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PROGRESS RELATING TO CIVILIAN APPLICATIONS DURING JANUARY, 1960

by

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Chief, Declassification Branch

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TABLE OF CONTENTS

TABLE OF CONTENTS
(Continued)

REPORTS RELATING TO CIVILIAN APPLICATIONS ISSUED DURING JANUARY, 1960

BMI-1396	"ML-1-1A Core Studies With the GCRE Critical Assembly", by Richard A. Egen, William S. Hogan, David A. Dingee, and Joel W. Chastain.
BMI-1400	"Metallurgical Studies of Niobium-Uranium Alloys", by John A. DeMastry, Frederic R. Shober, and Ronald F. Dickerson.
BMI-1401	"High-Temperature Irradiation of a Zirconium-Hydride-2 w/o Uranium Alloy", by Gerald E. Lamale, Alan W. Hare, Horatio H. Krause, Alan K. Hopkins, John H. Stang, Eugene M. Simons, and Ronald F. Dickerson.
BMI-1402	"Effect of Additions to Zircaloy on Hydrogen Pickup During Aqueous Corrosion", by Warren E. Berry, Earl L. White, and Frederick W. Fink.
BMI-1403	"Progress Relating to Civilian Applications During December, 1959", by Russell W. Dayton and Clyde R. Tipton, Jr.

A. REACTOR MATERIALS AND COMPONENTS (AEC-DRD)

R. F. Dickerson

The study designed to obtain additional information with respect to the electrical characteristics and thermal conductivity of fully oxidized solid solutions of U_3O_8 and La_2O_3 has been continued. Specimens containing varying amounts of La_2O_3 were heated for 1, 5, and 20 hr in 3200 F air. All showed weight losses much greater than those occurring in solid solutions of UO_2 and La_2O_3. Since the weight losses were random in nature, it is believed that reaction with the BeO substrate used to contain the sample during heating played an important part in the weight loss. A repeat experiment is planned. The possible pressure decomposition of U_4O_9 is being further studied in an attempt to explain why this most dense oxide of uranium should decompose under pressure.

The irradiation of 11 capsules containing specimens of Type 347 stainless steel is being continued in the ETR. Estimated exposures as high as 4.99×10^{21} nvt have been achieved to date. Two capsules designed to operate at 600 F as a result of gamma heating will be inserted in the WTR in February.

Cold-rolled sheets, 0.010 in. thick, of niobium-base binary alloys containing 1.84 w/o chromium, 3.21 w/o chromium, or 4.33 w/o zirconium and of a niobium-base ternary alloy containing 20.5 w/o titanium and 4.28 w/o chromium were used for weldability studies. Spot welds failed by tearing in the sheet surrounding the weld during peel tests, indicating that the weld metal was stronger than the sheet. Fusion welds in the 1.84 w/o chromium and 4.33 w/o chromium were as ductile as similar welds in unalloyed niobium. The alloys could all be considered weldable material.

The construction of the capsule designed for in-pile creep testing at 650 F will be initiated upon receipt of the necessary heaters and bellows. It is calculated that 600 w per in. will be dissipated while maintaining the specimen at 650 F. Gamma heating should supply 300 w per in. and the balance will be supplied by the electrical heaters. Internal-friction measurements are being used as one tool for determining the presence of strain aging in Zircaloy-2. The data obtained to date do not lend support to the presence of strain aging. On the other hand, tensile tests performed at room temperature and temperatures up to 650 F show a definite yield-point phenomenon after having been plastically strained, unloaded, and reloaded. The rapidity with which the material changes from a material with no yield point to one with a yield point implies a rapidly diffusing interstitial, or in other words a phenomenon similar to strain aging. Electron microscopy showed a preponderance of dislocations in annealed zirconium, which indicates incomplete annealing. These must be removed by longer annealing if evidences of strain aging are to be observed.

Attempts to use ellipsometry and electrical resistivity as tools for determining oxygen concentrations of less than 10 ppm in sodium have been delayed by equipment difficulties. The modifications to the mass spectrometer are nearly complete, and oxygen analysis will be initiated shortly. A cold-trapped sodium pumping loop will be used to supply sodium samples for the various studies. The oxygen level in the purification loop is maintained by cold trapping and can be determined by a vacuum-distillation sampler as well as a plugging-indicator bypass branch. Slow-speed friction and wear studies of sliding surfaces in liquid sodium have been reopened. The test apparatus is being placed in operating condition and materials for preparing the friction and wear specimens are also being obtained.

Valence Effects of Oxide Additions to Uranium Dioxide

W. B. Wilson, A. F. Gerds, and C. M. Schwartz

Solid-state studies of oxide additions to uranium oxides are being conducted for correlation with the high-temperature stability and physical properties of these solid solutions. Recent work has been directed toward fabrication of the "fully oxidized" solid solutions of U_3O_8 containing 40, 50, and 60 mole per cent La_2O_3. These materials were initially prepared by sintering U_3O_8 and La_2O_3 at 1200 C in air.

The 1200 C sintering of these materials resulted in relatively porous compacts. For this reason an additional sintering at 1750 C was performed in air for periods of 1, 5, and 20 hr. This final sintering is identical to the method previously used to evaluate the relative high-temperature stability of solid solutions of UO_2 containing La_2O_3, Y_2O_3, and other additions.

Weight losses following the 1750 C air sintering are presented in Table A-1.

TABLE A-1. WEIGHT LOSSES OF U_3O_8 CONTAINING OXIDE ADDITIONS AFTER HEATING IN DRY AIR AT 3200 F

Composition, mole per cent	Weight Loss(a) After Time Shown at 3200 F, per cent		
	1 Hr	5 Hr	20 Hr
U_3O_8-40 La_2O_3	12.1	1.75	7.8
U_3O_8-50 La_2O_3	+8.6	17.6	10.0
U_3O_8-60 La_2O_3	0.8	13.3	18.7
U_3O_8-25 Y_2O_3-25 MgO(b)	0.5	18.2	5.3

(a) Plus sign indicates weight gain.

(b) This composition was prepared in conjunction with previous work to reduce both the additive amount and the cross section for stable materials.

The results show that extensive weight losses occurred over that which has previously been experienced for the "reduced" solid solutions of UO_2 with additions. The losses appear to be of a random nature and are unrelated to either time or composition, and are believed to occur principally as the result of reaction with the BeO substrate used to contain the sample during firing. In addition, the relative porosity and presence of cracks may have resulted in further loss of material by fracturing.

A repeat experiment to minimize the reaction with the BeO is planned in order to obtain more reliable weight-loss data. X-ray diffraction and chemical analyses followed by studies of the electrical characteristics are under way on the compositions given in Table A-1.

High-Pressure High-Temperature Solid-State Studies

W. B. Wilson and C. M. Schwartz

An investigation is being conducted on the effects of ultrahigh pressure and high temperature on the uranium oxides and on reactions of the uranium oxides with various mixed oxides. A paper describing the results of the high-pressure investigation on the uranium oxides has been essentially completed. Experimental work was limited, during the month, principally to the effects of pressure and temperature on U_4O_9. The results are incomplete, at present. Modification of a new press for use with the high-pressure apparatus, which, when completed, will be used to continue the study of the effects of pressure on reactions, was undertaken.

Irradiation-Surveillance Program on Type 347 Stainless Steel

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

The program to determine the effects of fast-neutron irradiation (neutrons with energies greater than 1 Mev) on properties of AISI Type 347 stainless steel has continued. This program consists of the irradiation and postirradiation mechanical testing of subsize tensile, cyclic-strain fatigue, and impact specimens of Type 347 stainless steel. The specimens are being subjected to a range of neutron exposures at temperatures of about 120 and 600 F.

The program is being carried out for the purpose of providing backup information on the effects of irradiation on the KAPL C-33 loop and on other loops in the ETR constructed of Type 347 stainless steel. The KAPL loop will operate for about 3 years, during this time accumulating an integrated fast flux exposure of between 1.4 and 1.8×10^{22} nvt at a temperature near 600 F. At the present time information on mechanical-property changes in Type 347 stainless due to irradiation is available for exposures to 3.76×10^{21} nvt; the present program will provide information on properties between this exposure and that ultimately expected for the KAPL loop.

Eleven capsules are presently undergoing irradiation in the ETR. Eight capsules have been in operation since June, 1958, and three have been in operation since August, 1959. The total exposures obtained upon all eleven as of the end of ETR Cycle 25 (January 11, 1960) are shown in Table A-2. The estimates are based on ETR flux data obtained from areas adjacent to the capsules.

Three capsules will be irradiated at a temperature near 600 F. Two of the capsules are scheduled for insertion in the WTR in February. The third capsule will

TABLE A-2. CAPSULES PREPARED FOR THE TYPE 347 STAINLESS STEEL IRRADIATION-SURVEILLANCE PROGRAM

Capsule	Type of Specimens in Capsule	Proposed Irradiation Temperature, F	Approximate Removal Date[a]	Approximate Exposure at Time of Removal[b], nvt	Total Exposure as of January 11, 1960, nvt		Location	Remarks
					Top	Bottom		
BMI-24-1	Tensile and fatigue	600	January, 1959	1.55×10^{20}	--	--	BMI	Examined at BMI Hot-Cell Facility for melting
BMI-24-2	Tensile and fatigue	120	January, 1962	1.31×10^{22}	3.608×10^{21}	4.394×10^{21}	ETR K-8-NE	Being irradiated
BMI-24-3	Tensile and fatigue	600	--	--	--	--	BMI	To be irradiated in WTR
BMI-24-4	Tensile and fatigue	120	January, 1963	1.78×10^{22}	2.657×10^{21}	4.021×10^{21}	ETR K-8-SE	Being irradiated
BMI-24-5	Tensile and fatigue	600	--	--	--	--	BMI	To be irradiated in WTR
BMI-24-6	Tensile and fatigue	120	June, 1961	1.08×10^{22}	4.571×10^{21}	3.598×10^{21}	ETR K-8-NE	Being irradiated
BMI-24-7	Tensile and fatigue	600	--	--	--	--	BMI	To be irradiated in WTR
BMI-24-8	Tensile and fatigue	120	June, 1962	1.54×10^{22}	3.021×10^{21}	4.036×10^{21}	ETR K-8-SE	Being irradiated
BMI-24-9	Tensile and fatigue	600	--	--	--	--	BMI	To be irradiated in WTR
BMI-24-10	Tensile and fatigue	120	January, 1961	0.84×10^{22}	3.909×10^{21}	3.795×10^{21}	ETR K-8-SE	Being irradiated
BMI-24-11[c]	Tensile and fatigue	600	--	--	--	--	BMI	Damaged at ETR
BMI-24-12	Tensile and fatigue	120	June, 1960	0.61×10^{22}	4.994×10^{21}	4.424×10^{21}	ETR L-8-SE	Being irradiated
BMI-24-13	Impact	600	--	--	--	--	BMI	To be irradiated in WTR
BMI-24-14	Impact	120	June, 1962	1.54×10^{22}	4.293×10^{21}	4.368×10^{21}	ETR K-8-NW	Being irradiated

TABLE A-2. (Continued)

Capsule	Type of Specimens in Capsule	Proposed Irradiation Temperature, F	Approximate Removal Date(a)	Approximate Exposure at Time of Removal(b), nvt	Total Exposure as of January 11, 1960, nvt		Location	Remarks
					Top	Bottom		
BMI-24-15	Impact	600	--	--	--	--	BMI	To be irradiated in WTR
BMI-24-16	Impact	120	June, 1960	0.61×10^{22}	4.341×10^{21}	3.845×10^{21}	ETR K-8-NW	Being irradiated
BMI-24-17(c)	Tensile and fatigue	600	October, 1958	3.25×10^{20}	--	--	BMI	Examined at BMI Hot-Cell Facility after high temperature observed
BMI-24-18	Tensile and fatigue	120	--	--	7.07×10^{20}	1.102×10^{21}	ETR K-8-NW	Being irradiated for postirradiation annealing studies
BMI-24-19(c)	Tensile and fatigue	600	--	--	--	--	BMI	Fabricated to replace BMI-24-17; to be irradiated in WTR
BMI-24-20	Tensile and fatigue	120	--	--	8.16×10^{20}	1.102×10^{21}	ETR K-8-NE	Being irradiated for postirradiation annealing studies
BMI-24-21	Tensile and fatigue	600	--	--	--	--	BMI	Fabricated to replace BMI-24-1; to be irradiated in WTR
BMI-24-22	Tensile and fatigue	120	--	--	1.058×10^{21}	1.333×10^{21}	ETR L-8-SE	Being irradiated for postirradiation annealing studies

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(a) Based on 6-month lead on loop, plus 2 months for examination.

(b) Based on maximum fast flux at tube of 1.7×10^{14} nv for 6-month periods.

(c) Thermocouple-lead capsule.

be inserted in the reactor when more information concerning temperatures at various power levels is obtained. All three capsules are designed such that specimen temperatures are achieved by gamma heating of specimens and capsule components.

Development of Niobium-Base Alloys

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

Niobium and niobium-base alloys are potentially suitable materials for cladding applications in reactors operating at elevated temperatures. Being studied for possible use in the EBR are unalloyed niobium, niobium-1.84 w/o chromium, niobium-3.21 w/o chromium, niobium-4.33 w/o zirconium, and niobium-20.5 w/o titanium-4.28 w/o chromium. The properties of these alloys are being compared with those of a vanadium-11.7 w/o titanium-2.6 w/o niobium alloy.

All alloys being investigated were reduced approximately 80 per cent by forging at 2550 F in evacuated molybdenum cans. An attempt to cold roll the slabs to 0.030-in. sheet resulted in severe edge cracking. Tensile tests performed on cold-worked material of the niobium-1.84 w/o chromium, niobium-4.33 w/o zirconium, and niobium 20.5 w/o titanium-4.28 w/o chromium alloys indicate that they all have strengths superior to that of the vanadium-11.7 w/o titanium-2.6 w/o niobium alloy (which has shown properties acceptable for EBR use) at both 1200 and 1472 F.

Sections of all alloys were cut from the forged slab (2550 F) and these were rolled successfully at 800 F in air to 0.150-in. sheet. Further rolling studies at room temperature indicate that the most satisfactory sheet is produced if the warm-rolled (800 F) sheet is surface conditioned by belt sanding and annealing (1 hr at 2550 F) prior to cold rolling. Pieces of all alloys were cold rolled to 0.010-in. sheet to be used for welding studies.

The welding studies consisted of three tests to evaluate the weldability of the fabricated alloys: peel and tension shear tests of spot welds and guided bend tests of the butt welds. The spot-welded peel-test specimens were pulled apart with pliers; the tension-shear specimens were pulled apart in a tensile machine. In all cases, the spot welds failed by pulling a button out of one of the sheets, indicating that the spot weld is stronger than the base sheet material. The tension-shear strengths for the welds are listed in Table A-3.

To evaluate the fusion weldability, longitudinal butt welds were made and bent in the direction of the weld by the guided-bend-test procedure. This test consists of bending the welded sheet with rams of known radii. Each successive bend of the specimen is made with a ram of smaller radius. This procedure is followed until the weld cracks. The radius of the ram which produces cracking is divided by the thickness of the welds. This product is expressed as T, which is the minimum bend radius. Table A-3 lists the value T for alloys tested. In general, the smaller the value of T, the more ductile the material.

TABLE A-3. RESULTS OF TESTING OF ARC WELDS[(a)]

Alloy Composition, w/o	Tensile Shear Strength, lb	Minimum Bend Radius[(b)], T
Niobium (unalloyed)	170	18
Nb-1.84 Cr	177	18
Nb-3.21 Cr	186	9
Nb-4.33 Zr	185	18
Nb-20.5 Ti-4.28 Cr	235	1
V-11.7 Ti-2.6 Nb	194	12

(a) All welds appeared sound prior to testing.

(b) T = R/t, where R = radius of ram and t = thickness of welded sheet.

Additional ingots of niobium (unalloyed), and niobium-1 w/o chromium, niobium-3.0 w/o zirconium, niobium-4.5 w/o molybdenum, and niobium-10 w/o titanium-3.0 w/o chromium alloys were prepared and fabricated by forging at 1000 F in air. All the alloys were upset forged and rolled at 1000 F to approximately 70 per cent reduction. After warm rolling (1000 F), the rolled sheet was vapor blasted and rolled at 75 F to 0.035-in. sheet. All of the sheet exhibited moderate edge cracking, but fabrication was satisfactory otherwise. Tensile specimens have been prepared from this material and are being tested at 1200 F.

Development of Corrosion-Resistant Niobium Alloys

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

The evaluation of selected niobium alloys for possible service in pressurized-water reactors was continued. Earlier work has shown that good corrosion resistance and hot strength can be obtained in niobium with several alloy additions in amounts of 10 a/o or greater. Currently, alloys of lower total alloy content are being evaluated with the aim of obtaining improved fabricability along with adequate corrosion resistance and hot strength.

The corrosion results obtained to date in 600 and 680 F water and 750 F 1500-psi steam are summarized in Table A-4. Alloys containing more than 45 a/o zirconium or a ternary containing 28 a/o titanium-6 a/o chromium continue to appear most corrosion resistant, exhibiting adherent tarnish films and low weight gains. A 12 a/o vanadium alloy possesses the optimum combination of high-temperature strength, low neutron-absorption cross section, and adequate corrosion resistance.

