Report No. BMI-1473

UC-25 Metallurgy and Ceramics
(TID-4500, 15th Ed.)

Contract No. W-7405-eng-92

PROGRESS RELATING TO CIVILIAN
APPLICATIONS DURING
OCTOBER, 1960

by

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November 1, 1960

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REPORTS RELATING TO CIVILIAN APPLICATIONS
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- BMI-1466 "Measuring the Release of Short-Lived Fission Gases During Capsule Irradiations", by Charles W. Townley, Gilbert E. Raines, Ward S. Diethorn, and Duane N. Sunderman.
- BMI-1467 "Stabilizing Effects of Oxide Additions to Uranium Oxide", by Wendell B. Wilson and Arnold F. Gerds.
- BMI-1468 "Progress on Ceramic Coated Fuel Particles at Battelle".
- BMI-1469 "Progress Relating to Civilian Applications During September, 1960", by Russell W. Dayton and Clyde R. Tipton, Jr.

REACTOR MATERIALS AND COMPONENTS (AEC-DRD)

R. F. Dickerson

Results obtained from studies of the effect of pressure on reactions between U_3O_8 and Al_2O_3 indicate that at 1300 C and 60,000 atm the gamma modification of U_3O_8 reacts with Al_2O_3 to form a new phase. The data show that the phase of interest is produced at or near the equimolar composition, $UO_{2.67}-Al_2O_3$. It must be emphasized that no reaction between U_3O_8 and Al_2O_3 has been observed at ambient pressures.

The ETR has resumed operation and the ten capsules containing Type 347 stainless steel are again under fast-neutron bombardment. The capsules have been moved again and now reside in Position G-13. Unfortunately, no flux data are currently available for this position, although mapping is now in progress.

Niobium-base binary alloys containing 2.37 w/o chromium, 3.3 w/o zirconium, and 5.21 w/o vanadium, as well as a ternary alloy containing additions of 1.56 w/o zirconium and 1.09 w/o chromium, are promising candidates for advanced cladding material for the Experimental Breeder Reactor. All alloys have been cast into ingots weighing approximately 5 lb, and all have been successfully forged and rolled to 0.040-in. sheet at 540 C. All tensile testing is completed, and 650, 800, and 1000 C creep tests are in progress. Annealing for the recrystallization studies is under way, and 1-in.-diameter rod has been successfully rod rolled at 1200 C. Because of a change in program plans, thermal-expansion and thermal-conductivity measurements will not be accomplished.

The uncertainties connected with test reactor operating conditions coupled with lower than planned thermal conductivity of an MgO powder-filled annulus have resulted in modifications of the in-pile Zircaloy-2 creep capsule. Metal-finned heat-transfer media have been tried but still there are local zones where heat transfer is not properly accomplished. Attempts are in progress to relate with slip the striations in thin sections of Zircaloy-2 which are noted during examination by electron microscopy. Flat tensile specimens have been strained about 15 per cent and are being studied. Results to date indicate that slip in one system blocks slip on a second system at room temperature. This blocking should be temperature dependent.

Results of recent tests show that friction and wear of molybdenum is reduced in the presence of sodium. In the case of tungsten carbide the friction and wear characteristics are no different in or out of sodium; however, friction and wear of titanium carbide are much more severe in sodium than outside sodium. Since it appears that the surface films formed have a great effect on friction characteristics of materials, effort is under way to identify the presence and composition of surface films on a variety of materials.

High-Pressure High-Temperature Solid-State Studies

W. B. Wilson and C. M. Schwartz

An investigation is being conducted to determine the effects of combined ultrahigh pressure and high temperature on the oxides of uranium and on the reactions of uranium

oxides with various mixed oxides. Current work has been directed toward a study of the possible reactions of U_3O_8 and Al_2O_3 under pressure.

The initial phase of these studies was undertaken by subjecting U_3O_8 powder containing 25 and 50 mole per cent Al_2O_3 to various pressures and temperatures. X-ray diffraction examination of the products of these experiments indicated the possible formation of a reaction product which was not previously observed in the absence of pressure or associated with the high-pressure modifications of U_3O_8 .

In order to determine if pressure was required for the above reaction, work was undertaken at ambient pressure to produce a reaction between U_3O_8 and Al_2O_3 . All attempts to react U_3O_8 and Al_2O_3 by direct sintering, evaporation, or coprecipitation techniques resulted in free U_3O_8 and Al_2O_3 being detected, after air firing at 800 C.

A series of high-pressure high-temperature reactions was undertaken using the 800 C air-fired oxides prepared by the coprecipitation technique. These materials were heated to varying temperatures ranging from 1000 to 1500 C at 60,000 atm. At the lower temperatures the U_3O_8 was observed to transform to the gamma high-pressure modification which was coexistent with gamma alumina. At 1200 C, gamma U_3O_8 was observed coexistent with gamma alumina which was clearly transforming to alpha alumina. At 1300 C the unknown phase was observed. At still higher temperatures, i. e., above 1400 C, the dissociation of the uranium oxide toward lower oxygen/uranium ratios occurred, resulting in free alumina being formed. Ion-size consideration indicated that the reaction of alumina with the lower oxides of uranium is less likely to occur since the U^{+4} ion is considerably larger than U^{+6} .

Several samples were run at 15,000 atm under the same conditions of time and temperature as above. Analyses of the products indicated that only alpha U_3O_8 and gamma alumina, or alpha Al_2O_3 at higher temperatures, were produced. This result tends to suggest that at 60,000 atm the gamma modification of U_3O_8 is reacting at suitable temperatures to form the new phase which is observed.

A second series of samples was run at a constant pressure of 60,000 atm at a nominal temperature of 1300 C for periods of 1 to 2 hr each. The composition was varied to include $\text{UO}_{2.67}$ with 25, 50, and 75 mole per cent Al_2O_3 as well as the pure end members. The variation of the Al_2O_3 content served to produce an apparent variation of temperature, apparently by virtue of its greater thermal conductivity. However, the phase of interest was produced at or near the equimolar composition, $\text{UO}_{2.67}\text{-Al}_2\text{O}_3$.

Work will continue to confirm the validity of the reaction under investigation and to determine the conditions of temperature, pressure, and composition associated with the phase region of the reaction product. A second series of samples will be examined at slightly higher temperatures in an attempt to determine the precise conditions required to produce the reaction product without interfering phases. Assuming that it is possible to produce a single-phase material, appropriate analytical techniques will be employed to determine the nature of the reaction product and if possible its crystal structure.

Irradiation Surveillance Program on Type 347
Stainless Steel

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

A program is in progress to determine the effects of fast-neutron irradiation (neutrons with energies greater than 1 Mev) on the mechanical properties of AISI Type 347 stainless steel. The program consists of the irradiation and subsequent postirradiation mechanical testing of approximately 14 capsules containing subsize tensile, cyclic-strain fatigue, and impact specimens. Specimens will be exposed over a range of about 5.0×10^{21} to more than 1.5×10^{22} nvt at irradiation temperatures of about 120 and 600 F. Information obtained will be used to predict the in-reactor life of an operating ETR loop constructed of this material.

Currently, ten capsules are being irradiated in Position G-13 of the ETR, and two capsules are being irradiated inside fuel elements in the WTR, with a third capsule scheduled for WTR insertion at a later date. One capsule, BMI-24-14, was discharged from the ETR at the end of Cycle 28 (estimated fast-flux exposure was 5.7×10^{21} nvt). The results of mechanical tests on some of the specimens contained in this capsule were previously reported in BMI-1448. This irradiation exposure produced a decrease of 54 to 83 per cent in the energy necessary to cause fracture in room- and liquid-nitrogen-temperature tests of Charpy V-notch specimens and produced a substantial increase in bending resistance in subsize Izod specimens.

The ETR achieved a total of about 1360 MWD during Cycle 31, and is expected to operate for about 5000 MWD during Cycle 32 (termination date for Cycle 32 is October 31). No flux data are presently available for G-13, the position occupied by the surveillance capsules since Cycle 31, although a program to thoroughly map this region is in progress. The total estimated neutron exposures for both the ETR- and WTR-irradiated capsules as of the end of Cycle 30 were reported previously.

Present plans call for the discharge and subsequent testing of specimens of one of the maximum-exposure capsules early in 1961.

Development of High-Strength Niobium-Base Alloys

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

The Experimental Breeder Reactor, a sodium-cooled fast reactor, is currently operating at temperatures below the iron-uranium eutectic (725 C). In order to operate this reactor at temperatures above 725 C, a new cladding material must be substituted for the stainless steel currently in use. The absence of a eutectic reaction between uranium and niobium at temperatures up to 1000 C and the high strength at elevated temperatures of material producible by alloying make niobium alloys quite attractive for cladding applications. Prior research (BMI-1417) has shown that chromium, vanadium, and zirconium additions improve the elevated-temperature strength of niobium. Although the high melting points of these alloys indicated that they should retain their strengths well above 1000 C, an experimental study to confirm this was needed.

Niobium-base alloys containing 2.37 w/o chromium, -3.3 w/o zirconium, -5.21 w/o vanadium, and -1.56 w/o zirconium-1.09 w/o chromium have been double melted in a consumable-electrode arc-melting furnace and forged to 1/2-in.-thick slabs at 540 C. These slabs were subsequently rolled to 0.040-in. sheet at 540 C.

All specimens for creep- and tensile-property determinations were given a 1-hr anneal at 1450 C followed by furnace cooling. Metallographic examinations indicated that all four alloys were completely recrystallized by the above treatment.

The results of tensile testing of all four alloys were reported in BMI-1469. Evaluation of the data showed that very little change in yield strength of the alloys was experienced between 650 and 1000 C. On a weight percentage basis, chromium appears to be the most potent strengthener of niobium. Creep tests are in progress at 650, 800, and 1000 C.

Studies to determine the recrystallization temperature of material cold worked (reduction in thickness by rolling) 95 per cent and given a 1-hr anneal have been temporarily delayed. Specimens of each alloy being studied are being annealed at 982, 1038, 1093, 1149, 1204, 1260, and 1316 C. The recrystallization temperature will be determined by hardness measurements and then confirmed by metallographic examination.

Two-inch-diameter ingots weighing approximately 4 lb each of niobium-2 w/o chromium, -3.0 w/o zirconium, -5.3 w/o vanadium, and -2.0 w/o zirconium-1.0 w/o chromium alloys have been double melted by consumable-electrode arc melting. These have been rod rolled in stainless steel cans at 1200 C to 1 in. in diameter. Three-inch-long pieces of this rod have been sent to ANL for extrusion studies. The remaining rod will be rolled to sheet for tube-forming studies also at ANL.

Thermal-expansion and thermal-conductivity tests of the above-mentioned alloys will not be performed because of modifications of the program as originally planned.

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

F. R. Shober, P. B. Shumaker, A. P. Young, and C. K. Franklin

An investigation to study the creep properties of Zircaloy-2 during irradiation at elevated temperatures is being made. The purpose of this investigation is to compare total creep deformation obtained in a high flux of fast neutrons, neutrons having energies greater than 1 Mev, with that produced under similar stress and temperature conditions out of the reactor. Strain aging of Zircaloy-2 as it may pertain to the in-reactor experiment is being investigated. The instrumented capsule designed to contain the in-reactor experiment is presently undergoing modifications to increase the heat-transfer properties of the capsule. No new data have been obtained from the strain-aging studies.

A major problem in design of the capsule has been the control of the specimen test temperature. It is expected that a specimen temperature of 650 F can be achieved by a combination of gamma-energy heating and auxiliary electrical heat. Appropriate

control of the power to these auxiliary heaters will assure adequate temperature control. Originally, the design was based on the use of a 70-mil-thick MgO powder annulus as the principal barrier to heat flow from the specimen to the reactor coolant water. A thermal mock-up of the capsule with the MgO-filled annulus revealed that the heat transfer across the powder was inadequate. The experiment did not achieve a powder conductivity of $1.5 \text{ Btu}/(\text{ft})(\text{hr})(\text{F})$, the value employed for the design calculations. It was believed that a higher conductivity metal-finned annulus might be more serviceable since the gamma heating rate, particularly at reactor startup, will be higher than initially anticipated. However, thermal mock-up experiments with narrow, widely spaced fins designed for nominal metal-to-metal contact or very light interference fits with adjacent metal parts demonstrated an annulus conductance still inadequate for in-pile operation. Apparently, there were local zones in which heat-transfer-area contact was not properly established even though the machining tolerances were close. While total contact could be assured by constructing the fins for high interference fits, this could not be tolerated because of the high stresses induced in the various mating components.

In an attempt to bypass this difficulty with solid metal fins, an alternate scheme in which tubing replaces the solid fins is under investigation. Tubing, because of its relatively low resistance to crushing, can be designed for interference fits without the accompanying high component stresses. Final assembly of the in-pile capsule and an out-of-pile control capsule will begin as soon as a suitable heat-transfer combination has been determined.

Striations visible by electron microscopy in thin sections of cold-rolled Zircaloy-2 have been reported previously (BMI-1464). The striations were believed to be related to slip bands formed during rolling.

If striations in thin sections are related to slip bands, they should also appear in thin sections prepared from tensile samples. In a flat tensile sample, it should be possible by prepolishing one surface before straining to compare slip bands on the surface with striations in thin sections. For this purpose, a tensile sample of the type prepared for creep testing was prepolished and strained about 15 per cent. Electron micrographs of slip bands have been prepared from replicas of the polished surface after straining. Examination of thin sections from the strained sample is not complete.

