diffusion of uranium in uranium monocarbide is being studied using a tracer technique. Interest in diffusion rates in uranium monocarbide is based

F-4

TABLE F-1. STRENGTH OF UC AND U_2C_3

Material	Average ^(a) Transverse Strength, psi	Average ^(b) Compressive Strength, psi
U-4.7 w/o C	7,400	--
U-4.8 w/o C	8,900	54,100
U-4.9 w/o C	5,500	--
U-7.0 w/o C	13,300	78,300

(a) At least four tests run at each composition.

(b) Four test were run on 4.8 w/o carbon specimens;
two tests were run on 7.0 w/o carbon specimens.

on the consideration being given this material as a reactor fuel for high-temperature operation. Earlier in the study the rate of interdiffusion of uranium and carbon in uranium carbide was determined between 1600 and 2000 C. The techniques needed for measuring the self-diffusion rate of uranium in uranium monocarbide have been tested, and diffusion couples of uranium monocarbide have been analyzed. The analyses of these couples indicate that the self-diffusion coefficient for uranium in uranium monocarbide is about 100 times less than the interdiffusion coefficient determined previously. Currently, additional diffusion couples are being prepared and evaluated.

Diffusion couples for the current study are prepared by placing a 0.001-in. -thick metal foil of enriched uranium (93 per cent uranium-235) between two 0.25-in. -lengths of depleted uranium (0.04 per cent uranium-235) monocarbide rod. Preparation of couples for annealing includes wrapping the couple in tantalum foil and placing it in a degassed graphite jig for a bonding operation of 4 hr at 1400 C. After annealing for a suitable length of time at a higher temperature, the couple is sampled for analysis of uranium-235 by grinding the couple on 600-grit silicon carbide powder in the presence of a small amount of 0.1 N nitric acid solution.

In a diffusion couple annealed for 120 min at 2000 C, a diffusion zone approximately 0.018 in. in width was formed. Measurement of the uranium-235 present in the individual samples from this couple was obtained by measuring their gross gamma activity. It has been found that the samples generally contain less than 10 per cent uranium-235, and it is believed that in this range, the gross-gamma-counting method is more sensitive, and thus more satisfactory, than the neutron-activation method of analysis. Using these analyses, self-diffusion coefficients for uranium in uranium monocarbide were calculated. The results of these calculations for the two couples examined to date are as follows:

Temperature, C	Self-Diffusion Coefficient, D, cm ² sec
1800	$\sim 3 \times 10^{-9}$
2000	$\sim 1 \times 10^{-8}$

F-5 and F-6

Two other couples were annealed and examined metallographically. The couples appeared to be satisfactorily bonded and annealed. One couple was annealed for 240 min at 1900 C, and the other was annealed for 60 min at 2100 C. These couples will be sampled for analysis for uranium-235.

Future work will include the preparation and analysis of additional diffusion couples.

Irradiation Effects in UC

A. E. Austin, D. G. Freas, and C. M. Schwartz

The lattice strain and property changes induced in uranium monocarbide by neutron irradiation is presently being studied. Specimens of uranium monocarbide containing both natural and 10 per cent enriched uranium will be studied. The irradiations are to be carried out at low temperatures (500 to 600 F) and are to include burnups of 0.01 to 0.02 per cent of the uranium for specimens made of natural uranium and burnups of 0.02 to 1.0 per cent of the uranium for specimens containing 10 per cent enriched uranium. A total of about 36 specimens will be irradiated. The specimens will be about 1/2 in. long and 1/4 in. in diameter, and will be contained in NaK during irradiation to insure proper heat-transfer conditions.

The natural uranium specimens have been prepared by drop casting and have been radiographed to insure soundness. The inspected castings were stress-relief annealed for 1 hr at 1800 C. Each casting was cut into three pieces; including two specimens for irradiation and one control specimen. The preirradiation examinations of each specimen will include dimensional, density, and electrical-resistivity measurements and a final X-ray examination. A chemical analysis and a metallographic examination is to be performed on sections of each control specimen. Because of the small size of each irradiation specimen, it has been necessary to design a special electrical-resistivity apparatus which will give reliable measurements and which can be remotely operated.

The design and fabrication of the capsules and encapsulation of the specimens is to be completed during March. The capsules will be irradiated in the Battelle Research Reactor.

G-1

PHYSICAL RESEARCH (AEC-DR)

F. A. Rough

In research on methods of preparing high-purity crystals of UO_2 suitable for basic studies, some success has been achieved in producing crystals by two methods involving deposition from vapor. Work is also in progress to determine the feasibility of preparing crystals by crystal pulling from a fused bath of UO_2 and by zone melting.

Fusion Methods to Prepare Single Crystals of UO_2

W. P. Allred and B. Paris

Research on the growth of single crystals of UO_2 was continued during February. The major effort during this period was on the growth of crystals by pulling from a molten pool of UO_2 . A crucible consisting of an alloy of about 90 w/o tungsten, 5 w/o nickel, and 5 w/o copper was used to contain the melt. A 20-kw 450-kc r-f generator was used to heat the crucible. With the generator at full power the UO_2 was not completely molten. It was noted that the molten UO_2 wet the crucible. This observation is in direct contrast with the lack of wetting observed when pure tungsten is used.

Tantalum shields will be placed around the crucible. This should allow a higher temperature to be reached and thus provide for complete melting of the UO_2 samples. Further experiments on crystal pulling will use this arrangement.

A second method for the growing of large single crystals is under investigation. It is planned to zone refine the UO_2 in a horizontal tungsten tube. The power will be supplied by passing a high current through the tube. The zone will be formed by a tantalum shield which will surround a short section of the tube. The portion of the tungsten under the shield will be the hottest area of the tube. Proper adjustment of the current should melt the UO_2 beneath the shield while the remainder of the UO_2 ingot remains solid. The shield will then be moved along the ingot, thus producing a moving zone. In addition to the growth of crystals this system will be capable of purifying the UO_2 by zone refining. This equipment is being assembled and will be in operation during March.

Growth of UO_2 Crystals From the Vapor Phase

C. A. Alexander and R. B. Filbert, Jr.

During the past month the apparatus was modified to prevent the plugging that had been prevalent in earlier runs when using the reaction

to produce crystals of UO_2 . In a 100-hr run, several single crystals ranging in size

G-2

from 6 to 10 mm in length, as well as a large quantity of small cube-shaped crystals about 1 mm^3 in size, were grown from the vapor. Conditions for this run were a charge temperature of 1050 C and a flow rate of 2 cm^3 per min of water vapor, over the charge. The amount of water vapor admitted was the stoichiometric amount needed to react with the vapor at a temperature of 1050 C .

In addition to the above reaction, runs utilizing the evaporation of KF from a molten-salt solution of KF and UO_2 were made. To date, no crystals larger than 3 mm have been grown by this process, but the shape of the crystals grown in this manner is more regular than the larger ones grown from the $\text{UF}_4\text{-H}_2\text{O}$ reaction. For this reason, more runs are being planned utilizing KF as a solvent for UO_2 .

One KF run was made in which no appreciable amount of salt was evaporated from the solution. Instead, the temperature was gradually reduced at a rate of about 1 C per hr. The purpose of this run was to determine if crystalline UO_2 could be produced by a reduction in temperature of the solvent alone. Results of this run indicated that evaporation of the solvent salt was much more effective in producing crystals of UO_2 .

It is planned to continue experiments utilizing the $\text{UF}_4\text{-H}_2\text{O}$ reaction and evaporation of solvent during the next month in an effort to produce larger single UO_2 crystals.

H-1

RADIOISOTOPES AND RADIATION APPLICATIONS (AEC-OID)

D. N. Sunderman

Work is continuing on a series of four research programs undertaken for the Office of Isotopes Development. These programs are the development of radioactive-tracer quality-control systems, the use of intrinsic radioactive tracers for process control, the study of radiation-induced graft polymerization, and the nitration of hydrocarbons.

The quantitative determination of iron by titration to a radiometric endpoint was found to be very sensitive to the concentration of the tartaric acid complexing agent and the sodium acetate buffer. A mole ratio of additive to iron of 3.6 is necessary in the first case and of at least 40 in the second. The method is accurate if these conditions are maintained throughout the titration process. The development of a radiometric method for sulfate is under way, and initial results should be available within a month.

Construction of the iron-removal process equipment is under way. Lag time in procuring equipment will allow work to be directed toward further theoretical studies of intrinsic-tracer applications. The two avenues of study are, first, a search for applications similar in chemical process technology to the iron-removal process, and, second, an evaluation of the general applicability of intrinsic tracers to unit operations in process engineering.

A variety of long- and branched-chain esters of the methacrylate group was studied to determine the relationship of total radiation dose and free-radical-site formation to ester-group structure. It was found that higher site concentrations were obtained with the more highly branched structures for a given radiation dose. The analysis of gas resulting from irradiation of a sample of radiation-polymerized polymethylmethacrylate indicated the presence of monomer and methyl formate. The formation of monomer has been postulated by other investigators, but its presence has not been demonstrated experimentally in previous work.

The effect of ionizing radiation upon the nitration of hydrocarbons has been better defined by recent experiments. Although considerable difficulty is experienced in reproducing the yields of nitrocyclohexane, it appears certain that radiation effects are of minor importance in this system. Several further experiments with NO_2 as the nitrating agent are necessary to fully evaluate this system.

Development of Radioactive-Tracer Quality-Control Systems

R. Lieberman, C. W. Townley, R. A. Ewing, and
D. N. Sunderman

During February, work was completed on the determination of Fe_2O_3 using a complexometric titration with a radiometric endpoint.

The effect of various iron contents on the accuracy of the method as well as the effect of changes in the concentration of the tartaric acid and sodium acetate were

H-2

investigated. Solutions were titrated with both 0.05 M and 0.02 M EDTA. In all runs, $\text{Y}_2^{91}(\text{C}_2\text{O}_4)_3$ was used as the endpoint indicator. The volume of solution was 50 ml.

Using 0.05 M EDTA, it was found that the accuracy of the method decreased when the Fe_2O_3 content approached 74 mg. The use of 0.02 M EDTA introduced appreciable error in analysis, involving 7 and 15 mg of Fe_2O_3 .

It was also found that the analysis is dependent upon tartaric acid concentration, and accurate results could not be obtained in runs where the tartaric acid- Fe_2O_3 mole ratio differed significantly from 3.6. A large excess of sodium acetate buffer was also found to be necessary for accurate results. The sodium acetate- Fe_2O_3 mole ratio must be at least 40. Satisfactory results could not be obtained with quantities of sodium acetate lower than this. The results of these investigations are summarized in Table H-1.