TABLE A-4. SUMMARY OF CORROSION RESULTS OBTAINED ON NIOBIUM ALLOYS EXPOSED TO HIGH-TEMPERATURE WATER AND STEAM

	600 F Water		680 F Water		750 F Steam	
Alloy Addition (Balance Niobium), a/o	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
Commercial Niobium, Rocking-Hearth Melts						
Unalloyed Nb	280	-39.9	42(a)	Disintegrated	28(a)	Disintegrated
10.5 Zr	--	--	196(a)	0.67	182	-32.8
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	182	-38.5
4.67 W	--	--	196(a)	-29.3	--	--
9.56 W	--	--	7(a)	Cracked	--	--
2.45 Mo	--	--	196(a)	-7.10	98	Disintegrated in 98 days
5.20 Mo	--	--	196(a)	-1.30	182	-146.4
7.40 Mo	--	--	196(a)	0.62	--	--
4.42 V	--	--	196(a)	0.42	182	-13.1
6.59 V	--	--	196(a)	0.73	182	1.19
8.93 V	--	--	196(a)	0.59	182	1.10
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 in 98 days
9.41 Ti	--	--	196(a)	0.65	182	1.26
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	--	--
High-Purity Niobium, Consumable-Electrode Melts						
Unalloyed Nb	280	0.76	280	-7.40	224(a)	-54.0 Off test after 328 days
7.18 Mo	280	0.73	252(a)	Cracked	70(a)	Cracked
12.5 V	280	0.40	280	0.70	266	0.76

TABLE A-4. (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
46.8 Zr-5.06 Ti	280	0.30	252	1.07	238	2.54
11.2 Ti-3.2 Mo	280	0.44	252	0.66	238	0.46
18.8 Ti-8.7 Mo	280	0.22	252	0.66	238	0.53
9.9 Zr-9.4 V	168	0.26	168	0.29	154	-4.67
5.7 Zr-11.4 V	168	0.19	168	0.46	154	-1.57
9.1 Ti-6.3 Cr	168	0.16	168	0.49	154	-3.93
7.5 V	14	0.09	14	0.14	14	0.54
7.5 V-0.19 N	14	0.02	14	0.17	14	0.30
7.5 V-2.5 Ti	14	0.06	14	0.20	14	0.26
7.5 V-2.5 Mo	14	0.04	14	0.15	14	0.29
		High-Purity Niobium, Rocking-Hearth Melts				
Unalloyed Nb	--	--	193	-5.67	--	--
Unalloyed Nb	--	--	84(a)	Disintegrated in 84 days	--	--
Unalloyed Nb	--	--	168	0.18(b)	--	--
1.1 Zr	--	--	168(a)	Disintegrated	--	--
2.2 Zr	--	--	84	-14.8	--	--
5 Zr	--	--	168	-11.0	--	--
10.2 Zr	--	--	168	-0.02	--	--
40 Zr	--	--	168	0.73	--	--
65 Zr	--	--	168	-0.46 Edge cracking	--	--
75 Zr	--	--	168	1.04	--	--
90 Zr	--	--	168	1.28	--	--
3.2 Ti	--	--	196	-7.96	--	--
10.5 Ti	--	--	196	0.75	--	--
25.0 Ti	--	--	196	0.45	--	--
<0.02 Cr	--	--	196	-3.27	--	--
0.5 Cr	--	--	196	-0.80	--	--
0.5 Cr	--	--	168	-3.75	--	--
<0.08 Fe	--	--	168	-1.59	--	--
0.3 Fe	--	--	84	-10.4	--	--
10 Fe	--	--	28(a)	Cracked	--	--
10.9 Zr-5.1 Ti	--	--	168	0.61	--	--
25 Zr-5 Ti	--	--	168	-0.13	--	--
25 Zr-15 Ti	--	--	168	0.52	--	--
25 Zr-25 Ti	--	--	168	0.47	--	--
35 Zr-5 Ti	--	--	168	0.62	--	--
35 Zr-15 Ti	--	--	168	0.38	--	--
45 Zr-5 Ti	--	--	168	0.67	--	--

TABLE A-4. (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
10 Zr-5 Mo	--	--	168	0.19	--	--
35 Zr-5 Mo	--	--	168	0.52	--	--
45 Zr-5 Mo	--	--	168	0.87	--	--
35 Zr-5 Al	--	--	168	0.68	--	--
45 Zr-5 Al	--	--	168	0.62	--	--
10 Zr-5 Cr	--	--	168	0.54	--	--
45 Zr-5 Cr	--	--	168	0.47	--	--
10 Zr-5 Fe	--	--	168	0.49	--	--
2.5 V	--	--	196	1.06	--	--
2.0 V-2.5 Ti	--	--	168	0.85	--	--
2.0 V-2.3 Mo	--	--	168	1.04	--	--
2.2 V-0.54 Fe	--	--	168	0.64	--	--
1.8 V-<0.02 Cr	--	--	168	0.45	--	--
1.8 V-0.14 Al	--	--	168	0.86	--	--
2.5 V-2.5 Zr	--	--	84	-1.70	--	--
2.2 V-0.87 Ni	--	--	28(a)	-1.00	--	--
4.0 V-2.3 Zr	--	--	168	0.59	--	--
5 V-25 Zr	--	--	168	-1.10	--	--
5 V-35 Zr	--	--	168	-1.89	--	--
5 V-45 Zr	--	--	168	0.66	--	--
1 Ce	--	--	28	-4.03	--	--
1 Y	--	--	28	-5.16	--	--
5 Y	--	--	28	-17.38	--	--
1 Ni	--	--	14	-0.02	--	--
2.5 Ni	--	--	7	Broke in 7 days	--	--
5 Ni	--	--	7	Disintegrated in 7 days	--	--
1 Pd	--	--	28	-8.34	--	--
5 V-2.5 Ti	--	--	28	0.05	--	--
5 V-2.5 Mo	--	--	14	-0.01	--	--
5 V-2.5 Fe	--	--	14	0.10	--	--
5 V-2.5 Cr	--	--	28	0.36	--	--
5 V-2.5 Ni	--	--	14	0.35	--	--
5 V-2.5 Al	--	--	28	0.35	--	--
2.5 V-0.5 C	--	--	14	0.27	--	--
5 V-0.5 C	--	--	14	0.08	--	--
2.5 V-0.25 Ti-0.5 O	--	--	14	-0.02	--	--
5 V-0.25 Ti-0.5 O	--	--	14	0.02	--	--
2.5 V-0.25 Zr-0.5 O	--	--	14	0.06	--	--
5 V-0.25 Zr-0.5 O	--	--	14	0.05	--	--

TABLE A-4. (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
2.5 V-0.5 Ti-0.5 C	--	--	14	0.07	--	--
5 V-0.5 Ti-0.5 C	--	--	14	0.10	--	--
2.5 V-0.5 Zr-0.5 C	--	--	14	0.02	--	--
5 V-0.5 Zr-0.5 C	--	--	14	0.05	--	--
5 V	--	--	14	0.14	--	--
10 Zr	--	--	--	--	56	-1.73
10 Ti	--	--	--	--	56	0.39

(a) Off test.
(b) Losing weight.

Vanadium-zirconium and vanadium-titanium alloys with additions of carbon and oxygen interstitials for increased strength have been exposed 14 days to 680 F water. Weight gains obtained to date are low and compare favorably with those for similar alloys of lower interstitial content. These results are also included in Table A-4. They 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).

Corrosion testing of the alloys described in Table A-4 is being continued. Room- and/or elevated-temperature tensile and creep tests have been scheduled on samples of the following alloys (compositions given in atomic per cent, balance niobium):

5 V
5 V-2.5 Ti
5 V-2.5 Mo
5 V-2.5 Fe
5 V-2.5 Cr
5 V-2.5 AL
7.5 V
7.5 V-2.5 Ti
7.5 V-2.5 Mo
7.5 V-0.19 N
2.5 V-0.25 Ti-0.5 O
5 V-0.25 Ti-0.5 O
2.5 V-0.25 Zr-0.5 O
5 V-0.25 Zr-0.5 O
2.5 V-0.5 Ti-0.5 C
5 V-0.5 Ti-0.5 C
2.5 V-0.5 Zr-0.5 C
5 V-0.5 Zr-0.5 C
2.5 V-0.5 C
5 V-0.5 C

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

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

The creep strength of Zircaloy-2 at elevated temperature is to be determined and compared for two test conditions: in-reactor under a fast neutron flux (>1 Mev) and out-of-reactor. The test temperature for the initial tests will be 650 F. Strain aging has been reported to occur in Zircaloy at room temperature. Attempts are being made to detect and to study in greater detail the mechanism associated with the phenomenon. Tensile tests, internal-friction studies, and electron microscopy of thin film are being used as aids in the strain-aging studies.

The design of a capsule for creep testing at 650 F during irradiation has been completed and will be constructed upon receipt of the heaters and bellows from their respective suppliers. It is planned to stress sheet-type specimens by pressure-actuated bellows for periods of approximately 500 hr in capsules equipped with thermocouples and heaters. The in-pile test will be conducted in the WTR in a fast flux of approximately 1 x 10^{14} nv. The capsule, as designed, is theoretically capable of dissipating heat at the rate of 600 w per in. along the heated length, while maintaining the specimen at 650 F. It is expected that gamma heating during irradiation will supply 300 w per in. The necessary additional 300 w per in. will be supplied by the electric heaters. These have a total capacity of 1000 w per in.; this is sufficient to supply the total 600 w per in. during reactor shutdown periods.

A-13

Testing of a bellows unit for the stress-actuating feature of the capsule has been completed by the manufacturer with satisfactory results. Construction of the bellows for the anticipated capsules is now half completed. Electric heaters and control equipment have been ordered. Laboratory work investigating various fabrication and assembly problems and the fabrication of some capsule components will begin during February.

A series of experiments has been performed to determine if any internal-friction effects, due to strain aging, could be observed. Test specimens were strained 1 per cent in torsion at several temperatures, and the internal friction was measured as a function of time after straining. Test temperatures included 300, 325, 350, 377, and 400 C. If strain aging were occurring, it was thought that it would produce a reduction in internal friction with time or would be exhibited as an initial rise followed by a reduction. The results were similar at all test temperatures; that is, internal friction decreased with time, but was rather temperature insensitive. Strain-aging effects on internal friction should be temperature dependent, since diffusion is involved. These data do not lend support to the presence of strain-aging in Zircaloy-2.

In a series of tensile tests performed at room temperature and 450, 550, and 650 F on annealed (1 hr at 750 C in vacuum, furnace cooled), Zircaloy-2, the material showed a definite yield-point phenomenon after having been plastically strained, unloaded, and reloaded. Tensile specimens were prepared from the same material used for internal-friction studies. Upon each subsequent loading, whether loaded immediately or allowed to age at test temperature for 30 min before loading, 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 show a yield point implies a rapidly diffusing interstitial. These tests will be repeated using another lot of Zircaloy-2.

Electron microscopy has been continued on thin-sheet zirconium prepared from bar stock. This was done to eliminate the preferred orientation of rolled material. Attempts were made to observe dislocations in annealed zirconium. The material had been annealed in vacuum for 30 hr. The electron microscope showed what appeared to be a preponderance of dislocations, indicating incomplete annealing. After longer annealing times the specimens will be re-examined under the electron microscope.

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

A study is being made of new approaches to the problem of measuring the oxygen content of sodium. The objective is to prove the feasibility of rapidly monitoring the oxygen level in a large flowing-sodium system at oxygen concentrations below 10 ppm with a sensitivity of ±1 ppm. All previous methods lack the desired speed, and in most cases they also lack the required sensitivity. None of these previous techniques holds promise of the capacity for being developed readily into rapid and sensitive monitoring systems. Accordingly, a limited number of promising novel approaches are being studied to establish their feasibility on the basis of sensitivity.

In the initial stages, the feasibility studies are being conducted with sodium containing oxygen levels of 20 to 100 ppm. This makes the transfer of sodium from a single purification train to the various devices under study a less critical operation than would be the case with sodium at 10 ppm or below. Sodium containing two levels of oxygen is being used to predict the potential sensitivity of each technique at extremely low oxygen levels. Those techniques which show the most promise during this stage will be given advanced study with sodium containing oxygen in the 0 to 10-ppm range and at the same time will be evaluated for their adaptability to rapid monitoring.

Continued equipment difficulties have delayed the initial studies, and additional effort is needed before a comparison can be made as to their relative sensitivities.

Ellipsometry

The recent redesign of the sodium-containing cell, required for the polarizing-spectrometer study, has met with failure. The unsatisfactory flat window of the original design has been replaced with a prism cemented to the cell. Although vacuum checking of heated cells revealed no leaks, attempts to fill them with sodium resulted in leakage in the epoxy bond at the prism-cell body joint. New cells are being prepared in which the prism is mechanically sealed to the cell by means of a Teflon gasket. It is hoped that this design will avoid the apparent complication of thermal-expansivity differences. Also, this design has an additional advantage in ease of cleaning.

Electrical Resistivity

The design of the resistivity device, discussed in the previous report (BMI-1403), requires high-temperature brazed connections between a 0.014-in.-OD, thin-wall tube and 3/8-in. tubing – both of Type 304 stainless steel. Although the techniques worked out for making these connections have been proved satisfactory on a number of test pieces, the final assembly has shown leakage through this joint after three successive brazing attempts. Component parts are on hand, however, and, since each failure leads to additional improvements in technique, a leak-free unit should be available in the near future.

Polarographic Studies

During January the kinetics of sodium-drop formation were studied with glass capillaries. Results showed that the time to form a drop is extremely sensitive to the temperature of the sodium. The sensitivity results from the fact that "drop time" varies inversely with the fourth power of the radius of the capillary bore. As the temperature of the sodium decreases, sodium oxide precipitates on the bore surface, decreases the radius, and thereby increases the "drop time". With a calibrated capillary, only "drop time" and temperature would need to be measured. Thus, the technical feasibility for a simplified and sensitive "plugging indicator" has been demonstrated. There are indications, however, that the ultimate apparatus should be all-metal because the oxide precipitated in the present capillary could not be redissolved at temperatures up to 300 C. At higher temperatures, the sodium will undoubtedly react with the glass, further restricting the flow.

First work on polarography started with attempts to find a stable, conductive, organic electrolyte. Mineral oil, terphenyls, and tetradecane were chosen as solvents. The tetradecane is attacked by molten sodium. The terphenyls have a density greater than sodium, thus necessitating rising drops. The mineral oil, however, is less dense than sodium, permitting falling drops. Unfortunately, mineral oil has a resistivity of the order of 10^{12} ohm-cm. By additives, this has been reduced to the order of 10^9 ohm-cm. A resistivity of 10^7 ohm-cm or less is required, however, before actual polarographic studies can be attempted.

During February, a suitable conductive medium for polarographic studies will be sought, and oxidation-reduction reactions on sodium drops will then be studied as a function of oxygen content.

Mass-Spectrometry Study

The mass-spectrometer modifications are nearly completed. All material and equipment orders have been received and final assembly is in progress.

The sodium-transfer system consists of a specially prepared stainless steel tube which can be attached to the sodium-purification system. After evacuation, the tube will be helium purged and then filled with liquid sodium. The sodium will be cooled as rapidly as possible to minimize segregation of the oxide phase. After the sample tube is removed from the filling system, a screw-driven piston and a small-bore die will be added to convert the tube to an extrusion press. Precautions will be taken to prevent exposure of the sodium and accessory apparatus to the air during this step. In addition, after the sodium press is mounted in the mass spectrometer, the first few, possibly contaminated, samples will be discarded, and analyses of subsequent samples will be made until the results show that an equilibrium has been reached.

The size of the sample will be determined by the degree of rotation of the piston drive screw. Preliminary tests to establish the reproducibility of this technique will be made.

Fabrication of parts for the sodium press is completed. Ion-source modifications, including an oven for vaporizing the sodium, are nearly finished.

The first runs should be made within the next 2 weeks.

Purification-Loop Operation

A cold-trapped sodium pumping loop is being used to supply sodium samples to the various studies. The oxygen level in the purification loop is maintained by cold trapping and can be determined by a vacuum-distillation sampler as well as a plugging-indicator bypass branch. During January, modifications in loop operation have overcome the apparent problems of gas entrainment and oxide plugging which plagued previous attempts to operate for extended time at low cold-trap temperatures.

Improvements have been made in obtaining plugging-indicator data. Increased flow by substitution of a porous, sintered stainless steel disk for the original perforated plate has resulted in a larger flowmeter voltage. The substitution of a Brown servoamplifier system for the previous d-c amplifier has resulted in a more stable and noise-free output. The combined effect has been to increase the readability and reliability of the plugging-indicator data. In spite of this, the plugging temperature in a number of recent runs has differed by as much as 30 F from the cold-trap temperature. Instability in the X-Y plotter amplifier is suspected. Repairs and calibration are under way, and it is anticipated that the discordance between cold-trap and plugging temperatures will be reduced to ±15 F or less through continued experience with the present equipment.

The early data obtained from the vacuum-distillation runs indicated the need for modifications. Not only were the oxygen determinations considerably in error, but the device exhibited unpredictable thermal behavior during operation. The distillation apparatus has been modified to include a window so that the filling and distillation operation can be watched. In addition, a thin metal umbrella mounted on the end of a shaft can be rotated through a gland seal to deflect the initial charge of flushing sodium as well as protect the distillation cup from residual sodium dropping from the inlet tube or top of the distillation container during operation. A run made since the inclusion of these changes showed that, after the distillation was completed, additional sodium dropped from the presumably empty inlet tube. A similar occurrence would explain the peculiar behavior noted in several previous runs. It is expected that these recent changes will greatly enhance the reliability and precision of this device for future use.

Wear and Friction of Sliding Surfaces in Liquid Sodium

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

Apparatus for performing slow-speed friction and wear studies between a ball and flat specimen is being placed in operating condition. Materials for preparing the friction and wear specimens are also being obtained. These include: molybdenum, titanium carbide cermet (20 w/o cobalt binder), tungsten, and Type 347 stainless steel.

The initial friction experiments will be run using molybdenum specimens, in an environment made as oxygen free as possible. An attempt will be made to correlate specific microscopic areas of the wear track with stick-slip friction phenomena.

B-1

B. STUDIES OF FUEL ALLOYS (AEC-DRD)

R. F. Dickerson

Niobium-base binary alloys containing 10 to 60 w/o uranium have been exposed to 600 F water for 196 days. All specimens have a dull black oxide coating, but total weights are now beginning to decrease. Zirconium has been added to binary alloys containing 30 and 40 w/o uranium with the objective of improving the fabrication characteristics of the alloys. Stress-rupture testing of a niobium-10 w/o uranium alloy has continued through 800 hr at 1600 F under a stress of 40,000 psi, and a 20 w/o uranium alloy specimen is currently being tested at 2200 F under a stress of 20,000 psi. Design of capsules for irradiation studies has been initiated, and studies of the effects of oxygen on the composition limits of the gamma-immiscibility loop are in progress.

In order to insure availability of reasonably large sized ingots of thorium-uranium alloy with proper dispersion of finely divided uranium particles, melting studies have been initiated. These ingots are necessary for a source of material for irradiation testing. In addition, an investigation of the preparation of ThN is under way which involves a study of the effect of temperature and pressure on nitride formation. Preliminary creep data obtained at 600 C indicate that the addition of 1.5 w/o molybdenum or 2 w/o niobium to the thorium-10 w/o uranium alloy diminishes the creep rate at 3500 psi by a factor of 1/6 to 1/8. The rate exhibited by the 1.5 w/o molybdenum alloy stressed to 5500 psi is less than that of the thorium-10 w/o uranium alloy stressed to 3500 psi. Further creep data and high-temperature tensile data are being obtained on other alloys.

Development of Niobium-Uranium Alloys

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

Niobium-rich niobium-uranium alloys have been found to possess potential as high-temperature metallic fuels. It is necessary that the fabrication characteristics, mechanical and physical properties, corrosion behavior in various media, and irradiation properties be determined for the potential of these alloys to be realized.

Alloys containing from 10 to 60 w/o uranium were prepared using niobium containing from 0.03 to 0.07 w/o oxygen and from 0.02 to 0.74 w/o zirconium. The tensile properties, fabrication behavior, and corrosion behavior in air, water, steam, CO_2, and NaK of the alloys have been determined. No effect on the above-mentioned properties has been observed which can be attributed to the oxygen and zirconium variations of the alloys, although the present data are insufficient to determine if the corrosion behavior is affected.

Testing of niobium-uranium alloys in 600 F water has continued through 196 days; the resulting data are shown in Table B-1. All specimens being tested have a dull black oxide coating. The majority of the alloy specimens on test show total weight gains, although the weights are now decreasing.

TABLE B-1. CORROSION DATA FOR NIOBIUM-URANIUM ALLOYS IN 600 F WATER

Nominal Alloy Content (Balance Niobium), w/o	Impurity Content		Specimen Condition	Total Weight Change in 600 F Water in 196 Days, mg per cm^2
	Oxygen, ppm	Zirconium, w/o		
10 U	680	0.74	Fabricated	0.61
	1190	0.17	Fabricated	-1.12
	3170	0.02	Fabricated	-2.20
20 U	458	0.74	Fabricated	0.96
	523	0.17	Fabricated	1.90
	198	0.02	Fabricated	0.73
30 U	586	0.74	As cast	0.15
	669	0.17	Fabricated	-2.15
	165	0.02	As cast	-0.78
40 U	661	0.74	As cast	0.43
	579	0.17	As cast	0.20
	261	0.02	As cast	0.61
50 U	375	0.74	As cast	0.31
	334	0.17	As cast	0.40
	271	0.02	As cast	-0.31
60 U	471	0.74	As cast	-4.70
	273	0.17	As cast	-4.23
	192	0.02	As cast	-1.27
Zircaloy-2	--	--	--	0.28

Specimens of each alloy composition are currently being tested for compatibility with liquid sodium at 1500 F. These specimens will be examined after 500 hr of exposure.

Niobium-30 w/o uranium-10 w/o zirconium and niobium-40 w/o uranium-10 w/o zirconium alloys have been prepared by double consumable-electrode arc melting and are being encapsulated in molybdenum tubing prior to fabrication studies. It is hoped that the addition of zirconium will improve the fabrication characteristics of the alloys.

Stress-rupture testing of a niobium-10 w/o uranium alloy has continued through 800 hr at 1600 F under a stress of 40,000 psi. A niobium-20 w/o uranium alloy specimen is currently being tested at 2200 F under a stress of 20,000 psi.

Design of capsules for irradiation studies has been initiated. It is expected that specimens of the niobium-10, -20, and -30 w/o uranium alloys will be irradiated along with a niobium-uranium-zirconium alloy specimen of as yet undetermined composition. The irradiations are to be conducted to a 2.5 total a/o burnup at 1600 and 1800 F.

Alloy wire bars for a study of the effects of oxygen on the composition limits of the gamma-immiscibility loop in the niobium-uranium system have been homogenized and are being heat treated at temperatures ranging from 650 to 940 C. Phase limits will be evaluated on the basis of metallographic examination. Wire bars of uranium-niobium-zirconium ternary alloys containing 50 and 60 w/o uranium are being homogenized prior to heat treatment. The objective is to determine the effect of zirconium on the niobium-uranium immiscibility gap and on the stabilization of the body-centered-cubic solid solution.