The surface examination of the tensile sample indicated that, whereas slip on more than one slip system in a single grain occurs quite frequently, the slip bands do not generally intersect; and where they do appear to intersect, there is no offset. No offset has been observed either in striations in thin sections of cold-rolled Zircaloy-2 sheet. Apparently, slip on one system blocks slip on a second system at room temperature in Zircaloy-2. This blocking should be temperature dependent. The temperature dependence of creep properties may be related to the temperature at which slip bands can cross. If striation in thin sections from the tensile sample can be related to slip, it is planned to strain samples at elevated temperature to determine when slip bands do cross.

Radiation Stability of Boron-Containing Powders

R. A. Ewing and D. N. Sunderman

The objective of this program is to investigate the suitability of hafnium diboride as an alternate control material or burnable poison. Samples of irradiated specimens have been prepared for X-ray diffraction examination to determine the changes in unit-cell dimensions resulting from irradiation. Samples are also being prepared for lithium analyses to determine boron-10 burnup.

Friction and Wear of Sliding Surfaces in Sodium Environments

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

Molten sodium, when applied between the rubbing surfaces of two materials, has been shown to alter the friction and wear behavior exhibited by the materials when rubbed dry. For a molybdenum ball sliding across a molybdenum plate, friction and wear were reduced when sodium was present. For similar specimens of a tungsten carbide cermet (K-91) no such improvement was observed, and for a titanium carbide cermet (K-138A) friction and wear were greater with sodium. In the case of molybdenum, the observed friction and wear behavior have been correlated with the formation of a surface film of Na_2MoO_4 which functions as a lubricant. Recent efforts to identify similar films for the other materials tested have not yet been successful.

Efforts to identify the presence and composition of surface films on a variety of friction-test specimens are being continued. Electron-diffraction analyses of specimen surfaces exposed to the friction-test environments are in progress, and a re-analysis of the friction records is being performed. The latter procedure involves a comparison of the stick-slip behavior between the specimens rubbed dry and rubbed immersed in sodium. This technique may indicate the presence and extent of a surface film.

STUDIES OF ALLOY FUELS (AEC-DRD)

R. F. Dickerson

The study of niobium-uranium alloys is being continued.

Thorium aluminides, thorium beryllides, and thorium silicides have been successfully arc cast with resulting densities less than 2 per cent below the theoretical densities. In oxidation tests in dry air at 1200 F, ThBe_{13} showed only a negligible weight gain during a 2-hr test. Thorium and thorium-uranium aluminide, beryllide, carbide, and silicide specimens contained in argon-filled Vycor capsules survived a quench test from 1000 C into a brine-and-ice water solution. A number of these various compounds, some with ternary additions, are being corrosion tested in 350 C Santowax.

The study of the effect of zirconium and silicon additions to niobium-plutonium alloys is being continued. Thorium-5, -10, and -30 w/o plutonium and thorium-10 w/o zirconium-10 w/o plutonium alloys are being prepared by arc-melting techniques. The PuO_2 obtained for a proposed compatibility study has been characterized, and pellets have been made by pressing and sintering. The green density of the material was about 60 per cent of theoretical, but after sintering for 1 hr at 1700 C the density increased to 90 per cent of theoretical.

Development of Niobium-Uranium Alloys

J. A. De Mastry, D. P. Moak, A. A. Bauer, and R. F. Dickerson

A program is under way to develop niobium-uranium alloys for reactor applications at elevated temperatures.

It is planned to investigate the fabrication characteristics of alloys containing 30 w/o or more uranium. The creep strength of niobium-10, -20, and -30 w/o uranium alloys and a niobium-20 w/o uranium-20 w/o zirconium alloy will be determined at 1600 and 2000 F. Testing will start as soon as specimens have been fabricated.

An irradiation program is under way. One capsule has been assembled and is ready for irradiation. A second capsule is currently being assembled. Each capsule contains five fuel-pin specimens which will be irradiated at temperatures of 1600 and 1800 F.

Development of Uranium-Containing Thorium Compounds

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

Thorium and thorium-uranium compounds are being investigated with the aim of developing a high-temperature fuel suitable for use in a thermal breeder reactor. Compounds having a low thermal-neutron cross section, a high melting point, and a high

thorium density have been chosen for study. They are: Th_2Al , ThBe_{13} , ThB_4 , ThC , ThC_2 , ThN , Th_3P_4 , ThS , Th_3Si_2 , and ThSi , and these same compounds with a 10 a/o addition of uranium. Some of the important properties of the more refractory thorium compounds from which the above compounds were selected for study are presented in Table B-1.

The results of density determinations on several arc-cast thorium and thorium-uranium compounds are shown below:

	<u>Density, g per cm^3</u>
Th_2Al	9.59
$(\text{Th}_9\text{U})_2\text{Al}$	9.89
ThBe_{13}	4.10
$(\text{Th}_9\text{U})\text{Be}_{13}$	4.28
Th_3Si_2	9.65
$(\text{Th}_9\text{U})_3\text{Si}_2$	9.87
ThSi	8.95
$(\text{Th}_9\text{U})\text{Si}$	9.05

The densities of the arc-cast Th_2Al , ThBe_{13} , Th_3Si_2 , and ThSi (see Table B-1) are less than 2 per cent under their theoretical densities, thus showing that the arc-casting technique is yielding high-density material.

Dry-air oxidation tests are being performed at 1200 F. Preliminary results show that Th_2Al and ThSi had average weight gains of $144 \text{ mg}/(\text{cm}^2)(\text{hr})$ and $268 \text{ mg}/(\text{cm}^2)(\text{hr})$, respectively. The compound ThBe_{13} showed a negligible weight gain during a 2-hr test.

Thermal-shock tests were performed by quenching specimens, encapsulated in argon-filled Vycor capsules, from 1000 C into a brine-and-ice water solution. All of the thorium and thorium-uranium aluminides, beryllides, carbides, and silicides tested survived this test without severe cracking. This type of test is approximately equivalent to an air cool from 1000 C.

Presently thorium-uranium monocarbide and dicarbides with small additions of either molybdenum, niobium, silicon, or zirconium and thorium and thorium-uranium aluminides, beryllides, and silicides are being tested in 350 C Santowax.

Hot-hardness measurements will be made, and corrosion resistance in boiling water, CO_2 , and NaK will be determined. The thermal-expansion, thermal-conductivity, and strength characteristics evaluated in compression of the most promising compounds will be determined.

TABLE B-1. PROPERTIES OF SOME HIGH-MELTING-POINT THORIUM COMPOUNDS

Compound	Thorium Content		X-Ray Density, g per cm ³	$\frac{\sum_a \text{Th in ThX}}{\sum_a \text{ThX}}$	Approximate Melting Point, C	Crystal Structure	Uranium Counterpart	Thorium Density, g per cm ³ of compound
	w/o	a/o						
Th ₂ Al	94	66.7	9.67	0.983	1660	Tetragonal	None	9.09
ThAl ₂	81	33.3	7.00	0.941	1620	Hexagonal	UAl ₂	5.69
ThBe ₁₃	66.5	7.15	4.10	0.997	>1600	Face-centered cubic	UBe ₁₃	2.72
ThB ₄	84.4	20	8.45	0.974 ^(a)	2500	Tetragonal	UB ₄	7.13
ThB ₆	78	14.3	6.4	0.962 ^(a)	2195	Cubic	None	4.98
ThC	95.2	50	10.7	0.999	2600	Face-centered cubic	UC	10.2
ThC ₂	92.0	33	8.96	0.999	2700	Monoclinic	UC ₂	8.25
ThN	94.3	50	11.56	0.801	2630	Face-centered cubic	UN	10.9
Th ₃ P ₄	85	42.9	8.56	0.966	--	Body-centered cubic	U ₃ P ₄	7.28
ThO ₂	91.2	33.3	10.03	0.999	3300	Cubic	UO ₂	9.12
ThS	84.7	50	9.6	0.893	2200	Face-centered cubic	US	8.06
Th ₃ Si ₂	92.6	60	9.81	0.988	--	Tetragonal	U ₃ Si ₂	9.09
ThSi	89.2	50	8.99	0.981	--	Orthorhombic	USi	8.02

(a) Cross section calculated assuming use of boron-11.

A Study of Plutonium-Containing Fuel Alloys

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

A project aimed at developing irradiation-resistant plutonium-bearing fuel alloys is being conducted. Selection of alloy systems to be initially investigated was based on irradiation behavior of similar uranium alloys. These alloy systems include niobium, thorium, uranium-molybdenum, and uranium-niobium alloys.

The investigation of binary niobium-plutonium alloys has been halted as metallographic examination of these alloys indicates very limited solid solubility and a tendency for the plutonium to concentrate at the grain boundaries. Such a structure would normally not be considered attractive in a fuel element. Ternary additions of silicon and zirconium are being studied in an effort to capitalize on the attractive high-temperature properties of niobium. Niobium-25.6 w/o plutonium-6.1 w/o silicon, niobium-10 w/o zirconium-10 w/o plutonium, and niobium-30 w/o zirconium-10 w/o plutonium alloys are being prepared by an arc-melting and drop-casting technique. It is hoped that the silicon will tie up the plutonium as PuSi_2 to produce a niobium-30 volume per cent PuSi_2 dispersion-type alloy. The zirconium additions are expected to increase the solubility of plutonium in niobium and possibly result in a high-strength solid-solution alloy.

Thorium-5, -10, and -30 w/o plutonium and thorium-10 w/o zirconium-10 w/o plutonium alloys are also being prepared by arc-melting techniques. Extensive solid solubility of plutonium in thorium is expected to improve the strength of thorium. Zirconium additions should further enhance the mechanical properties of thorium and may improve corrosion resistance.

Other alloys to be prepared and studied include uranium-10 and -15 w/o molybdenum-5, -10, -15, and -20 w/o plutonium, and uranium-20 w/o niobium-5, -10, -15, and -20 w/o plutonium. Both uranium-molybdenum and uranium-niobium alloys exhibit good elevated-temperature strength, and it is hoped plutonium additions will not seriously affect these properties. Quaternary additions of zirconium to the uranium-niobium-base alloys will also be investigated.

Preliminary investigations of these alloy systems will consist mainly of metallographic examination, hardness testing, and tensile testing. Effects of heat treatment on structure and hardness will also be determined. Alloys exhibiting promising combinations of microstructure (solid solutions or finely dispersed structures) and strength will be investigated further.

Compatibility of PuO_2

S. J. Paprocki, D. L. Keller, and W. M. Pardue

It is the purpose of this program to study the compatibility of PuO_2 with prospective cladding and diluent materials. The experimental procedures will consist of heat treating compacts containing a mixture of PuO_2 powder and the material in question and then following the appearance of new phases or reaction products by X-ray diffraction

measurements, metallographic examination, and changes in the physical condition of the compacts. Abutted surfaces of PuO_2 and the candidate material will also be studied after extended heat treatments.

Initial studies were concerned with calculations of the standard free-energy change for possible PuO_2 -metal reactions at temperatures of 1000 to 2000 K. These results, summarized in BMI-1455, provided a preliminary screening of the various materials.

In order to best interpret the compatibility tests and the sintering performance of PuO_2 , some effort was expended in characterizing samples of the 500 g of PuO_2 powder earmarked for this program. Standard screen analysis and petrographic examination have revealed that the average particle size is approximately 9μ with a maximum size of 64μ . The grain size varies from submicroscopic to 3μ in diameter. The particle shape is generally spherical with essentially zero porosity. Two extraneous phases were tentatively identified as UO_2 - PuO_2 solid solution (0.1 volume per cent) and Pu_2O_3 (10 volume per cent).

During the past month, pellets were pressed and sintered to determine the sinterability of this particular PuO_2 powder. The as-pressed green density increased with the compacting pressure and varied with the type of organic binder used. An average value was 60 per cent of theoretical. After sintering in argon for 1 hr at 1700 C, the density increased to approximately 90 per cent of theoretical.

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

J. B. Melehan, R. H. Barnes, R. L. Ritzman,
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Studies of fission-gas release in single crystal and sintered UO_2 as determined by postirradiation heating and by continuous in-pile measurement continued as well as studies of the pore structure of sintered UO_2 . These studies are intended to provide an understanding of the mechanisms of release of fission gas in UO_2 .

Diffusion in UO_2

Studies involving fission-gas release from uranium dioxide during postirradiation heating were continued. Xenon-133 release from a sintered uranium dioxide pellet having a theoretical density of 89.2 per cent was measured at temperatures of 1207 and 1440 C. An identical experiment with a similar specimen is scheduled to determine the reproducibility of the release measurements. Results of this and other recent experiments are summarized in Table C-1. The results of the measurements of xenon-133 release from sintered uranium dioxide are expressed in terms of an apparent diffusion coefficient ($D' = D/a^2$), which is the absolute diffusion coefficient divided by the square of the equivalent sphere radius. These data are being analyzed further to correlate them with the diffusion data from the single crystals and data from other laboratories by means of the information being determined on the releasing surface area. This method of expression of the data is used for convenience, since the release involved is small and is surface dependent for all of the test conditions. Thus, the term, "a", should be looked upon as a measure of the surface-to-volume ratio of the specimen.