TABLE H-1. EFFECTS OF REAGENT CONCENTRATIONS ON Fe_2O_3 DETERMINATIONS

Iron Added as Fe_2O_3 , mg	Molarity of EDTA	Mole Ratio		Initial pH	Iron Found as Fe_2O_3 , mg	Absolute Error, per cent
		Tartaric Acid: Fe_2O_3	Sodium Acetate: Fe_2O_3			
7.36	0.0200	3.60	43.2	5.0	7.50	1.90
		3.60	43.2	5.0	7.61	3.40
14.72	0.0200	3.60	43.2	5.0	14.72	0.00
		3.60	43.2	5.0	15.09	2.51
		3.60	43.2	5.0	15.04	2.17
		3.60	43.2	5.0	14.52	-1.36
36.81	0.0499	3.60	43.2	5.0	14.68	-0.27
		3.60	64.8	5.2	37.02	0.57
		6.06	64.8	4.9	--(a)	--
		3.60	21.6	4.6	--(a)	--
		1.80	43.2	5.2	--(a)	--
		2.02	21.6	4.7	--(a)	--
73.62	0.0499	3.60	43.4	5.0	74.13	0.69
		3.60	43.4	5.0	76.82	4.35
		2.88	35.8	5.0	67.26	-8.65
		1.94	21.6	4.8	--(a)	--

(a) Endpoint could not be observed.

Several values presented in the table of results of iron determinations given in last month's report (BMI-1409) were in error. The Fe_2O_3 concentrations presented in that table were high by a factor of 2. The corrected values are presented in Table H-2. These determinations were made with a 50-ml solution volume, a tartaric acid- Fe_2O_3 mole ratio of 3.60, and a sodium acetate- Fe_2O_3 mole ratio of 43.2. The best results were obtained under these conditions. The Fe_2O_3 concentration recommended for optimum accuracy is 20 to 40 mg of Fe_2O_3 per 50 ml.

Work has begun on a radiometric method for the determination of sulfate in portland cement. Strontium-89 will be used to precipitate $\text{Sr}^{89}\text{SO}_4$. The observed change in radioactivity of the sample as the result of precipitation will be used to determine the sulfate content. It is expected that this method will be applicable directly to cement samples, with no prior separations necessary.

H-3

TABLE H-2. PRECISION AND ACCURACY OF EIGHT Fe_2O_3 DETERMINATIONS(Iron added, expressed as $\text{Fe}_2\text{O}_3 = 37.12$ mg.)

Determination	Fe_2O_3 , mg	Deviation From Average, mg	Absolute Error, per cent
1	36.82	-0.40	0.81
2	37.14	-0.08	0.05
3	37.29	+0.07	0.46
4	37.22	0.00	0.27
5	37.33	+0.11	0.57
6	37.81	+0.59	1.86
7	37.17	-0.05	0.13
8	37.02	-0.20	0.27
Average values	37.22	± 0.18	--
Standard deviation	--	± 0.29	--

An investigation of other industries in which radioisotopes might be applied to quality control has begun. This investigation will continue in order to select an industry for study when work has been completed with the cement industry. The final choice will be made after consultation with the Office of Isotopes Development.

Use of Intrinsic Radioactive Tracers for Process Control

J. L. McFarling, P. Gluck, J. F. Kircher, and D. N. Sunderman

During February, work has continued on the construction of the model iron-removal process system. Several of the equipment items are ready for installation; however, delays in delivery of other items have slowed construction. These delays are not expected to be long or to seriously affect the experimental program as planned. Construction will be accelerated during the coming month. In the meantime, emphasis has been shifted to the theoretical phases of the program. Attention has been directed toward other applications of radiotracer-controlled iron removal similar to the ammonia-leach nickel-refining process. This phase of the program is being broadened by surveying the general field of hydrometallurgy in an attempt to uncover potential intrinsic radiotracer applications in addition to iron removal. The present approach is to investigate the use of any applicable intrinsic radiotracer to facilitate the removal of critical impurities in various hydrometallurgical processes. It is felt that the radio-tracer techniques developed during the present engineering study of iron removal will be directly applicable to the majority of potential applications uncovered in the investigative survey.

The study of chemical engineering unit operations to determine general conditions for application of intrinsic radiotracers is just beginning. This phase of the program will receive increased emphasis during the coming month.

Future work will be concerned with completion of the model process as quickly as possible and continuation of the theoretical studies.

Graft-Polymerization Studies

I. S. Ungar, W. B. Gager, J. F. Kircher, and R. I. Leininger

During February the investigation of the mechanism of formation and decay of radiation-induced free radicals continued. A study of the effect of total dose on site formation was made for polypropylmethacrylate (PPMA), polyisopropylmethacrylate (PisoPMA), polyisobutylmethacrylate (PisoBMA), and poly-secondary-butylmethacrylate (PsecBMA). Sites per monomer unit versus total dose were plotted for each of the samples. The results from PisoBMA and PsecBMA were compared with those previously found for polybutylmethacrylate (PBMA) and poly-tertiary-butylmethacrylate (PtertBMA). It was found that at a given dose the site concentration forms a series where $P_{tertBMA} > P_{secBMA} > P_{isoBMA} > PBMA$. In other words the greater the branching of the ester chain, the greater the site concentration for a given dose. It was also found that the less highly branched esters reached their maximum site concentration at lower doses, and the concentration decreased sooner with increasing dose. Similar results were obtained for PPMA and PisoPMA. The PPMA had fewer sites at a given dose than PisoPMA, and reached its maximum site concentration at a lower dose.

During this month, a sample of radiation-polymerized polymethylmethacrylate (PMMA) was irradiated to approximately 4×10^7 rads, and the vapors produced were analyzed by gas chromatography. Two esters, methylmethacrylate and methyl formate, were identified. The presence of monomer is significant since its presence has often been postulated but not proven. Methyl formate, which has previously been reported, was found in 25 times the concentration of methylmethacrylate. It thus becomes apparent that the action of gamma irradiation is not limited to chain scission. The presence of methyl formate and a two-order-of-magnitude increase in site concentration between PBMA and PtertBMA indicates that the ester group is attacked and that the nature of the ester group may determine to a large extent the ease with which sites are formed.

During the next month work will continue to determine the mechanism of radical formation in methacrylates. The vapors from irradiated PMBA's will be analyzed. An attempt will be made to correlate the products of irradiation with the number of sites formed and the shape of the EPR spectrum.

Information has been received that a short paper based on this work has been accepted for presentation at the Gordon Conference on Radiation Chemistry this summer.

Nitration of Hydrocarbons

M. J. Oestmann, G. A. Lutz, E. J. Kahler,
and J. F. Kircher

During February, the effect of radiation on the nitric acid-cyclohexane system was investigated under conditions of more efficient mixing of the reactants. Seven irradiation and thermal experiments were completed. Analytical results are reported in Table H-3.

TABLE H-3. PRODUCT ANALYSES OF NITRIC ACID-CYCLOHEXANE EXPERIMENTS^(a)

Experiment	Temperature, C	Time, hr	Gamma Dose, 10 ⁶ rads	Moles of Gas Produced	Analysis of Recovered Organic Solution, w/o					Adipic Acid or Other Solids
					C ₆ H ₁₁ NO ₂	(C ₆ H ₁₁) _m (NO ₂) _n	(C ₆ H ₁₁) ₂	C ₆ H ₁₁ OH	C ₅ H ₁₀ C=O	
56	60	20	3.8	0.007	0.75	--	--	Trace	Trace	0.54
57	60	45	--	0.014	1.4	0.06	0.08	0.09	0.02	2.20
54(b)	60	45	8.6	0.018	4.5	0.21	0.11	0.20	0.04	1.92
55(b)	60	45	--	0.017	3.3	Trace	Trace	0.06	Trace	1.81
59	60	65	12.4	0.018	1.1	0.14	0.44	0.25	0.13	1.95
53	60	65	--	0.019	1.6	--	--	0.13	0.02	0.96
58	110	20	--	0.029	3.9	--	0.23	0.18	0.04	1.11
46	60	45	8.6	0.020	1.4	0.12	0.05	0.17	0.02	1.67

(a) All experiments were conducted in glass vessels using a 10/1 mole ratio of cyclohexane/nitric acid.

(b) Reaction mixture consisting of nitric acid and cyclohexane spiked with 1.4 w/o C₆H₁₁NO₂.

H-5

A 10/1 mole ratio of cyclohexane/nitric acid was used in all experiments. The glass reaction vessels used were designed to keep the magnetic stirring bar centered. Good stirring was assured in these experiments. All experiments except 58 were conducted at 60 C for periods of 20 to 65 hr. The corresponding gamma doses were from 3.8×10^6 to 1.2×10^7 rads.

Table H-3 shows the approximate moles of gas produced during each reaction. Gas samples from Experiments 56, 59, and 58 were analyzed by mass spectrometry. The major component was nitrous oxide, amounting to about 55 volume per cent. From proposed reaction mechanisms*, this is the expected result. Smaller yields of nitrogen, nitric oxide, carbon dioxide and monoxide, and hydrogen were obtained. Less than 1 volume per cent each of argon, methane, oxygen, and water was obtained.

The distribution of product yields was about the same in each experiment. There was, however, a slight increase in the yield of oxidized products obtained with an increase in reaction temperature from 60 C (Experiment 56) to 110 C (Experiment 58).

To confirm the results obtained previously on the radiation stability of nitrocyclohexane, Experiments 54 and 55 were conducted for 45 hr using cyclohexane spiked with 1.4 w/o nitrocyclohexane prior to reaction. Approximately the same yields were obtained in the spiked reactions as in the unspiked reactions (Experiments 46 and 57). Therefore, no significant degradation of nitrocyclohexane occurred during radiation or heating.

The accumulated data show poor reproducibility in product yields from either irradiation or nonirradiation experiments. In comparing irradiation and thermal experiments, the range of nitrocyclohexane yields overlaps even for experiments with different exposure times. This may be due to difficulty in obtaining identical experimental conditions and errors in the analytical procedure. Consequently, no definite conclusion as to the effect of radiation on nitration can be drawn. It appears, however, that radiation does not produce large effects on nitration under the particular experimental conditions used.

Five experiments using NO_2 as the nitrating agent were completed. Reactions were conducted at 60 C with a 10/1 mole ratio of cyclohexane/nitrogen dioxide. Results of these experiments will be reported as soon as they become available. Next month additional NO_2 experiments will be continued.