Development of Thorium-Uranium Alloys

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

Development of thorium-uranium-base alloys with improved radiation stability and corrosion resistance is the objective of a program currently in progress. The effect of thorium purity, casting methods, and fabrication on the size and distribution of uranium-rich particles is being investigated, as is the effect of alloy additions on the properties of the binary alloys. Alloy compositions are thorium-uranium base with additions of molybdenum, niobium, and zirconium and zirconium in conjunction with niobium. The preparation and properties of thorium carbides and nitrides and thorium-uranium carbides and nitrides are also being studied.

Melting studies have been expanded from laboratory-scale to large-scale melts. Techniques for preparing finely divided, homogeneously dispersed alloys of uranium in thorium will be investigated using the thorium-10 w/o uranium composition.

The type of melting (vacuum-induction versus arc melting), the purity of starting materials, the pour temperature, and the cooling rate during solidification will be considered in terms of their effect on particle size and homogeneity. It is anticipated that the effects of melt stirring and the use of alloy additives such as niobium and molybdenum will be investigated also. During the period that materials are being secured, a search of the literature is being made for means of securing a uniform dispersion of uranium particles in the cast ingot.

In addition to the thorium-uranium alloy study, an investigation of the preparation of ThN is under way, involving a study of the effect of temperature and pressure on nitride formation. The thermodynamic characteristics of the thorium-nitrogen reaction will be investigated before additional experimental preparations are attempted.

Metallographic examination of thorium-uranium-zirconium alloys heat treated for 24 hr at 800 C and water quenched has disclosed particles of the uranium-zirconium gamma phase in a pearlitic thorium-zirconium matrix. No noticeable structural differences are apparent in alloys of the same composition prepared from two different grades of thorium (iodide and Ames) and cast by two different melting methods (induction and arc cast). Heat-treatment studies of the alloy material will continue, and the recrystallization behavior will be investigated. The effect of heat treatment on thorium carbides and thorium-uranium carbides will be studied also.

Preliminary creep data on thorium-uranium alloys obtained at 600 C are presented in Table B-2. Examination of the secondary-creep rates shows that the addition of 1.5 w/o molybdenum or 2 w/o niobium to the thorium-10 w/o uranium alloy diminishes the creep rate at 3500 psi by a factor of 1/6 to 1/8. The creep rate exhibited by the thorium-10 w/o uranium-1.5 w/o molybdenum alloy stressed to 5500 psi is less than that shown by the binary alloy stressed to 3500 psi. Further creep data and also high-temperature tensile data are being obtained on alloys of thorium-5, -10, -15, and -20 w/o uranium and thorium-10 w/o uranium with additions of 1.5 w/o molybdenum, 2 w/o niobium, 10 w/o zirconium, and 10 w/o zirconium-2 w/o niobium.

TABLE B-2. SECONDARY-CREEP DATA OBTAINED FROM ANNEALED THORIUM-URANIUM-BASE ALLOYS

Nominal Composition (Balance Thorium), w/o	Test Temperature, C	Stress, psi	Time, hr	Minimum Creep Rate, per cent per hr
10 U	600	1500	232.0	0.0026
10 U	600	3500	238.9	0.0100
10 U	600	4000	149.1	0.0135
10 U-1.5 Mo	600	3500	(a)	0.0013
10 U-1.5 Mo	600	5500	(a)	0.0085
10 U-2 Nb	600	3500	231.6	0.00174

(a) Test is still in progress.

C-1

C. 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

Results of various studies designed to support experiments of fission-gas release from UO_2 during and after irradiation are reported. No results have been obtained on fission-gas release from UO_2, but initiation of experiments, both in-pile and in the radiochemical laboratory after irradiation, is imminent.

Characterization of Sintered UO_2 and Model of Gas Release

Structural studies of single cyrstals and sintered polycrystalline UO_2 bodies are being continued as an aid to understanding the mechanism of fission-gas release during irradiation and postirradiation heat treatment. There has been no new information accumulated during the past month. Additional samples are being processed for pre- and postirradiation examination of surface structure. Also, specimens are being examined microscopically to determine the size distribution of grain-boundary pores after various sintering treatments.

A series of surface-area measurements by gas adsorption is in progress to establish the effects of various (fabrication and pretreatment) conditions upon the B.E.T. surface areas.

Five additional groups of sintered specimens are being prepared to represent variations in microstructure, density, and specific surface area. These specimens are to be used in a program to correlate density, open porosity, and microstructure with surface-area measurements. Fabrication conditions for the five groups of specimens will be as follows:

Ball-Milling Time, hr	Oxidation Temperature, F	Forming Pressure, psi	Sintering Temperature[(a)], F
2	--	40,000	2,600
2	--	40,000	2,800
2	--	40,000	3,000
15	--	20,000	3,000
15	1,000	20,000	3,000

(a) In hydrogen for 1 hr.

Future work will include preparation of additional specimens from preoxidized Davison UO_2 powder. The objective is to develop techniques for fabricating uniform specimens that will be useful in determining the effect of open porosity on fission-product retention. Work thus far indicates that this is a feasible objective.

C-2

Diffusion in UO_2

The phase of the program to study the release of fission gas from uranium dioxide during postirradiation heat treatment is continuing.

Physical measurements have been completed on ten flat plates of single-crystal uranium dioxide. These specimens, which will be used for fission-gas-diffusion measurements, are currently being encapsulated for irradiation as needed. The fission-gas-release measurements with this series of specimens are scheduled to begin in February.

Preparation for In-Pile Study

The in-pile apparatus for study of fission gas continuously during irradiation was inserted in the BRR. It was removed, however, because of difficulties in obtaining adequate vacuum before the helium flow was initiated. The complete unit, including beam-tube furnace, helium-gas-purification system, fission-gas-collection systems, and monitoring equipment, are in working order, and reinsertion in the BRR is expected at the next shutdown.

Sintered specimens are on hand for initial in-pile studies.

As an aid to interpretation of in-pile experiments, mathematical solutions for the release of radioactive gases, based on the "equivalent-sphere" model, taking into account the diffusion of the precursor, are being worked out. So far, only the case for equilibrium has been considered where the diffusion coefficients of the parent (iodine) and daughter (xenon) are equal, the release rate of the daughter is the same as for the case where it is assumed that the parent does not diffuse for a certain condition. The condition is that the parameter,

$$\mu_1 >> \mu_2 ,$$

where, by definition,

$$\mu = \frac{a^2\lambda}{D}$$

and,

a = equivalent-sphere radius

λ = decay constant

D = diffusion coefficient

Subscripts 1 and 2 = parent and daughter, respectively.

Further consideration is being given to the mathematics of the problem.

D. GENERAL FUEL-ELEMENT DEVELOPMENT (AEC-DRD)

S. J. Paprocki

Fabrication techniques are being developed and physical and mechanical properties are being determined for cermet fuel materials consisting of 60 to 90 volume per cent of UO_2 dispersed in stainless steel and various refractory-metal matrices. These are promising fuel materials because they incorporate a relatively high volume loading of UO_2 and embody many of the properties of dispersion fuels.

Solid-phase bonding techniques are being developed for niobium and molybdenum. A satisfactory process for the self-bonding of niobium has been developed. The process involves pretreatment of niobium surfaces with a solution consisting of 65 parts of nitric acid and 35 parts of hydrofluoric acid followed by gas-pressure bonding at 2100 F for 3 hr utilizing a helium pressure of 10,000 psi.

A study is being made of the kinetics of solid-phase bonding processes involving temperature and pressure. The most recent experiments have been concerned with the relationship of pressure to obtain intimate contact and the effect of grain orientation on subsequent grain growth across the bond interface.

Fabrication of Cermet Fuel Elements

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

Cermets of 80 volume per cent UO_2 dispersed in stainless steel, molybdenum, chromium, and niobium are currently being fabricated by pressure-bonding techniques for physical and mechanical testing.

A cermet rod of 80 volume per cent UO_2-molybdenum has been pressure bonded for 3 hr at 2350 F under a helium gas pressure of 10,000 psi with a resulting density of 94.4 per cent of theoretical. The 5/8-in.-diameter by 2-1/4-in.-long cermet rod is currently being end capped for thermal-conductivity measurements. In addition, an 80 volume per cent UO_2-niobium cermet rod is being prepared for thermal-conductivity measurements. The niobium-matrix cermet was prepared by compacting pellets from niobium-coated UO_2 powder at 30 tsi, followed by vacuum sintering for 1-1/4 hr at 3270 F. Sintered core densities were approximately 76 per cent of theoretical. The pellets were then inserted in a tantalum tube and pressure bonded for 3 hr at 2100 F under a helium gas pressure of 10,000 psi. The core density obtained is estimated at 83 per cent of theoretical. The specimen is now being machined for thermal-conductivity measurements.

One-inch-square by 0.100-in.-thick sintered compacts of 80 volume per cent UO_2-chromium and -niobium are being pressure bonded for 3 hr at 10,000 psi at temperatures ranging from 1900 to 2200 F. The cores were prepared by compacting at 30 tsi followed by vacuum sintering for 1/2 hr at 2550 F. Two cores of 80 volume per cent UO_2-chromium have been pressure bonded for 3 hr at 10,000 psi at a temperature of 2100 and

2200 F. Estimated core densities are, respectively, 77 and 81 per cent of theoretical. Core densities were 71 per cent of theoretical before pressure bonding. Currently, a series of molybdenum 1 by 1 by 0.100-in. cermets are being prepared for pressure bonding. All 1 by 1-in. cermets will be used to obtain modulus-of-rupture and thermal-cycling data.

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

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

Investigations are being conducted to develop techniques for cladding fuel elements with niobium or molybdenum by the gas-pressure-bonding process. Evaluation of specimens is based on the quality of bond obtained and the properties of the cladding material after bonding. Ceramic and cermet fuels of various configurations are to be clad with these metals.

Niobium specimens, having surfaces pickled in a solution of 65 parts nitric acid and 35 parts hydrofluoric acid, pressure bonded at 2100 F for 3 hr have exhibited excellent bonds; however, in some areas of these specimens, a row of small, isolated voids contained completely within grains has been noted along the original interface. Several specimens were prepared to determine if these apparent voids were caused by diffusion or residual contamination not removed by the surface preparation. Flat-plate specimens with bonding surfaces properly cleaned by pickling and with as-rolled surfaces were bonded at 2100 F for 3 hr at 10,000 psi. After bonding, the specimens were sectioned; one section was examined in the as-bonded condition, while the other section was cold rolled 25 per cent and annealed at 2100 F for 1 hr to produce recrystallization of the niobium. Examination of the as-bonded sections revealed a significant amount of contamination in the as-rolled specimen and only occasional contamination or voids within grains of the pickled specimen. After the cold-rolling and recrystallization treatment, the contamination was still present in the bond line of the as-rolled specimen The bond line could not be located in the pickled specimen after this treatment; consequently, it is apparent that the spherical areas within the grain are diffusion voids and not contamination. These results would indicate that some niobium grains should be expected to contain diffusion voids in specimens prepared by gas-pressure bonding. Further studies are being made with high-temperature annealing of bonded specimens to verify these results.

Specimens of niobium-clad uranium dioxide-21 w/o alumina exhibited an increase in oxygen content and subsequent cladding embrittlement caused by the reaction of the niobium cladding with the alumina in the core. To verify this, two specimens were prepared, one containing pure uranium dioxide against the niobium and the other containing pure alumina. The specimens were bonded at 2100 F and 10,000 psi for 3 hr in protective containers. Bend tests made on these specimens revealed the niobium next to the uranium dioxide was completely ductile; however, the niobium next to the alumina was embrittled during the bonding operation. Chemical analyses are now being conducted to determine the extent of the alumina-niobium reaction that is encountered with these conditions.

Self-bonding of molybdenum has been accomplished at 2300 F and 10,000 psi for 3 hr. Bonds obtained under these conditions have proven to be inconsistent in quality and strength. Specimens bonded at higher temperatures possess consistently better bonds; however, the ductility of the cladding material is severely decreased. In an effort to better understand the embrittlement problem, a series of molybdenum specimens was annealed at 2300, 2450, 3300, and 3600 F at a pressure of 1×10^{-5} mm of mercury. Pieces of as-received material, material rolled at 90 deg to the original rolling direction, and as-bonded specimens were included in each run. Results of these anneals indicate the as-received molybdenum will remain ductile only in the original rolling direction after recrystallization. All other specimens studied were completely embrittled at temperatures of 2450 F and higher. The reason for the residual ductility in the rolling direction is the fibered-type grain structure in that direction. This permits fewer grain boundaries to align themselves, and, therefore, it is less conducive to intergranular fracture. From these results, it is apparent that in order to achieve ductile molybdenum-clad specimens, it would be necessary to use cladding that will retain its ductility in all directions at elevated temperatures. The present technology of molybdenum does not offer a solution to this problem; consequently, the emphasis on the remainder of the program will be on niobium cladding of cermets and ceramics.

Factors Affecting Pressure Bonding

G. W. Cunningham and J. W. Spretnak

An investigation is being made of the mechanism and kinetics of solid-phase bonding of metals under application of heat and pressure. Most of the specimens are being prepared with OFHC copper. Two factors under consideration at this time are the relationship of hot hardness to the pressure required to place the metal surfaces in intimate contact at any particular temperature and the effect of grain orientation on grain growth across the interface.

In order to determine the range of pressures required to establish the pressure-hot hardness relationship described above, a series of specimens has been bonded at pressures of 1000 to 6000 psi and at temperatures of 300 C to very near the melting point (1083 C). Contrary to previously reported results, it appears that in the plastic region (above approximately 450 C) it will be necessary to bond specimens at pressures below 2000 psi in order to obtain specimens with large areas not in contact at the interface. The gage presently used on the hot press unit is not accurate below 2000 psi; therefore, the establishment of a relationship between required bonding pressure and hot hardness at temperatures above 450 C will be delayed until a more accurate gage is obtained. However, several series of specimens are now being bonded at temperatures of 400 C and below.

Preliminary studies are continuing on the effect of grain orientation on grain growth across the interface. Large-grained specimens are prepared by annealing in vacuum at 1000 C. Specimens have been bonded at 1000, 925, and 500 C. In the high-temperature region above 900 C, grain growth is very rapid and grain growth across the interface cannot be slowed to a low enough rate to effectively study the effect of grain orientation. In the specimens bonded at 1000 C and 3000 psi, complete grain growth across the interface occurred in less than 1 min. After 5 min, the only indication of the bond line was a line of isolated pores. Almost no grain growth occurred in the specimen pressed at 500 C. A series of specimens will be prepared at 500 C to determine whether the progress of the grain growth across the interface can be followed at that temperature.

E. 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

The objective of this program is to develop a fabrication technique which will maintain or improve the quality of ceramic, cermet, and dispersion fuel elements and, at the same time, reduce the manufacturing costs. The gas-pressure-bonding technique was selected as a promising method of fabrication for achieving these objectives. The ceramic, cermet, and dispersion fuel systems were selected for investigation since these fuel systems offer the greatest potential for achieving a high burnup over a range of temperatures and also offer an excellent opportunity for a substantial cost reduction by the utilization of pressure bonding. The study is concerned with the refinement and further development of the gas-pressure-bonding process to accomplish simultaneous densification and cladding of these fuels with stainless steel.

Uranium Dioxide Compaction Studies

Compaction studies in this program have been directed toward developing high-density green compacts of uranium dioxide which will 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 powder types and several powder mixtures, centrifugal packing, ultrasonic compacting, presintering, and binder removal. At the present, preliminary investigations on the effects of initial particle sizes and stoichiometry on the final density have been initiated.

A study of ideal particle-size distributions for optimum packing was continued. The cold-compaction characteristics of both fused and special dense UO_2 powders consisting of the ideal size distribution for cubic packing were determined. The size distribution for these powders was as follows: 1.25 w/o minus 325 mesh, 6.25 w/o minus 150 plus 200 mesh, and 92.5 w/o minus 60 plus 80 mesh. Densities of 82.3, 83.8, and 83.9 per cent of theoretical were attained in pressing the fused powders at 40, 45, and 50 tsi. Similarly, the special dense powders yielded densities 77.0, 78.1, and 79.3 per cent of theoretical at the respective compacting pressures. These values are somewhat lower than those attained with the ideal rhombohedral packing density, which resulted in densities of 84.7, 85.3, and 85.5 per cent of theoretical at 40, 45, and 50 tsi.

Gas-Pressure Bonding

The gas-pressure-bonding studies include the determination of the densification characteristics of various types and combinations of UO_2 powders both in the form of tapped powders and cold-pressed pellets, the evaluation of various fuel-element shapes for the simultaneous densification and cladding of the UO_2 powders, and the investigation of various fuel-element shapes containing high-density UO_2 cores which cannot be produced by other conventional fabrication methods. Investigations are also being conducted to obtain a more complete evaluation of the gas-pressure-bonded UO_2 and the stainless steel bonds obtained by gas-pressure bonding.

E-2

A preliminary investigation was conducted to demonstrate the feasibility of pressure bonding an edge-welded stainless steel-clad UO_2 compartmentalized flat plate. The components were machined from 10- and 100-mil as-rolled Type 304 stainless steel sheet. The UO_2 cores measuring, 0.100 by 0.2495 by 1.500 in., possessed a high initial density. The plates measured 0.120 by 1.875 by 5.500 in., with each of the nine cores being individually compartmentalized. Examination of the plates revealed that such elements can be prepared with excellent dimensional control. Some cracks were apparent in the UO_2 cores in these plates; however, this effect was attributed to assembly. Metallographic examination of all stainless steel interfaces revealed excellent bonds with 90 to 100 per cent grain growth. These bonds withstood both mechanical bend and peel tests. The relative strength and ductility of the cladding and the strength of the bonds between compartments were established by burst tests. In this test, compartments of an element are subjected to increasing internal pressures until rupture of the cladding occurs. Pressures of 5400 and 5600 psi were required to produce rupture with cladding deflections of 0.074 to 0.085 in. being recorded before rupture occurred. Examination of the specimen after burst testing revealed that none of the bonds between the cladding and ribs of the elements failed during the tests. In view of these results, the gas-pressure-bonding process appears to be a feasible method for fabricating stainless steel-clad UO_2 compartmentalized flat plates.

Ten tubes containing compacts of several mixtures of various particle sizes of fused and special dense UO_2 were pressure bonded to establish the effect of such mixtures upon densification. The tubes were heated to 1200 F in vacuum to remove the binder prior to sealing and then gas-pressure bonded at 2100 F for 3 hr at 10,000 psi. These specimens are being evaluated.

The corrugated rod and tube element design appears to be the most promising of those being considered to minimize surface roughness, warpage, and dimensional nonuniformity which occurs when pressure bonding elements containing tapped powders or cores of a low density. Initial pressure-bonded elements of this design did not permit a complete evaluation of the design due to faulty welds. Other designs are also being investigated which may improve dimensional properties of the clad elements.

Permeability and thermal-conductivity tests of pressure-bonded UO_2 are presently being conducted. Further work will concern a more complete evaluation of the fuel-element designs and the pressure-bonded UO_2.

F. DEVELOPMENT OF URANIUM CARBIDE (AEC-FUEL CYCLE)

F. A. Rough and W. Chubb

Uranium monocarbide and the other uranium carbides, as well as alloys based upon uranium monocarbide, are being developed under the auspices of the AEC Fuel-Cycle Development Program. These submetallic materials represent alternatives to the ceramic-type reactor fuels based on uranium dioxide. The uranium carbide-type fuels are known to have advantages over uranium dioxide with respect to density and conductivity, and they may have advantages with respect to cost of fabrication, irradiation resistance, and ease of reprocessing. The present program of research is intended to further define the properties of the uranium carbides and their alloys with respect to techniques for production of shapes, with respect to their physical and mechanical properties, with respect to their chemical behavior in contact with solids, liquids, and gases, and with respect to their mechanical behavior in a neutron flux.

Efforts during the first 10 months of this program have resulted in scaling up the casting process from 80-g ingots to 5000-g ingots. The density of castings containing 4.8 w/o carbon has been found to be 13.5 g per cm^3; the density of castings containing 7.0 w/o carbon has been found to be 12.4 g per cm^3. The densities and resistivities of alloys containing up to 9 w/o carbon have been surveyed. The compatibility of carbides with a number of common metals and with Santowax R has been surveyed. The rates of interdiffusion of uranium and carbon in uranium carbides have been measured. The addition of alloying elements to uranium carbides has been attempted, and a promising class of cast carbide cermets has been discovered.