TABLE C-1. XENON-133 DIFFUSION IN SINTERED URANIUM DIOXIDE

Specimen	Specimen Density, g per cm ³	Integrated Thermal-Neutron Exposure, nvt	Material Surrounding Specimen During Irradiation	Diffusion Temperature, C	Diffusion Heating, Time, min	Apparent Diffusion Coefficient ($D' = D/a^2$), sec ⁻¹
1	8.65	2.04×10^{16}	UO_2 powder	1402	330	2.68×10^{-10}
2	8.71	2.04×10^{16}	UO_2 powder	1185	285	1.75×10^{-9}
	8.71	2.04×10^{16}	UO_2 powder	1393	311	4.26×10^{-9}
3	9.85	1.27×10^{16}	UO_2 powder	1201	390	1.04×10^{-10}
	9.85	1.27×10^{16}	UO_2 powder	1398	390	8.64×10^{-9}
4	9.85	1.27×10^{16}	UO_2 powder	1404	390	1.05×10^{-8}
5	9.93	2.18×10^{16}	UO_2 powder	1197	330	1.02×10^{-12}
	9.93	2.18×10^{16}	UO_2 powder	1401	390	1.17×10^{-11}
6	10.02	2.18×10^{16}	UO_2 powder	1399	390	6.56×10^{-13}
7	10.64	1.77×10^{16}	UO_2 powder	1406	390	4.27×10^{-12}
8	10.61	1.77×10^{16}	UO_2 powder	1202	390	2.65×10^{-15}
	10.61	1.77×10^{16}	UO_2 powder	1397	390	7.30×10^{-14}
	10.61	1.77×10^{16}	UO_2 powder	1546	240	5.57×10^{-13}
9	9.77	3.54×10^{15}	None	1207	390	3.68×10^{-11}
	9.77	3.54×10^{15}	None	1440	390	7.55×10^{-9}

The preliminary analysis of the data from all the single-crystal experiments that have been performed to date has been completed. The results of the measurements of xenon-133 diffusion from small flat-plate specimens of Norton single-crystal uranium dioxide along with the specimen details are summarized in Table C-2.

TABLE C-2. XENON-133 DIFFUSION IN SINGLE-CRYSTAL URANIUM DIOXIDE

Specimen	Heat Treatment	Specimen Density, g per cm ³	Integrated Thermal-Neutron Exposure, nvt	Diffusion Temperature, C	Diffusion Heating, Time, min	Diffusion Coefficient, cm ² per sec
1	None	10.93	5.8×10^{17}	1205	530	8.43×10^{-18}
2	None	10.94	2.7×10^{17}	1400	345	1.71×10^{-17}
3	(a)	11.01	7.3×10^{17}	1390	362	1.72×10^{-17}
4	(b)	10.91	5.4×10^{17}	1300	650	3.45×10^{-17}
5	None	10.92	1.2×10^{18}	1200 1310 1405	330 330 330	6.15×10^{-18} 1.41×10^{-17} 2.02×10^{-17}
6	None	10.98	9.7×10^{17}	1400	360	2.14×10^{-17}

(a) As-polished specimen was heated under vacuum for 3 hr at 1800 C, removed from the furnace for metallographic examination, and heated under vacuum at 1800 C for an additional hour. The specimen was then repolished and heated in hydrogen for 1 hr at 600 C.

(b) As-polished specimen was heated under vacuum for 3 hr, repolished, and then heated in hydrogen for 1 hr at 600 C.

The diffusion coefficients for the single-crystal material were determined from the relationship between the fraction of xenon-133 released and diffusion time using solutions of Fick's diffusion equation for a flat plate. An activation energy of about 30 kcal per g mole was determined from the diffusion coefficients in Table C-2 for the diffusion of xenon-133 in single-crystal uranium dioxide. It should be recognized, though, that this value is based on a limited amount of data at the lower temperatures. Also, the activation energy applies only for material not heat treated, since no data on heat-treated samples are included below about 1400 C. It is probable that the activation energy determined on heat-treated or annealed material would be considerably different than that given here for the arc-melted material.

Characterization of UO₂ and Model of Gas Release

To accurately determine the diffusion coefficients of fission gases in UO₂, the total surface area which is releasing the fission gases must be known. Therefore, measurements have been made to determine the surface area of sintered UO₂ pellets with varying densities, by using quantitative metallographic techniques. In addition, other measurements such as grain size are of interest for a series of specimens.

During the last month, the grain size and the mean free path between the voids in the sintered UO_2 specimens were determined, and the surface areas of the internal pores were calculated. The results are given below:

Specimen Group	Density, per cent of theoretical	Surface Area, cm^2 per cm^3	Grain Size, 10^{-4} cm	Mean Free Path, 10^{-4} cm
1	92.6	4044	3.68	9.1
2	94.5	2340	11.1	16.2
3	96.3	1592	14.4	24.3
4	98.6	748	23.7	52.8

There seems to be a linear relationship between density and internal surface area for all specimens except Group 1, which has a larger internal surface area than expected.

Of course, these specimens contain both open and closed porosity, and during a diffusion experiment only the gases which are released into the open pores will be detected. Thus, attempts were made to distinguish between open and closed porosity. To determine the open porosity, the UO_2 pellet was impregnated with a plastic which contains a fluorescent indicator. When the specimen is viewed under a mercury-vapor lamp, the open porosity can be determined. This type of measurement is being continued on a series of samples.

In-Pile Fission-Product Release

Measurements are continuing on the release of fission gases from UO_2 during irradiation. Experiments just completed have consisted of 3-day exposures of sintered UO_2 (92 per cent of the theoretical density) at 500 and 1000 F. Gas samples collected during irradiation were analyzed for the krypton-85m, krypton-87, xenon-133, and xenon-135 isotopes. In the 500 F experiment, the quantity, R/B (the ratio of isotope release rate to isotope production rate) varied between 3×10^{-4} and 3×10^{-3} . At 1000 F, the R/B values were in the range 1×10^{-4} to 3×10^{-3} with an apparent consistent increase to a saturation value within a matter of several hours. The gas release mechanisms operating in the two experiments are being considered, and additional analytical analyses of the radiochemical data are being made. The effect of surface area on gas release rate will be studied in the next experiment using single-crystal UO_2 with surface areas about 10^{-2} to 10^{-3} times that of the sintered material studied to date. From this experiment it is expected that the release from the surface recoils from the surrounding components will predominate and can be measured. Thus, a separation of any release originating from surface recoils can be made from the releases observed in the other experiments.

GENERAL FUEL-ELEMENT DEVELOPMENT (AEC-DRD)

S. J. Paprocki

Irradiation tests are planned for UO_2 cermet fuels incorporating 20 volume per cent of chromium, molybdenum, and niobium. The specimens will be irradiated at a surface temperature of 1600 F and maximum central-core temperature of approximately 0.7 of the melting point of the contained refractory metal.

Techniques are being developed for the fabrication of UN-base cermet fuels. Preliminary tests indicate that the UN is compatible with stainless steel at 2400 F and molybdenum at 3000 F; however, a reaction was observed with niobium at 3000 F.

A study is being conducted with the objective of developing low-cost fabrication techniques for preparing tubular fuel elements. A stainless steel-clad stainless- UO_2 fuel element has been selected as a reference element; however, it is planned to develop methods that will be applicable for the fabrication of other fuel systems.

Irradiation of Cermet Fuel Elements

S. J. Paprocki, G. W. Cunningham, D. E. Lozier, and E. O. Fromm

Irradiation-capsule studies are being initiated to evaluate the resistance to radiation damage of bodies composed of UO_2 -refractory metal cermets; the metals being utilized are chromium, molybdenum, and niobium. According to present specifications, the experimental bodies will be cylinders fueled with 80 volume per cent UO_2 of a high enrichment. A prime irradiation parameter is that the in-pile center-line temperatures be approximately 0.7 of the melting point of the particular refractory metal present. This stipulation, together with the high thermal conductivities of the dispersions, will make it necessary to conduct the irradiations in a high thermal-neutron flux. Unusually high rates of fission heat production will result from this particular combination. Target burnups for the irradiations will be in the range of 5 per cent of the total uranium atoms present.

It is anticipated that the irradiation can commence in late spring. Therefore, at the present time emphasis will be placed on capsule design, procurement of reactor space, and determination of specimen-fabrication procedures.

It is probable that a total of 12 specimens, four of each cermet, will be irradiated in two capsules. Because of the high thermal fluxes required (in the 3 to 4×10^{14} nv range), it is planned to use the ETR as the irradiation facility. Preliminary calculations indicate that the heat flux-temperature parameters can be satisfied by the use of single-wall capsules with sodium or NaK as the heat-transfer medium.

The major problem concerning the fabrication of specimens involves the selection of cladding. It is difficult to produce tubing of the proper size from the metals chromium, molybdenum, and niobium, and the substitution of other materials creates a

problem in the matching of thermal-expansion coefficients. A ferritic stainless steel is being considered as a cladding, and specimens are being prepared to determine whether core-cladding separation will occur during thermal cycling similar to reactor conditions. In the event that stainless steel is used, it may be necessary to incorporate a void area in the specimen to contain any fission gases which may be released. The cladding temperature will be approximately 1600 F, and the strength of the stainless may be too low to contain the fission gases without some void volume.

Fabrication of Cermet Fuel Elements

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

Techniques of producing UN cermets having a density 90 per cent of theoretical and containing 60 to 90 volume per cent UN are being investigated. As fabrication procedures are established, specimens will be prepared for physical-property measurements.

Currently a number of matrix metals are being mixed with UN for sintering at 4 hr and 2250 F in a vacuum to obtain some idea of compatibility. Cores containing UN and chromium, molybdenum, niobium, and tungsten have been green compacted and sintered. A microscopic examination of the tungsten-containing core could not be made because of its low density. A microscopic examination of the chromium- and niobium-containing specimens indicated some reaction; however, the degree and amount of reaction could not be judged because of the high porosity of the cores. No melting was detected. The molybdenum-containing specimen indicated complete compatibility. Additional UN cores containing chromium, molybdenum, niobium, and Type 347 stainless steel have been prepared for pressure-bonding studies. Because of the higher densities anticipated by the gas-pressure-bonding technique, a better analysis of compatibility is expected.

In addition, a 60 volume per cent UN-Type 347 stainless steel green-pressed cermet has been vacuum sintered for 3-1/2 hr at 2400 F. Green and sintered densities were 68 per cent of theoretical. A microscopic examination of the core indicated the temperature may have been higher as the stainless steel appeared to have melted. The UN particle surfaces were rounded, indicating some solution in the stainless matrix. Similar UN cores with matrix metals of molybdenum and niobium showed no density change after vacuum sintering for 1 hr at 3000 F. A microscopic examination of the UN-molybdenum cermets indicated complete compatibility under these conditions. Microscopic examination of the UN-niobium core indicated the formation of a nitride of niobium and the solution of uranium in the niobium or the solution of UN in the niobium. Additional UN cermets are being prepared for sintering studies.

Development of Low-Cost Tubular Fuel Elements

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

The tubular fuel-element configuration is generally conceded to be one of the most desirable for high-power-density reactors. Since more and more reactor applications

call for high-power-density capability, this program was initiated with the objective of developing techniques for the production of low-cost tubular elements of consistent quality.

Two general approaches are currently under investigation. The first of these is essentially the assembly of concentric components and the consolidation of these components by processes not involving extensive deformation. The principal problem areas with this approach have been in the fabrication of the core component and in the consolidation step.

Two processes have been shown to be feasible for the consolidation step. These processes are high-temperature high-pressure gas autoclaving and explosive consolidation. Since the explosive technique appears to offer the potential of being the lower cost of the two, most of the effort on consolidation is being concentrated on this technique. In experiments conducted to date, the cores have been produced by merely pouring blended powders in the annulus between the concentric cladding tubes and welding the assembly closed in a vacuum by electron-beam techniques. Macroscopic evaluation after compaction has revealed a minor amount of bridging or areas of lower as-loaded density. These compact to thinner sections. It is felt that these difficulties can be circumvented by employing vibratory techniques as developed by Hanford and others or by using cold binder extrusion techniques. Recent experiments with polyvinyl alcohol as a binder indicate that it has great promise for extrusion techniques. Tests are currently in progress to determine optimum binder-filler ratios, and burnout characteristics.

The second approach to low-cost tubular elements has been to adapt commercial techniques for producing tubing by roll bonding sheets of material in controlled areas and then expanding the unbonded areas by hydraulic, pneumatic, or mechanical means into tubes connected by thin fins. Currently the details of these commercial processes are being determined and samples are being obtained.

Factors Affecting Pressure Bonding

G. W. Cunningham and J. W. Spretnak

A study has been made of void formation and growth in the interfacial bond region of pressure-bonded copper specimens. Both metallography and microradiography are being used to evaluate the results.

The equation

$$P + \frac{2\gamma}{r} = kTN \ln \frac{N_v}{N_{v_0}}$$

is useful for qualitative predictions of the number of voids in the bond region. In this equation, which has been used by other workers for the study of void formation in copper-nickel diffusion couples, P is the applied hydrostatic pressure, γ is the surface tension, r is the radius of the pore, k is Boltzman's constant, T is the absolute temperature, N is the atomic density, N_v is the actual vacancy concentration, and N_{v_0} is the equilibrium vacancy concentration.

For a nucleus of a particular radius in a matrix with a constant vacancy concentration, there will be an equilibrium pressure which will maintain the void at that size. If pressure is decreased, the void will capture vacancies and grow in size to the new equilibrium size. If pressure is increased, the void should collapse. In the system under study, it is likely that the vacancy concentration will decrease regardless of the pressure, since other vacancy sinks such as grain boundaries should be operative. Thus, decrease in pressure could be counteracted over a period of time by a decrease in vacancy concentration. However, it would be expected that an increase in pressure would decrease the number of voids since the critical size of the void for growth would be increased, and a smaller number of nuclei would be available. The size of the void is more likely to be controlled by the rate of vacancy transport than the equilibrium size. That the predicted behavior is observed is indicated in Table D-1. For the cases illustrated, both the number of voids and their size are less under the application of pressure.