*Topchiev, A. V., Nitration of Hydrocarbons, Pergamon Press, New York (1959), pp 167-168.

I-1 and I-2

VOID-DISTRIBUTION AND HEAT-TRANSFER
STUDIES (AEC-EURATOM)D. V. Grillot, R. O. Wooton, M. F. Peter,
H. M. Epstein, and J. W. Chastain

During February experimental measurements of voids resulting from subcooled boiling continued. The ranges of parameters used in the tests were the following:

Pressure	600 to 1200 psia
Reynolds number	Up to 52,500
Heat flux	0.2×10^5 to 2.0×10^5 Btu/(hr)(ft ²)

The over-all stability of the tests has improved greatly, but the effects of long-term drifts are still noticeable. However, the drifting is slow enough so that its effect on relative measurements is nearly negligible.

A specially designed chamber with a high beta-to-gamma sensitivity ratio is being aged at present and should be ready for trial operation next month. This system should give better statistical data in a given time period than the Geiger counter now in use.

J-1

MATERIALS DEVELOPMENT AND EVALUATION (HAPO)

F. R. Shober

The creep properties of annealed and of 15 per cent cold-worked Zircaloy-2 are being investigated under a number of conditions of stress and temperature. Studies to develop a AgBr fuel-element leak detector for use in water-cooled reactors have been concluded.

Experimental work on the development of a thermal-neutron-flux monitor is being discontinued. In a program to develop corrosion-resistant welding alloys for use with Hastelloy F, 12 new alloys have been selected. The selection was based on the corrosion and metallurgical evaluation of the first series of alloys. Melting and fabrication will be initiated in March.

Mechanical Properties of Zirconium Alloys

L. P. Rice and J. A. VanEcho

This program is concerned with determining creep properties of Zircaloy-2 sheet material under various conditions of stress and temperature. Such information is necessary in the design of pressurized process tubes, the Plutonium Recycle Program, and other applications at Hanford utilizing the Zircaloy-2 alloy.

The program consists essentially of three phases. Phase I is concerned with investigating stress-rupture, long-time creep, conditions of time and deformation for onset of third-stage creep, and the effect of time and stress on the ductility of 15 per cent cold-worked material. Temperatures of interest are 550, 650, and 750 F. Stresses range from those causing rupture in a few hours to very low stresses resulting in creep rates of about 0.000001 per cent per hr. Tests may run for as long as 15,000 to 20,000 hr in obtaining such low creep rates. In this and the other phases of the present program, all tests are made in vacuum of the order of 0.001 to 0.0001 mm of mercury.

Phase II is concerned with the effect of temperature cycling on creep properties and ductility of annealed and of 15 per cent cold-worked Zircaloy-2 sheet. Temperatures of interest here are 550 and 650 F. The cycle consists of 6 days at temperature and 1 day at room temperature with the specimen continuously under stress. Times involved are 3000 hr per test at temperature or 3500 hr of total time. The stresses chosen result in creep rates of about 0.000005 to 0.0005 per cent per hr.

This phase of the program has just lately been expanded to include determining the effect of stress as well as temperature cycling on the creep properties and ductility of annealed Zircaloy-2. Both stress and temperature are being cycled 1 day out of 7 with the stress reduced to zero and the temperature lowered to room temperature. Otherwise, the procedure is similar to that of the temperature-cycled tests.

Phase III of the program is much the same as Phase I but involves annealed rather than cold-worked material. Conditions of interest include temperatures of 550, 650, and 750 F and stresses that will result in rupture as well as creep rates as low as 0.000001 per cent per hr. Maximum test times are expected to be in the vicinity of 10,000 hr.

Thermal-Neutron-Flux Monitoring System

D. R. Grieser, P. M. Steinback, J. W. Lennon, and W. H. Goldthwaite

The objective of this program is the development of a power-balancing-type thermal-neutron flux monitor.

The program has concentrated on the development of small, reliable, and stable uranium-bearing elements required for insertion into the flux probe. An examination of the experimental evidence garnered from systematic fabrication and electrical-property studies of $\text{MoSi}_2\text{-Al}_2\text{O}_3\text{-UO}_2$ ceramic elements strongly suggests that suitable probe elements cannot be produced with this specific combination.

Experimental work has been suspended, and a topical report is being written. Concurrently, a number of alternate materials combinations and approaches are being evaluated for possible use in the development of a power-balancing device.

Development of Corrosion-Resistant Welding Alloys for Use With Hastelloy F to Contain Decladding Solutions

C. L. Peterson, P. D. Miller, J. D. Jackson, A. M. Hall,
R. E. Monroe, and W. K. Boyd

Based on the corrosion and metallurgical evaluations of the first 12 experimental alloys, a second list of 12 experimental alloys to be melted at BMI was agreed upon during a conference at HAPO. This list is tentative, in that some of the proposed alloys may not be amenable to forging and rolling. The melting program will be carried out during March. The emphasis on the new list of alloys has been changed to considering these alloys, themselves, as possible container materials and not as welding alloys for vacuum-melted Hastelloy F. Consequently, only self-welded coupons of these alloys will be employed in the subsequent corrosion evaluations in boiling Sulfex and Niflex solutions.

K-1

DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS (SRL)

R. J. Carlson and N. E. Daniel

Stress-rupture tests at 200 C using stresses of 7,000 to 11,000 psi have been conducted on an aluminum-35 w/o uranium binary alloy and on an aluminum-35 w/o uranium alloy containing 2 and 3 w/o tin or zirconium. The time to failure under these variable conditions has been from less than 1 hr to more than 200 hr.

Differential-thermal-analysis data indicate the ternary eutectic temperature for the aluminum-uranium-nickel alloy is 621 C. From these data and data obtained from binary alloys of the elements, it is believed that either iron or silicon is effective in depressing the eutectic temperature when nickel is present.

Preparation of Aluminum-Uranium Alloys

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

Aluminum-uranium alloys containing concentrations of uranium in excess of 20 w/o are of interest as reactor fuels if satisfactory casting and fabricating techniques can be developed. The final product sought is a fuel element in the form of a hollow cylindrical tube clad inside and outside with aluminum. Improvements in both the casting and fabricating characteristics of aluminum-35 w/o uranium alloys have been observed when suitable ternary additions are made to the alloy. The improved casting characteristics are attributed to the increased fluidity imparted to the alloys by the additions, and to the smaller compound particle size exhibited by the alloys containing these additions. The improvements in fabricating characteristics are attributed to the inhibition of the UAl_3 -to- UAl_4 reaction. Two additions which are capable of complete retention of UAl_3 under normal casting conditions are tin and zirconium. The program in progress is concerned with the effects of up to 3.0 w/o tin and zirconium on the castability, the corrosion resistance, and the mechanical properties of the aluminum-35 w/o uranium alloy.

Hot-hardness and elevated-temperature tensile tests have been completed, and the data from these tests have been used to determine the stresses and temperatures of subsequent creep and stress-rupture tests. It was decided that stress-rupture data should be obtained at 200 C, with the initial stress approximating the ultimate strength of the alloys. The results of the stress-rupture tests conducted under these conditions were reported in the previous report. Recently, tests have been conducted using reduced stresses at 200 C. The results from these tests are given in the tabulation below.

K-2

Alloy Composition	Stress, psi	Time to Failure, hr	Elongation, per cent	Reduction in Area, per cent
Al-35 w/o U-2.0 w/o Sn	11,000	0.9	19.5	23.0
	8,500	57	9.8	14.4
Al-35 w/o U-3.0 w/o Sn	10,000	1.4	17.9	24
	7,000	130	11.4	18
Al-35 w/o U	11,000	6	8.9	15.8
	9,500	227	8.9	14.3
Al-35 w/o U-2.0 w/o Zr	11,000	4.7	14.6	22.3
	11,000	3	20.3	24.7
	9,000	57	18.7	21.7
	9,000	92.5	17.1	24.2
Al-35 w/o U-3.0 w/o Zr	11,000	3.2	15.4	22.9
	10,000	6.3	18.7	28.9
	8,500	48	13.8	7.9

With the data available from the stress-rupture tests it is now possible to initiate creep testing. It is planned to conduct these tests at 200 C also for periods up to 1000 hr. The effects of two or more elemental additions on the structure of the aluminum-35 w/o uranium alloy is to be evaluated by metallographic examination of 1-in. -diameter by 8-in. -long castings.

A Study of the Aluminum-Uranium-Nickel Eutectic Temperature

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

A program is being conducted to determine the ternary eutectic temperature in an aluminum-13 w/o uranium-5.6 w/o nickel alloy prepared from commercial materials. Previously, a ternary eutectic temperature of 624 ± 2 C was observed for a similar alloy prepared with high-purity components.

Alloys have been prepared for study by induction melting in a zirconia crucible and casting into copper molds. Spectrographic analysis of the aluminum employed in the alloy indicated impurities to be 0.36 w/o iron, 0.07 w/o silicon, 0.01 w/o copper, and 0.006 w/o manganese. The nickel contained 0.1 w/o cobalt, 0.01 w/o silicon, 0.001 w/o copper, and less than 0.003 w/o manganese. The uranium was commercial grade.

Differential thermal-analysis data indicate the ternary eutectic temperature for an alloy prepared with this material is 621 C. Very close temperature agreement was observed on all analytic runs for this alloy. In addition, the aluminum-13.1 w/o uranium and aluminum-5.7 w/o nickel binary eutectic alloys exhibited eutectic temperatures of 640 and 629 C, respectively. In alloys prepared from pure materials, both these temperatures are 640 C. From these data, it would appear that the eutectic-depressing impurity is effective only when nickel is present. It appears probable that either iron or silicon is the impurity responsible for the lowered melting point.

Additional thermal data are being obtained by a comparative differential technique to verify the temperature differences observed.

L-1 and L-2

REFLECTIVE-INSULATION STUDIES (KAISER)

L. J. Flanigan and H. R. Hazard

Studies of reflective-insulation materials suitable for use in the Experimental Gas-Cooled Reactor are being conducted. Suitability of the insulation will be judged by its freedom from distortion or other damage occurring during thermal cycling and rapid decompression and by its insulating value. Previously reported work includes completion of the conceptual design of the heat-transfer apparatus, the decompression chamber, and the thermal-cycling equipment and initiation of the detailed designs.

In February, detailed design of the thermal-cycling equipment was completed and construction started. The heat-transfer apparatus design is about 80 per cent complete, and construction of components has been started. The vessel to be used for decompression studies has been designed, and construction drawings are being prepared.