During the last month, efforts to produce a high-density shape by powder-metallurgy techniques were continued, and it was shown that the uranium-propane reaction results in an unusually fine powder containing almost 4.7 w/o carbon. This powder sinters to a higher density than the coarser powders. The use of chromium metal as an aid to sintering also seems to hold some promise.

Casting studies have resulted in improved control over the composition of the product. In the skull-casting operation, homogeneity of the skull is a problem, but homogeneity of the castings is not, with the result that composition control has been a problem. During the last month, however, a 2-in.-diameter casting containing 5.3 w/o carbon was prepared.

Alloy additions to uranium monocarbide have resulted in markedly improved properties. Additions of 1 to 10 w/o of Mo_2C, TiC, or UC have apparently decreased the corrosion rate of UC in Santowax R at 350 C by a factor of about 50 times, giving weight losses of the order of 10 mg/(cm^2)(week). Additions of 10 w/o of molybdenum, niobium, or tantalum (as metal) have produced castings with transverse rupture strengths in excess of 30,000 psi (the maximum stress applied).

Preliminary results indicate that the rate of self-diffusion of uranium in uranium monocarbide is about 100 times lower than the rate of interdiffusion of uranium and carbon. This is not too surprising, since there is an atomic-mass difference of 20 times and an atomic-volume difference of 5 times between uranium and carbon. Further work is needed to confirm this relation.

F-2

Alternate Fabrication Methods for UC

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

Various methods of preparing UC powder and dense UC bodies by powder-metallurgy techniques are being investigated.

Near-stoichiometric UC powder has been made by reacting uranium powder with propane at 1000 F. The total carbon content was 4.68 w/o, while the free carbon was 0.32 w/o. An X-ray diffraction analysis of this product revealed a very strong but diffuse pattern of UC and a very faint unknown phase.

The particle-size distributions of various uranium carbide powders (from uranium-propane reaction, uranium-methane reaction, commercial sources, and crushed arc-melted buttons) are being measured by Coulter-counter and microscopic techniques In the Coulter counter, approximately 4 x 10^4 particles from each of three representative samples are measured for each material. The sample is suspended in an alcohol-base solution by means of a high-speed stirrer and drawn upward through a 100-μ orifice by a pump. The change in resistance of the solution and particles going through the orifice is statistically related to the weight distribution of various sizes. The microscopic technique used consisted of estimates of various size factors as observed in three photomicrographs from each sample. The results of these measurements are shown in Table F-1. Further efforts to characterize the different uranium carbide powders will include a measurement of their surface area and their subgrain size, since each might likely influence their sinterability.

TABLE F-1. COMPARISON OF PARTICLE-SIZE DISTRIBUTION OF URANIUM CARBIDE POWDERS PRODUCED BY VARIOUS METHODS USING THE COULTER COUNTER AND MICROSCOPIC TECHNIQUES[(a)]

Specified Particle Size, μ	Average Amount Above Specified Size, w/o					
	Coulter Counter			Microscope		
	A	B	C	A	B	C
5	99	93	85	--	--	--
10	85	62	30	77	57	--
15	62	30	4	--	--	--
20	40	10	--	59	25	--
25	22	3	--	--	--	--
30	10	--	--	46	18	--
35	5	--	--	--	--	--
40	2	--	--	31	11	--
50	--	--	--	14	0	--
60	--	--	--	0	0	--

(a) Description of samples:
A - arc-melted uranium carbide, 4.83 w/o carbon, 1970 ppm oxygen, crushed and screened through 325 mesh (44 μ).
B - uranium carbide produced by uranium-methane reaction, 5.98 w/o carbon.
C - uranium carbide produced by uranium-propane reaction, 4.68 w/o carbon.

In Table F-2, preliminary sintering data are reported for several different types of uranium carbide powders. Although all compacts were pressed at 40 tsi, a considerable variation in green density was noticed. After sintering for 1 hr at 1900 C, the uranium-propane powder showed the greatest densification. Until metallography is available, the results are tentative in that the density differences may be only a reflection of the amount of free uranium formed during vacuum sintering.

A compact that was surrounded by degassed graphite was sintered in a static argon atmosphere in an attempt to reduce the formation of free uranium. Although metallographic data for this specimen are not available, it appears that little densification occurred during sintering.

Since previous work demonstrated a rapid rate of pore shrinkage in the presence of liquid chromium (liquid-phase sintering), a uranium carbide compact containing 10 volume per cent chromium was prepared and sintered in three steps - 30 min at 1030 C, 50 min at 1930 C, and 45 min at 2000 C. Practically all the chromium was lost during sintering. Analyses showed less than 0.05 w/o to be present as compared with the 5.54 w/o added initially. Even though the sintered compact was quite porous, the sintering process progressed considerably in that neck growth and grain growth between particles was quite extensive. Further tests are planned in which the process is followed in more detail just below and above the melting point of chromium.

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. During the past month, the preparation and improvement of large cast shapes of uranium carbide prepared by skull-type arc melting was continued. Sound, homogeneous castings have been made by melting and pouring into a heated mold. Until recently the carbon content of these castings has been around 6 w/o carbon, whereas the desired composition is 5 w/o carbon.

As suspected and reported last month, segregation existed in the uranium carbide skull in which the melts were made. It was found to contain over 6 w/o carbon near its top, making it impossible to pour castings of 5 w/o carbon. The skull composition was adjusted by making additions of unalloyed uranium. A series of large 2-in.-diameter castings has been made in order to decrease the segregation of carbon in the skull. These castings were analyzed and contain on the average 3.2 w/o carbon. Following the removal of excess carbon in this manner, the skull was restored to normal operating size with charges of metallic uranium and 3.5 w/o carbon. A 2-in.-diameter casting made from this skull was found to be homogeneous at a composition of 5.3 w/o carbon. While this is slightly above the desired carbon content, unusual carbon electrode erosion sufficient to cause the observed increase in carbon content occurred during the homogenization and outgassing of the new skull.

TABLE F-2. RESULTS OF SINTERING TREATMENTS ON VARIOUS URANIUM CARBIDE COMPACTS

	UC Powder Analysis, ppm			Carbon, w/o			Sintering Data			Density, g per cm^3	
Method of Production	Oxygen	Nitrogen	Hydrogen	Total	Free	Mesh Size	Environment	Time, min	Temperature, C	Green	Sintered
Arc melted and crushed	790	--	15	4.83	0.03	-325	Vacuum	60	1900	8.82	9.82
Arc melted and crushed	790	--	15	4.83	0.03	-325	Vacuum	60	1900	8.82	9.92
Commercial (Numec)	1900	3110	24	4.89	<0.01	-100 +325	Vacuum	60	1900	9.22	9.43
Commercial (Numec)	1900	3110	24	4.89	<0.01	-100 +325	Vacuum	60	1900	9.51	9.77
Uranium plus methane	6900	950	350	5.98	0.16	-325	Vacuum	60	1900	7.07	10.23
Uranium plus methane	6900	950	350	5.98	0.16	-325	Vacuum	60	1900	7.40	10.62
Uranium plus propane	5600	1600	46	4.68	0.32	-325	Vacuum	60	1900	7.11	11.40
Commercial (Numec)	1900	3110	24	4.89	<0.01	-100 +325	Vacuum	45	2000	8.47	10.09
Commercial (Numec)(a)	1900	3110	24	4.89	<0.01	-100 +325	Vacuum	45	2000	8.47	10.12
Arc melted and crushed	790	--	15	4.83	0.03	-325	Static argon and degassed graphite	30	1910	9.28	8.37

(a) 10 volume per cent chromium metal added to promote sintering.

Research during the next month will involve continued work on composition control and production of 3/4-in.-diameter by 8-in.-long cylindrical shapes of uranium carbide with a nominal content of 5 w/o carbon. A hopper feed and several other devices are being considered to increase the number of castings that can be produced in the skull-type arc furnace in a given time.

Metallurgical and Engineering Properties of Uranium Monocarbide

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

The present study of uranium carbides is concerned with the definition and improvement of the properties of these materials. Past work has partially defined the effects of impurities and carbon content upon density, rupture strength, electrical resistivity, and corrosion resistance. An investigation of the effects of ternary additions and heat treatment on the above properties as well as compatibility and thermal properties is being continued.

Ternary additions of 1 w/o or 10 w/o Mo_2C, TiC, or VC to uranium monocarbide had little effect on the transverse rupture strength of the material. The corrosion rate of uranium monocarbide in Santowax R at 350 C has been reduced by these additions from 500 $mg/(cm^2)(week)$ to less than 10 $mg/(cm^2)(week)$. Like amounts of Al_4C, Cr_3C_2, NbC, TaC, and ZrC resulted in lesser improvements in corrosion resistance. Work is in progress to determine the optimum amount of the above carbides in UC, U_2C_3, and UC_2 and their effect on other physical and mechanical properties, as well as upon corrosion resistance in other media and upon compatibility with possible cladding materials.

Additions of 10 w/o molybdenum, niobium, or tantalum to UC resulted in transverse rupture strengths of greater than 30,000 psi. The effects of lesser and greater amounts of the above additions as well as of titanium and vanadium are being investigated with respect to compatibility and physical, mechanical, and corrosion properties.

To define the thermal properties of UC_2, U_2C_3, and UC, specimens are being prepared for thermal-conductivity, specific-heat, and linear thermal-expansion measurements. Work is also being continued on the effects of annealing temperature on UC and U_2C_3.

Uranium Monocarbide Diffusion Studies

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

Diffusion rates in uranium monocarbide are of interest in predicting the elevated-temperature behavior of this material since it is a potential nuclear fuel for reactors operating at elevated temperatures. The rate of interdiffusion of uranium and carbon in uranium carbide at a series of temperatures has been determined and reported previously. Diffusion couples of uranium monocarbide have been bonded successfully, and the techniques necessary for determining the rates of self-diffusion of uranium in uranium monocarbide have been proved adequate. Currently the self-diffusion of uranium in uranium monocarbide is being studied. A couple has been evaluated and additional couples are being processed.

In the current study, the diffusion couples consist of a 0.001-in.-thick foil of enriched uranium (93 per cent uranium-235) metal sandwiched between two 0.25-in. lengths of depleted uranium (0.04 per cent uranium-235) monocarbide rod. The experimental procedure includes wrapping the couple in tantalum foil and placing it in a degassed graphite jig, bonding this assembly in vacuum for 4 hr at 1400 C, and, finally, diffusion annealing. After annealing, the couple is sampled for neutron-activation analyses for 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 evaluating a diffusion couple annealed for 90 min at 1800 C, a diffusion zone approximately 0.008 in. in width was found to have formed. To determine the presence and approximate percentage of uranium-235 present in the individual samples, their gross gamma activity was measured. The results obtained indicated that the techniques developed for the study are reliable. Since the diffusion zone was of insufficient size to give an adequate number of analytical samples, subsequent couples will be prepared using longer annealing times. A self-diffusion coefficient for uranium in uranium monocarbide has been calculated using the gross gamma analytical data from this couple. The value obtained was approximately 100 times lower than the value of the interdiffusion coefficient which was determined previously.

A couple has been bonded for 120 min at 2000 C. Metallographic examination of the couple indicates that it is satisfactorily bonded and annealed. This couple will be sampled for neutron-activation analyses.

Future work will include the preparation of additional diffusion couples and the neutron activation of samples from these diffusion couples in the BRR.

Irradiation Effects in UC

A. E. Austin, D. G. Freas, C. M. Schwatz,
and D. Stahl

An investigation of the effect of fission and irradiation on the structural and physical properties of UC is in progress. Specimens, 1/4 in. in diameter by 1/2 in. long, of unenriched uranium carbide with a nominal 5.0 w/o carbon content are being prepared for irradiation. They will be encapsulated in sets of six and exposed in the BRR at low temperatures (less than 500 F) and to low burnups (0.01 to 0.02 a/o of uranium). Enriched specimens will also be prepared and irradiated to higher burnup. The purpose of these experiments is to attempt to obtain measurements of strain and crystal-structure disorder for correlation with burnup and physical properties. For this purpose, it is planned to use X-ray diffraction measurements and electron microscopical observation. The additional specimens in the capsule will provide material for subsequent annealing heat treatments. In order to eliminate any strain developed in the arc-casting operation and possible transformation during postirradiation heat treatments, the specimens are being annealed at about 1600 C.

Continued examination by electron microscopy is being made of UC specimens irradiated to high burnups for the purpose of investigating the previously reported (BMI-1403) disappearance of the UC_2 precipitate. Macroreplicas from these specimens will be made by coating with cellulose acetate film wet with methyl acetate and drying in a desiccator box containing barium oxide. These negative replicas will then be replicated again using a cellulose nitrate film to give a thin positive replica for electron microscopy. Further examination of replicas so made from unirradiated UC specimens has revealed a fine granular structure which is an effect of interaction in this double-replica process. Other positive-replica processes are being investigated to minimize this artifact structure.

G. PHYSICAL RESEARCH (AEC-DR)

F. A. Rough

In the study of the mechanism of migration of hydrogen in zirconium under a temperature gradient, the determination of the needed diffusion coefficients has been completed, and work is in progress to conduct an experiment for comparison with theoretical predictions of hydrogen migration. The sample to be studied is pressure bonded in stainless steel.

Several methods of preparation of UO_2 single crystals by deposition from vapor and by various fusion methods have been studied. The methods being studied now are: (1) the deposit of crystals in a molten potassium fluoride-UO_2 bath as the potassium fluoride is volatilized off; and (2) pulling of UO_2 crystals from a molten UO_2 bath contained in tungsten.

Fusion Methods to Prepare Single Crystals of UO_2

W. P. Allred and B. Paris

During January, research was continued on the problem of growing large single crystals of UO_2 by fusion methods. To date, several approaches to this problem have been undertaken. The use of the electron-beam floating zone has been investigated; it appears that the vapor pressure of the material is too high for the use of this equipment. In a study of floating-zone melting by use of induction heating, problems of coupling to the ingot with r-f current have been encountered.

The UO_2 has been melted using a resistance-heated tungsten spiral. The UO_2 was in direct contact with the tungsten, however. Attempts to melt a zone in the UO_2 rods without direct contact with the tungsten failed because the tungsten became ductile at the high temperatures and sagged out of shape.

It was interesting that although the molten UO_2 shorted across the tungsten windings, no shorting of the current path was noticed. It is suspected that the resistivity of molten UO_2 is still too high for efficient heating by r-f current. It was also noticed that no visible reaction of the UO_2 with tungsten occurred. This observation leads to a very encouraging possibility. It is possible that single crystals can be grown by pulling the crystals from the melt such as is done with many of the semiconductor materials. A tungsten crucible would be used to contain the melt. It is probable that the UO_2 can also be zone refined in a tungsten boat, and that the single crystals can be grown at the same time. Adaptation of a crystal puller is being made to exploit this possibility.

G-2

Thermal Migration of Hydrogen in Zirconium

J. W. Droege, W. M. Albrecht,
W. D. Goode, and H. H. Krause

A basic study of the migration of hydrogen in zirconium under the influence of a thermal gradient is being conducted. A new diffusion cell, prepared by pressure bonding, has been completed. Diffusion coefficients for hydrogen in delta zirconium hydride have been determined, both as functions of temperature and of hydrogen concentration.

Thermal Diffusion

Improved pressure-bonding techniques have made it possible to fabricate a diffusion cell with adequate bonding of the zirconium hydride to the stainless steel container. The test element previously employed showed a good metallurgical bond, so identical conditions were used in producing the new unit. Helium pressure of 10,000 psi was applied at 1800 F for 2 hr, followed by 1 hr at 1900 F. During the pressure bonding the composition of the hydride changed from $ZrH_{1.02}$ to $ZrH_{0.90}$, indicating a loss of hydrogen through the stainless steel cladding at these temperatures. The diffusion experiments for this study will be conducted at lower temperatures and with sufficient partial pressure of hydrogen outside the element to prevent any loss of hydrogen during the runs.

The diffusion cell is now being incorporated in the vacuum system and fitted with heating and cooling coils. It will then be placed in the gradient furnace, and calibration experiments at constant temperature will be carried out. Thermal diffusion will be measured in terms of hydrogen concentration at the faces of the zirconium hydride specimen when a one-dimensional temperature gradient is applied.

Diffusion Coefficients

The experimental determinations of diffusion coefficients for hydrogen in zirconium hydride in the ranges 500 to 750 C and 60 to 65 a/o hydrogen have been completed. It was found that the diffusion coefficients determined by the steady-state and time-lag methods were in fair agreement. However, the values obtained from the time-lag method were judged to be the more accurate. Diffusion coefficients obtained by the time-lag method since the last report (BMI-1398) are given in Table G-1.

The results of eight determinations showed that there was no definite trend in the values of the diffusion coefficients with hydrogen concentration. Therefore, the diffusion coefficients for hydrogen in zirconium hydride may be expressed by

$$D\ (\text{cm}^2\ \text{per sec}) = 804\ \exp\left[-(35{,}300 \pm 1{,}100)/RT\right],$$

as determined by the least-squares method from time-lag data.

TABLE G-1. DIFFUSION COEFFICIENTS FOR HYDROGEN IN ZIRCONIUM HYDRIDE

Temperature, C	Average Composition, a/o hydrogen	Diffusion Coefficient, D, 10^{-6} cm^2 per sec
600	60.9	0.82
600	65.3	0.96
700	64.7	11
750	63.8	25

A paper on the diffusion of hydrogen in beta zirconium and zirconium hydride is being prepared.

Growth of UO_2 Crystals From the Vapor Phase

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

Experimental effort this past month has been directed toward producing UO_2 crystals from the reaction of $UF_4(g)$ and $H_2O(g)$. The apparatus employed consisted of a modified transpiration vapor-pressure apparatus which employed two separate carrier-gas flow systems. Pure argon was introduced at one end of the apparatus and flowed over the molten UF_4 and carried the vapor to another region where a flow of water vapor in argon impinged upon the gaseous UF_4. Many small red crystals of UO_2, approximately 0.1 mm in size, were found in the vapor deposition. A serious problem in this experiment was the plugging of the water-vapor inlet line due to deposition of mixed UO_2 and UF_4 at the orifice. A larger orifice should permit runs of several days' duration, however.

From the observations covered in last month's report (BMI-1403), where it was noted that UO_2 crystals were found in the UF_4 charge, it was decided to attempt some runs using a solution of UO_2 in a readily soluble fluoride. Evaporation experiments with 10 mole per cent of UO_2 in molten KF were made this month and have yielded the largest crystals of UO_2 produced so far. The procedure employed in these experiments has been to load a shallow nickel boat with the 10 mole per cent charge and flow argon over the charge to remove the KF vapor. The crystals formed were rectangular in shape and were growing into the solution from the surface. Crystals as large as 3 by 2 by 0.5 mm have been grown through evaporation to date. For next month, it is planned to continue vaporization and evaporation experiments in an effort to obtain larger crystals.

H. RADIOISOTOPES AND RADIATION APPLICATIONS (AEC-OID)

D. N. Sunderman

The program of research in the fields of radioisotope applications and radiation chemistry is continuing. Radioisotope applications include the use of radiotracer techniques in quality control and the study of intrinsic tracers for process control. Radiation-chemistry studies are evaluating the structural factors in radiation-induced graft polymerization and the effect of phase upon the nitration of hydrocarbons.

The complexometric titration of iron with EDTA using a radiometric endpoint was shown to be capable of a precision and accuracy of better than ±1 per cent standard deviation. Results obtained on eight analyses with about 75 mg of Fe_2O_3 indicated a maximum absolute error of 1.84 per cent, with the average value better than 0.5 per cent.

Intrinsic-tracer work is being directed toward the assembly of equipment for the small-scale engineering demonstration of the technique. Major pieces of equipment have been ordered. Earlier effort on the application of intrinsic tracers to unit operations has been reactivated to make up for a lag in experimental work.

Radiation-induced free-radical site formation has been studied for poly-tertiary-butylmethacrylate, polycyclohexylmethacrylate, and polymethyl-α-chloracrylate. Substituents both on the ester chain and the α-carbon influence the rate of formation of sites as a function of radiation dose. Preliminary results also indicate that the total dose may affect the lifetime of radicals, at least in polymethylmethacrylate.