TABLE D-1. EFFECT OF PRESSURE ON VOID CONCENTRATION^(a)

Surface Roughness, μ in.	Annealing Pressure, psig	Voids Per Cm^2	Maximum Void Diameter, 10^{-4} cm
500 - 500	0	3,130	26.4
500 - 500	4000	30	9.9
125 - 8	0	2,080	33.0
125 - 8	4000	1,550	13.2
8 - 8	0	3,480	13.2
8 - 8	4000	328	6.6

(a) Specimens were bonded at 1500 F and 10,000 psi for 1 min and then annealed 30 min at 1500 F and the pressure indicated.

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

S. W. Porembka

This program was established to refine and further develop the gas-pressure-bonding technique for the fabrication of low-cost ceramic, cermet, and dispersion fuel elements. Since this technique permits both the densification and cladding of fuel in a single bonding operation, it appears attractive as a means for maintaining or improving fuel-element quality while reducing costs. In the program to date, the major portion of the effort has been devoted to the ceramic fuel system as it offers the greatest potential in future applications. However, it is expected that the techniques developed will be applicable to cermet and dispersion systems with a minimum of modification.

The behavior of various types of uranium dioxide has been established through a detailed study of their cold-compacting and pressure-bonding characteristics. The effects of pressure bonding on the properties of Type 304 stainless steel cladding have also been evaluated. These results have permitted the selection of optimum conditions for achieving acceptable final oxide densities in high-integrity bonded fuel elements. Current efforts are directed toward developing preliminary process specifications for the more promising pressure-bonded fuel elements.

Uranium Dioxide Compaction Studies

S. J. Paprocki, D. L. Keller, and J. B. Fox

Various uranium dioxide powders have been investigated with respect to developing high-density green cores which will attain a high density and good dimensional control upon subsequent pressure bonding. Current efforts concerning a more detailed evaluation of the compaction characteristics of fused and ceramic uranium dioxide mixtures are nearly completed. This study is directed toward optimizing both the mixture and compaction characteristics for preliminary process specifications. In conjunction with this investigation, the corresponding pressure-bonding characteristics will also be optimized.

Four specimens of UO_2 which were bonded for 3 hr at 2100 F and 10,000 psi were subjected to a quantitative metallographic analysis in an effort to establish the correlation among various density-measurement techniques. This procedure involves point counting from a grid superimposed on photomicrographs of the specimens taken at a magnification of 250 diameters. Photomicrographs were taken on two different fields of each specimen and the density of each field was determined. The results of these measurements are given in Table E-1, where a reasonable correlation between metallographic and pycnometer techniques was realized.

TABLE E-1. COMPARISON OF DENSITY-MEASUREMENT TECHNIQUES FOR PRESSURE-BONDED URANIUM DIOXIDE

Specimen	Material	Density, per cent of theoretical				
		Metallographic Counting ^(a)			Radiograph Measurement	Pycnometer Determination
		Field A	Field B	Average		
69S	70 w/o fused UO ₂ - 30 w/o ceramic UO ₂	96.2	95.3	95.8	92.1	97.8
67	50 w/o fused UO ₂ - 50 w/o ceramic UO ₂	98.2	98.7	98.5	90.14	99.5
42S	Special dense UO ₂ (ideal cubic)	89.3	92.7	91.0	80.8	90.7
34B	Special dense UO ₂ (-140 +200)	85.5	85.4	85.5	87.0	89.7

(a) There were two fields of each specimen; each with three different orientations of grid.

Gas-Pressure-Bonding Studies

S. J. Paprocki, E. S. Hodge, and C. B. Boyer

Investigations to develop specifications for the gas-pressure bonding of different basic fuel-element shapes utilizing high-density green-pressed oxide cores are being continued. Type 304 stainless steel-clad UO₂ rod and flat-plate elements and flat-plate assemblies have been pressure bonded with excellent dimensional tolerances when high-density cores are used; however, when green-pressed oxide cores are used dimensional control has been found to be very dependent on the method of compaction and on the initial oxide density. Current efforts concern the minimizing of the effects of densification upon the different fuel-element shapes where green-pressed cores are utilized.

The results of the effects of bonding conditions upon the stoichiometry and density of ceramic UO₂ are given in Table E-2. This study is being continued to include the effect of different binders and methods of binder removal upon the stoichiometry and density of this oxide.

Subsize components for both rod and flat-plate elements have been prepared to establish preliminary process specifications with regard to dimensional control. Later, large-scale elements will be fabricated.

TABLE E-2. EFFECTS OF GAS-PRESSURE-BONDING CONDITIONS UPON THE STOICHIOMETRY AND DENSITY OF CERAMIC-GRADE UO_2

Tube ^(a)	Pressure-Bonding Conditions			Oxygen/Uranium Ratio	Density ^(b) , g per cm ³
	Time, hr	Temperature, F	Pressure, psi		
PB-77	3	2100	10,000	2.011	10.6
PB-78	3-1/2	2100	10,000	2.006	10.9
PB-79	4	2100	10,000	2.014	10.7
PB-80	3	2150	10,000	2.016	10.8
PB-81	3	2200	10,000	2.002	10.9
PB-82 ^(c)	3	2150	10,000	2.008	10.6
PB-83 ^(c)	3-1/2	2100	10,000	2.000	10.9
PB-84 ^(d)	3	2150	10,000	2.006	10.7
PB-85 ^(d)	3-1/2	2100	10,000	2.004	10.8
PB-86	3	2050	10,000	2.008	10.7

(a) The oxide was tamp packed into these tubes without the use of binders.

(b) Densities are by pycnometer measurement.

(c) Tubes PB-82 and PB-83 contained a 75 per cent ceramic-25 per cent fused oxide mixture.

(d) Tubes PB-84 and PB-85 contained a 50 per cent ceramic-50 per cent fused oxide mixture.

DEVELOPMENT OF URANIUM CARBIDE (AEC-FUEL CYCLE)

F. A. Rough and W. Chubb

Uranium carbides and their alloys are being developed for the AEC as a part of the Fuel-Cycle Development Program. An integrated program of research on the preparation and properties of uranium carbides is in progress at Battelle Memorial Institute. This research and development program includes: (1) studies of the properties of hot-pressed uranium carbide powders, (2) development of skull-melting and casting techniques for carbides, (3) determination of the mechanical and physical properties of uranium carbides at room and elevated temperatures, (4) measurements of the rates of diffusion of uranium and carbon in uranium carbides, and (5) studies designed to produce basic information on the nature of radiation-damage processes in uranium carbide.

Plus 16-mesh uranium monocarbide shot obtained from Vitro Laboratories was evaluated as a possible material for arc-melting stock. An analysis disclosed that the material was very pure and contained only 188 ppm oxygen.

Metallographic and X-ray examinations of as-cast uranium-vanadium-carbon alloys disclosed the presence of vanadium metal, uranium metal, and uranium monocarbide in certain substoichiometric alloys. It follows that uranium will displace carbon from vanadium carbide at the melting point of uranium carbide.

A tracer technique has been devised for measuring the rate of self-diffusion of carbon in uranium carbide. A diffusion couple in which one of the UC components is enriched in carbon-14 will be employed. Because of the short range of the beta rays from carbon-14 in uranium carbide, analyses for carbon-14 can be made on the carbide surface after the sample has been prepared by grinding. Removal of an additional 0.0005 in. will prepare the sample for the next analysis.

Alternate Fabrication Methods for UC

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

Economical methods for fabricating useful uranium carbide components by powder-metallurgy techniques are being studied.

It was found in previous work on this program that a continuous grain-boundary uranium phase was present invariably in high-density vacuum-sintered compacts. This phase could be eliminated by substituting a static argon sintering atmosphere for the vacuum; however, maximum densities attainable were on the order of 80 to 85 per cent of theoretical. Analysis of these argon-sintered compacts suggested that nitrogen and oxygen were in the UC lattice. It is planned to measure the effect of this nitrogen and oxygen on the high-temperature properties of UC with compacts that have been further densified by hot pressing.

Several compacts prepared from 25- μ uranium-4.64 w/o carbon powder were sintered 4 hr at approximately 1700 C under static argon for use in these tests. Average

density was 9.7 g per cm³. If microscopic examination shows that no free uranium is present, these compacts will be densified by hydrostatic hot-pressing techniques and prepared for testing. In the event that grain-boundary uranium is present, higher carbon and/or nitrogen starting material will be used. This latter material has been prepared as arc-melted buttons.

Varying quantities of a phase tentatively identified as U₂C₃ have been observed in uranium carbide cores hot pressed at 1470 C. One such core, the powder of which contained 4.94 w/o carbon, was vacuum heat treated 1 hr at 1800 C after the hot pressing. The U₂C₃-like phase was not present after this treatment. This behavior is what one would expect of U₂C₃.

In an attempt to reduce UO₂ contamination and simplify handling procedures, it is planned to densify UC of varying mesh fractions by hydrostatic hot-pressing techniques.

Melting and Casting Techniques for Uranium-Carbon Alloys

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

In the interest of developing techniques for the production of high-quality cast shapes, efforts are being devoted to the problem of purity and composition control for large volumes of uranium carbide arc melted and cast by skull techniques.

Efforts to determine the effects of different grades of graphite upon control of composition are being continued. A new water-cooled radiation shield has been installed in the skull furnace, and several melts have been made using vacuum-outgassed commercial graphite electrode tips and charge additions. Splashing of the UC melt and resulting tip erosion were less by visual observation with the outgassed commercial graphite than with untreated graphite. The castings produced in these melts were sound and had a good surface. Several more melts using vacuum-outgassed graphite will be made, and the carbon analyses of the castings and the weight lost by each electrode will be considered in evaluating the control of composition produced with this graphite.

Two pounds of plus 16-mesh uranium carbide shot were received from the Vitro Corporation. Chemical analysis of this shot showed 4.78 w/o carbon and 0.061 w/o nitrogen. Spectrographic analysis of the shot showed <0.05 ppm beryllium, 1 ppm boron, <10 ppm calcium, <1 ppm cobalt, 2 ppm copper, 15 ppm iron, <1 ppm magnesium, 4 ppm molybdenum, 8 ppm nickel, 50 ppm silicon, and <100 ppm tungsten. Vacuum-fusion gas analysis of the shot showed 188 ppm oxygen and 7 ppm hydrogen. Metallographic examination of the shot showed variations from one piece of shot to another from approximately 4 to 6 w/o carbon. There was some segregation within certain pieces of the shot, usually in the form of small areas containing large amounts of free uranium. No UO₂ was visible in the shot. This product has considerable promise as a charge material for skull-type arc melting and casting. Castings prepared from this material will be made in the near future.

Research next month will be continued on defining the merits of different grades of graphite and their effect on control of composition of skull-melted and cast uranium carbide. AGOT and TSF reactor grade graphite will be studied by observing carbon-content fluctuations of the cast UC prepared with these different graphites under the same melting conditions.

Mechanical and Physical Properties of Uranium Monocarbide

G. G. Chesmar, R. W. Endebrook, E. L. Foster, and R. F. Dickerson

Of primary interest in this research program are the strength, hardness, and thermal conductivity of uranium carbides at elevated temperatures. Further, the effects of alloy additions, coatings, and heat treatment on these properties are of considerable interest.

A UC-20 w/o vanadium alloy has been prepared. The alloy was examined by metallography and by X-ray diffraction, and the phases found were uranium monocarbide, uranium, and vanadium. These findings are in conflict with the existing phase diagram as developed from previous data and published in BMI-1441. The data show that if carbon is introduced into the uranium-vanadium system the uranium has a greater affinity for carbon than the vanadium.

Determinations of the high-temperature properties of uranium-carbon alloys are being initiated. Preparations are being made to obtain high-temperature thermal-conductivity and hardness measurements on uranium monocarbide. A thermal-conductivity specimen (3 in. in diameter by 1 in. thick) is required. Initial attempts to cast such a specimen resulted in a casting containing transverse cracks. Further attempts will be made. Since specimen soundness is a prerequisite in the determination of thermal conductivity, procedures of casting and molding are to be varied during November in order to find a means of producing a sound specimen. Because thermal shock is a source of specimen imperfection, heated molds are to be utilized as a means for reducing shock during casting.

In addition, specimens will be prepared for hot-hardness determinations. Plans are also being made to obtain thermal-expansion and -resistivity measurements in the 1000 to 1600 C temperature range.

Uranium Monocarbide Diffusion Studies

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

Gross gamma counting is being used to study the self-diffusion rate of uranium in uranium monocarbide. Diffusion rates in uranium monocarbide are of interest in predicting the elevated-temperature properties of this material. Previously in this study the rate of interdiffusion of carbon and uranium was determined between 1600 and 2000 C using graphite and liquid uranium saturated with carbon. Currently, the rate of self-diffusion of uranium in uranium monocarbide is being studied using a diffusion couple consisting of a foil of enriched uranium sandwiched between two sections of depleted uranium monocarbide rod, and a technique using carbon-14 is being considered from an experimental point of view for studying the rate of self-diffusion of carbon in uranium monocarbide.

A diffusion couple annealed for 3840 min at 1700 C was analyzed for enriched uranium using gross gamma counting of the 0.184-Mev gamma ray. The diffusion coefficient for the self-diffusion of uranium in this couple was calculated as 6.8×10^{-11} cm² per sec. A preliminary calculation of the activation energy for self-diffusion of

uranium in uranium monocarbide was found to be 82,000 cal per mole using the available data. An additional couple is being prepared for annealing for 480 min at 2000 C.