In March construction of the thermal-cycling equipment will be completed, and the decompression chamber will be built. Design of the heat-transfer apparatus will be finished and fabrication continued.

M-1 and M-2

PROBLEMS ASSOCIATED WITH THE RECOVERY OF
SPENT REACTOR FUEL ELEMENTS (ORNL)Corrosion Studies of the Fluoride-Volatility Process

P. D. Miller, C. L. Peterson, W. N. Stiegelmeier, and F. W. Fink

Sections from a titanium steam coil, part of which failed during service in an ORNL Purex acid-recovery evaporator, were examined. A metallographic study showed no significant structural differences which could account for the fracture. It has been concluded that failure was the result of a mechanical rubbing action by a vertical tube in the assembly.

Study of the Effects of Irradiation on Cladding-
and Core-Dissolution Processes

R. A. Ewing, H. B. Brugger, and D. N. Sunderman

Sulfex Process

Analyses of the dissolutions of the ORR-irradiated Consolidated Edison power-reactor prototype pins have been completed. Results of these studies are contained in a topical report in preparation.

Preliminary Sulfex dissolution experiments of a new batch of "cold" Consolidated Edison prototype pins, of composition more nearly approaching design specifications, have begun. These experiments are to provide reference data for the subsequent dissolution studies of MTR-irradiated high-density fuel pins. Dissolution tests of the irradiated pins can begin as soon as these are released from their present postirradiation studies.

Darex Process

No Darex dissolutions were conducted during the month.

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N-1 and N-2

VARIABLE-MODERATOR REACTOR CRITICAL-ASSEMBLY STUDIES (ATL-A)

R. A. Egen, L. H. Bettenhausen, W. S. Hogan,
D. A. Dingee, and J. W. Chastain

Preparation for the VMR critical-assembly studies continued. The major core components arrived from Advanced Technology Laboratories (American-Standard) and were assembled. The liquid-handling systems were completed, and the control apparatus was nearly all installed.

The first shipment of fuel pins was received. A fuel element was partially assembled to determine future assembly procedures.

A final review of the experimental program was made with ATL personnel, and the tests were sequentially arranged in accordance with their plans for use of the data.

The remainder of the fuel is expected in March. Initial criticality experiments will follow shortly after receipt of the fuel.

O-1

URANIUM CARBIDE AS A REACTOR FUEL (AI)

F. A. Rough

The examination of a capsule of specimens containing nominal 4.8 w/o carbon irradiated to about a 5000-MWD/T uranium burnup has been completed. The results are comparable with those for 4.6 w/o carbon and 5.0 w/o carbon specimens having similar burnups. Some cracking and only minor dimensional density changes were observed.

One additional capsule of specimens is being irradiated to an estimated uranium burnup of 20,000 MWD/T U.

Irradiation of Uranium Monocarbide

D. Stahl, J. H. Stang, and W. H. Goldthwaite

During February, specimen center-line temperatures for Capsule BMI-23-4, which contains uranium monocarbide specimens, were in the 1050 to 1100 F range. The capsule is scheduled for discharge from the MTR at the end of Cycle 140 (mid-June, 1960) for a total irradiation of 26 MTR cycles and a target burnup of 20,000 MWD/T of uranium. This capsule is the final one of this series.

Postirradiation Examination of Uranium Monocarbide

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

The primary objective of this program is to investigate the effects of neutron irradiation and fission-product damage on uranium monocarbide, and to evaluate this material as a possible fuel for sodium-cooled reactors.

As reported previously, the postirradiation examination of specimens from Capsules BMI-23-3 and BMI-23-5 has been completed. It was found that the uranium-5.0 w/o carbon specimens in BMI-23-3 and the uranium-4.6 w/o carbon specimens in BMI-23-5, after being irradiated to nominal uranium burnups of 10,000 MWD/T and 5000 MWD/T, respectively, did not suffer any serious irradiation damage. However, the microscopic examination of the 5.0 w/o carbon specimens revealed a general depletion of the UC_2 structure, due possibly to neutron irradiation, or to a thermal gradient during irradiation.

Examination of Capsule BMI-23-6 was initiated at the BMI Hot-Cell Facility. This capsule, containing specimens with nominal compositions of uranium-4.8 w/o carbon, was irradiated six MTR cycles to estimated burnups of approximately 0.69 a/o uranium or 5100 MWD/T of uranium and 0.72 a/o uranium or 5400 MWD/T of uranium for the top and bottom 2-in. specimens, respectively.

O-2

Upon arrival at the Hot-Cell Facility, the outside dosimeter wires were removed, the capsule was punctured, and the released fission gas was collected in small glass vials for subsequent analyses. Macroexamination of the specimens revealed very little evidence of radiation damage. The specimens generally were in good condition, but contained a few cracks. Some of the NaK had remained in cracks in the surface areas of the specimens. In areas where the NaK had not been completely removed from the cracks, it appeared that the NaK was reacting in air, causing flaking and gradual widening of these cracks.

Microexamination of these specimens revealed some evidences of radial cracking, but less cracking than was observed in the specimens of higher carbon content from the BMI-23-1, BMI-23-2, BMI-23-3, and BMI-23-5 Capsules. A comparison of the post-irradiation structures with the preirradiation structures of the same specimens showed no appreciable differences in grain size. The irradiated structures appeared to be completely normal, and typical of structures of the uranium-4.8 w/o carbon alloy.

Tables O-1 and O-2 list the pre- and postirradiation measurements of dimensions and density for Capsule BMI-23-6.

TABLE O-1. DENSITY OF URANIUM-4.8 w/o CARBON SPECIMENS IN CAPSULE BMI-23-6 BEFORE AND AFTER IRRADIATION

Specimen	Specimen Density, g per cm ³		Density Change, per cent
	Preirradiation	Postirradiation	
AI-41 (top)	13.536	13.380	-1.15
AI-46 (bottom)	13.857	13.640	-1.57

TABLE O-2. DIAMETER MEASUREMENTS OF URANIUM-4.8 w/o CARBON SPECIMENS IN CAPSULE BMI-23-6 BEFORE AND AFTER IRRADIATION

Specimen	Before Irradiation	Specimen Diameter, in.					Diameter Change, per cent
		Reading 1	Reading 2	Reading 3	Reading 4	Average	
AI-41 (top)	0.3738	0.3756	0.3759	0.3751	0.3755	0.3756	+0.5
AI-46 (bottom)	0.3758	0.3779	0.3774	0.3775	0.3779	0.3777	+0.5

O-3 and O-4

An examination of the above data indicates that the uranium-3.8 w/o carbon (stoichiometric) specimens behaved very well at the average surface and core temperatures shown in Table O-3.

TABLE O-3. AVERAGE SURFACE AND CORE TEMPERATURES OF URANIUM-4.8 w/o CARBON SPECIMENS IN CAPSULE BMI-23-6

Specimen	Temperature, F	
	Surface	Core
AI-41 (top)	900	1370
AI-46 (bottom)	565	900

The relatively small changes in density and diameter indicate negligible effects of irradiation on the dimensional stability of UC at this composition.

Burnup data based on radiochemical analyses and dosimetry will be forthcoming in the near future.

P-1

PEBBLE-BED REACTOR MATERIALS (S & P)

W. S. Diethorn and W. H. Goldthwaite

In support of the Pebble-Bed Reactor program Battelle is studying the mechanical properties and fission-gas retention of fueled-graphite spheres. The program consists of laboratory evaluation, fabrication of coated fuel particles, and irradiation-capsule studies.

LABORATORY EVALUATION OF FUELED-GRAPHITE SPHERES

M. C. Brockway and A. K. Smalley

Behavior in impact and compression tests and the coating integrity of coated fueled-graphite spheres are being studied with specimens received from commercial vendors. Most of the work this month was devoted to the evaluation of candidate spheres for Sweep Capsule SP-5. Work in the permeability and self-welding studies will be postponed until laboratory evaluation in preparation for this capsule is completed.

FABRICATION DEVELOPMENT OF Al_2O_3 -CLAD UO_2 FUEL PARTICLESSintered Cladding

A. K. Smalley and W. H. Duckworth

Work in this phase of the program has been terminated.

Vapor-Deposited Cladding

T. E. Cook, J. M. Blocher, and A. K. Smalley

The preparation and evaluation of UO_2 having a vapor-deposited Al_2O_3 cladding are continuing. Batches containing both natural and enriched UO_2 have been prepared and incorporated into graphite spheres for fission-gas release studies.

One natural batch having a cladding thickness of $40\ \mu$ was thermally cycled between room temperature and 2500 F in air, and then weighed. A sample was heated in air in a covered platinum crucible for 15 min, and then allowed to cool rapidly by removing the crucible from the furnace. The sample went through nine of these heat-quench cycles. A small weight loss, comparable to that reported last month (BMI-1409)

P-2

for a similar sample heated in air for 1 hr at 1200 F, was observed after the third cycle. No further weight change was observed after the remaining six cycles. Microscopic examination of the sample after the ninth cycle revealed cracks in about 1 per cent of the particles. Plans are under way to thermally cycle clad particles in an inert atmosphere to determine the effect of heating and quenching.

An enriched batch, in which cladding thicknesses ranged from 50 to 60 μ , was prepared this month. The cladding was applied in five steps. Unlike the case with the natural particles prepared last month, three distinct layers were detected in the cladding. Petrographic analysis of the particles showed that the laminar cladding ranged in thickness from 50 to 60 μ . Chemical analyses showed that the clad particles contain 36.0 w/o of UO_2 . The lattice parameter of the UO_2 (X-ray diffraction) is 5.4710 ± 0.0005 A, equivalent to an oxygen-to-uranium ratio of 2.008 ± 0.005 . The Al_2O_3 is corundum (α - Al_2O_3), having an average crystal size well below 5 μ . The average crystal size of the UO_2 is about 5 μ . The refractive index corresponds very closely to that of stoichiometric UO_2 . Metallographic examination and spectrographic analysis are in progress.

Carburization Studies

A. F. Gerds

A heat treatment (2500 F) of baked compacts containing vapor-deposited Al_2O_3 -clad UO_2 particles in graphite matrices is in progress. Specimens will be examined metallographically after 170, 500, and 1000 hr to determine the compatibility of these coated particles with graphite.