Nitration experiments have been conducted to determine the efficiency of previous results which indicated an inhibition of nitration by the presence of ionizing radiation. These experiments pointed up the importance of phase contact, which, if not carefully controlled, could give rise to the differences obtained in radiation and control experiments. The addition of nitrocyclohexane to the reaction vessel prior to irradiation has shown that very little will be degraded by the irradiation conditions.

Development of Radioactive-Tracer Quality-Control Systems

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

Work was continued on the determination of iron in portland cement by means of a complexometric titration with a radiometric endpoint. The procedure under study consists of an EDTA titration with $Y_2{}^{91}(C_2O_4)_3$ as an endpoint indicator.

A series of determinations was carried out using 26.0 mg of iron (III) in solution as the nitrate. Tartaric acid (120 mg) was added to maintain the iron in solution, and 10 ml of 1 M sodium acetate was added to buffer the solution at pH 5.0. Approximately 200 mg of $Y_2{}^{91}(C_2O_4)_3$, having a specific activity of 7.5×10^{-4} μc per mg, was added to the mixture. A standard EDTA solution (0.05 M) was used to titrate the iron (III).

After each addition of EDTA the solution was stirred at least 10 min and then radioassayed. The counting rate of the solution was plotted as a function of the volume of EDTA added, and a sharp increase in counting rate was observed as the endpoint was passed.

The results of iron determinations made in this manner are given in Table H-1. The average of eight analyses is 74.45 mg of Fe_2O_3 with a standard deviation of 0.33 mg. The calculated quantity of Fe_2O_3 in each sample was 74.25 mg. The precision and accuracy of the method are both within 0.5 per cent.

TABLE H-1. PRECISION AND ACCURACY OF EIGHT IRON (III) DETERMINATIONS

Determination	Fe_2O_3, mg	Deviation From Average, mg	Calculated Fe_2O_3, mg	Error, per cent
1	73.63	-0.82	74.25	0.84
2	74.27	-0.18	74.25	0.003
3	74.58	+0.13	74.25	0.44
4	74.43	-0.02	74.25	0.24
5	74.66	+0.15	74.25	0.55
6	75.62	+1.17	74.25	1.84
7	74.35	-0.10	74.25	0.13
8	74.03	-0.42	74.25	0.30
Average values	74.45	±0.37	--	--
Standard deviation	--	±0.33	--	--

Future work on the iron analysis will include studies of the effects of interfering ions, reagent concentrations, iron concentration, and other variables. The development of a radiometric method for aluminum will also continue.

Use of Intrinsic Radioactive Tracers for Process Control

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

During January, the major effort on this program was spent on the construction of the model iron-removal process system. The over-all design has been completed and all major hardware items have been ordered. Actual assembly of the apparatus will proceed as the equipment is received.

Early in this program the unit operations of chemical engineering were surveyed briefly to help choose the initial radiotracer application. The unit operations will now be studied in detail. The object of this study is to determine the conditions for intrinsic radiotracer applications to each unit operation. From this study applications to specific industrial processes will be suggested.

Further study of the electrolytic zinc process suggests that there is very little advantage to be gained from intrinsic radiotracer control of the iron-impurity removal step. The concentration of iron impurity in the electrolyte is apparently not critical, and its removal presents no unusual problems. However, some other elements such as germanium and antimony are very injurious if present, even in small amounts. It might be feasible, therefore, to use intrinsic tracers of these impurities in zinc-electrolyte purification.

Next month, the evaluation of alternate applications of iron removal will continue, and the detailed analysis of unit operations for intrinsic tracer applications will be initiated. Construction of the process model will be carried forward as the equipment becomes available.

Graft-Polymerization Studies

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

During the past month the investigation of the formation and decay of radiation-induced free radicals progressed. A study of the effect of total dose on site formation was made for poly-tertiary-butylmethacrylate (PtertBMA), polycyclohexylmethacrylate (PCHMA), and polymethyl-α-chloroacrylate (PM-α-ClA). A comparison of the plots of sites per monomer unit for polybutylmethacrylate (PBMA) and PtertBMA indicated that PtertBMA at the same total dose contained more active sites than PBMA. For example, at a total dose of $\sim 10^7$ rads PBMA contained $\sim 9 \times 10^{-4}$ site per monomer unit, while PtertBMA contained $\sim 5.5 \times 10^{-2}$ sites per monomer unit. A similar comparison of PM-α-ClA with polymethylmethacrylate (PMMA) indicated that PM-α-ClA contained more sites at a given total dose than did PMMA. At a total dose of 1×10^7 rads PMMA contained $\sim 5 \times 10^{-3}$ site per monomer unit while PM-α-ClA contained $\sim 5 \times 10^{-2}$ site per monomer unit. From the work described above and that discussed in the previous monthly report (BMI-1403), it may be seen that substituents on the ester chain as well as on the α-carbon of methacrylates exert an influence on the number of sites formed for a given radiation dose. It is not known, at this time, whether these differences in numbers of sites are due to differences in ease of formation or in stability of the radicals.

The last monthly report (BMI-1403) disclosed that at high total dosages ($>10^7$ rads) the number of sites per monomer unit leveled out and started to decrease with increasing dose. Several possible explanations were advanced, including the possibility of the existence of two or more species of free radicals. Samples of PMMA were irradiated to 4×10^5 and 5×10^7 rads, and their half-lives were determined. Preliminary data indicated that the half-life of PMMA radicals formed by irradiation to 4×10^5 rads was roughly four times that formed by irradiation to 5×10^7 rads.

During this month samples of PMMA with molecular weight varying from ~4000 to ~15,000 were irradiated. A plot of sites per monomer unit versus total dose demonstrated a difference in the rate of site formation. In general it appeared that the higher-molecular-weight samples had a higher rate of site formation.

During the next month work will continue on the study of the mechanism of radiation-induced free radicals in methacrylates. Samples of polypropyl-, isopropyl-, butyl-, secondary butyl-, isobutyl- and tertiary butylmethacrylates will be irradiated to determine the effect of branching of the ester chain on site formation. Preparations are being made to look for methylmethacrylate, by gas chromatography, in the gases formed during irradiation. The presence of monomer would help explain the change of EPR spectra and the apparent shortened half-life of radicals formed by higher dosages.

Nitration of Hydrocarbons

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

During January, major effort was devoted to an investigation of the apparent inhibiting effect of radiation on nitration. Fourteen irradiation and thermal experiments were completed with the nitric acid-cyclohexane system in the liquid phase. Analytical results have been obtained for eleven of these experiments are summarized in Table H-2.

All experiments were conducted at 60 C using a 10/1 mole ratio of cyclohexane to nitric acid. Samples were irradiated from 20 to 65 hr, corresponding to gamma doses from 3.8×10^6 to 1.2×10^7 rads.

A series of experiments was conducted at 20-hr exposure times to confirm the yields obtained previously and also to check the reproducibility of experimental conditions between the irradiation and nonirradiation experiments. Irradiation and thermal experiments (45 and 52, respectively) were conducted in a watertight pool apparatus. These experiments show a slight increase in nitrocyclohexane yield in a radiation field. However, these yields are lower than that obtained in an otherwise similar experiment (43) conducted in the constant-temperature oil bath. Thermal experiments (41 and 42) conducted in the pool apparatus with no stirring between the two liquid phases also produced lower yields. Good stirring can easily be assured in the oil-bath experiments but not in those conducted in the enclosed apparatus used in the pool.

The fourth column of Table H-2 shows the approximate moles of gas produced in these experiments. Again, any difference between thermal and irradiation experiments depends on the efficiency of mixing the two liquid phases. Results show a slight increase in the moles of gas produced with increasing exposure time, as expected.

The stability of nitrocyclohexane in a radiation field was investigated by spiking cyclohexane with 1.6 w/o nitrocyclohexane prior to reaction with nitric acid. The results obtained with spiked reaction mixtures (Experiments 48 and 49) are essentially the same as those obtained with unspiked reaction mixtures (Experiments 45 and 43). Therefore, nitrocyclohexane is not significantly decomposed under these experimental conditions.

The accumulated data indicate that the low yields of nitro products in irradiation experiments, relative to the nonirradiation experiments, could be due to the lack of efficient mixing of cyclohexane and nitric acid. Higher yields were always obtained when mixing efficiency was increased. The fact that mixing could be observed in thermal runs but could not be observed or assured in the irradiation experiments would account

TABLE H-2. PRODUCT ANALYSIS OF NITRIC ACID-CYCLOHEXANE EXPERIMENTS[a]

Experiments	Time, hr	Gamma Dose, 10^{-6} rads	Moles of Gas Produced	Analysis of Recovered Organic Solution, w/o						Comments
				$C_6H_{11}NO_2$	$(C_6H_{11})_m(NO_2)_n$	$(C_6H_{11})_2$	$C_6H_{11}OH$	$C_5H_{10}C{=}O$	Adipic Acid or Other Solids	
45	20	3.8	0.013	0.27	--	--	Trace	Trace	0.005	Pool
52	20	--	0.010	0.18	--	--	Trace	Trace	0.035	Pool
43	20	--	(b)	1.2	--	--	Trace	Trace	1.64	Oil bath
41	20	--	(b)	0.06	--	--	--	--	--	Pool – poor stirring
42	20	--	(b)	0.10	--	--	Trace	Trace	--	Pool – no stirring
48(c)	20	3.8	0.013	2.0	Trace	Trace	Trace	Trace	0.006	Pool
49(c)	20	--	0.017	2.7	0.05	Trace	0.05	0.02	1.41	Oil bath
50	30	--	(b)	1.4	0.07	Trace	0.11	0.03	1.91	Oil bath
46	45	8.6	0.020	1.4	0.12	0.05	0.17	0.02	1.67	Pool
47	65	12.4	0.023	1.9	0.14	Trace	0.18	0.03	2.69	Pool
44	65	--	0.021	2.6	--	--	0.15	Trace	2.02	Oil bath

(a) All experiments were conducted at 60 C using a 10/1 mole ratio of cyclohexane/nitric acid in glass vessels.

(b) Not determined.

(c) Reaction mixture consisting of nitric acid and cyclohexane spiked with 1.6 w/o $C_6H_{11}NO_2$.

for the generally higher yields obtained in the thermal runs and explain the apparent inhibitory effect of radiation on the nitration reaction.

During the next month experiments will continue with the cyclohexane-nitric acid system to investigate the effect of radiation under conditions or more efficient mixing of the reactants. Experiments will be initiated employing NO_2 as the nitrating agent. From these experiments it may be possible to determine whether the type of nitrating agent influences the effect of radiation on nitration.

I. VOID-DISTRIBUTION AND HEAT-TRANSFER STUDIES (AEC-EURATOM)

Roger O. Wooton, David V. Grillot, Harold M. Epstein,
M. Frederick Peter, and Joel W. Chastain

During the past month, methods of eliminating the problem of two-phase flow instabilities were studied. Several approaches to the problem were attempted, and the final solution was twofold in nature.

First, it was discovered that the present system is extremely sensitive to power fluctuations. To overcome this difficulty, the experiments will be run at night to minimize line-voltage variations. The second alteration, a change in throttling techniques, has eliminated all major instabilities in the loop. This second alteration involves a technique of balancing the throttling of the system at both the inlet and outlet of the test section.

These modifications have made it possible to obtain meaningful data, and it is anticipated that these experiments will be continued throughout the next month.

Elimination of major experimental difficulties permitted attention to be given to methods of interpreting and correlating experimental data. Information obtained in correlating data from initial test conditions will indicate which parameters have greatest influence on void distribution. These parameters will be used as the bases for determining new test conditions which will be studied during the coming month.

J. MATERIALS DEVELOPMENT AND EVALUATION (HAPO)

F. R. Shober

The effect of repeated temperature cycles on the creep properties of annealed Zircaloy-2 is being investigated, as well as the long-term creep properties at 290, 345, and 400 C. Tests are planned in which both the stress and the temperature will be cycled. In studies to develop a AgBr fuel-element leak detector for use in water-cooled reactors, work during the month involved: (1) continuation of the fission-product decontamination studies, (2) initiation of experiments to determine the solubility of AgBr in water at water-flow rates of 0.1 gal per min, and (3) measurement of solubility of AgI at flow rates from 0.1 to 1 gal per min.

The use of $MoSi_2$-Al_2O_3-UO_2 ceramic sensing probe in studies to develop a thermal-neutron-flux monitoring system has been limited due to the erratic electrical behavior of the material. Several new approaches for producing uranium-bearing resistors are being surveyed. A study of the corrosion properties of welding alloys for Hastelloy F has been continued. Self-welded and unwelded specimens of these alloys have been exposed in boiling Sulfex and Niflex solutions.

Mechanical Properties of Zirconium Alloys

L. P. Rice and J. A. VanEcho

The long-term program for determining the creep and stress-rupture properties of Zircaloy-2 sheet material has continued. During January a specimen in one test ruptured, and three thermal-cycling tests were discontinued. Current tests are being conducted with annealed material at 290, 345, and 400 C.

Since the last report period, one annealed Zircaloy-2 specimen being creep tested at 345 C (650 F) has failed after 7184 hr. During the same period three cyclic tests were discontinued after 3000 hr. In Table J-1 the creep behavior of temperature-cycled specimens of annealed Zircaloy-2 is summarized to date. Results of the non-cycled creep tests on the annealed material at comparable stresses are still incomplete. Therefore, the effect of the thermal cycle is still not definitely known. Indications are, however, that the creep rate is increased by cycling at both 290 and 345 C.

In the near future, some additional cyclic tests will be made on the annealed material where the stress, as well as the temperature, will be cycled. In these tests, the stress will be lowered to zero every 6 days and at the same time the temperature will be reduced to room temperature.

TABLE J-1. SUMMARY OF CYCLIC-TEMPERATURE[(a)] CREEP DATA ON ANNEALED ZIRCALOY-2 SHEET STRESSED IN THE LONGITUDINAL DIRECTION

Specimen	Stress, psi	Deformation at Indicated Time, per cent: Loading	500 Hr	1000 Hr	2000 Hr	3000 Hr	Final	Load Off	Total Time, hr	Minimum Creep Rate, per cent per hr
					290 C (550 F) Tests					
Zr-A-24	20,000	2.91	4.08	4.11	4.15	4.18	4.18	4.01	3018	0.00004
Zr-A-19	17,000	1.48	1.84	1.87	1.90	1.93	1.93	1.79	3030	0.000025
Zr-A-1	14,000	0.08	0.14	0.15	0.16	0.17	0.17	0.11	3022	0.000012
					345 C (650 F) Tests					
Zr-A-23	17,000	1.58	1.91	1.98	2.10	2.21	2.21	2.08	3031	0.00012
Zr-A-22	15,000	0.93	1.04	1.09	1.18	1.26	1.26	1.14	3032	0.000085
Zr-A-4	12,500	0.19	0.37	0.46	0.59	0.68	0.69	0.58	3105	0.000075

(a) Cycle consists of 6 days at temperature (290 or 345 C) and 1 day at room temperature.

Development of a Fuel-Element Leak Detector

J. E. Howes, Jr., T. S. Elleman, and D. N. Sunderman

Investigations this month on the development of an isotopic-exchange leak-detection system have included: (1) continuation of the fission-product decontamination studies, (2) initiation of experiments to determine the solubility of AgBr in water at water-flow rates of 0.1 gal per min, and (3) measurement of the solubility of AgI at flow rates from 0.1 to 1 gal per min.

The decontamination studies are part of the general investigation of techniques for preventing fission-product buildup on AgBr columns. The AgBr solubility determinations at a water-flow rate of 0.1 gal per min have been carried out to supplement existing solubility data which were taken at much higher water-flow rates. The AgI solubility data will be used to evaluate the usable lifetime of an exchange column containing AgI rather than AgBr.

In the fission-product decontamination studies it was found that fission products in acid solution were retained to a much lesser extent by AgBr columns than fission products in neutral solutions. When fission products in approximately 0.3 N HNO_3 were passed through an AgBr column, only 0.18 per cent was retained. This retention percentage is about 20 times lower than the fission products retained from runs made in neutral solution. Also, it was discovered that fission products could be easily washed off AgBr columns by an acid rinse. A wash with 300 ml of 6 N HNO_3 was found to remove about 99 per cent of the fission-product activity normally retained by an AgBr column. These results indicate that it should be possible to maintain a low fission-product background on an operating AgBr column by periodically washing the column with acid.

Experiments are under way to determine the rate of dissolution of AgI as a function of flow rate and the rate of dissolution of AgBr at a low flow rate. Results of these experiments will give information concerning the usable lifetime which can be expected from AgBr and AgI columns at various flow rates.

The direction of future investigations on the program will depend on the results of the program review which is currently being conducted.

Thermal-Neutron-Flux Monitoring System

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

In work to adapt a thermal-neutron-flux monitoring device (BMI-1083) for use in the Hanford reactors, no procedure for consistently preparing a $MoSi_2$-Al_2O_3-UO_2 ceramic electrical heating element with the properties desired has been found. Accordingly, new material combinations are being considered.

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

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

Self-welded specimens of the 12 experimental alloys and unwelded specimens of vacuum-melted Hastelloy F have been exposed to boiling Sulfex and Niflex solutions for five 24-hr periods.

The Sulfex solutions were aerated at a rate of about 0.5 ml of cylinder air per ml of solution per min. As anticipated, this resulted in roughly a tenfold increase in corrosion rates over similar exposures in nonaerated solutions. No borderline passivity was shown, the specimens being active each period. The rates are shown in Table J-2.

Even though considerably higher rates were measured in the aerated Sulfex solutions the weldments were not selectively attacked. The attack on all specimens was fairly uniform, and the corrosion rates, except for Alloy 1, were sufficiently similar that there is little basis for choosing one alloy over another for Sulfex use.

The weldments of submerged specimens of Alloys 1, 2, 7, and 8 were perforated by three 24-hr-exposure periods to boiling Niflex solutions. After five periods, the weldments of Alloys 9 and 12 showed severe attack. The same type of attack occurred with Hastelloy F specimens welded with these alloys, as mentioned in BMI-1403. However, weldments with Alloys 3, 4, 5, 6, 10, and 11 did not show selective attack. Furthermore, there was no increased attack in the heat-affected areas of the parent metal. This was unlike the situation with Hastelloy F specimens welded with these alloys where, in each case, heavy attack occurred in the heat-affected zone. These experimental alloys corroded uniformly and, although the rates were high, the results with self-welded Alloys 3, 4, 10, and 11 are encouraging in that they are no higher than those with the unwelded specimens.

TABLE J-2. CORROSION RATES OF SELF-WELDED SPECIMENS OF EXPERIMENTAL ALLOYS AFTER EXPOSURE TO SULFEX OR NIFLEX SOLUTIONS

Alloy(a)	Corrosion Rate(b), mils per month	
	Sulfex Solution	Niflex Solution
1	28	82(c)
2	17	102(c)
3	19	37
4	17	41
5	15	58
6	18	61
7	19	102(c)
8	15	145(c)
9	16	54
10	20	35
11	21	37
12	16	61
Hastelloy F (unwelded)	16	69

(a) For alloy compositions, refer to Table A-3, BMI-1398.

(b) The corrosion rates given are the average values for five 24-hr periods of exposure of duplicate liquid-phase specimens to the boiling solutions. The Sulfex solution was aerated.

(c) These weldments were perforated after three 24-hr periods, and the average values are based on this exposure.

Specimens of Alloys 1 and 4 in the as-cast condition were exposed to boiling Niflex solution. The rate for Alloy 1 was about ten times as high as measured on the unwelded, rolled specimen, while the rate for Alloy 4 was about double. This helps explain why the weldments made with Alloy 1 are attacked so severely. It is evident that niobium additions are not desirable. A few preliminary experiments have been made in Niflex solutions with other alloys such as Haynes 21, Illium 98, and CD4MCu. Using the data collected thus far as a guide, another group of 12 experimental compositions is being assembled.

K. DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS (SRL)

R. J. Carlson and N. E. Daniel

Creep and stress-rupture tests have been initiated on an aluminum-35 w/o uranium binary alloy and on ternary alloys containing 2 and 3 w/o tin or zirconium. Data from stress-rupture tests conducted at 200 C with stresses of 11,000 to 14,000 psi have been obtained. The effects of impurities on the eutectic temperature of the aluminum-uranium-nickel ternary system are being investigated. Initial results indicate that the lowering of the aluminum-nickel eutectic temperature is reflected in subsequent lowering of the ternary eutectic temperature.

Preparation of Aluminum-Uranium Alloys

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

Previous experience with the aluminum-35 w/o uranium alloy has shown that improvements in casting and fabricating characteristics can be obtained when suitable ternary additions are used. Among the additions which appear to improve the alloy are tin and zirconium. The improvements noted can be attributed to the increase in the matrix-to-compound ratio that is brought about by the inhibition of the UAl_3-to-UAl_4 reaction and also to decreases in the particle size of the UAl_3. Before alloys containing these additions are used it is desirable to determine the effects of the additions upon the mechanical properties, the corrosion resistance, and the castability of aluminum-35 w/o uranium alloy. The program in progress is concerned with the effects of additions of up to 3.0 w/o tin and zirconium.

Creep and stress-rupture tests have been initiated on the aluminum-35 w/o uranium binary alloy and on ternary alloys containing 2 and 3 w/o tin or zirconium. From the yield and tensile data obtained at elevated temperatures it was decided that the most meaningful stress-rupture data could be obtained at 200 C. The initial tests were conducted at 200 C with stresses approximating the ultimate strength of the alloys. The data obtained from these tests are shown in the tabulation below.

Alloy Composition	Stress, psi	Time of Failure, hr	Elongation, per cent	Reduction in Area, per cent
Al-35 w/o U-2.0 w/o Sn	12,350	0.15	15.4	16.8
	11,000	0.9	19.5	23.0
Al-35 w/o U-3.0 w/o Sn	12,050	0.2	14.6	20.0
Al-35 w/o U	14,000	0.025	6.5	9.7
	11,000	6.0	--	--
Al-35 w/o U-2.0 w/o Zr	13,450	0.1	16.2	18.8
	11,000	4.7	--	--
	13,400	0.116	19.5	30.7
Al-35 w/o U-3.0 w/o Zr	12,350	0.0016	16.2	29.5
	13,400	0.133	13.0	19.0

More complete data will be available next month from tests using lower stresses.

To evaluate the effects of tin and zirconium on the coating characteristics of lower uranium alloys, aluminum-25 w/o uranium alloys containing these additions have been prepared in the form of hollow cylindrical extrusion billets. These billets were prepared utilizing a two-hole graphite pouring-cup rotating over a stationary vertical mold at approximately 16 rpm. The melts were poured at 2150 F into a steel mold with a steel core. Water cooling was used to hasten cooling and to impart directional solidification to the castings. The billets are being evaluated by chemical analyses and radiographic examination of selected segments.

Creep and stress-rupture data will be of prime interest in future work. The emphasis will continue to be placed upon alloys containing 2 and 3 w/o tin or zirconium. Future alloy studies will be concerned with the effects of two or more elemental additions on the structure of the aluminum-35 w/o uranium alloy.

A Study of an Aluminum-Uranium-Nickel Eutectic Temperature

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

The effects of impurities present in commercial-grade materials on the ternary eutectic temperature of the aluminum-uranium-nickel system is being investigated. The ternary eutectic temperature was previously determined to be 624 ± 2 C when very pure materials were employed for study. There is some feeling that impurities present in commercial-grade materials may lower this temperature. The problem is of importance in diffusion bonding of aluminum-clad uranium fuel elements which contain a nickel diffusion layer. As sufficient diffusion probably occurs in some areas to allow all three components to be in contact, the bonding operation must be restricted to temperatures below that of the ternary eutectic or cladding failure may result.

Alloys corresponding to the low-melting binary and ternary eutectic compositions of the systems involved have been prepared by induction-melting techniques. These are aluminum-13.3 w/o uranium, aluminum-5.7 w/o nickel, and aluminum-13 w/o uranium-5.6 w/o nickel. The aluminum used in preparing these alloys corresponds to commercial 2S aluminum, while the nickel is the product of an electrolytic operation and is of unknown purity.

Initial differential thermal-analysis data for the aluminum-13 w/o uranium-5.6 w/o nickel alloy indicate eutectic melting began at approximately 621 C. This is 3 C lower than the 624 C temperature observed for a similar alloy prepared using high-purity materials. Eutectic temperatures observed for the binary aluminum-uranium and aluminum-nickel eutectic alloys prepared from commercially pure materials were 640 and 629 C, respectively. With very pure starting materials, both binary eutectic temperatures occur at 640 C. From these results, it would appear that the lowering of the aluminum-nickel eutectic temperature is reflected in the subsequent lowering of the ternary eutectic temperature.

Additional differential thermal-analysis data will be obtained to confirm these temperatures.

L-1 and L-2

L. REFLECTIVE-INSULATION STUDIES (KAISER)

L. J. Flanigan and H. R. Hazard

An investigation of the characteristics of reflective-insulation materials suitable for use in the Experimental Gas-Cooled Reactor is being carried out at Battelle to provide design data for the prototype. Suitability of the insulation is to be judged by its freedom from distortion or other damage occurring during thermal cycling and rapid decompression, and by its insulating value.

Work on the project was started early in January. Conceptual design on the full-temperature and pressure heat-transfer apparatus, the decompression chamber, and the thermal-cycling equipment has been completed, and detailed design was started.

In February, detailed design of the test apparatus will be completed, and construction will be started.

M. 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. Stiegelmeyer, and F. W. Fink

The corrosion of the ORNL Mark II fluorinator was described in BMI-1381 and BMI-1377. The vessel and components were constructed largely from "L" Nickel. Rather severe intergranular corrosion was observed, particularly at areas in contact with the liquid. Recent metallographic measurements of grain size have indicated that a slight amount of grain growth occurred in the lower portions of the vessel. It was concluded, however, that the accelerated attack in these areas was caused by greater impurity concentrations and slightly elevated temperatures.

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

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

Sulfex Process

No Sulfex dissolution experiments were conducted during the month. Most of the analytical results are now available on the four previous Sulfex tests of ORNL-irradiated Consolidated Edison fuel pins; these are contained in Tables M-1 and M-2.

Where shattering appeared to be minimal or absent, losses of uranium to the cladding solution immediately after decladding were approximately the same as those observed in similar runs on unirradiated pins, a few tenths of a per cent. Thorium losses, though also low, 0.1 per cent of less, were higher than in the cold tests, and only slightly lower than uranium losses. The data indicate that plutonium losses under these conditions were also in the same general range (0.1 to 0.3 per cent) and were fairly consistent. No significant dissolution selectivity is apparent from these data.

All losses increased with prolonged digestion of the dejacketed core pellets in the cladding solution. Where shattering was present, losses above 1 per cent were frequent.

Plutonium analyses of core solutions are not yet completed. Those available are in fair general agreement with the calculated values and with the burnup analyses. These latter indicate that burnups were in the 175 to 300-MWD/T core range, slightly lower than anticipated.

Darex Process

Darex dissolution tests were conducted during the month on the remaining two ORNL-irradiated Consolidated Edison pins (A-42 and A-115). Procedure for decladding

TABLE M-1. ANALYSES OF CLADDING SOLUTIONS FROM SULFEX DISSOLUTIONS OF IRRADIATED CONSOLIDATED EDISON FUEL PINS

			Cladding Solutions								
			Uranium		Thorium		Plutonium				
Run	Pin	Dissolution Time, hr	Concentration, g per liter	Loss[a], per cent	Concentration, g per liter	Loss[a], per cent	Concentration, mg per liter	Loss, per cent	Iron, g per liter	End-Cap Residue, g	Remarks
Sulfex S-1	A-103	3	0.089	0.77	1.26	0.51	0.14	0.53[b]	42.3		Severe shattering
		6	0.080	0.69	1.05	0.43	--	--	34.6		
		8	0.119	1.8	1.66	1.2	--	--	42.0	6.36	
Sulfex S-2	A-117	3	0.020	0.26	0.20	0.12	0.030	0.17[b]	25.4		Passivated; 8 M H_2SO_4 used
		6	0.082	1.6	0.73	0.70	--	--	18.1		Pin still partially clad
		3	0.073	1.4	0.65	0.57	0.13	1.1[b]	20.8	Nil	Fresh charge 8 M H_2SO_4
Sulfex S-3	A-108	3	0.0089	0.12	0.11	0.068	0.019	0.12[c]	49.8		Shattering not observed
		6	0.017	0.30	0.14	0.12	--	--	40.2	4.29	
Sulfex S-4	A-196	3[d]	0.0161	0.21	0.15	0.095	0.033	0.14[c]	65.0		Shattering not observed
		6	0.092	1.6	0.95	0.81	--	--	46.4	1.51	

(a) Based on estimated input of 1.75 g of uranium and 37.0 g of thorium.

(b) Based on estimated 4.0 mg of plutonium input.

(c) Based on total plutonium found in core solutions.

(d) Sample taken directly from dissolver; all other samples filtered and withdrawn from the product receiver.

TABLE M-2. ANALYSES OF CORE SOLUTIONS FROM SULFEX DISSOLUTIONS OF IRRADIATED CONSOLIDATED EDISON FUEL PINS

			Uranium		Thorium		Plutonium				Burnup	
Run	Solution	Volume, ml	Concentration, g per liter	Weight, g	Concentration, g per liter	Weight, g	Concentration, g per liter	Weight, g	Iron, g per liter	Sulfate, g per liter	Uranium, a/o	Core, MWD/T
Sulfex S-1[a]	First core	85	6.1	0.52	111	9.4	--	--	0.21	--	0.46	~195
	Second core	50	--	--	28.5	1.4	--	--	--	--	--	--
	Composite[b]	--	--	--	--	--	--	--	--	--	--	--
Sulfex S-2	First core	140	8.7	1.22	137	19.2	--	--	0.17	3.4	--	--
	First wash[c]	50	--	--	23	1.2	--	--	0.14	2.1	--	--
	Second core	150	2.5	0.37	54	8.1	--	--	--	5.9	0.47	~200
	Second wash	50	--	--	--	--	--	--	--	1.0	--	--
	Second composite	200	--	--	37	--	--	--	0.02	--	--	--
Sulfex S-3	First core	145	--	--	252	36.6	--	--	--	1.2	--	--
	Second core	50	--	--	37	1.9	--	--	--	1.2	--	--
	Composite	245	6.4	1.56	120	--	0.0149	0.0037	0.26	1.1	0.42	~175
Sulfex S-4	First core	145	--	--	230	33.2	--	--	--	1.6	--	--
	Second core	65	--	--	32	2.1	--	--	--	1.7	--	--
	Composite	250	6.5	1.62	123	--	0.0222	0.0056	0.11	1.5	0.70	~300

(a) Dissolver fractured at start of core dissolution; dissolution continued with 12.6 g of recovered core material.

(b) Sample lost.

(c) Washed to remove thorium sulfate coating from core pellets.

followed that recommended by the Chemical Technology Division of ORNL. Principal deviations from the procedure used in the Sulfex process are cold initial acid (500 ml of 5 M HNO_3-2 M HCl) and a shorter dissolution time (1 hr).

The dejacketed pellets were digested with the cladding solution for an additional 3 hr to investigate the effect of digestion time on losses of fuel and fertile materials to the cladding solution. In previous runs, interim cladding-solution samples were passed through the same glass-frit filter used for the main cladding and core solutions and withdrawn from the product receiver. Interim samples were therefore undiluted; the final sample was diluted with the washings. To eliminate any danger of contamination of cladding-solution samples by possible traces of core solution held up in the apparatus from previous experiments, the sampling procedure was modified, beginning with these tests. Cladding-solution samples are now being withdrawn directly from the dissolver with a pipette. All samples are therefore on an undiluted basis.

Analytical results for uranium and thorium losses to the Darex cladding solutions are shown in Table M-3.

TABLE M-3. URANIUM AND THORIUM LOSSES TO DAREX DECLADDING SOLUTION

		Dissolution	Uranium			Thorium		
Run	Pin	Time, hr	Concentration, g per liter	Weight, g	Loss[a], per cent	Concentration, g per liter	Weight, g	Loss[a], per cent
Darex D-5	A-42	1	0.089	0.037	2.1	1.0	0.42	1.1
		4	0.163	0.065	3.7	2.4	0.95	2.6
Darex D-6	A-115	1	0.0026	0.0011	0.063	0.028	0.012	0.032
		4	0.0065	0.0026	0.15	0.053	0.021	0.057

(a) Based on estimated input of 1.75 g of uranium and 37 g of thorium.

Although uranium and thorium losses to the cladding solution approximately doubled with an additional 3-hr contact, in both experiments, a more significant variation was the difference, by a factor of about 30, between corresponding samples from the two. This large variation appears to have been the result of some fragmentation of the core of Pin A-42, whereas Pin A-115 appeared to be free from fragmentation. Visually, most of the core pellets of Pin A-42 appeared to be intact; however, the small amount of fines filtered out of the final cladding solution (perhaps 1 to 2 g) appeared to be extremely fine, to an extent that filtration was markedly hampered. Dissolution of these fines was evidently appreciable.

It is not possible to compare Darex and Sulfex losses from these data. Uranium losses in Run D-5 were higher than in any of the previous four Sulfex tests; conversely, uranium losses in Run D-6 were lower than any heretofore observed. Thorium losses followed the same general pattern. Shattering, or absence of shattering, appears to be a more significant variable than the nature of the dissolvent.

CETR Pins

Several CETR pins, irradiated 2 cycles at the MTR(~4000 MWD/T), are scheduled to be subjected later to dissolution studies. In a previous report (BMI-1391) it had been noted that a cross-sectional sample of one of these pins, taken for a burnup analysis, had been found to be almost totally insoluble in core dissolvent (<1 per cent in ~20 hr).

Only approximately 1 per cent was dissolved after an additional 26-hr digestion in refluxing 13 M HNO_3 core dissolvent. However, further examination has revealed that this specimen is not ThO_2-UO_2, but Al_2O_3 (presumably from an end plug). These data indicate that if Al_2O_3 end plugs are used, a heel will accumulate in full-scale dissolvers which will have to be mechanically removed from time to time.

Dissolution studies of the MTR-irradiated CETR pins are scheduled to begin as soon as they are released from their present program. Some companion studies on similar "cold" pins may also be carried on.

N. 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 research with the VMR critical assembly continued during the past month. Most of the electrical and plumbing installation was completed.

The experimental program was reviewed, and preparations were made for special experiments. Depleted uranium foil (0.04 w/o uranium-235, 5 mils thick) was procured and the composition verified by an irradiation analysis. This fuel will later be used in measuring fast-fission effect and resonance escape probability. A "double-fission" chamber was constructed to perform calibrations required for fast-fission-effect measurements. This chamber will permit simultaneous irradiation of both a depleted and natural uranium foil to a known number of fissions. The foils will be used to calibrate Battelle's counting facilities in terms of activity per fission in uranium-235 and uranium-238.

It is expected that final components and uranium dioxide fuel pins will be received during the coming month. Initial attempts at criticality will follow.

O. URANIUM CARBIDE AS A REACTOR FUEL (AI)

F. A. Rough

Examination of uranium-5 w/o carbon specimens irradiated to about 10,000 MWD/T has shown that the UC_2 phase is destroyed in the central portions of the specimens. The resulting damage does not appear to be serious; however, these results suggest that a composition near stoichiometric (4.8 w/o carbon) is desirable.

Irradiation at the MTR of the final capsule of specimens in this program is due to be completed in June.

Irradiation of Uranium Monocarbide

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

Of a total of six capsules originally involved in this program for the irradiation of cylindrical specimens of UC, only one, BMI-23-4, is still being irradiated at the MTR. Capsule BMI-23-6, the next-to-last capsule in the series, was discharged during December and arrived at the Battelle Hot-Cell Facility during January for postirradiation inspections.

The irradiation of Capsule BMI-23-4 is continuing in the A-27-SE position of the MTR for a target burnup of 20,000 MWD/T of uranium. The capsule is scheduled for discharge at the end of MTR Cycle 140 (June 20, 1960) for a total irradiation of 26 MTR cycles. Specimen center-line temperatures as estimated from thermocouples still operating were in the 1050 to 1100 F range during January.

Postirradiation Examination of Irradiated Uranium Monocarbide

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

A program is currently being conducted to determine the changes induced in the properties of uranium monocarbide by irradiation in a test reactor. In order to study these changes, specimens for six irradiation capsules were prepared, encapsulated, and irradiated in the MTR.

During the past month, postirradiation microexamination of specimens from Capsules BMI-23-3 and BMI-23-5 having nominal compositions of 5.0 and 4.6 w/o carbon, respectively, was completed. Capsule BMI-23-3, containing specimens of uranium-5 w/o carbon, was inserted into the A-28-NE position at the MTR for a total irradiation of 12 cycles. Average mean core temperature was about 1150 F, and the average mean surface temperature was about 750 F. The capsule was irradiated for a total of 7288 megawatt-days with burnups based on dosimetry analyses of approximately 10,000 and 10,500 MWD/T of uranium for the top and bottom specimens, respectively.

O-2

Capsule BMI-23-5, containing specimens of uranium-4.6 w/o carbon, was inserted into the MTR for a total irradiation of six cycles, with average mean core and surface temperatures of 1325 and 880 F. The capsule was irradiated for a total of 3583 megawatt-days with burnups based on dosimetry analyses of 5300 and 5250 MWD/T of uranium for the top and bottom specimens, respectively. Examination of the photomicrographs of specimens from both capsules indicated there was a larger amount of general cracking in the specimens from Capsule BMI-23-3, possibly due to the longer irradiation of these specimens. Further examination revealed a general depletion of the Widmanstätten pattern of UC_2 in the structure going from the outside toward the center of the specimens. In the center of the specimens no UC_2 could be detected. It appears that neutron irradiation or a temperature gradient within the specimens caused this depletion. The possibility exists that the irradiation could cause the UC_2 to go into solution in the UC matrix, or that the UC_2 migrated out of the specimen because of the thermal gradient during irradiation. It is interesting that this phenomenon was observed in both the top and bottom specimens in this capsule and also, as previously reported, in the top specimen of Capsule BMI-23-2 which was irradiated to 4600 MWD/T of uranium.

Examination of the specimens taken from Capsule BMI-23-5 revealed no evidences of changes in structure in both the as-polished and as-etched conditions. Both the etched and as-polished photomicrographs show the UC matrix with the excess uranium clearly outlined in the UC grain boundaries.

Capsule BMI-23-6, containing specimens with a nominal composition of uranium-4.8 w/o carbon, arrived at the BMI Hot-Cell Facility after being irradiated at the MTR for six cycles. At this time, the capsule is being prepared for postirradiation examinations and measurements.

P. PEBBLE-BED REACTOR MATERIALS (S & P)

W. S. Diethorn and W. H. Goldthwaite

Research in support of the development and evaluation of spherical fueled-graphite fuel elements for the Pebble-Bed Reactor program is in progress at Battelle. Candidate fuel elements, 1-1/2 in. in diameter, prepared by several vendors and varying in composition and method of manufacture, are being evaluated in laboratory tests and irradiations designed to obtain information on their structural integrity and fission-product retention.

LABORATORY EVALUATION OF FUELED-GRAPHITE SPHERES

M. C. Brockway and A. K. Smalley

The resistance to impact and compression, coating integrity, self-welding, and coating permeability are being studied with coated spheres supplied by commercial vendors.

During the month of January the rate of effort in this phase of the program was low. No runs were scheduled in the permeability study. The self-welding test apparatus was modified to reduce oxygen contamination in the helium purge gas used in this high-temperature study.

FABRICATION DEVELOPMENT OF Al_2O_3-CLAD UO_2 FUEL PARTICLES

Sintered Cladding

A. K. Smalley and W. H. Duckworth

In a continuation of the work reported last month (BMI-1403), clad particles prepared from mixtures of UO_3 and UO_2 were evaluated.

Batches containing 55 to 75 w/o of UO_3 were studied. Clad particles in which the initial fuel composition was 65 w/o or more of UO_3 showed relatively low weight gains in the oxidation test. Less than 5 w/o of the UO_2 in the sintered particles was oxidized when samples were heated at 1200 F in air. The better batches of clad particles contained 25 w/o UO_2. Above a UO_2 content of 25 w/o the oxidation resistance fell off rapidly.