A technique has been devised for studying the rate of self-diffusion of carbon in uranium monocarbide. It is planned to use a diffusion couple consisting of two cylinders of uranium monocarbide bonded together by placing a 0.001-in.-thick foil of uranium between them and heating 4 hr at 1400 C. One cylinder would be enriched in carbon-14. It would have a specific carbon-14 activity of 80 to 100 μc per g, which is about 1000 times the minimum detectable activity, thus providing a means of analyzing a decreasing activity gradient across a diffusion zone produced during annealing. It is planned to determine the rate of self-diffusion of carbon in the temperature range of 1600 to 2000 C for times sufficient to produce 0.025 to 0.030 in. of diffusion zone. The couple will be prepared for analysis by grinding away successive portions of the couple, starting at the natural (carbon-12) end of the couple. The couple will be shortened by grinding away 0.0005 to 0.0009-in. increments of the length of the couple. Measurement of the beta activity of the carbon-14 will be made on the exposed face of the couple after each slice is removed. The beta activity of carbon-14 has a range of 0.0009 in. in uranium monocarbide, with 90 per cent of the measured activity being emitted by the material in a layer within 0.0005 in. of the surface. Carbon-14 for conducting this study of the rate of self-diffusion of carbon in uranium monocarbide has been ordered.

Future work will include the analysis of the couple being prepared for annealing at 2000 C and the initiation of the study to determine the rate of self-diffusion of carbon in uranium monocarbide using carbon-14 as a tracer.

Effects of Radiation on UC

D. G. Freas, A. E. Austin, J. E. Gates,
C. M. Schwartz, and R. F. Dickerson

Natural-Uranium Studies

An investigation of radiation-induced changes in the structural and physical properties of UC composed of natural uranium with about 5.0 w/o carbon has been completed. The specimens were irradiated in two NaK-filled capsules in the BRR to burnups of 0.01 and 0.03 a/o of the uranium at surface temperatures of less than 500 F.

The changes in dimensional properties of these specimens were found to be small, and only slight decreases in density were measured. The electrical resistivity increased by almost a factor of three after irradiation to both 0.01 and 0.03 a/o burnup of the uranium. Since the resistivity values did not show a hyperbolic-type increase with increasing burnup, it was thought that saturation of displaced interstitial atoms and vacancies in the lattice was attained during irradiation. X-ray diffraction studies of the crystal lattice of specimens from both burnup levels also indicated that saturation of displaced interstitial atoms and vacancies had occurred. Metallographic examinations of specimens from both levels of burnup failed to reveal any structural changes. The electron-microscopy examination of a specimen with 0.01 a/o burnup of the uranium showed no detectable changes as compared with an unirradiated control specimen. The specimen with 0.03 a/o burnup was not examined. No further research with the UC specimens composed of natural uranium is presently planned.

Enriched Uranium Studies

A study of the radiation stability of three types of 10 per cent enriched uranium-carbon fuel materials is continuing. Specimens of uranium-5.0 w/o carbon, uranium-6.7 w/o carbon, and uranium-8.5 w/o carbon alloys were encapsulated in four NaK-filled capsules and inserted in the BRR during the shutdown of September 19 and 20, 1960. During the first reactor cycle, the neutron flux was found to be approximately 20 per cent lower than had been anticipated, causing the specimen temperatures to be several hundred degrees lower than desired. However, a mixture of inert gases has been incorporated into the outer gas annuli of the capsules thus raising the specimen temperatures to a more desirable level. Although the design specimen temperatures (1400 F) may be attained, the intended burnup levels will probably not be achieved in the allotted time.

PHYSICAL RESEARCH (AEC-DR)

F. A. Rough

The study of growth of UO_2 single crystals continued.

Growth of UO_2 Single Crystals

J. D. Jackson and C. A. Alexander

During the past month, construction of the tungsten resistance furnace for growing of UO_2 single crystals was completed, and temperature corrections for eye ports were determined.

It was found that a temperature of 2750 C can be achieved and maintained within the resistance element when it is surrounded by successive tungsten, tantalum, and molybdenum shields.

Experiments in the new furnace were carried out with sintered UO_2 pellets with an oxygen/uranium ratio less than 2.01. In one run, the UO_2 charge was contained in a tantalum crucible which was supported by a 5-mil tungsten cylinder. After holding the charge at an indicated temperature of 2550 C in a pressurized selenium atmosphere for about 15 min, the crucible was lowered out of the hot zone. Upon cooling it was found that complete melting did not take place in the time interval and that a small hole had formed in the tantalum crucible and some molten UO_2 had flowed down between the crucible and the tungsten support. No evidence of attack was seen on the tungsten. Examination of the frozen UO_2 indicated that crystalline grains about a millimeter in size had grown, but melting occurred mainly on the UO_2 surface.

In another experiment, a tungsten crucible was substituted for the tantalum. A higher temperature, between 2600 and 2700 C, was maintained for 1/4 hr. However, only limited surface melting was observed.

It may be that higher temperatures or a longer time at temperature is necessary to maintain liquid-phase UO_2 . The present experiments were terminated because of overheated power leads. When this limitation is corrected, higher temperatures or longer runs will be possible.

RADIOISOTOPE AND RADIATION APPLICATIONS (AEC-OID)

D. N. Sunderman

Research is continuing in the areas of intrinsic-tracer applications to process control and the effect of structural factors upon radiation-induced graft polymerization.

A series of dynamic experiments on the process-model system shows the high sensitivity of the intrinsic-tracer concept in indicating changes in operating conditions. The system has been used to demonstrate radiotracer control of iron removal from a nickel-refinery stream. Momentary changes in precipitation and filtration efficiency give unequivocal indications at the effluent monitor as soon as the iron-rich material enters it. Changes which would not receive notice in intermittent sampling are very important to quality control of the nickel product, and the intrinsic tracer technique can detect such changes in time for corrective action to be taken. The dynamic experiments complete the study of the iron-removal step. Further experimentation will be directed toward an evaluation of other precipitation systems with physical characteristics differing markedly from the iron hydroxide system.

Further irradiation experiments with high-molecular-weight PMMA indicate that the effect of molecular weight upon active-site formation may have been the result of incomplete monomer removal. The higher EPR spectral resolution for high-molecular-weight material is still observed with monomer-free material. Preliminary work with polymethyl acrylate indicates a much higher threshold dose for obtaining measurable concentrations of active sites. The use of iodine as a free-radical scavenger will be studied in the next few weeks.

Use of Intrinsic Radioactive Tracers for Process Control

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

During October, the final phase of the program to demonstrate radiotracer control of iron removal from a nickel-refinery stream was completed.

The emphasis during the month was on carrying out dynamic experiments with the process-model system. During each experimental run, various changes were made in process operating conditions to simulate accidental occurrences which might take place under plant conditions. These deliberate changes consisted of (1) suddenly reducing the ammonia input to the reactor, (2) suddenly opening one of the filter-control valves, or (3) deliberately introducing some feed solution into the filtrate.

The effect of some of these changes on two of the experimental runs is shown by the data in Table H-1. The effect of reducing the ammonia input to the iron precipitator increases the iron content of the filtrate, but some time elapses before this effect is noticeable. On the other hand, the effect of suddenly increasing the slurry input to the filter is almost immediately detected by the radioassay system, as would be expected. The factor determining the response time of the radioassay system to some change in process conditions is just the time it takes for material at the point of change in the process to travel to the detector.

TABLE H-1. DATA FROM EXPERIMENTS WITH THE PROCESS-MODEL SYSTEM FOR IRON REMOVAL

Iron Analysis of Feed Solution, g per liter	Count Rate in Feed Solution, cpm	pH of Iron Precipitation Reactor	Time Since Start of Run, min	Changes in Process Conditions	Count Rate in Filtrate, cpm $\pm \sigma$	Radioactivity Reduction Ratio	Iron Reduction Ratio Shown By Analysis
Experiment 1							
0.536	49,000 $\pm$ 1,000	4.5	120	--	410 $\pm$ 12	119 $\pm$ 4	97.5 $\pm$ 2
		4.5	150	--	315 $\pm$ 10	155 $\pm$ 5	141 $\pm$ 4
		4.5	165	--	440 $\pm$ 12	111 $\pm$ 3	105 $\pm$ 4
		4.5	195	Ammonia input reduced	280 $\pm$ 10	174 $\pm$ 6	212 $\pm$ 8
		4.0	210	--	325 $\pm$ 11	150 $\pm$ 5	--
		4.0	225	--	395 $\pm$ 11	124 $\pm$ 4	122 $\pm$ 5
		4.0	270	--	500 $\pm$ 12	97 $\pm$ 2	93 $\pm$ 3
		4.0	285	--	545 $\pm$ 13	90 $\pm$ 2	--
		4.0	315	Slurry valve into filter opened full	585 $\pm$ 13	84 $\pm$ 2	--
		4.0	318	--	1125 $\pm$ 24	43.5 $\pm$ 1	48.3 $\pm$ 1.3
		4.0	322	--	1705 $\pm$ 25	28.7 $\pm$ 0.4	31.3 $\pm$ 0.4
		4.0	326	--	1765 $\pm$ 25	27.7 $\pm$ 0.4	29.9 $\pm$ 0.5
		4.0	330	--	1930 $\pm$ 27	25.3 $\pm$ 0.3	24.8 $\pm$ 0.6
		4.0	335	Slurry valve closed	1810 $\pm$ 26	27.0 $\pm$ 0.4	--
		4.0	345	--	1240 $\pm$ 24	39.6 $\pm$ 0.7	43.5 $\pm$ 1.1
Experiment 2							
0.778	84,000 $\pm$ 1,500	5.0	330	--	725 $\pm$ 33	115 $\pm$ 5	150 $\pm$ 5
		5.0	345	--	700 $\pm$ 32	120 $\pm$ 5	--
		5.0	346	Feed-solution bypass valve opened	700 $\pm$ 32	--	--
		5.0	348	Valve closed	5450 $\pm$ 90	15.4 $\pm$ 0.2	15.3 $\pm$ 0.1
		5.0	350	--	3850 $\pm$ 75	21.8 $\pm$ 0.4	20.6 $\pm$ 0.2
		5.0	361	--	825 $\pm$ 35	102 $\pm$ 4	105 $\pm$ 5

The second set of data shown in Table H-1 was taken toward the end of another experimental run. In this case the bypass valve on the feed line was opened for 1 min, introducing feed solution directly into the filtrate stream. Within 1 min after opening this valve the activity count in the filtrate had jumped by a factor of about four, and within 2 min the activity was eight times its original value, after which it dropped off rapidly.

It is to be noted that within 15 min after the bypass valve was momentarily opened, the filtrate activity had returned to almost the same level as before the incident. It is clear that in a case such as this, a routine analysis of the stream conducted every 30 min could have missed the incident entirely, whereas the radioassay system showed almost immediately and quite precisely the amount of excess iron introduced into the filtrate.

Very good agreement was found between the reduction ratios given by the radioassay system and those given by iron analysis of filtrate samples. In most cases the two methods agreed to within 15 per cent and differed by only 25 to 30 per cent in the few extreme cases.

Future work will consist of demonstrating that the intrinsic radiotracer control of filtration-purification is applicable to a wide variety of different systems. Work next month will consist of delineating typical processes useful for demonstration purposes and then beginning an experimental demonstration of one of the process applications. It is expected that the process-model system constructed to study iron removal, as well as the techniques developed in that study, will be employed in these future experiments with little or no modification.

Graft-Polymerization Studies

F. A. Sliemers, W. B. Gager,
J. F. Kircher, and R. I. Leininger

During September, evidence had been obtained which indicated that the efficiency of site formation in PMMA decreases with increasing molecular weight in the high-molecular-weight (2 to 4×10^5) range. Further, these high-molecular-weight polymers gave EPR spectra having much better resolution than had been obtained with low-molecular-weight ($< 2 \times 10^4$) PMMA. Based on earlier work, these findings suggested the possibility that the monomer had not been removed completely from the polymers.

To determine if monomer doping actually occurred, several additional samples of these polymers were heated to 100°C in vacuum, irradiated, and subjected to EPR measurements. These later results indicated that the observed molecular-weight effect may have been caused by monomer doping. The EPR spectral resolution, however, appeared to be a true function of molecular weight. Additional work is planned in this area.

An investigation of the influence of total dose on site formation for polymethylacrylate (PMA) has been initiated. Samples have been irradiated for total doses in the range 3×10^5 to 3×10^7 rads. The EPR spectrum associated with this polymer is quite different from that observed with the corresponding methacrylate polymer. Moreover, it appears that much higher total doses will be required to obtain measurable numbers of

H-4

free-radical sites. No indication of the total number of sites was obtainable below approximately 10^7 rads.

During November, the work described above will be continued. In addition, the work will include experiments in which iodine will be used as a free-radical scavenger in an attempt to further clarify the mechanism of free-radical formation.

ASSISTANCE TO AEC-EURATOM PROGRAM

H. M. Epstein and D. L. Keller

Studies concerned with obtaining information on void-content and heat-transfer relationships in forced-convection subcooled boiling water and with the development of uranium mononitride as a possible fuel material are under way for the Joint US-Euratom Research and Development Board. Both programs are directed toward obtaining data that will be useful in efforts to achieve more economical nuclear power.

VOID-DISTRIBUTION AND HEAT-TRANSFER STUDIES

R. O. Wooton, F. G. Peter, J. Foglia,
H. M. Epstein, and J. W. Chastain

This program is being conducted to investigate void distributions and slip ratios over a range of conditions of interest in light-water reactors.

During the past month, additional experimental data were obtained to investigate an unusual variation or "hump" in the slip ratio-versus-pressure curve at pressures around 900 psi. In this work the pressure was varied in 20-psi steps between 850 psi and 1050 psi while the mass flow and heat flux were held constant. The slip ratios were calculated from the new data and compared with the previous results. In general, the experimental evidence indicates that the slip ratio is not a simple function of the pressure over much of the range studied; rather, it exhibits an instability of ± 25 per cent. The existence of a hump in the slip-ratio curve at the high-pressure end of the unstable region has been tentatively confirmed. However, further study is necessary to determine if the slip ratio is actually a smooth function of the pressure outside this range.