FISSION-PRODUCT RELEASE FROM FUELED-GRAPHITE SPHERES

W. S. Diethorn

In this phase of the program coated, fueled-graphite spheres and clad UO_2 particles are neutron activated and then studied for fission-gas release during postirradiation heating. Spheres showing good fission-gas retention are advanced to the in-pile irradiation study. The in-pile study includes high-flux static capsules and low- and high-flux sweep capsules.

Neutron-Activation Studies

H. S. Rosenberg

Two batches of natural, vapor-deposited Al_2O_3 -clad UO_2 particles and graphite spheres containing particles from these batches were neutron activated this month (Tables P-1 and P-2). Alpha assays of the unirradiated particles in an internal, flow

P-3

TABLE P-1. NEUTRON-ACTIVATION RESULTS FOR NATURAL Al_2O_3 -CLAD UO_2 PARTICLES

Cladding Thickness, μ	Postirradiation Heat-Treatment Temperature, F	Time at Temperature, min	Fission-Gas Release, per cent of xenon-133
40	1500	144	1.3×10^{-4}
	1500	60	$<3.7 \times 10^{-3}$
	2000	50	$<3.7 \times 10^{-3}$
	Total		3.7×10^{-3}
20	1850	100	$<1.7 \times 10^{-2}$
	2250	75	1.7×10^{-2}
	2400	135	1.6×10^{-2}
	Total		3.3×10^{-2}

TABLE P-2. NEUTRON-ACTIVATION RESULTS FOR FUELED-GRAPHITE SPHERES(a)

Sphere	Cladding Thickness, μ	Postirradiation Heat-Treatment Temperature, F	Time at Temperature, min	Fission-Gas Release, per cent of xenon-133
437	20	1650	80	$<8.1 \times 10^{-4}(b)$
		1800	50	1.6×10^{-3}
		2200	60	4.9×10^{-3}
		2350	30	2.6×10^{-2}
		Total		3.3×10^{-2}
449	40 - 60	1500	270	7.5×10^{-4}
		1900 (cooled to 600 F)	60	$<9.4 \times 10^{-4}(b)$
		1900 (cooled to 600 F)	60	$<9.4 \times 10^{-4}(b)$
		1900	135	$<9.4 \times 10^{-4}(b)$
		Total		9.4×10^{-4}

(a) Containing vapor-deposited Al_2O_3 -clad natural UO_2 .

(b) No detectable release, upper limit.

P-4

proportional counter show that uranium contamination in the cladding surface is probably responsible for the xenon-133 release listed at the lowest temperatures in Table P-1. The totals are higher and suggest diffusion release of xenon-133.

Results in Table P-2 show that the claddings are not damaged during sphere fabrication and that thermal-cycling of the spheres does not damage the claddings.

In-Pile Capsule Experiments

D. Stahl, G. E. Raines, and W. H. Goldthwaite

SP-4

The BRR irradiation of this static capsule containing six fully enriched graphite spheres is operating satisfactorily with individual specimen-surface temperatures ranging from 1500 to 1800 F. The planned termination date of this experiment is mid-March, when the estimated fission burnup will be 6 to 7 w/o of the uranium-235. The spheres will be examined in the Battelle Hot-Cell Facility.

SP-5

The target date for BRR insertion of this high-flux sweep-capsule experiment, originally scheduled for mid-March, may be delayed until April. The schedule will depend on specimen availability.

Based on previous sweep-capsule experience in this program, it is planned to follow the release of krypton-87, krypton-88, and krypton-85m and xenon-133 and xenon-135 during the in-pile history. A trap containing steel mesh will be placed in the exit line of the capsule in order to catch solid daughter fission products resulting from the decay of very-short-lived fission gases during residence of the latter in the trap. Radiochemical analyses of the washings from this trap should provide indirect estimates of the rate of release of short-lived gases. A search for the daughters of krypton-89, xenon-140, and xenon-141 is planned.

The capsule and trap are well into the fabrication stage.

SPF-1

Irradiation of this low-flux furnace capsule containing a single UC₂ fueled-graphite sphere with a pyrolytic carbon coating was terminated during January. Latest results of the gas sampling were reported in BMI-1409. The capsule is now being stored in the pool of the BRR. There are no plans to examine the specimen.

P-5 and P-6

SPF-2

Furnace Capsule SPF-2, containing a natural UO_2 fueled-graphite sphere, coated with silicon carbide, was inserted in the BRR on February 10. Gas samples were collected at 200, 1000, 1500, and 1900 F. Fission-gas retention is excellent. Very small amounts of unidentifiable fission-gas species were released below 1500 F. Krypton-87, krypton-88, and krypton-85m and xenon-133 and xenon-135 were found in sufficient amounts in the 1500 and 1900 F samples, collected as long as 4 hr, to permit identification and assay. Krypton-88 is uncertain. The fission-gas assays will be completed in early March and reported next month. Based on the known detection sensitivity for krypton-87, the estimated R/B for this species at 1000 F is about 10^{-6} . One or two additional gas samples are planned at 1900 F before termination of this experiment.

SPF-3

Irradiation of Furnace Capsule SPF-3, containing a fueled-graphite sphere (499, Table P-2), is planned in March. The fuel is minus 100 plus 140-mesh natural UO_2 clad with 40 to 60 μ of vapor-deposited Al_2O_3 . The scheduling and in-pile time of this capsule will depend on the availability of the gas train presently committed to the SP-5 capsule experiment.

7
6

Q-1

TANTALUM AND TANTALUM ALLOYS FOR LAMPRE APPLICATIONS (LASL)

J. H. Stang

In studies of tantalum and tantalum-base alloys being conducted for the LAMPRE program at Los Alamos Scientific Laboratory, recent activity has included the processing of a group of arc-melted binary tantalum alloys to strip stock, additional determinations of the physical properties of irradiated tensile specimens of tantalum, and X-ray lattice-parameter measurements of tantalum-oxygen materials to investigate interstitial solid solubility. The latter are the first experimental data obtained in the phase-identification study of tantalum.

Irradiated tantalum has been found to exhibit markedly different properties than the unirradiated material. The alterations include higher strength and hardness, although ductility is not seriously impaired. To check dosimeter-based tantalum-to-tungsten conversions during the irradiations, chemical analyses for tungsten have recently been made on irradiated samples, tungsten contents indicated by the two methods are, for no apparent reason, considerably different.

Development of Container Materials for LAMPRE Applications

D. C. Drennen, C. J. Slunder, and J. G. Dunleavy

As reported in BMI-1409, 16 button specimens of binary tantalum alloys were produced during January; the alloying additions were hafnium, iron, silicon, thorium, and zirconium. These buttons are now being processed to 0.030-mil strip to provide specimens for LASL corrosion tests. As a first step, the buttons were cold pressed (19 to 40 per cent reduction) to a thickness of about 0.26 in. to break up the cast structure; the pieces were then machined to remove surface contaminants. Generally, the pressing behavior of the alloys was good. However, the tantalum-1 w/o silicon alloy cracked immediately upon application of the load. No attempt was made to press the tantalum-3 w/o silicon alloy since it cracked into several pieces during arc casting.

The processing of the workable alloys is continuing. Concurrently, sections of the arc-cast button specimens are being prepared for microstructural examination and hardness determination.

Effect of Irradiation Damage of Tantalum

C. K. Franklin, F. R. Shober, and R. F. Dickerson

A summation of room-temperature tensile-property data for both irradiated and unirradiated tantalum was presented in BMI-1407. Increases in strength and decreases in ductility were evident as a result of irradiation. Unirradiated tantalum annealed at

Q-2

1400 C for 1 hr showed an 0.2 per cent offset yield strength of about 30,000 psi. After irradiation, the material showed yield strengths as high as 84,000 psi for specimens irradiated for seven MTR cycles. A decrease in tensile ductility was also observed in the irradiated material. However, the irradiated specimens exhibited no brittle behavior when tested in bending.

Increases in the hardness of the material also accompanied irradiation. Hardness impressions were made on both unirradiated and irradiated samples with a Tukon hardness tester, using a Knoop diamond indenter and a 100-g load. The unirradiated tantalum showed an average of hardness of 103 KHN, while the irradiated specimens showed average hardnesses of 274 and 309 KHN for irradiations of four and seven cycles, respectively.

Samples of the irradiated material were dissolved, and the presence of tungsten was determined by colorimetric analysis. The results of this test indicate that the conversion of tantalum to tungsten was not as great as expected. Only 0.7 w/o tungsten was formed in the tantalum during the four-cycle irradiation period, and only about 1.2 w/o tungsten was formed in seven cycles. Dosimeter analyses indicated that sufficient thermal fluxes were present to produce at least twice these amounts. This discrepancy between the chemical analyses for tungsten and the amount of tungsten predicted by dosimeter analyses is not fully understood.

Dosimeter analyses will be made to determine the total fast-neutron flux accumulated during the irradiation periods.

Precipitate Phase Identification and Interstitial-Type Solid
Solubility in Tantalum

J. B. Schroeder, D. A. Vaughan, and C. M. Schwartz

The interstitial solid solubility of carbon, nitrogen, and oxygen in tantalum is being investigated over the temperature range 500 to 1500 C. The composition range of interest will include supersaturated specimens in order to obtain the initial precipitate phases for identification. The purpose of these studies is to aid LASL in understanding the role of interstitial impurities in the corrosion of tantalum by plutonium fuel mixtures.

The X-ray lattice-parameter determinations on the oxygen series containing 0 to 4 a/o addition were completed. The experimental values were consistently larger than those reported in the literature* and those previously measured at BMI for high-purity tantalum.

Additional analytical work is in progress to explain these data. Carbon, hydrogen, and nitrogen are considered to be the most likely contaminants. The preparation of the nitrogen series is being delayed until the present analytical work is completed.

*Wasilewski, R. J., "The Solubility of Oxygen in, and the Oxides of, Tantalum", J. Am. Chem. Soc., 75, 1001-2 (February 20, 1953).

R-1 and R-2

DEVELOPMENTAL STUDIES FOR THE PWR (BAPD)

R. W. Dayton

Further studies of the preparation of large flat-plate PWR fuel plates will be conducted by Bettis, and no further work has been done.

Fabrication of Large-Scale PWR-Type Fuel Plates

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

Bettis is presently considering the preparation of Zircaloy-2-clad compartmented uranium dioxide fuel plates for PWR Core 2 on a production basis. The feasibility of preparing small-scale fuel plates of this type, approximately 15 in. long and 15 core compartments wide by gas-pressure bonding, has been established. During the initial process study, the length of the specimens was increased from 15 to 33 to 40 in., while the width was held constant. Favorable results were also achieved with these larger scale specimens. The final scale-up to full size is being investigated at Bettis for production considerations; consequently, it is not planned to further study bonding of large-scale PWR-type fuel plates.