Clad particles having an OD of 1000 μ are being incorporated into a graphite sphere in a study of fabrication problems. Following completion of this latter work, the program on sintered claddings will be terminated.

Carburization Studies

A. F. Gerds

Sintered Al_2O_3-clad UO_2 particles, dispersed in two different carbon matrices, were heated at 2500 F for 1000 hr in helium to determine the compatibility of these combinations of materials. Metallographic examination showed varying degrees of reaction, depending on both the grade of carbon and the binder.

Specimens composed of Al_2O_3-coated UO_2 particles dispersed in AGOT graphite, with Barrett pitch as binder, proved to be the most compatible combination tested. The Al_2O_3-coated particles retained their integrity, but a slight reaction with the graphite was observed, producing voids in the Al_2O_3-graphite interface.

With AGOT graphite the BV1600 resin binder, a severe reaction was observed. The Al_2O_3 cladding disappeared, leaving a void around the UO_2 seed. A small amount of Al_2O_3 was found at the void surface. The UO_2 seed retained its characteristic color and appearance under the microscope.

A severe reaction was also observed with Texas 55 coke and Barrett pitch binder. A void surrounded the seed, and only a small residue of Al_2O_3 was found adjacent to the coke. The UO_2 seed was transformed to a dense metallike material which appeared to contain three phases. The identity of these three phases is unknown.

Vapor-Deposited Cladding

T. E. Cook and J. M. Blocher

Particles of clad UO_2 are being prepared by vapor deposition of Al_2O_3 in a new program initiated this month. Samples of MCW UO_2 shot between 100 and 140 mesh of normal enrichment were coated by the hydrolysis of Al_2Cl_6 vapor in a fluidized bed of the shot.

Two batches containing 20- and 40-μ coatings have been prepared. Samples from both batches showed very good oxidation resistance on heating in air at 1200 F for 5 hr. During the first hour of heating a small weight loss was observed, but no further weight change occurred during the remaining 4 hr. The reason for the weight loss is not known. One possibility is the loss of absorbed moisture.

A sample of the 20-μ batch will be incorporated into several graphite spheres for further evaluation.

P-3

FISSION-PRODUCT RELEASE FROM FUELED-GRAPHITE SPHERES

W. S. Diethorn

Fueled-graphite spheres are being investigated in neutron-activation experiments, and sweep-gas and static irradiation capsules.

Neutron-Activation Studies

H. S. Rosenberg

Two fueled-graphite spheres coated with 2- and 5-mil dense carbon coatings were neutron activated this month. Results are reported in Table P-1.

TABLE P-1. NEUTRON-ACTIVATION RESULTS

Specimen	Postirradiation Heat-Treatment Temperature, F	Time at Temperature, min	Fission-Gas Release, per cent of xenon-133
FA-20, No. 429 (2 mils)	1700	70	$<4 \times 10^{-3}$(a)
	2000	100	$<4 \times 10^{-3}$(a)
FA-21, No. 2 (50 mils)	1650	75	$<4 \times 10^{-3}$(a)
	2300	80	4×10^{-3}

(a) No detectable release.

In-Pile Capsule Experiments

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

SP-4. This capsule continues to operate satisfactorily.

SP-5. Sweep Capsule SP-5 is being assembled for BRR irradiation in mid-March.

SPF-1. The SPF-1 capsule experiment was continued on January 5. The capsule was operated at 1900 F and 150 F. This latter temperature was achieved by simply turning off the capsule heater. A total of four gas samples were collected at these two temperatures. The data are summarized in Table P-2.

TABLE P-2. SUMMARY OF SPF-1 DATA

Gas Sample	Collection Date, 1960	Flux, n/(cm^2)(sec)	Temperature, F	R/B[(a)]				
				Krypton-87	Krypton-88	Krypton-85m	Xenon-135	Xenon-133
13	1-5	1.7×10^{10}	1900	9.2×10^{-3}	3.7×10^{-3}	7.6×10^{-2}	7.4×10^{-2}	6×10^{-2}
14	1-7	1.7×10^{10}	1900	11×10^{-3}	5.6×10^{-3}	2.6×10^{-2}	2×10^{-2}	8.7×10^{-2}
15	1-7	1.7×10^{10}	150	0.38×10^{-3}	Nil	0.15×10^{-2}	5.4×10^{-3}	1.9×10^{-2}
16	1-8	1.7×10^{10}	150	0.16×10^{-3}	Nil	0.038×10^{-2}	1.5×10^{-3}	5×10^{-3}

(a) R = the number of atoms released per second from the sphere.
B = the number of atoms produced per second in the sphere.

Sample 13 was collected about 2 hr after capsule startup. Sample 15 was collected 1.5 hr after the heater was turned off. Krypton-88 was below the limit of detection in Samples 15 and 16.

All data from this capsule are currently being analyzed in an attempt to develop a fission-gas release model. A topical report covering this experiment is planned.

SPF-2. This furnace capsule has been delayed in the final stage of assembly. It is expected that this capsule will be inserted in the BRR during February.

Q. TANTALUM AND TANTALUM ALLOYS FOR LAMPRE APPLICATIONS (LASL)

J. H. Stang

Studies involving tantalum and tantalum alloys are being conducted in assistance to the LAMPRE program at Los Alamos Scientific Laboratory. During January data of particular interest were obtained on the mechanical properties of irradiated tantalum. The findings indicate that the irradiation exposures received produced a marked increase in the strength of tantalum and some loss of ductility. The specimens involved were irradiated at the MTR in a position reportedly having high flux but relatively low fast flux. By doing this, it was expected that effects from the formation of vacancy-interstitial combinations associated with atomic displacements would be kept to a minimum and that the principal modification of the material would be the conversion of tantalum to tungsten.

Development of Container Materials for LAMPRE Applications

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

The difficulty encountered in December with the erosion of the tungsten electrode during the arc melting of binary tantalum alloys has been overcome. Sixteen button specimens, weighing 200 g each, with the following additions have been prepared by arc melting under a partial pressure of helium:

0.5, 1.5, 3, and 6 w/o thorium
1.0 and 3.0 w/o iron
1.0 and 3 w/o silicon
0.5, 1.5, and 3 w/o hafnium
0.5, 1.5, and 3.0 w/o zirconium.

Radiographic inspection has shown that, with the exception of the alloys containing from 1.5 to 6 w/o thorium, the alloys are basically sound. In the case of the three thorium-containing alloys, there are many small voids present which were not eliminated by remelting.

The button specimens are being processed to 0.030-in. strip for plutonium-alloy corrosion testing at LASL.

Effect of Irradiation Damage of Tantalum

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

During January, mechanical-property tests were conducted at the Battelle Hot-Cell Facility on samples of tantalum which had been encapsulated and irradiated at the MTR at a fairly low temperature. One capsule was irradiated for 4 MTR cycles while the other was irradiated for 7 MTR cycles. The exposures were selected to produce a conversion of tantalum to tungsten of 1-1/2 and 3 w/o, respectively.

Table Q-1 presents tensile-property data now on hand for the irradiated specimens and for a specimen of unirradiated annealed tantalum. Comparison of the data indicates a substantial increase in the strength of the material as a result of irradiation. Corresponding increases are also noticed in the 0. 2 per cent offset yield strength. While the irradiated tantalum showed increases in strength, some loss in ductility of about 40 per cent was experienced in the annealed, unirradiated tantalum, and after irradiation to 1.57×10^{21} nvt the elongation decreased to about 7 per cent.

In an attempt to evaluate the ductility further, both the unirradiated and irradiated tantalum specimens were tested by simple bend tests. All specimens were bent 90 deg and examined. No evidence of cracks in or near the bending zone was found. Final results of these tests indicate that irradiation effectively increases the strength of the material without producing a prohibitive loss in ductility.

It is planned to conduct hardness determinations on both the irradiated and unirradiated specimens and chemical analyses on the irradiated specimens to determine the tungsten conversion.

Precipitate Phase Identification and Interstitial-Type Solid Solubility in Tantalum

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

The identification of precipitate phases in tantalum known to be detrimental to its corrosion resistance is continuing. The oxygen-charged samples have been prepared, annealed at 1500 C, and are being analyzed by X-ray diffraction and metallography. No results are available at this time.

If good correlation is obtained with the published data*, the oxygen studies will be greatly simplified. It should be pointed out that at 1070 C the published work reports 9 a/o in solid solution while one sample annealed at about 2000 C contained approximately 25 a/o in solid solution. This work also indicated that the only oxides formed were Ta_2O_5 and Ta_2O.

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

TABLE Q-1. ROOM-TEMPERATURE TENSILE PROPERTIES OF UNIRRADIATED AND IRRADIATED TANTALUM

Tantalum Specimen	Total Integrated Flux Based on Dosimetry Analysis, nvt	Intended Composition, w/o	Ultimate Tensile Strength, psi	0.2 Per Cent Offset Yield Strength, psi	Elongation at Maximum Load, per cent	Elongation in 1 In., per cent	Reduction in Area, per cent
Unirradiated	--	Unalloyed Ta	42,000	30,000	--	40	68
Irradiated	7.8×10^{20}	Ta + 1.5 W	71,010	70,000	0.23	12	68
	7.8×10^{20}	Ta + 1.5 W	65,878	62,900	1.10	21	72
	7.8×10^{20}	Ta + 1.5 W	71,333	66,400	0.61	10	61
	7.8×10^{20}	Ta + 1.5 W	70,666	63,900	0.49	11	60
	1.57×10^{21}	Ta + 3.0 W	87,837	84,932	0.44	6	59
	1.57×10^{21}	Ta + 3.0 W	87,012	81,818	0.90	7	62
	1.57×10^{21}	Ta + 3.0 W	83,098	80,563	0.11	7	77
	1.57×10^{21}	Ta + 3.0 W	87,328	78,000	0.41	7	58

R. DEVELOPMENTAL STUDIES FOR THE PWR (BAPD)

R. W. Dayton

Studies of the preparation of large fuel plates containing UO2 cores have been continued. The properties of the bonded plates appear satisfactory, although the metallographic appearance of the bonds indicates the need for further study.

Fabrication of Large-Scale PWR-Type Fuel Plates

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

The feasibility of preparing small-scale Zircaloy-clad uranium dioxide PWR Core 2-type compartmented fuel plates approximately 15 in. long and 15 core compartments wide by gas-pressure bonding has been established. Process-development studies for the preparation of large-scale specimens of this type on a production basis are now being conducted. During the present process studies, the length of the specimen was increased from 15 to 33 to 40 in. while the width was held constant.

The burst, leak, and corrosion tests as well as the metallographic examination of the 40-in. -long specimens previously bonded have been completed. The results of the burst tests revealed that there was no intercompartmental leakage and that in all cases the cladding ruptured in a ductile fashion over the core area at an average burst pressure of 3300 psi. The results of the corrosion tests revealed that after a 1-week exposure to 680 F water there was no growth in any of the defected compartments. After this corrosion test, the specimens were again tested at 700 psi for intercompartmental leakage. No intercompartmental failure was observed. The Zircaloy-2 cladding also possessed the shiny black film associated with good-quality Zircaloy-2. The metallographic results indicated that while the Zircaloy-to-Zircaloy bonds were generally good, there was, occasionally, evidence of some contamination or voids at the bond interface which may have been caused by residual helium which was introduced during the welding and not removed by the evacuation or by small particles of contamination introduced during belt abrading. Some oxygen contamination of the cladding was also observed as a result of core-to-cladding reaction at the corners of a few cores. Because of the random nature of this reaction product, it is believed that it was caused by a nonuniform crystalline carbide coating on a few of the uranium dioxide cores.

S-1

S. 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

Ceramic-type fuels 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 are being considered as potential fuels for the MGCR. A program to study the stability of these materials after various radiation exposures at specimen-surface temperatures of about 1500 F is in progress. The evaluation of beryllium oxide and graphite-matrix specimens includes the irradiation, examination, and evaluation of 24 specimens contained in four instrumented capsules. Each of the four capsules is equipped with electrical heaters and six thermocouples. Each capsule contains six specimens, two of each fuel composition. Each specimen is composed of four pellets, approximately 0.222 in. in diameter by 0.250 in. long, sealed in Type 316 stainless steel tubing under a protective helium atmosphere.

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

The three additional capsules have been in the MTR since the beginning of Cycle 125 and have just completed their ninth irradiation cycle, Cycle 133. During Cycle 133, two of the electrical heaters did not operate in Capsule BMI-31-1, resulting in lower over-all temperatures for this capsule. It is believed that the trouble encountered was external to the capsule itself. An examination of the power-supply controls and lead wires will be made during the shutdown period between Cycles 133 and 134. The remaining two capsules are operating satisfactorily.

A summary of the temperature and heater-power requirements of each capsule during MTR Cycle 133 is shown in Table S-1. Except for the heater difficulty mentioned previously, all three capsules are operating routinely. Capsules BMI-31-2 and BMI-31-3 each have four operating thermocouples and three heaters. Capsule BMI-31-1 at present has three operating thermocouples and only one heater; however, the two other heaters are expected to be made operational during the shutdown prior to Cycle 134.

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

TABLE S-1. TEMPERATURE AND ELECTRICAL-HEATER POWER CONSUMPTION FOR CAPSULES BMI-31-1, BMI-31-2, AND BMI-31-3 DURING MTR CYCLE 133

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	1000	--	--	1420[b]	1390	1215	--
BMI-31-2	2395	1415	--	1450[b]	--	1500	1475
BMI-31-3	2800	1410	1500[b]	--	1500	--	1460

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

(b) Controlling thermocouple.

A single capsule containing Al_2O_3-UO_2 samples has been removed from the BRR core after being irradiated for six reactor cycles. The activity of the gas in the thermocouple-lead tube was approximately 10 microcuries per ml; analysis showed that this activity was caused principally by the presence of xenon-133, indicating a leak of fission gas from the inner capsule to the outer shell and lead tube. It was necessary to remove this fission gas from the lead tube by pumping and cold trapping before the lead tube could be severed from the capsule. It is expected that the capsule will be shipped to General Atomic for hot-cell examination during the first week in February.

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.

S-3 and S-4

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 was started during January to determine the feasibility of preparing uranium and thorium carbide powders and subsequently to coat these powders with carbon, zirconium carbide, or niobium carbide films by vapor-deposition techniques. Parallel efforts on both preparation and coating are being carried out.

Thermodynamic calculations were performed to provide information on the feasibility of preparing uranium carbide by various reactions involving uranium halides, hydrocarbons, and reductants. There appear to be a number of reactions which look promising, and several of these reactions will be investigated. No attempt to prepare thorium carbide will be undertaken until the feasibility of preparing uranium carbide has been demonstrated experimentally.

T. DEVELOPMENTAL STUDIES FOR THE SM-2 (ALCO)

S. J. Paprocki

The work discussed 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.

The current studies involve the fabrication of full-scale fuel plates and the evaluation of the core homogeneity of these plates by radiography, metallography, and analyses for uranium and boron. Segregation of UO_2 was observed in the initial full-scale fuel plates. A study of blending and compacting procedures revealed that homogeneous dispersions can be produced by control of the blending process. By the proper control of the sintering and rolling cycle, the loss of boron has been limited to less than 10 per cent.

At the present time, reference and alternate fuel specimens are being irradiated in three MTR capsules and one ETR capsule. A second capsule, fully instrumented with thermocouples and auxiliary heaters, has been shipped to the ETR.

Materials Development

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

Reference materials have been selected and fabrication techniques are being evaluated for use in preparing fuel plates and absorber plates for the SM-2 reactor.

Fuel Materials

Fuel plates for the SM-2 reactor will contain a composite core with a 22-in.-long fuel section and a 1/2-in.-long suppressor section. The fuel core contains 26 w/o UO_2 and 1.2 w/o ZrB_2 dispersed in a Type 347 stainless matrix. The suppressor contains 17 w/o Eu_2O_3 dispersed in an elemental 18-9 stainless matrix. This 0.030-in.-thick core is clad with 0.005-in.-thick Type 347 stainless steel. Full-size fuel plates containing fully enriched UO_2 have been prepared for critical-assembly tests at Alco. Other fully enriched plates will be prepared as materials become available for testing in the SM-1 core. Both full-size and small-scale specimens containing natural and depleted UO_2 are being prepared to evaluate fabrication techniques.

Uranium analyses have been made on two full-size fully enriched fuel plates, and the results are shown in Table T-1. These analyses show a variation greater than the 2 per cent allowed on SM-1 plates, but this variation is not sufficient to warrant any major change in the fabrication procedure. However, one or two plates in this first series of full-size plates were segregated to the extent that uranium-rich and uranium-poor areas could be detected on radiographs. These plates were subjected to a thorough metallographic examination and chemical analysis. The results of chemical analyses on one of the plates is shown in Table T-2. In this case, a variation of 11.5 per cent occurred. The samples used were only one-third of the size used in the standard tests, and, consequently, the variation is greater than would be expected in the standard test, but the variation is still excessive.

TABLE T-1. URANIUM ANALYSES ON SECTIONS OF FULL-SIZE PLATES

Plate	Section[a]	Uranium Analysis, w/o
EAR-2	F	15.5
	G	15.3
	H	15.3
	I	16.1
	J	16.3
EAR-21	F	15.5
	G	16.2
	H	16.1
	I	16.3
	J	15.2

(a) 1 by 1-in. sections cut on a diagonal traverse across the core.

TABLE T-2. URANIUM ANALYSES ON A SEGREGATED PLATE

Plate	Section	Appearance	Uranium Analysis, w/o
AR-43	A	Normal	16.0
	B	Dark	14.2
	C	Normal	17.1
	D	Dark	15.5
	E	Dark	15.2
	F	Normal	16.1

The results listed above led to an investigation of the blending and compacting procedures, and, subsequently, the conclusion was reached that excellent dispersions of spherical UO_2 can be produced with several different binders and that there are no blending difficulties sufficient to preclude the use of spherical UO_2 powder in the SM-2 fuel plates. In general, it appears that blending difficulties are caused by insufficient amounts of a binder, poor methods of adding the binder, and poor handling and die-loading techniques. For the binders investigated (lauryl alcohol, Ceremul "C", and camphor-methanol) approximately 1/2 w/o binder appeared to be a satisfactory quantity. A greater amount of lauryl alcohol was required, since this binder is more viscous and, consequently, more difficult to disperse throughout the blend. If the binder is added to the blend, it should be added after the powders have been mixed dry, and it should be fluid enough to be quickly dispersed throughout the blend without causing balls of agglomerates to form.

A theoretical approach to the problem would demand that the heavier particles (UO_2) be coated with an organic substance with adhesive properties. It is interesting to observe that when UO_2 is dipped in camphor-methanol and evaporated to dryness, leaving a thin coat of camphor, excellent uniformity can be achieved in UO_2-stainless dispersions. If the powders as blended are slightly moist, less care is required in handling and loading the powders in the die. However, tapping and excessive leveling of the powders should be avoided.

The major problem area in the fabrication of SM-2 plates has been boron retention. Sintering in vacuo essentially eliminates boron loss during sintering, but a 5 to 10 per cent loss can occur during roll cladding. Under these conditions, the boron content can be maintained within 5 per cent of the specified loading, which is acceptable from a physics standpoint, but work is being continued in an effort to reduce the loss to a lower level. Boron losses at various stages in the process have been measured and are listed in Table T-3. Calculated losses based upon the diffusion rate of boron being the controlling factor are also listed. From these data, it is evident that the diffusion of boron is very rapid and that the rate-controlling process is either the rate of boron removal at the surface or the rate of dissolution of the ZrB_2. Because of the fact that ZrB_2 particles show only a slight reaction and the fact that the boron content of the cladding is very low (0.002 w/o boron), it is postulated that the rate-controlling factor is the dissolution of ZrB_2. Additional tests are in progress to prove or disprove this hypothesis. It will be noted that the boron loss is greater than that predicted for the SM-2 plates. In this case, an annealing time of 15 min rather than the normal 5 min was used, and a stainless powder containing three times the normal amount of oxygen was used. This was accomplished to intentionally accentuate boron losses in these tests. Had the normal 5-min anneal been used, it can be calculated that boron losses would have been approximately 10 per cent.