Investigation of the Bankoff* flow parameter, K , has continued. Data taken in the past month yield values of K within ± 5 per cent of a correlation of previous data. Values of K were also calculated for data taken by Egen, et al.** at 2000 psi. These high-pressure runs gave K values slightly less than one, which substantiates the pressure dependence of K predicted by the Russian workers*.

A correlation of void fractions in the subcooled boiling region, developed by Griffith, et al.***, is being studied.

*Novak, Z., "On the Variable-Density Single-Fluid Model for Two-Phase Flow", J. Heat Transfer, p 255 (August, 1960).

**Egen, R. A., Dingee, D. A., and Chastain, J. W., "Vapor Formation and Behavior in Boiling Heat Transfer", BMI-1163 (February 4, 1957).

***Griffith, P., Clark, J. A., and Rohsenow, W. M., "Void Volumes in Subcooled Boiling Systems", ASME Paper No. 58-HT-19 (March 27, 1958).

DEVELOPMENT OF URANIUM MONONITRIDE

D. L. Keller

In the program for the development of uranium mononitride several methods of preparing high-density bodies suitable for irradiation tests and mechanical-property tests are being studied. In vacuum-sintering studies at 2000 to 2100 C there were significant losses of nitrogen from UN, while at 1800 C little densification of the coarse UN particles occurred. Hot-pressing studies carried out in steel tubes at 1290 C produced UN bodies of 92 per cent of theoretical density.

In the melting program high-pressure furnace equipment is being readied for experimental work. In the meantime experiments are being conducted to determine the time required to complete the reaction between liquid uranium and nitrogen maintained at 3 atm.

Powder-Metallurgy Studies

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

In order to provide high-density UN bodies for irradiation testing, the sintering and hot pressing of UN powders are being investigated.

As reported last month (BMI-1469), approximately 1.5 kg of UN powder with an average particle size of 66μ was prepared by the reaction of uranium metal and nitrogen. Nitrogen analysis by a modified Kjeldahl method of a representative sample of this UN powder showed a nitrogen content of 5.47 ± 0.03 w/o. Uranium mononitride powder is now being prepared from uranium hydride to obtain a finer particle size.

Of the various binders employed for green pressing, a saturated solution of Carbowax 6000 in methyl alcohol has given the best results. A preliminary evaluation of the corrosion effects of various solvents failed to yield significant data.

Initial attempts to vacuum sinter this relatively coarse UN powder at temperatures up to 1800 C were unsuccessful because of a lack of densification. At higher temperatures, 2000 to 2100 C, melting was observed. Metallographic examination of the specimens revealed particles of UN surrounded by a continuous matrix of free uranium. Thus the decomposition pressure of nitrogen above UN becomes significant at the temperatures which may be required for sintering. Calculations with available thermodynamic data are of little aid in establishing the true equilibrium pressures because of inaccuracies in the known data. A series of experiments will be run at various temperatures and partial pressures of nitrogen in an attempt to effect the sintering of UN powder without decomposition.

Two of these dense compacts of UN particles dispersed in a uranium matrix were heated at 1300 C under 1 atm of nitrogen for 1 and 6 hr. Metallographic examination of the specimen heated for 1 hr showed about a 5-mil penetration of the surface with a light blue-gray phase, probably a higher nitride of uranium. The specimen heated for 6 hr

showed much deeper penetration, about 40 mils. It had also lost its structural integrity; numerous cracks were surrounded by this higher nitride phase, indicating continuity of the cracks with the surface.

Control of the nitrogen content is strongly dependent on control of nitrogen pressure above the UN. Attempts will be made to ascertain the pressure-temperature relationship and to determine if the pressure range is so small and critical as to forestall practical control.

With regard to the direct sintering of higher nitrides in the hope of achieving an activated sintering process similar to the sintering of uranium from uranium hydride, as well as a reduction in the cost of powder preparation, a mixture of 10.3 g of U_2N_3 and 4.7 g of uranium powder prepared by the decomposition of UH_3 was pressed at 50 tsi and heated under vacuum to 1475 C for 1 hr. The specimen was powdery and fragile, and little or no sintering occurred. Since temperatures in excess of the melting point of uranium were achieved with no visible evidence of melting, diffusion of nitrogen from the higher nitride into the 20 to 21 volume per cent of free uranium powder originally present in the compact probably occurred. It was not possible to show this metallographically because of the high porosity and fragileness of the specimen.

Specimens of U_2N_3 have been pressed at 50 tsi and slowly raised to 1700 C while evacuating without break-up of the specimens. After 1/2 hr at 1700 C, they appeared to be more sound than UN powder sintered at 1800 C for 2 hr. These specimens were quite porous and good metallographic results could not be obtained.

In another fabrication technique, a series of cold-compacted uranium nitride cores has been isostatically hot pressed at 1290 C to yield densities of 84 to 92 per cent of theoretical based on a theoretical density of 14.32 g per cm^3 for uranium mononitride. The variable in this series was particle size, with various mesh fractions from minus 20 to minus 325 mesh being used. The higher densities were observed with a particle size less than 175 μ or 100 mesh. The green cores were sealed in evacuated stainless steel tubes after removing the zinc stearate binder and then hot pressed under a 5-tsi pressure for 3 hr at 1290 C. Densities were determined by measurements from core X-rays. The powder used was considered to be of a quality inferior to that now being accumulated; however, metallographic examination of cross sections through these cores showed, in most cases, a predominantly one-phase structure, namely UN. No extraneous grain-boundary phases such as uranium were present, although variable quantities of a non-metallic phase, possibly a higher nitride, were observed. The quantities of this phase were estimated to vary from less than 1 volume per cent to 5 volume per cent in the different cores. This phase was present as widely separated discrete particles approximately 70 μ in diameter.

A solid bond was consistently observed between the nitride core and the stainless cladding; however, this bonding was accompanied by a 30- μ reaction zone.

It is planned to use the high-quality UN powder now available to pinpoint optimum mesh sizes and hot-pressing conditions. Temperatures up to 1400 C will be employed in conjunction with containers more refractory than stainless steel.

Melting and Casting

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

Interest in uranium mononitride as a potential high-integrity reactor fuel has led to a research program to investigate feasible methods of preparation. The portion of the program reported here deals with melting and casting techniques, and has for its main objective the development of procedures for the preparation of high-density bodies suitable for mechanical- and physical-property and irradiation tests.

A cursory investigation of the thermochemistry of the reaction of uranium with nitrogen in an arc furnace indicated the need for a pressure furnace capable of operation at several atmospheres of nitrogen pressure. In order to obtain more reliable data pertinent to the preparation of UN, certain facets of the melting operation are to be investigated in existing equipment during the period that high-pressure equipment is being readied. Experiments are presently being conducted to determine the time necessary to complete the reaction at an arbitrarily chosen pressure of 3 atm (abs). Purified nitrogen is being used (less than 50 ppm oxygen) and pressures are being maintained at 3 atm manually throughout melting and cooling cycles. The melts are to be held molten for 4, 6, 8, 10, and 12 min, and, for uniformity of procedure, each melt will be turned upside down once after one-half of the allotted reaction time. Arc conditions of amperage and voltage are to be roughly the same for each melt. Buttons produced in this experiment are to be analyzed for nitrogen and for impurities such as tungsten (from the electrode) and copper (from the hearth).

Buttons which have already been produced have the appearance of gray iron. They are brittle, but not nearly so brittle as comparable, stoichiometric UC buttons. Yttrium-treated tungsten electrodes showed variable erosion, and definite gains in weight after the melting operation. A melting test is scheduled to compare weight-gain and erosion characteristics of an untreated tungsten electrode.

After the results of the reaction-time experiment have been evaluated, a second experiment is planned in which pressure will be the variable. Melts are to be reacted with purified nitrogen at pressures varying from 1/4 to 3 atm (abs). Materials obtained will be compared with those produced by melting stoichiometric UN (prepared by powder techniques) under like nitrogen pressures and melting times. Materials generated are to be examined and compared for nitrogen and impurity contents.

MATERIALS DEVELOPMENT AND EVALUATION (HAPO)

F. R. Shober

The electrical and thermal properties of UO_2 are being measured before and after irradiation. In an investigation of the creep properties of Zircaloy-2 at temperatures from 290 to 400 C, tests on cold-worked material and on annealed materials are continuing under several dynamic test conditions.

A program directed toward the development of a metallic container for the fused chloride-electrolytic fuel-recovery process was evaluated by corrosion tests on approximately 30 metals and alloys.

Thermal Conductivity and Electrical Conductivity of UO_2

C. F. Lucks and H. W. Deem

Measurements have been completed on an unirradiated UO_2 disk specimen identified as Fabrication Specimen A. These measurements have not yet been reduced for reporting. During October no work was done on the vibrationally compacted UO_2 powder specimen.

Difficulties have been experienced with the specimen-measuring thermocouples in the hot-cell thermal-conductivity apparatus. The thermocouples have been changed and measurements are in progress on a second specimen of the second burnup series. The first specimen of this set showed a larger scatter in the data than expected. Measurements in progress should show if the apparatus is operating satisfactorily.

Mechanical Properties of Zirconium Alloys

J. A. VanECHO and L. P. Rice

This is a long-range program of determining the creep properties of Zircaloy-2 sheet in the temperature range of 550 to 750 F. Tests extending beyond 25,000 hr are being performed on 15 per cent cold-worked material and approaching 15,000 hr on annealed material. The effects of a temperature cycle alone and of a temperature and stress cycle combined on the creep strength of Zircaloy-2 sheet are also being investigated.

Twenty creep machines are being utilized on this program with 20 tests now in progress. The September report (BMI-1469) contains a summary of all tests now in progress. Since most of the tests are of a long-time nature, there is actually very little change occurring from one month to the next. Therefore, the status of the various creep tests is essentially the same as reported for September.

To review the over-all program, a stress of 37,500 psi at 550 F on the 15 per cent cold-worked material resulted in failure in 10,882 hr. A stress of 35,000 psi on the

same material resulted in a test that was discontinued in 15,000 hr without reaching the third stage of creep.

At 650 F, in a 30,000-psi test on the 15 per cent cold-worked material, failure occurred in approximately 7300 hr. A similar test at 15,000 psi is still in progress after about 25,000 hr with no indication of imminent failure.

In the annealed material, the difference between a stress that will cause early failure and one that will result in a long-time creep test is less than in the cold-worked material. This effect is more evident at the lower temperature of 550 F than it is at 750 F. At 550 F, a stress of 25,000 psi causes failure in about 0.3 hr; but a slightly lower stress of 22,500 psi results in a test with a very low creep rate (about 0.000006 per cent per hr) that is still in progress after 13,500 hr.

At 650 F, a stress of 22,500 psi caused the failure of a specimen in 7184 hr. At a lower stress, 20,000 psi, a test has been in progress for 11,000 hr but with failure imminent. At 750 F, failures have occurred in 48.7, 1035.7, and 3663.5 hr, and a test at 12,500 psi, although in progress for 11,000 hr, should produce failure shortly.

The effect of the temperature cycle on both the cold-worked and the annealed Zircaloy-2 sheet seems to be to accelerate the creep rate. A combined stress and temperature cycle appears to produce a greater creep rate than no cycle but a lower creep rate than temperature cycle alone. The combined stress and temperature cycle is imposed only on annealed material. This cycle consists of reducing the temperature to 80 F 1 day per week and at the same time reducing the stress to zero.

Materials of Construction for the Fused Chloride-Electrolytic UO₂ Process

E. L. White, P. D. Miller, C. L. Peterson, and W. K. Boyd

This research program is being directed toward developing a metallic container for fused chloride-electrolytic fuel-recovery process.

Coupons of various candidate metals and alloys are suspended in Vycor tubes holding an equimolar melt of sodium and potassium chlorides at 750 to 800 C. The molten salt containing U₃O₈ is first sparged with chlorine and oxygen for 4 hr. Then a sparge of chlorine only is used for an additional 4 hr.

To date, about 30 different metals and alloys have been evaluated. None has shown rates of attack of less than 1 in. per yr. Accelerated attack is found at interface areas and at locations adjacent to the sparge tube. Platinum, Hastelloy D, Hastelloy B, and Alumel have shown the best resistance.

Evaluations of different alloys, selected to illustrate the effects of various components, are being continued.

REFINING THE GRAIN SIZE OF URANIUM
BY VIBRATIONAL ENERGY (SRL)

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

Vibrating energy - sonic and ultrasonic - has proved useful in producing relatively fine-grained structures in cast metals and has been investigated to a limited extent as a means of accelerating processes and altering the structure in materials in the solid state. Although it is possible to effect grain refinement by the heat treatment of alpha-worked uranium, the desired core-to-cladding interface and uniform cladding thickness are quite often unattainable because of the large grain size or the preferred orientation of the structure. It is believed that the use of vibratory energy in conjunction with the heat treatments may result in a smaller grain size in which preferred orientation is substantially decreased or eliminated. Therefore, a program of research concerned with the application of vibratory energy as a means of attaining the randomly oriented fine-grained structure necessary in uranium coextrusion blanks has been initiated.

Twenty-four specimens of unalloyed uranium hot alpha rolled and then gamma annealed have been heat treated at temperatures from about 30 C above to 10 C below the alpha-to-beta transformation temperature for periods of time up to 20 min. Alpha-rolled specimens have also been heat treated at 550 C for up to 20 min. Each heat treatment has been conducted both with and without the application of ultrasonic energy. In those cases where ultrasonic energy has been utilized, a 20-kc 2.5-kw energy source and a 10-kc 2.5-kw energy source have been employed in conjunction with a laminated nickel transducer coupled directly to the uranium specimens. Metallographic examination of these specimens is in progress.