This completes the work on this project.

S-1

DEVELOPMENT OF MATERIALS FOR THE MGCR AND HTGR (GA)

W. C. Riley

Research on core materials for both the MGCR and HTGR is in progress at Battelle. For the MGCR, the major effort is on development and evaluation of UO_2 dispersions in BeO or Al_2O_3 and dispersions of UC or UC_2 in graphite. Currently, the evaluation consists mainly of static-capsule irradiation in the MTR.

For the HTGR, vapor deposition is being investigated as a method of preparing and coating uranium and thorium carbide powder.

High-Burnup Irradiation Effects in Fuel Materials

W. E. Murr, N. E. Miller, J. E. Gates, and R. F. Dickerson

A program is in progress that is designed to study the radiation stability of ceramic-type fuels after various levels of burnup at specimen-surface temperatures near 1500 F. The fuels, of interest to the MGCR concept, include specimens composed of about 20 volume per cent uranium dioxide in beryllium oxide, 20 volume per cent uranium dioxide in aluminum oxide, 20 volume per cent uranium monocarbide in graphite, and 20 volume per cent uranium dicarbide in graphite.

The evaluation of beryllium oxide-matrix and graphite-matrix specimens consists of the irradiation and examination of 24 specimens in 4 capsules equipped with heaters and thermocouples for maintaining and monitoring specimen temperatures. Each capsule contains two specimens of each type of fuel; each specimen is composed of four cylindrical pellets approximately 0.222 in. in diameter by 0.250 in. long sealed in Type 316 stainless steel tubing under a helium atmosphere.

The initial capsule of this series was irradiated in the Battelle Research Reactor to specimen burnups of about 1.1 to 1.7 uranium a/o at surface temperatures ranging from 1300 to 1550 F. Postirradiation examination revealed that all six specimens were in good physical condition. Detailed information concerning the examination was reported in BMI-1366 and BMI-1377.

The three additional capsules have been in the MTR since the beginning of MTR Cycle 125 and have just completed their tenth irradiation cycle, Cycle 134. The electrical-heater difficulties reported for Capsule BMI-31-1 during MTR Cycle 133 have been corrected, and all three heaters and three of the six original thermocouples in the capsule are again in operation. The remaining two capsules are operating satisfactorily with all three heaters and four of the six thermocouples functional.

Table S-1 gives the thermocouple temperatures and heater-power requirements for maintaining the capsules in the desired temperature range during MTR Cycle 134. Capsule BMI-31-3 is located in the lowest flux position and requires the largest amount of auxiliary heat to operate near 1500 F.

S-2

TABLE S-1. TEMPERATURE AND ELECTRICAL-HEATER POWER CONSUMPTION FOR CAPSULES BMI-31-1, BMI-31-2, AND BMI-31-3 DURING MTR CYCLE 134

Capsule	Electrical-Heater Power Consumption, w	Thermocouple Reading ^(a) , F					
		No. 1	No. 2	No. 3	No. 4	No. 5	No. 6
BMI-31-1	960	--	--	1490 ^(b)	1420	1215	--
BMI-31-2	2200	1380	--	1445 ^(b)	--	1410 ^(c)	1410
BMI-31-3	2880	1440	1510 ^(b)	--	1480	--	1420

(a) Specimen-surface temperatures are calculated to be 25 to 30 F higher.

(b) Controlling thermocouple.

(c) Temperature is erratic.

It is planned to retain all three capsules in the MTR at least until the end of Cycle 143 (approximately August 1, 1960). At this time, the specimens in Capsule BMI-31-1 will have approximately 20 to 25 uranium a/o burnup, and the specimens in Capsule BMI-31-2 and BMI-31-3 will have approximately 12 and 9 uranium a/o burnup, respectively.

A single instrumented capsule containing $\text{Al}_2\text{O}_3\text{-UO}_2$ samples was irradiated in the BRR for six reactor cycles. This capsule has been sent to General Atomic for hot-cell examination.

Carbon-Transport Corrosion Studies

N. E. Miller, D. J. Hamman, J. E. Gates, and W. S. Diethorn

Selected metal and graphite specimens have been exposed to radiation in helium-filled quartz tori designed to promote convective flow of the helium and gaseous impurities past the specimens. All experimental work has been completed, and a topical report is being prepared.

Preparation and Coating of Uranium and Thorium Carbide Powders

J. H. Oxley, R. B. Landrigan, A. C. Secrest,
C. F. Powell, and J. M. Blocher, Jr.

An experimental program is under way to determine the feasibility of preparing uranium and thorium carbide powders and, subsequently, to coat these powders with carbon, niobium carbide, or zirconium carbide films by vapor-deposition techniques. Parallel efforts on both preparation and coating are being carried out.

S-3

The design and construction of the equipment were completed in February, and exploratory studies on the carbon deposition have been initiated. A substitute for uranium and/or thorium carbide powder is being used in this initial phase of the coating study.

It is planned to use a uranium tetraiodide feed material in the initial experiments to prepare uranium carbide. This material was selected because uranium iodides are the most volatile of the uranium halides and can be most readily converted to carbides, and because the common reductant iodides (i. e. , MgI_2 , ZnI_2 , etc.) are the most volatile of the reductant metal halides. The data in Table S-2 show the relative advantages of the iodides. However, if experimental results suggest that a shift to a uranium chloride or fluoride feed would be advantageous, the program will be modified immediately. Uranium tetraiodide has been ordered from a commercial vendor, and a lot of this material has been prepared at Battelle to minimize loss of time due to extended delivery dates for this item.

TABLE S-2. FREE-ENERGY CHANGES FOR REACTIONS INVOLVING URANIUM MONOCARBIDE^(a)

Reaction	Melting Points, C		Boiling Points, C		Free-Energy Change ^(b) at Indicated Temperature, kcal					
	UX ₃	MX ₂	UX ₃	MX ₂	727 C	927 C	1127 C	1327 C	1527 C	1727 C
UF ₃ + C + 3/2 H ₂ → UC + 3 HF	1427	--	2277	--	+67	+56	+44	+32	+21	+10
UCl ₃ + C + 3/2 H ₂ → UC + 3 HCl	1116	--	1727	--	+70	+61	+51	+42	+33	+24
UBr ₃ + C + 3/2 H ₂ → UC + 3 HBr	752	--	1576	--	+69	+63	+57	+50	+44	+41
UI ₃ + C + 3/2 H ₂ → UC + 3 HI ^(c)	757	--	1427	--	+55	+46	+38	+30	+24	+20
UF ₃ + C + 3/2 Zn → UC + 3/2 ZnF ₂	1427	872	2277	1497	+58	+58	+62	+65	+72	+75
UCl ₃ + C + 3/2 Zn → UC + 3/2 ZnCl ₂	1116	365	1727	732	+35	+32	+29	+27	+24	+21
UBr ₃ + C + 3/2 Zn → UC + 3/2 ZnBr ₂	752	394	1576	697	+28	--	--	--	--	--
UI ₃ + C + 3/2 Zn → UC + 3/2 ZnI ₂	757	446	1427	624	- 4	--	--	--	--	--
UF ₃ + C + 3/2 Mg → UC + 3/2 MgF ₂	1427	1263	2277	2227	-71	-71	-70	-61	-54	-48
UCl ₃ + C + 3/2 Mg → UC + 3/2 MgCl ₂	1116	712	1727	1418	-29	-31	-34	-21	-21	-21
UBr ₃ + C + 3/2 Mg → UC + 3/2 MgBr ₂	752	710	1576	1230	-32	-31	-30	-30	--	--
UI ₃ + C + 3/2 Mg → UC + 3/2 MgI ₂	757	650	1427	927	-40	-42	-43	-44	--	--
CH ₄ → C + 2H ₂	--	--	--	--	- 5	-10	-15	-21	-26	-32

(a) Sources of data:

Brewer, L., and Krikorian, J., "High-Temperature Studies: I. Reactions of the Refractory Silicides With Carbon and With Nitrogen; II. Thermodynamic Properties of the Carbides; III. Heat of Formation of the 3μ State of C₂ from Graphite", UCRL-2888 (April, 1955). (Uranium monocarbide.)

Farr, J. D., Huber, E. J., Head, E. L., and Holley, C. E., "The Preparation of Uranium Monocarbide and Its Heat of Formation", J. Phys. Chem., **63**, 1455 (1959). (Uranium monocarbide.)

Glassner, A., "The Thermochemical Properties of the Oxide, Fluorides, and Chlorides to 2500°K", ANL-5750 (1957). (All metal fluorides and chlorides.)

Katz, J. J., and Rabinowitch, E., Chemistry of Uranium, McGraw-Hill Book Company, New York (1951), Paper No. 58 by MacWood, G. E. (Uranium bromides and iodides.)

Quill, L. L., The Chemistry and Metallurgy of Miscellaneous Materials: Thermodynamics, McGraw-Hill Book Company, New York (1950), Paper No. 6 by Brewer, L., Bromley, L. A., Gilles, P. W., and Lofgren, N. L. (Metal bromides and iodides other than uranium.)

Rossini, F. D., Cowie, P. A., Ellison, F. O., and Browne, C. C., "Properties of Titanium Compounds and Related Substances", ONR ACT-17 (1956). (Hydrogen halides, atomic halides.)

(b) The free-energy changes are given for the reactants and products in their normal phases at the indicated temperatures.

(c) HI dissociates to 1/2 H₂ + I at the higher temperatures, so the net reaction is actually more favorable than indicated by the estimated free-energy change. A similar but less pronounced dissociation of ZnI₂ and MgI₂ will also take place at high temperatures.

T-1

DEVELOPMENTAL STUDIES FOR THE SM-2 (ALCO)

S. J. Paprocki

The work presented in this section is being conducted in assistance to Alco Products and is concerned with the development of fuel, absorber, and suppressor materials for the SM-2.

Prototype fuel plates are being fabricated for subsequent irradiation testing in a WTR loop and in the SM-1 reactor core. The fabricated plates will be shipped to Alco for welding into composite elements. Studies concerned with retention of boron as a burnable poison are continuing. These studies include investigation of the effects of fabrication variables and heat treatments in an effort to determine the mechanism of boron loss.