TABLE T-3. BORON LOSS IN VARIOUS STAGES DURING ROLL CLADDING

Specimen	Condition	Boron Loss, per cent Measured	Boron Loss, per cent Calculated
27G	Green	0.00	--
27S	Sintered at 2150 F for 2 hr	1.61	--[(a)]
27	Hot rolled four passes at 2200 F	5.01	5.90
29	Hot rolled seven passes at 2200 F	8.60	12.90
31	Hot rolled seven passes and annealed 15 min at 2200 F, pickled, and cold rolled 20 per cent	18.60	27.30
33	Same as 31 and annealed 1 hr at 2050 F	19.60	74.00

(a) Since sintering losses are controllable, losses at this stage are not considered in subsequent calculations.

As shown in Table T-4, boron losses also appear to be related to the type of stainless steel matrix. It should be noted that silicon decreases as boron losses increase. Nickel would be expected to have a detrimental effect, since ZrB_2 as well as the other borides investigated react badly with pure nickel at temperatures used in fuel-plate fabrication. However, the loss shows direct correlation only with silicon content. As the silicon decreases, oxygen increases, and oxygen is known to promote boron losses. Additional chemical analyses and tests will be required to ascertain the major cause of different boron losses in these specimens.

The effect of oxygen on ZrB_2 and boron losses has been demonstrated by oxidizing ZrB_2 in air for 10 min at 1000 F and roll cladding specimens containing the oxidized ZrB_2. Oxidation of the ZrB_2 in air caused a change in composition from 19. 2 to 18. 2 w/o boron. Upon sintering and roll cladding, a 33. 4 w/o boron loss occurred compared with a 9. 0 w/o boron loss for a similar specimen containing standard ZrB_2.

TABLE T-4. EFFECT OF MATRIX COMPOSITION ON BORON LOSS

Matrix Composition (Balance Nominally Iron), w/o				
Chromium	Nickel	Silicon	Carbon	Boron Loss, per cent
16. 62	12. 18	1. 29	0. 03	9. 04
18. 96	10. 76	1. 21	0. 05	10. 65
15. 95	13. 51	1. 15	0. 03	13. 70

The use of metal-coated ZrB_2 particles is also being investigated as a means of reducing boron loss. Zirconium diboride powders with coatings of 2 to 3 μ, 4 to 5 μ, and 7 to 8 μ of niobium have been prepared by the fluidized-bed process and have been used to fabricate test specimens. Boron analyses have been received only for the specimen containing ZrB_2 with a 2 to 3-μ coating, and, although the coating was beneficial, a 3. 47 per cent boron loss occurred. It is believed that thicker coatings will reduce the loss to a lower level. Additional analyses are being made on specimens containing ZrB_2 with heavier niobium coatings. Also, an attempt is being made to coat ZrB_2 with chromium and tungsten.

Development of Control and Suppressor Materials

Presently, the most promising control and suppressor material for the SM-2 is a dispersion of Eu_2O_3 in an elemental-powder 18-9 stainless steel. An evaluation of corrosion screening tests has indicated that the air-fired monoclinic Eu_2O_3 available in the desired particle size from Heavy Minerals is a more desirable material than the cubic Eu_2O_3 purchased as a fine powder and fired to obtain a monoclinic crystal structure in a larger particle size. A 14-day corrosion test at 600 F in degassed water has resulted in no weight gain or dimensional change for the specimens containing the Heavy Minerals monoclinic Eu_2O_3 in an elemental-powder 18-9 stainless steel matrix. Similar specimens containing Eu_2O_3 which had been fired in hydrogen and crushed to the desired particle size showed a weight gain and an increase in thickness over the defect of approximately 30 per cent for specimens containing 0. 01 w/o silicon and 20 per cent for specimens having a 10 w/o silicon content in the Eu_2O_3.

Additional corrosion screening tests are being conducted to evaluate a prealloyed Type 347 stainless steel matrix containing 1. 2 w/o silicon and, also, vacuum sintering techniques on both elemental powder and prealloyed matrices. After a 24-hr test period at 600 F in degassed water, both types of vacuum-sintered specimens have increased approximately 3 per cent in thickness, whereas the hydrogen-sintered specimen containing an elemental matrix has remained unchanged. The specimens have been returned to the autoclave for the completion of a 14-day test.

A study has been initiated to determine the optimum fabrication techniques for the control plates. Rolling reductions of 10, 8, 6, and 4 to 1 in thickness will be made and evaluated. A study will also be conducted to determine the feasibility of making the core in one unit rather than stacking the core in three sections separated by a foil of the matrix composition as done in the PM-2A.

Full-size plates will be made after the fabrication techniques have been established.

Encapsulation Studies

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

The irradiation stability of reference SM-2 fuel specimens and alternate fuel materials is being investigated in capsule irradiations. Three noninstrumented capsules (BMI-32-1, BMI-32-2, and BMI-32-3) are being irradiated in core positions of the MTR. The first of seven lead capsules, BMI-32-4, is being irradiated in the ETR.

Preliminary results of a heat-transfer and temperature-distribution analysis performed at the IBM-704 installation at NYU tend to confirm the original design calculations. Specimen-surface temperatures computed by the two methods for a specific set of practical conditions are within 15 F. The IBM-704 analysis shows that the maximum temperature difference between specimen surfaces and adjacent thermocouples is approximately 270 F. This indicates that the specimens in BMI-32-4, wherein maximum thermocouple readings of approximately 400 F have been observed, are being irradiated within the desired temperature limits.

Analysis of the nuclear mock-ups which were run at the MTR and ETR to determine neutron-flux perturbation factors has yielded the following information to date: (1) the peak unperturbed flux for the ETR capsule, BMI-32-4, is probably about 12 per cent lower than quoted, (2) peak unperturbed fluxes for the MTR capsules appear to be as much as 10 per cent higher than quoted, (3) accrued specimen burnups for each of the four capsules as computed from mock-up data agree within acceptable limits with values computed from reactor-quoted flux data.

The second instrumented capsule, BMI-32-5, was sent to the ETR on January 28. Preshipment checkouts indicated satisfactory operation of all heaters and thermocouples. Capsule BMI-32-6 is approximately 50 per cent assembled, and the remaining four lead capsules now planned are about 10 per cent assembled.

U. 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

In addition to the BeO-UO_2 fabrication studies, the irradiation-effects program, and the critical-assembly experiments being conducted in the materials development program, a study concerned with the modification of Hastelloy X has recently been initiated. Through small alloy additions, it is hoped that improved corrosion and mechanical properties will be obtained after extended exposure (10,000 hr) to a nitrogen-1/2 volume per cent oxygen atmosphere at 1750 F.

Fabrication of BeO-UO_2 Fuel Pellets

H. D. Sheets and A. K. Smalley

Pellets of BeO-25 volume per cent UO_2 are being prepared for loop and capsule exposures.

Previously, 24 pellets about 1/16 in. in diameter and 3/32 in. high were prepared for capsule irradiation in the BRR. Bulk density of the sintered pellets ranged from 5.06 to 5.11 g per cm^3 (about 98 per cent of theoretical density). Chemical analyses of six pellets from the same lot indicated that the UO_2 content ranged from 59.3 to 59.9 w/o (27.5 to 27.8 volume per cent) UO_2 and that the oxygen-to-uranium ratio of the UO_2 was about 2.05. The oxygen-to-uranium ratio calculated from lattice-parameter measurements was 2.03. Spectrochemical analysis of two specimens showed the following metallic impurities:

Impurity	Amount Present, ppm
Magnesium	700
Silicon	400
Iron	100
Copper	60
Titanium	300
Nickel	200
Calcium	100

Impurity	Amount Present, ppm
Aluminum	400
Boron	3
Manganese	10
Chromium	60
Molybdenum	10
Zinc	5-10
Cobalt	5

Metallographic examination of a sintered pellet showed that the BeO was fine grained (under 40 μ) and was the continous phase. The UO_2 particles were 40 μ and less in diameter and were multicrystalline. Most of the voids occurred at and around the UO_2 particles, although there were some relatively large (40 to 60 μ) voids in the BeO. No evidence of cracks or laminations was found.

Fabrication of the 2500 specimens needed for loop exposures now is under way. Fabrication conditions are the same as for the capsule specimens. To date, about 2000 pellets have been prepared. This work will be continued.

Encapsulation Studies

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

Irradiation of Specimens Containing UO_2 Dispersed in BeO

The irradiation capsule containing six Hastelloy X-clad specimens of UO_2 dispersed in BeO was inserted in Position 45 of the BRR core for the cycle which started on January 13. Without auxiliary electrical heat, the specimen-surface temperatures as monitored by adjacent thermocouples were in the 1300 to 1400 F range at full reactor power. However, the addition of approximately 1100 w of electrical heat to the system resulted in thermocouple temperatures ranging from 1564 to 1685 F; the latter temperature is estimated to indicate a specimen-surface temperature of 1750 F, which is the capsule design point.

Almost two irradiation cycles have been completed with the temperatures fairly steady and the heater performance satisfactory. While the over-all experiment appears to be progressing well from the thermal viewpoint, the fact that the temperatures are low without auxiliary heat is puzzling. The capsule design seems fairly straightforward with regard to heat transfer, and the capsule geometry was selected on the basis of flux data obtained from well-dosimetered nuclear mock-up runs. The net situation does not make it possible to predict burnup rates with the degree of certainty that was anticipated. One interpretation of the available information indicates that a total burnup of approximately 0.7 per cent of the uranium-235 is being accumulated per reactor cycle; it is estimated that this can be subject to an error of about ±20 per cent.

Irradiation of Specimens Containing MCW Spherical UO_2 Dispersed in Stainless Steel

This program has the objective of comparing the irradiation stability of MCW spherical and ORNL hydrothermal UO_2 (30 w/o highly enriched) dispersed in stainless steel. Two stainless steel-clad specimens of each type are being irradiated in Capsules BMI-33-1 and BMI-33-2 now in the MTR.

Capsule BMI-33-1 was inserted in Position A-40-SW for Cycle 131, which started on November 25, 1959. This capsule is scheduled to be discharged on March 28, 1960, following a six-cycle exposure. Recorded thermocouple temperatures of 1200 F maximum indicate that the specimen center-line temperatures are at approximately 1350 F maximum. Fission burnup levels of approximately 15 per cent should be achieved during the six cycles.

Capsule BMI-33-2, identical in design to BMI-33-1, was inserted in Position A-6-NE for Cycle 133, which started on January 8, 1960. The quoted thermal-neutron flux in this position is significantly higher than that for A-40-SW, and thus, in this capsule, specimen center-line temperatures quite close to the desired specimen center-line desired, 1650 F, should have been achieved. Recorded temperatures during the first cycle, however, indicate that the specimens were at approximately the same temperatures as those in BMI-33-1. Therefore, a request to move BMI-33-2 to a higher flux position was submitted, and the desired repositioning was carried out during Cycle 134 shutdown (January 25 to 29). The new position, A-6-NW, has a quoted neutron flux approximately 25 per cent higher than that of A-6-NE.

Effects of Irradiation

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

A study of the radiation stability of fuel-element materials for compact gas-cooled reactors includes: (1) the evaluation of encapsulated solid and annularly loaded UO_2 specimens clad with Inconel, and (2) the evaluation of encapsulated specimens of UO_2 dispersed in graphite and clad with Inconel 702, Hastalloy X, or Carpenter 20 Cb, and (3) the evaluation of in-pile-loop subassemblies containing PWR-type fuel pins of solid UO_2 clad with Inconel.

Capsule Program

The burnup of four sections taken from three different specimens contained in Capsule BMI-27-2 has been determined by isotopic analysis. These sections were removed as follows: one section was removed from the top end of Specimen GE-solid 9, which was the top specimen in the capsule; one section each was removed from the top end and from the center of the fueled section of Specimen MC-solid 7, which was the third specimen from the top of the capsule; and one section was removed from the center of Specimen GE-solid 11, which was the fourth specimen from the top of the capsule. The results are reported in Table U-1. It should be noted that, on the basis of the data on Specimen 7, there was very little difference between the burnup of the end and center sections of the fueled pins. This indicates that there was very little end effect and that temperature gradients along the specimen were therefore probably low.

TABLE U-1. BURNUP RESULTS FROM SPECIMENS IRRADIATED IN CAPSULE BMI-27-2

Specimen	Location of Analyzed Section	Uranium Burnup Determined by Mass Spectrography[(a)], a/o
GE-solid 9	Top pellet	0.9
MC-solid 7	Top pellet	1.7
MC-solid 7	Center pellet	1.6
GE-solid 11	Center pellet	3.9

(a) Burnup determined from uranium-235 ratio.

Fission-gas-release data revised on the basis of burnups obtained from isotopic analyses indicate that approximately 6.4 per cent (6.7×10^{-2} ml) of the theoretical volume of gaseous fission products formed during irradiation was released from Specimen GE-solid 11 and that essentially no gaseous fission products were released from the fuel contained in Specimen MC-solid 7.

These results complete the evaluation of specimens contained in Capsules BMI-27-1 and BMI-27-2.

The examination of specimens irradiated in Capsule BMI-29-1 has been terminated. This capsule contained six specimens of 8 w/o UO_2 dispersed in a graphite matrix, each approximately 1.0 in. long by 0.260 in. in diameter, and clad with 0.025-in.-thick Hastalloy X, Carpenter 20 Cb or Inconel 702 tubing. The capsule was irradiated in the MTR to a burnup calculated from neutron dosimetry data of about 8.6 uranium a/o at specimen-surface temperatures of between 1600 and 1775 F. All specimens appeared in good condition, and no density or dimensional changes were observed.

In-Pile-Loop Subassemblies

The metallographic examination of sections of fuel pins from Subassembly 1B-1 αT and chemical analysis of the spacer from Pin 16 has been completed. The microstructures of the longitudinal section of the spacer from Pin 16 and the transverse section from Pin 15 were found to be very similar to the eutectic structure observed in the transverse section from Pin 16. Chemical analyses of the spacer and cladding of Pin 16 indicated that 0.5 w/o sulfur was present. It is believed that sulfur contamination produced the observed eutectic structure, thus causing failure of the fuel pins. These results complete the evaluation of this subassembly.

GCRE Critical-Assembly Experiments

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

The ML-1-1B critical assembly is being constructed. During the past month the core components were assembled with the exception of the structure being constructed to support the scissor-acting control blades.

The assembly will be completed and checked out during the next month. Initial attempts at criticality can be started as soon as the fuel is received at Battelle.

Alloy Modification of Hastelloy X

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

The present cladding choice for the GCRE is Hastelloy X. Exposure of the cladding material to a nitrogen-1/2 volume per cent oxygen atmosphere at 1750 F has resulted in a loss in ductility (from 40 per cent total elongation to 12 per cent total elongation) after 5000 hr of exposure. This decrease is not serious; but it is believed that further exposure (an additional 5000 hr) might result in a further loss in ductility which could lead to cladding failure. It is thought that either grain growth or redistribution of second phases (including carbide formation) may be responsible. The loss of ductility due to grain growth may be remedied by adding a grain refiner such as yttrium. The addition of niobium or yttrium may result in better distributions of the second phases, or allow the carbides to be retained in a ductile niobium or yttrium matrix within the grain boundary. Therefore, the effect of alloy additions which may improve the ductility after exposure to the nitrogen-1/2 volume per cent oxygen atmosphere and which also may improve the oxidation resistance of the cladding material is being studied. The effect of variations in the base-alloy composition is also being investigated. In addition, a study of the mechanism of oxidation and the effect of alloying on the mechanism is planned.

A list of the alloys to be prepared is shown as Table U-2. Alloys are being melted in an Al_2O_3 crucible and poured at 2900 F into a Zirconite mold. This operation is being accomplished in a vacuum-induction furnace. Initially 1000 g was charged. The ingots will be forged at 2100 F and rolled at the same temperature to 0.050-in. sheet. After fabrication a portion of the 0.050-in. sheet will be sent to AGN for corrosion testing. The remaining sheet will be fabricated into tensile specimens for testing at 75, 1000, 1750, and 1850 F in air.

Melting has been completed and samples are being removed from each ingot for chemical analyses and metallographic examination. Fabrication will follow as soon as the necessary samples are obtained.

TABLE U-2. NOMINAL COMPOSITION OF MODIFIED HASTELLOY X-TYPE ALLOYS

Alloy	Nominal Composition, w/o									
	Ni	Cr	Mo	W	Fe	Mn	Si	C	Y	Nb
61	45	21	10	0.7	20	1.0	1.0	0.1	0	0
62	45	21	10	0.7	19.5	1.0	1.0	0.1	0.5	0
63	45	21	10	0.7	19	1.0	1.0	0.1	1.0	0
64	45	21	10	0.7	20	1.0	1.0	0.01	0	0
65	45	21	10	0.7	19.5	1.0	1.0	0.01	0.05	0
66	45	21	10	0.7	19.0	1.0	1.0	0.01	1.0	0
67	45	21	10	0.7	18	1.0	1.0	0.1	0	2
68	45	21	10	0.7	17.5	1.0	1.0	0.1	0.5	2
69	45	21	10	0.7	17.0	1.0	1.0	0.1	1.0	2
70	55	21	10	0.17	10	1.0	1.0	0.1	0	0
71	55	21	10	0.17	9.5	1.0	1.0	0.1	0.5	0
72	55	21	10	0.17	9	1.0	1.0	0.1	1.0	0
73	55	21	10	0.7	10	1.0	1.0	0.01	0	0
74	55	21	10	0.7	8	1.0	1.0	0.1	0	2
75	45	25	10	0.7	15	1.0	1.0	0.1	0	0
76	45	25	10	0.7	14.5	1.0	1.0	0.1	0.5	0
77	45	25	10	0.7	14.0	1.0	1.0	0.1	1.0	0
78	45	25	10	0.7	20	1.0	1.0	0.01	0	0
79	45	25	10	0.7	18	1.0	1.0	0.1	0	2

U-7

IN-PILE-LOOP PROGRAM

G. A. Francis

The two in-pile recirculating gas loops reported here are installed at the Battelle Research Reactor and at the Engineering Test Reactor. Both units are being used in support of the AGN program for the Army Reactors Branch of the AEC. The BRR loop is scheduled to start operation with a pin-type fuel subassembly in late February or early March. The ETR loop should go into operation near the same time with a plate-type fuel subassembly.

BRR Loop Program

S. J. Basham and W. H. Goldthwaite

During January preparation for operation of the AGN test specimen designated 1B-2T was continued. The information relative to hazards was received, and a report was completed. It is now being reviewed by the appropriate Battelle committee.

The continuous loop gas-monitoring system was installed, and calibration was started. This calibration will be based on data obtained by introducing different fission gases into the chamber. The evaluation of data should be completed during February.

Plans call for receipt of the 1B-2T late in February, with experiment operation to begin after flux mapping is completed.

ETR Loop Program

E. O. Fromm, J. V. Baum, D. E. Roop,
and G. A. Francis

The initial fuel subassembly to be irradiated in the ETR loop is designated 1-3P and is a plate-type subassembly similar to that used in the initial GCRE. The irradiation of this specimen should begin in late February or early March. This date is contingent upon reactor scheduling, loop-blower performance, and satisfactory operation of the loop itself.

A third blower, designated Blower A, was shipped to the site during January, and is now installed in the loop system. Blower operation at the site has been limited to the Blower B unit. One belt failure has been experienced after approximately 554 hr of operation. The exact cause of the failure has not been determined. Study of the failure includes operation of a unit under similar conditions in Battelle's laboratory and an evaluation of the application by the belt supplier, who is investigating the possibility of the belt being defective.

The blower now on test at Battelle is designated Blower D. It has operated approximately 600 hr and has been subjected to five startups that simulate conditions at the reactor. Plans call for continuation of this test until a need for the unit at the reactor is recognized.

The change to loop control based on variable flow has been checked at low heater powers. The check appears satisfactory but is not conclusive and will be continued next month. The initial heater control has been used in these tests. As a result of the heater-control study, the supplier agreed to furnish a new unit. This has been received at the site and is now ready for installation.

It has been requested that the 1-3P be installed during the first normal reactor shutdown in February so that irradiation will begin on the subsequent startup.

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

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

A study has been initiated to determine the corrosion resistance of thorium and uranium and certain uranium alloys during storage under atmospheric conditions. Preliminary studies are under way in the laboratory to establish the exposure techniques for the evaluation program. The actual exposure tests will start soon after the material becomes available, probably early in March.

RWD/CRT:all