Future work will be concerned with the completion of eight additional heat treatments utilizing the 10-kc vibratory power source and with the metallographic examination of the specimens generated during the course of the program. The evaluation will be concerned with the relative grain sizes and the uniformity of the grain size exhibited by the ultrasonically treated specimens when compared with similar specimens heat treated without the use of vibratory energy.

COATED-PARTICLE FUEL MATERIALS (AEC-DRD)

R. W. Dayton

A program on the development of coated-particle fuel concepts was initiated during the month of October. Practical methods have been demonstrated for coating small particles of fissile or fertile materials with various thin, impervious refractory coatings. Irradiation studies of a graphite body containing 100- μ uranium dioxide particles coated with 40 μ of alumina have shown that such a fuel has good fission-product-retention properties. The purpose of this new development program is to provide fundamental data on various coated-particle systems of possible interest, including Al_2O_3 , BeO , carbon, and others.

Comprehensive programs in several areas of research were defined during the month of October, and experimental studies of some phases of the work have been begun.

A study to develop abrasion-resistant coatings for UO_2 particles as a possible fuel material for the Fluidized-Bed Reactor is also under way.

Evaluation of Commercial Products

T. S. Elleman and D. N. Sunderman

A program for the testing and evaluation of coated particles submitted by commercial manufacturers is being prepared. When complete, this program will be submitted to interested manufacturers. As presently visualized, the tests will be intended to demonstrate coating integrity, contamination, and strength.

Particle-Coating Studies

M. F. Browning and J. M. Blocher, Jr.

Chemical vapor-deposition reactions have been shown to give refractory coatings on nuclear-fuel particles having good density and fission-product retention. Such characteristics have been found for alumina coatings on UO_2 and pyrolytic carbon coatings on UC_2 .

Considerable information has been obtained on the application of these coatings in a fluidized bed of the fuel particles. However, only a limited understanding of the effects of coating variables on the properties of the coatings has been developed. To obtain such an understanding will be the purpose of this program. This will lead to optimization of the coating properties and will permit a better estimate to be made of the economics of the coating process.

It is anticipated that research will be done initially with alumina, beryllia, and pyrolytic carbon coatings. Alumina and beryllia can be conveniently obtained by hydrolysis of the respective chlorides:

Pyrolytic carbon can be obtained by the pyrolysis of a hydrocarbon such as acetylene:

Prior work has indicated that the temperature of coating formation is an important factor in determining the properties of the coating. This variable will be the first to be studied. Other variables, including (1) the choice of reaction, (2) reactant concentration, (3) gas-flow rate, and (4) fluidized-bed inventory, will be studied to the degree that they are shown to be significant. However, before this program is initiated, minor equipment modifications to allow improvement in the alumina and beryllia coatings will be investigated.

Attention will be directed immediately to eliminating the formation of, or at least the effects of, finely divided dust particles resulting from homogeneous reaction away from the surface of the coated particles, or incomplete hydrolysis. This material can plug the fluidized-bed reactor and terminate a run, making it necessary to apply thicker coatings stepwise, with possible incidence of interlayer voids. Particles of dust can be fed back into the coatings, resulting in islands of porous material. Particularly troublesome in the beryllia coating preparation is the deposition of partially coated particles in the layer of loosely sintered dust collected above the fluidized bed.

Determination of the properties of the coated particles prepared during processing evaluation will be done primarily by the following techniques: metallography (light and electron microscopy), testing of oxidation resistance (where applicable), determination of resistance to leaching in hot nitric acid, measurement of alpha emission, compressive-strength determination, testing of resistance to thermal cycling, and chemical analysis. It is expected that these more routine procedures will be supplemented, where indicated, by examinations by X-ray diffraction and electron-probe X-ray analysis techniques, and by evaluations involving neutron activation with postirradiation heating.

Ceramic-Matrix Studies

M. C. Brockway and W. H. Duckworth

Initial irradiation studies will be concerned with both matrix-supported and unsupported coated fuel particles. The initial matrix material will be a synthetic graphite. The performance of the graphite-matrix fuel element will, in addition to other factors, depend on the properties of the specific graphite matrix and the effect of dispersed fuel on matrix properties. Matrix properties of interest include stability to irradiation at elevated temperatures, thermal conductivity, density, strength, fabricability, and impermeability.

Current graphite technology will be used to select starting materials and fabrication conditions having greatest promise of producing good dispersions of coated fuel particles in graphite. Experiments will be made, first on unfueled material, and then on material containing fuel, to demonstrate that the selection of materials and conditions is proper for producing a strong fuel material with a uniform dispersion of fuel, and

without cracking the fuel particles. When the conditions for preparing representative bodies of suitable quality have been shown to be satisfactory, selected dispersion bodies will be evaluated for use in the irradiation program.

Fuel-Irradiation Studies

W. H. Goldthwaite

Irradiations of UO_2 particles coated with $40\ \mu$ of Al_2O_3 and dispersed in 1-1/2-in.-diameter graphite spheres have indicated that the Al_2O_3 coatings are effective in minimizing the release of short- and long-lived gaseous fission products. Changes in the release characteristics have occurred with increasing burnup, but over-all the results have been encouraging. On the basis of the data already available on these Al_2O_3 -coated particles, it is possible to select important parameters for study in the next series of irradiation experiments.

It is believed that, at temperatures below the limiting temperatures set by the Al_2O_3 -graphite reactions, release of fission products through an undamaged $40\text{-}\mu$ Al_2O_3 coating by diffusion or permeability will be negligible. However, if the coatings fracture or are otherwise damaged, release of fission products may be high. Therefore, it is reasonable to expect that the greatest immediate improvement in the performance of Al_2O_3 -coated particles can be accomplished by preventing fracture of the coatings.

An irradiation program has been planned to study the importance of porosity and matrix support on the integrity of $40\text{-}\mu$ coatings of Al_2O_3 on 100 to $200\text{-}\mu$ UO_2 . Twenty-four specimens will be irradiated in sealed capsules and subsequently examined and evaluated by hot-cell techniques. The 24 specimens will include all combinations of the following variables ($3 \times 2 \times 2 \times 2$):

A. Porosity

- (1) Porous UO_2 and dense Al_2O_3 with $10\text{-}\mu$ porous layer adjacent to UO_2 surface
- (2) Porous UO_2 and dense Al_2O_3
- (3) Dense UO_2 and dense Al_2O_3

B. Matrix Support

- (1) Particles in graphite matrix
- (2) Particles unsupported

C. Temperature

- (1) 300 F
- (2) 1800 to 2000 F

D. Burnup

- (1) 20 to 25,000 MWD/TONNE
- (2) 40 to 50,000 MWD/TONNE.

The hot-cell evaluations will include visual and metallographic examination of particles and matrix for temperature- and radiation-induced changes in integrity, structure, and dimensions, the mobility of nonvolatile fission products and uranium, and reactions between the Al_2O_3 and the graphite. Oxidation or acid leaching of bare particles may be used to detect small flaws in the coatings. Heat treatments, during which the fission products are swept by an inert gas to activated charcoal traps for analysis, will be employed to study the diffusion and permeability characteristics of the coatings after radiation.

The specimen type which appears, from postirradiation examinations, to have performed best at the two temperatures will be studied further in sweep-capsule irradiations to high burnup at several different temperatures. Sweep-capsule irradiations permit determinations of fission-product release during the irradiation. The specimen surface is swept continuously during the irradiation with inert gas. Sampling of the inert gas stream at frequent intervals provides direct information on the release of long-lived gaseous fission products. At less frequent intervals, the stream can be diverted to stainless steel-mesh-filled traps located next to the capsule. Nonvolatile long-lived daughter products of short-lived volatile fission products which are released from the specimen decay and plate out on the stainless steel mesh as they are formed. Radiochemical analysis of the mesh from different locations in the trap indicates the release rate of the short-lived gaseous precursors, giving information pertinent to reactor-system contamination by the daughters and further insight into the mechanism of release from the specimen.

Fission-Gas-Release Studies

T. S. Elleman and D. N. Sunderman

A program is currently being carried out to improve the accuracy of fission-gas analyses and to determine what factors are of major importance in obtaining reproducible results. Specifically, the program will involve the standardization of laboratory spectrometers with individual fission gases covering a range of gamma-ray energies, the experimental study of techniques for resolving complex gamma-ray spectra, and the determination of fission-gas distribution on the charcoal sample trap for typical collection times and temperatures.

Equipment is presently being assembled for the first phase of this program, which will involve the standardization of gamma-ray spectrometers for fission-product xenon-133. It is planned to have considerably improved fission-gas analysis techniques available for use in conjunction with the static-irradiation-capsule studies, as well as with the subsequent sweep-gas irradiation-capsule studies.

Abrasion-Resistant Coatings for UO₂ Particles

M. F. Browning, R. B. Landrigan, J. H. Oxley,
and J. M. Blocher, Jr.

A brief investigation was initiated for The Martin Company to determine the promise of chromium and alumina as abrasion-resistant coatings for 0.075-in. cylindrical urania particles. This fuel material is under study for the Fluidized-Bed Reactor.

One lot of urania cylinders was coated with 16 to 17 μ of chromium and another lot with 137 to 206 μ of alumina. The chromium coating was deposited in a rotating basket at 325 C by the thermal decomposition of chromium dicumene, while the alumina coating was formed at 950 C by the hydrolysis of aluminum chloride in a fluidized bed of the urania particles.

In a 5-hr abrasion test, it was found that the chromium coating reduced the rate of weight loss by a factor of 20 relative to the uncoated particles. By contrast, the alumina coating gave no significant improvement in abrasion resistance.

These and other coating materials are being considered for further examination.

PROBLEMS ASSOCIATED WITH THE RECOVERY OF SPENT REACTOR FUEL ELEMENTS (ORNL)

R. A. Ewing and C. L. Peterson

Losses of thorium and uranium in Sulfex decladding of a high-burnup (~15,000 MWD/T) Consolidated Edison prototype fuel pin were 0.02 and 0.05 per cent, comparable to those for 1000-MWD/T fuel pins. Thorium losses were also low for Darex decladding, 0.06 per cent, but uranium losses were significant, approaching 1.5 per cent. This uranium loss is about five times greater than the average uranium loss obtained when low-burnup Consolidated Edison fuel pins are declad by the Darex process.

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

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

Dissolutions of High-Burnup Consolidated Edison Fuel Pins

Analyses are continuing on the solutions obtained from the dissolutions of the first two high-burnup (15,000 to 25,000 MWD/T) prototype Consolidated Edison fuel pins. Although thorium and uranium analyses of the core solutions are not yet available, approximate losses to cladding solutions may be calculated from the known core weights and their reported composition of 95 w/o ThO₂-5 w/o UO₂. These data are summarized in Table M-1.

TABLE M-1. CUMULATIVE THORIUM AND URANIUM LOSSES DURING DECLADDING OF HIGH-BURNUP CONSOLIDATED EDISON FUEL PINS

Decladding Time, hr	Core Weight, g	Thorium			Uranium		
		Concentration, mg per ml	Total, mg	Loss, per cent	Concentration, mg per ml	Total, mg	Loss, per cent
<u>Sulfex Run 6</u>							
1	47.798	0.023	4.4	0.011	0.00024	0.045	0.002
3		0.069	~10.3	0.022	0.0077	1.2	0.055
<u>Darex Run 7</u>							
1	49.244	0.074	29.5	0.060	0.079	30.4	1.42
3		0.095	35.6	0.072	0.127	47.5	2.20

Upon completion of Sulfex decladding (3 hr) thorium and uranium losses were comparable, and both were quite low. Losses were very little different than those observed for Sulfex decladding of the previously examined CETR fuel pins, which had a much lower burnup (600 to 1000 MWD/T).

The type of decladding solution used had little or no effect on the losses of single-valence thorium. Thorium loss was only 0.06 per cent upon completion of Darex decladding (1 hr). However, uranium losses to an oxidizing Darex solution were about 25 times greater than to a nonoxidizing Sulfex solution, approaching 1.5 per cent. Comparable losses for the lower burnup CETR pins were in the 0.2 to 0.3 per cent range. Densities and uranium valences were comparable for both types of pins, suggesting that the increased loss of uranium is the result of the higher level of irradiation. The absence of change with Sulfex decladding suggests also that the oxidation state of the uranium may be the principal parameter.

After completion of the chemical analyses of these first two experiments, a second pair of high-burnup pins is to be dissolved.

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

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

A critical-assembly experimental program is being conducted for The Advanced Technology Laboratories of the American-Standard Corporation as part of a long-range program to evaluate the technical and economic feasibility of the Variable-Moderator Reactor (VMR) concept.

A series of short experiments was performed to evaluate $P(t)$, the ratio of fission-product gamma activity per fission of uranium-235 to that of uranium-238. This time-dependent function is required in evaluating δ^{28} , the ratio of uranium-235 to uranium-238 fissions in the reactor fuel. This determination completed the experimental phase of the program.

The compilation of the final report progressed throughout the month. Data were put in final form, intercore compilations of data were assembled, and error analyses were performed.

URANIUM CARBIDE AS A REACTOR FUEL (AI)

E. L. Foster

Atomics International's interest in the development of fuels for higher temperatures has led to support of research on the effects of radiation on uranium monocarbide. Specimens prepared by arc melting and casting techniques have been irradiated to burnups estimated to be as high as 22,000 MWD/T. Postirradiation examinations have shown that the general appearance was good, with only slight surface and volume changes being noted.