Prototype and alternate SM-2 fuel materials are being irradiated in the MTR and ETR. At the end of March, it is calculated that the burnup will range from about 40 to 70 a/o of contained uranium-235. The capsules will be discharged and returned to Battelle for postirradiation evaluation. Additional capsules instrumented with both thermocouples and auxiliary heaters have been shipped to NRTS for insertion in the ETR.

Materials Development

S. J. Paprocki, D. L. Keller, G. W. Cunningham, A. K. Foulds,
and D. E. Lozier

Fabrication techniques are being developed for use in preparing fuel plates and absorber plates for the SM-2 reactor. Full-size stainless-clad stainless- UO_2 dispersion fuel plates containing fully enriched UO_2 are also being prepared for incorporation in the SM-1 spike core and half-size plates are being prepared for insertion in a WTR loop.

Fuel Materials

The major effort on this program is being directed toward the fabrication of 38 full-size fuel plates and 9 half-width fuel plates which will be used to fabricate fuel elements for insertion into the SM-1 reactor core and a WTR in-pile loop. The composition and dimensions of the fuel-plate cores are listed in Table T-1. Unless otherwise indicated, all plates will contain a 1/2-in. -long by 0.030-in. -thick suppressor section of 16 w/o EU_2O_3 dispersed in an elemental iron-18 w/o chromium-9 w/o nickel matrix. The width will be identical to the fuel section. The full-size fuel plates will be clad with 0.005-in. -thick Type 347 low-cobalt low-tantalum stainless steel. A 500-lb heat of this steel has been prepared from electrolytic iron, chromium, and nickel and a special high-purity niobium. Five 100-lb ingots were cast, forged, and rolled to plate and sheet. Picture-frame packs and cover plates have been machined for fuel-plate preparation. Complete analyses of the fabricated material have not been obtained, but it is known that specifications regarding cobalt and tantalum content have been met. The

TABLE T-1. FUEL PLATES FOR IRRADIATION TESTING IN SM-1 AND WTR REACTORS

Number of Plates	Designation	Fuel	Testing Reactor	Burnable Poison ^(a)	Suppressor	Active Core Size
2	Core II reference	24.2 w/o Geneva UO ₂	SM-1	B ₄ C	None	2.650 by 0.030 by 22 in.
22	SM-2 reference	26 w/o spherical UO ₂	SM-1	ZrB ₂	Eu ₂ O ₃	Ditto
2	SM-2 reference, no boron	26 w/o spherical UO ₂	SM-1	Boron-stainless picture frame	None	Ditto
2	SM-2 reference	26 w/o Geneva UO ₂	SM-1	ZrB ₂	Eu ₂ O ₃	Ditto
2	SM-2 green compact	26 w/o spherical UO ₂	SM-1	ZrB ₂	Eu ₂ O ₃	Ditto
2	SM-2 reference	26 w/o spherical UO ₂	SM-1	NbB ₂	Eu ₂ O ₃	Ditto
2	SM-2 reference	26 w/o spherical UO ₂	SM-1	Coated ZrB ₂	Eu ₂ O ₃	Ditto
2	SM-2 reference UN	24.3 w/o UN	SM-1	ZrB ₂	Eu ₂ O ₃	Ditto
2	SM-2 reference UN	24.3 w/o UN	SM-1	No boron	Eu ₂ O ₃	Ditto
9	SM-2 reference	26 w/o spherical UO ₂	WTR	ZrB ₂	Eu ₂ O ₃	16 by 1.490 by 0.030 in.

(a) Boron content is 0.212 w/o except for the B₄C specimen for which boron content cannot be predicted.

T-3

required maximum loading of cobalt cannot exceed 0.025 w/o and the maximum loading of tantalum cannot exceed 0.01 w/o.

Because of close tolerances required on the suppressor end of the fuel plate, an evaluation of the rolling schedule has been made in an attempt to select a roll schedule that will result in an optimum core structure with minimum tail effects at the suppressor end. The results are shown in Table T-2. The plates rolled at 2000 F or a combination of 2000 and 2200 F were rolled primarily to study the effect on core structure. Since more stringering and fracturing of the UO_2 occurred, it is not proposed to use any of these combinations. Best results regarding suppressor tail effects are obtained when the suppressor goes through on the leading end of the first pass. If the plate is not flipped and reversed on each pass, the tail effects even on the leading edge become accentuated. Also, since one end of the plate is constantly rolled at a lower temperature, a difference in core structure is possible. Since an optimum core structure is obtained when heavy reductions are used on the first pass, it appears desirable to maintain at least a 30 per cent reduction in thickness on the first pass. As indicated in Table T-2, minimum tail effects are obtained when the 30 per cent reduction is used on the first two passes. Very light reductions should produce less tail effects, but an optimum core structure would not be obtained. More passes and longer fabrication times are expected to produce greater boron losses.

Compacts are being blended, pressed, and sintered, and fabrication of full-sized fuel plates has been initiated. Fabrication of the nine WTR plates is in progress and is expected to be completed within a few days.

Studies on boron loss during roll cladding have continued. It has been postulated that the rate of dissolution of ZrB_2 is the rate-controlling factor. To gain additional evidence that the rate of boron removal from the surface is not rate controlling, a series of two specimens was fabricated in which one specimen was pickled to remove scale after each pass. Both specimens were air cooled after each pass and were held in the furnace for an identical period of time. The pickled specimen lost 9.14 per cent boron while the unpickled specimen lost 6.5 per cent boron. The difference in loss can probably be attributed to the removal of boron-containing scale and metal after each pass. If the rate of boron removal at the surfaces were rate controlling, it would be expected that boron loss would be very great on the pickled surface, since the boron is removed after each pass and since oxygen in the atmosphere is more readily available to combine with the boron diffusing to the surface during annealing treatments. Additional studies using boron-stainless alloy cores are being made to verify the assumption that boron removal is not the rate-controlling factor in boron loss.

Comparison of boron loss after a 24-hr anneal at 2050 F indicates that the rate of boron diffusion is not the controlling factor. After a standard fabrication procedure, a 5.65 per cent boron loss was observed and after the additional 24-hr anneal on a similar specimen a 17.8 per cent boron loss was observed. This loss is approximately 0.4 per cent boron per hr at 2050 F, compared with a calculated 15 per cent minimum loss per hr based on diffusion data.

Specimens have also been prepared to study the effect of binders on boron loss. As indicated in Table T-3, there does not appear to be any definite relationship between boron content and the type or amount of binder used in these fabricated plates.

TABLE T-2. END EFFECTS CAUSED BY VARIATIONS IN ROLLING

Plate	Rolling Temperature, F	Rolling Direction	Rolling Reduction, per cent ^(a)			Core End Effects			
						Suppressor End		Stem End	
			First Pass	Second Pass	Balance	Feathering, in.	Tailing, in.	Feathering, in.	Tailing, in.
81	2000	Stem first	40	20	20	0.03	0.22	0.13	--
82	2200	Stem first	30	30	--	--	--	--	--
	2000	--	--	--	20	0.02	0.1	0.04	--
83	2200	Stem first	40	--	--	--	--	--	--
	2000	--	--	20	20	--	0.1	0.07	--
84	2200	Stem first	40	20	20	--	0.1	0.07	--
85	2200	Stem first	30	30	20	--	0.15	0.05	--
86	2200	Suppressor first	40	20	20	0.06	--	--	0.1
87	2200	Suppressor first	30	30	20	0.04	--	--	0.1
88 ^(b)	2200	Suppressor first	40	20	20	0.02	--	--	0.2
89 ^(b)	2200	Suppressor first	30	30	20	0.16	--	--	0.17

(a) 0.25-in. cores given a total reduction of about 8.3:1.

(b) All packs flipped and reversed between hot-rolling passes, except Plates 88 and 89. These were rolled suppressor first each time. A cold reduction of 18 per cent followed hot rolling.

T-5

Metallography has not been completed, and an accurate estimation of boron loss cannot be made until core thickness is measured. However, over the range of values expected, it appears that there has been very little boron lost.

TABLE T-3. EFFECT OF BINDER ADDITIONS ON BORON CONTENT OF FABRICATED PLATES

Binder Added ^(a)	Boron Content ^(b) , w/o
None	0.190
<1/2 w/o camphor-methanol	0.199
1/2 w/o camphor-methanol	0.201
1 w/o camphor-methanol	0.198
1/2 w/o Ceremul "C"	0.204
1/2 w/o Carbowax 6000	0.192

(a) Amount indicated is weight per cent of core.

(b) Includes approximately 5-mil-thick cladding on two surfaces.

Encapsulation Studies

J. F. Lagedrost, W. E. Murr, and J. H. Stang

Reference and alternate SM-2 fuel-test specimens are being irradiated in the MTR and the ETR. The target specimen-surface temperatures are in the 600 to 650 F range. Target burnups range from about 40 uranium a/o for the ETR capsule to about 70 uranium a/o for the MTR capsules.

The three noninstrumented capsules (BMI-32-1, BMI-32-2, and BMI-32-3) being irradiated in the MTR core are scheduled to be discharged on March 28, 1960. The first instrumented capsule, BMI-32-4, is also scheduled to be discharged from its ETR beryllium-reflector position about March 28, 1960. The last operating thermocouple in this capsule failed when the reactor returned to power following a scram on February 7. Prior to the scram the couple had shown temperatures in the 400 to 405 F range which indicated that specimen-surface temperatures were in the neighborhood of 650 F. Actually, these surface temperatures are about the same as those at the start of the irradiation. This consistency of temperature has not been expected in view of the estimated rate of depletion of fuel. At the presently estimated burnup of approximately 30 uranium a/o, a general temperature-level reduction of at least 50 F would be anticipated. It may be that the thermocouple did not read correctly, that the neutron flux may have increased in proportion to the fuel burnout, or that the boron added as a burnable poison is behaving in an ideal fashion to smooth out burnup.

The four capsules will be returned to the Battelle hot cell during the first 2 weeks of April for postirradiation examination. It is expected that the capsules will be opened and examination of the specimens will be essentially complete by mid-June, 1960.

T-6

Three of the six remaining instrumented capsules have been assembled and sent to the ETR. Of these, BMI-32-5 is scheduled for insertion during Cycle 28 shutdown (late March or early April), and BMI-32-6 and BMI-32-8 will probably be inserted during the following shutdown. Capsule BMI-32-9 will be completed early in March, and BMI-32-7 and BMI-32-10 probably by mid-April.

U-1

GAS-COOLED REACTOR PROGRAM (AGN)

D. L. Keller

Studies for Aerojet-General Nucleonics directed toward the development of compact gas-cooled reactors are reported in this section. The activities on the various tasks are reported under "Materials Development Program" and "In-Pile-Loop Program".

MATERIALS DEVELOPMENT PROGRAM

D. L. Keller

The fabrication of UO_2 -BeO fuel pellets for various reactor experiments is continuing. Chemical analyses, density measurements, and recovery percentages are reported on the first group of approximately 3000 pellets.

Further progress of in-pile capsule irradiations is reported; however, no hot-cell evaluation was performed during February.

Construction has been completed on the ML-1-IB critical assembly, and final adjustments are being made in preparation for receiving the fuel from AGN around the first of April.

One-kilogram ingots of modified Hastelloy X alloys were induction melted, forged at 2200 F, and rolled at 2200 F to 0.060-in. sheet. The effect of yttrium and niobium additions as well as the effect of varying the base-alloy composition will be evaluated from the standpoint of mechanical properties and corrosion resistance. The latter work will be performed at AGN.

Fabrication of BeO- UO_2 Fuel Pellets

H. D. Sheets and A. K. Smalley

The objective of this program is to prepare BeO ceramics containing fuel-enriched UO_2 for loop exposure.

Fabrication of 2966 acceptable hollow pellets 0.200 ± 0.030 in. high with an OD of 0.1745 ± 0.0015 in. and an ID of 0.055 in. was completed during February; 82.6 per cent of the pellets sintered were acceptable. Of the pellets sintered 10.4 per cent were outside diametral tolerance, 4.8 per cent were chipped, and 2.2 per cent were rejected because of internal flaws detected by radiography. Bulk-density measurements on specimens chosen from each lot indicated that the bulk density of the sintered pellets ranged from 4.90 to 5.09 g per cm^3 (about 94 to 98 per cent of theoretical). Chemical analyses

U-2

for UO_2 content on 15 pellets ranged from 58.0 to 60.1 w/o, with an average of 59.1 w/o. The UO_2 in these same pellets had an oxygen-to-uranium ratio ranging from 2.00 to 2.05, as determined by a weight-gain technique. Metallographic examination revealed that these pellets were similar in microstructure to the previously prepared capsule specimens.

Fabrication of an additional 4000 pellets of the same dimensions, but with slightly higher fuel loading, for additional experiments will be started as soon as UO_2 fuel can be procured.

Encapsulation Studies

J. H. Stang, J. F. Lagedrost, and D. W. Nicholson

Irradiation of Specimens Containing MCW Spherical UO_2 Dispersed in Stainless Steel

The irradiation stability of MCW spherical and ORNL hydrothermal UO_2 (30 w/o highly enriched) dispersed in stainless steel is under investigation. Two plate-type specimens of each type are being irradiated in Capsules BMI-33-1 and BMI-33-2 now in the MTR.

The specimens in BMI-33-1 are undergoing a six-reactor-cycle exposure at center-line temperatures in the 1075 to 1275 F range (calculated from thermocouple data). Estimated fission burnup levels of 12 to 15 a/o will be achieved at the March 28, 1960, scheduled discharge date. Capsule BMI-33-2, in which specimen temperatures during its first reactor cycle were approximately the same as those in BMI-33-1, was moved to a higher flux position for three additional cycles. Specimen center-line temperatures after the move were in the 1400 to 1650 F range (calculated). This capsule is also scheduled for discharge on March 28, 1960, by which time its specimens should reach burnup levels in the 9 to 11 a/o range.

UO_2 -BeO Irradiation Studies (AGN)

This program involves studies of irradiation effects on Hastelloy X-clad dispersions of highly enriched UO_2 in BeO. Six small cylindrical specimens, contained in Capsule AGN-BRR-II, are being irradiated at a nominal 1725 F surface temperature in the BRR core.

The capsule is scheduled to be discharged from the reactor about March 7, 1960, after its fourth 2-week cycle. The two electric heaters and five of the six thermocouples originally present continue to operate satisfactorily. During the second and third cycles, the center two specimens were maintained at design temperature (nominally, 1725 F at the specimen surfaces) 84 per cent (450 hr) of the reactor operation time; the corresponding minimum specimen-surface temperature was about 1600 F. These temperatures are estimated from data provided by intracapsule thermocouples

U-3

located very close to the specimens. During the remaining 16 per cent of the time, lower temperatures were observed because (1) the reactor was operated at reduced power, or (2) the auxiliary-heat-input circuit was opened by a momentary temperature excursion above 1750 F, the maximum allowable specimen-surface temperature.

Operation during the fourth cycle has been similar to that observed during the second and third cycles. It is estimated that the total burnup of the specimens will reach 2-1/2 to 3 a/o at discharge.

Effects of Irradiation

J. H. Saling, W. E. Murr, J. E. Gates, and R. F. Dickerson

No hot-cell evaluation was performed during February.

GCRE Critical-Assembly Experiments

J. W. Ray, R. G. Matthews, W. S. Hogan,
D. A. Dingee, and J. W. Chastain

Construction has been completed on the ML-1-1B critical assembly. During the past month the supports for the scissors-acting control blades were fabricated and assembled. In addition, final checkouts and adjustments were made on liquid-level indicators, neutron-source drive, and control-rod drives and position-limit switches.

Next month the fuel-element containers and fuel-storage area will be cleaned up in preparation for receipt of fuel around the first of April. Final adjustments will be made on reactor instrumentation and controls, and the interlock system will be checked.

Alloy Modification of Hastelloy X

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

Hastelloy X is the material that has been chosen for application as the fuel cladding in the reactor. Exposure of this material to a nitrogen-1/2 volume per cent oxygen atmosphere at 1750 F has resulted in a loss of ductility (from 40 per cent total elongation to 12 per cent total elongation) after 5000 hr. Further exposure could lead to more serious losses in ductility and possible cladding failure. It is believed that either grain growth or carbide precipitation may be responsible for the loss of ductility observed. The loss in ductility due to grain growth may be remedied by adding a grain refiner such as yttrium. The embrittling effect of carbides in the grain boundaries can be counteracted by niobium or yttrium additions which may produce a more favorable phase distribution or act as a ductile matrix for carbide precipitation.

U-4

The effect of alloying additions which may improve the ductility of Hastelloy X after exposure to the nitrogen-1/2 volume per cent oxygen atmosphere and which also may improve the oxidation resistance of the material is being studied. Niobium and yttrium are being added for this purpose. The effect of variations in the base-alloy composition is also being studied. Chromium and nickel contents are being increased at the expense of the iron present in the alloy, and the carbon content is being varied. The base composition is nickel-21 w/o chromium 20 w/o iron-10 w/o molybdenum-0.7 w/o tungsten-1.0 w/o manganese-1.0 w/o silicon-0.1 w/o carbon.

One-kilogram ingots were induction melted in alumina crucibles and poured at 2900 F into Zirconite molds. These were forged at 2200 F to slabs and then rolled at 2200 F to 0.060-in. sheet. All of the alloys except one, containing a 1 w/o yttrium addition and 25 w/o chromium instead of 21 w/o, were successfully reduced 95 per cent. This alloy fractured during forging; a duplicate is being prepared.

A portion of the fabricated sheet has been sent to Aerojet General for corrosion testing. Tensile specimens for testing at 75, 1000, 1750, and 1850 F in air are being prepared from the remaining sheet.

Samples for metallographic examination and chemical analyses have been obtained from the as-cast ingots and are being processed.

IN-PILE-LOOP PROGRAM

G. A. Francis

During the period covered by this report, activity on the loop program was continued. At the Battelle Research Reactor, preparation for operation of the recirculating in-pile gas loop was continued, and a change was made in experiment plans. The Engineering Test Reactor loop fuel-specimen irradiation was delayed by mechanical problems associated with blower operation.

BRR Loop Program

W. G. Rieder, S. J. Basham, and W. H. Goldthwaite

Preparation for operation of the loop was continued during February. The major activities were associated with calibration of the new gas-monitoring system and the performance of a flux run.

The continuous loop gas-monitoring-system calibration will be based on data obtained by introducing fission gases into the sensing chamber. The actual calibration runs have been completed, and the data are being reduced. The calibration will be completed and the sensitivity of the device will be established during initial irradiation of a

U-5 and U-6

test specimen. It is planned that the instrument will be used as a reactor scram-actuating device during future tests.

A fuel subassembly designated 1B-2T was received from Aerojet General Nuclear and was used in a flux run. The data will be reduced and reported in the next regular progress report.

A change in the AGN's irradiation plans has set the next irradiation to start during March. The specimen will be a pin-type unit instrumented for thermal cycling. The test will run for approximately 1000 hr and will be followed by a similar specimen instrumented for engineering information.

ETR Loop Program

E. O. Fromm, J. V. Baum, A. M. Rockwood, and G. A. Francis

Problems encountered both at the reactor site and during blower tests at Battelle have resulted in a change of irradiation plans. Shortly before the scheduled insertion of the 1-3P fuel subassembly, a belt failure occurred on the operating blower at the reactor site. At Battelle a belt failure occurred after 680 hr of operation.

To summarize, since the blower modification last October, there have been three belt failures. These occurred after 554, 503, and 680 hr of operation at design conditions. The reactor cycle is approximately 500 hr, and the loop system has three blowers in position at all times with one operating.

Since the belt performance was not acceptable, the element was not inserted during February. It has been decided to make minor drive modifications based on experience and the belt manufacturer's recommendations. It is planned that the 1-3P will be inserted at the end of the next ETR operating cycle if the blower modifications are completed and the test of a modified blower at Battelle has been under way 550 to 600 hr. Present project activity is being directed toward this modification and operation.

Other activity during February included the installation of a new loop heater control at the site. The control was furnished by the original supplier and appears to function satisfactorily. At Battelle the blower test was continued. The unit on test which has not been modified has run 1300 hr without regreasing. An inspection after 610 hr of belt operation revealed no indication of incipient failure. This test indicates that a belt drive system would be satisfactory for use during specimen irradiations. The test will be continued until the modified system can be prepared for operation.

12 11 50

V-1 and V-2

A STUDY OF THE CORROSION RESISTANCE OF THORIUM AND
URANIUM UNDER ATMOSPHERIC CONDITIONS (UCRL)

E. F. Stephan, P. D. Miller, and F. W. Fink

Preliminary work has started to determine the corrosion resistance of thorium, uranium, and certain uranium alloys during storage under atmospheric conditions.

Equipment is being assembled to evaluate these materials under a variety of laboratory conditions simulating several stages of storage. Preliminary studies will be made with bare and coated coupons which will be available late in March.

RWD:CRT:all