Postirradiation Examination of Uranium Monocarbide

A. W. Hare and R. F. Dickerson

The postirradiation examination of 12 cylindrical, 2-in. -long uranium carbide specimens is nearing completion. The specimens were irradiated to burnups of 1400 to 22,000 MWD/T of uranium and at core and surface temperatures of from 880 to 1600 F and 540 to 915 F, respectively. Details of the postirradiation examination have been previously reported.

During the past month, a metallographic examination of two specimens and a thermocouple tip from the last capsule, the highest burnup capsule, was initiated. These examinations will be completed during the period.

PEBBLE-BED REACTOR MATERIALS (S&P)

B. W. Dunnington

Battelle is investigating graphite spherical fuel elements in support of the Pebble-Bed Reactor project. A major requirement of the fuel is good fission-product retention. A promising approach to the fission-product-release problem is to coat the individual fuel particles with dense alumina or carbon. Evaluation of coated UO_2 and UC_2 particles has been receiving the most emphasis in the program during the past few months.

Experiments with Sweep Capsules SP-5 and SPF-3, containing fully enriched and natural Al_2O_3 -coated UO_2 particles in graphite, respectively, were completed this month. Postirradiation examination of the sphere in SP-5 will be carried out as part of the Battelle Coated-Particle Program. The rate of fission-gas leakage from the fully enriched sphere remained about the same during October. Burnup is 6 a/o of the uranium. Fission-gas leakage from the natural sphere has shown a negligible temperature effect up to 1900 F.

IN-PILE-CAPSULE EXPERIMENTS

G. E. Raines and W. H. Goldthwaite

Candidate fuel elements for the Pebble-Bed Reactor are being irradiated in low-flux and high-flux sweep capsules. In the low-flux capsules fission-gas release from natural fueled-graphite spheres is studied at temperatures up to 1900 F. The effect of burnup on fission-gas release from fully enriched spheres is studied at a single temperature (~1500 F) in the high-flux capsules.

SP-5

Irradiation of this high-flux sweep capsule in the BRR was completed in October. Specimen description is given in BMI-1464. Analysis of sweep helium passed over the sphere during irradiation gives release rates for krypton-85m, krypton-87, krypton-88, xenon-133, and xenon-135. Only one sphere, FA-22, No. 471, is being operated as a sweep specimen.

Sphere FA-22, No. 471, contains 500,000 100 to 125- μ -diameter particles of fully enriched UO_2 each coated with 40 μ of Al_2O_3 . During October, sphere-surface temperature was in the range of 950 to 1150 F, with center temperatures about 150 F higher. On completion of the experiment as of November 1, this sphere had been irradiated for 160 days for an estimated uranium-235 fission burnup of 6 w/o. R/B values are listed in Table P-1. These values are about the same as the values determined in September.

TABLE P-1. FISSION-GAS-RELEASE DATA FOR SPECIMEN NO. 471 IN SP-5

Sample Collection Date	Estimated Surface Temperature, F	R/B ^(a)				
		Krypton-85m	Krypton-87	Krypton-88	Xenon-133	Xenon-135
10-3-60	1150	3.4×10^{-4}	1.9×10^{-5}	7.5×10^{-5}	1.7×10^{-2}	1.9×10^{-4}
10-12-60	1150	6.3×10^{-4}	2.7×10^{-5}	1.1×10^{-4}	9.7×10^{-3}	2.3×10^{-4}
10-20-60	950	4.1×10^{-5}	5.9×10^{-6}	1.2×10^{-5}	6.9×10^{-4}	2.2×10^{-5}
10-21-60	950	5.1×10^{-5}	6.3×10^{-6}	1.6×10^{-5}	1.5×10^{-4}	1.9×10^{-5}

(a) R/B, the fractional release ratio, is the ratio of the number of atoms per sec being released to the number of atoms per sec being generated.

SPF-3

The low-flux Sweep Capsule designated SPF-3 contains natural Al_2O_3 -coated UO_2 particles in a graphite sphere. Coating thickness is 40μ . Specimen designation is FA-22, No. 449. With the exception of uranium-235 enrichment, this sphere is similar to the FA-22 sphere in SP-5. Fission-gas release is being studied in the 300 to 1900 F range in this temperature-controlled capsule.

R/B values are reported in Table P-2. Examination of the R/B values show slight evidence of a temperature effect.

TABLE P-2. FISSION-GAS-RELEASE DATA FOR SPECIMEN NO. 449 IN SPF-3

Sample Collection Date	Specimen Temperature, F	Neutron Flux ^(a) , nv	R/B ^(b)				
			Krypton-85m	Krypton-87	Krypton-88	Xenon-133	Xenon-135
10-6-60	1895	3×10^{11}	4.6×10^{-6}	9.2×10^{-7}	1.7×10^{-6}	9.0×10^{-6}	2.7×10^{-6}
10-12-60	1035	3×10^{11}	1.7×10^{-7}	(c)	(c)	1.4×10^{-6}	2.6×10^{-7}
10-27-60	1800	3×10^{11}	5.2×10^{-5}	1.6×10^{-5}	2.2×10^{-5}	2.8×10^{-4}	5.6×10^{-5}
10-28-60	1790	3×10^{11}	1.8×10^{-5}	3.6×10^{-6}	1.1×10^{-5}	9.5×10^{-5}	1.1×10^{-5}

(a) Neutron flux is determined from an external dosimeter wire exposed during a representative period of the operation.

(b) R/B, the fractional release ratio, is the ratio of the number of atoms per sec being released to the number of atoms per sec being generated.

(c) These species were below sensitivity.

TANTALUM AND TANTALUM ALLOYS FOR LAMPRE APPLICATIONS (LASL)

J. H. Stang

The research at Battelle under the auspices of the LAMPRE development program at Los Alamos Scientific Laboratory terminated at the end of October. Activity during October was, in the case of the alloy-development program, restricted to rounding out a fund of metallurgical data for the final group of binary tantalum alloys and, in the case of the X-ray and metallographic studies of tantalum, restricted to refining prior interstitial-element solubility data.

Topical reports summarizing information obtained in the two programs covered in the present section are in preparation. Also in preparation is a topical report describing details of the tantalum-irradiation program which Battelle conducted for Los Alamos and completed late this past summer.

Development of Container Materials for LAMPRE Applications

D. C. Drennen, C. J. Slunder, J. G. Dunleavy,
and A. M. Hall

A study to determine the optimum annealing temperatures for 13 binary tantalum-base alloys, which were sent to LASL for annealing, has been completed. Optimum annealing temperatures were selected on the basis of both hardness data and the results of metallographic studies. The annealing temperatures and hardness data for these alloys are presented in Table Q-1.

TABLE Q-1. EFFECT OF ANNEALING TEMPERATURE ON THE HARDNESS OF TANTALUM-BASE ALLOYS^(a)

Composition (Balance Tantalum)										Optimum ^(b)
Amount, w/o				Vickers Hardness (10-Kg Load) at Indicated Temperature						Annealing
Alloy	Element	Charged	Analyzed	As						Temperature,
				Rolled	2400 F	2600 F	2800 F	3000 F	3200 F	F
71	Al	0.5	<0.001	177	122	95	92	92	95	2800
72 ^(c, d)	Al	1.0	<0.001	171	127	105	102	100	108	2800
76	Be	2.0	0.01	208	154	127	118	109	109	3200
78	Ce	2.0	<0.10	139	115	100	96	100	109	3200
80	La	2.0	<0.03	162	132	98	107	105	103	3000
82	Sc	1.46	<0.1	179	160	130	122	118	129	3000
84	U	2.0	1.2	186	132	118	111	112	115	2800
86	Ti	2.0	0.01	168	104	105	107	103	106	3200
101	Ti	6.0	0.34	187	140	118	120	125	131	2800
104	--	--	Unalloyed Ta	193	126	118	114	117	114	3200
105 ^(d)	--	--	Unalloyed Ta	120	83	95	91	89	95	2600
109 ^(e)	Y	6.0	0.001	161	151	157	146	117	139	2800
110 ^(f)	Y	6.0	<0.001	181	140	165	173	179	173	2800

(a) The 0.030-in. -thick strip specimens were vacuum annealed for 30 min.

(b) Recommend annealing for 1 hr.

(c) Remelted with yttrium-conditioned tungsten electrode tip.

(d) Small area on tantalum button fused to copper furnace wall during one melting cycle; it is possible that contamination could have occurred during removal of alloyed area from the button.

(e) Segregated.

(f) Possible contamination during annealing.

In general, the hardness of the alloys changed with annealing temperature in an expected manner. However, there was an inconsistency in the behavior of the two yttrium-containing alloys. These two alloys were dissimilar in microstructure. Alloy 109 was segregated and contained a large amount of inter- and intragranular second phase, whereas Alloy 110 consisted of a homogeneous single-phase structure. There is a possibility that Alloy 110 was contaminated during annealing. Analyses of the annealed alloys will be conducted at Los Alamos.

It was difficult to select the optimum annealing temperature for the alloys containing scandium, titanium, and yttrium because of an elongated grain structure and/or excessive banding.

Chemical analyses for the alloying element were conducted on the materials in the as-rolled condition. In general, the results given in Table Q-1 indicate that, as expected, retention of most of the alloying additions was quite low. The best recovery was obtained with the uranium-containing alloy.

Hardness and grain-size data for these materials in the as-cast condition are given in Table Q-2. Several of the alloys were harder than unalloyed tantalum, even though chemical analyses indicated that very little of the particular alloying addition was retained. It is possible that even minor amounts of these additions produce marked hardening in tantalum. It is also possible, of course, that the higher hardness was caused by gaseous contamination during arc melting.

TABLE Q-2. HARDNESS AND GRAIN-SIZE DATA FOR AS-CAST TANTALUM ALLOYS

Alloy	Charged Composition (Balance Tantalum), w/o	Hardness, VHN	Grain Diameter, mm
71	0.5 Al	98	2.3
72	1.0 Al	134	3.0
73(a)	0.5 B	288	(b)
74(a)	1.0 B	376	(b)
76	2.0 Be	147	(b)
78	2.0 Ce	81	2.1
80	2.0 La	100	0.88
82	1.46 Sc	105	1.71
84	2.0 U	109	1.65
86	2.0 Ti	85	0.82
101	6.0 Ti	116	1.28
104	Unalloyed Ta	107	3.18
105	Unalloyed Ta	78	3.00
109	6.0 Y	96	(b)
110	6.0 Y	150	0.68

(a) Not fabricable.

(b) Grain size not determined.

Tensile properties were determined on another group of tantalum alloys in the vacuum-annealed condition and are listed in Table Q-3. These data indicate that the tensile strength of tantalum can be increased significantly by alloying. Increasing additions of the alloying elements employed resulted in an increase in tensile strength without a great change in ductility.

TABLE Q-3. ROOM-TEMPERATURE TENSILE PROPERTIES OF VACUUM-ANNEALED TANTALUM ALLOYS^(a)

Alloy	Composition (Balance Tantalum), w/o	Annealing Temperature, F	0.2 Per Cent Offset Yield Strength, 10 ³ psi	Ultimate Tensile Strength, 10 ³ psi	Elongation in 1 in., per cent
87	3.0 Th	3200 ^(b)	33.80	55.05	13
90	1.5 Th	3200 ^(b)	32.90	49.65	14
91	1.0 Fe	2600	29.35	36.55	15
92	3.0 Fe	2600	34.55	40.05	17
95	0.5 Hf	3000	37.70	48.10	24
96	1.5 Hf	3000	34.80	49.30	25
97	3.0 Hf	3000	48.80	62.15	22
98	0.5 Zr	3000	39.95	50.90	18
99	1.5 Zr	3000	46.70	61.80	24
100	3.0 Zr	3000	71.45	86.35	18
103	Unalloyed Ta	2600	32.05	37.20	21

(a) The 0.030-in. specimens were vacuum annealed 1 hr.

(b) Vacuum annealed 1 hr at 2900 F plus 1 hr at 3200 F.

Precipitate-Phase Identification and Interstitial-Type
Solid Solubility in Tantalum

D. A. Vaughan and C. M. Schwartz

X-ray diffraction and metallographic studies on the solid solubilities and initial precipitate phases in the tantalum-carbon, tantalum-nitrogen, and tantalum-oxygen systems have been carried out at Battelle to aid LASL in understanding the effects of these interstitials on the corrosion resistance of tantalum in plutonium fuel mixtures. Previous reports gave the preliminary results on specimens annealed at 1500 and 1000 C. During the past month these results were refined, the specimens annealed at 500 C were analyzed, and a topical report was under preparation. Table Q-4 presents a summary of the results obtained on this program.

TABLE Q-4. SOLID-SOLUBILITY LIMIT AND PRECIPITATE PHASES IN THE TANTALUM-CARBON, TANTALUM-NITROGEN, AND TANTALUM-OXYGEN SYSTEMS

System	Solid Solubility, a/o			Precipitate Phase
	1500 C	1000 C	500 C	
Ta-C	0.17	<0.07	--	Ta ₂ C
Ta-N	3.70	2.75	1.8	Ta _x N (complex cubic, a ₀ = 10.09 Å)
Ta-O	3.65	2.95	2.5	Ta ₂ O ₅ (low-temperature allotropic modification)

FLOW STUDIES FOR THE PWR (BAPD)

R. W. Dayton

Experimental studies of flow in a model of PWR, Core 2, were concluded this month, after obtaining certain required data.

Reactor Flow Studies

L. J. Flanigan and H. R. Hazard

Air-flow studies in a 0.4-scale model of the PWR-2 are being conducted at Battelle to provide design data for the prototype. Previously reported work includes design of the model, completion of the test apparatus, and initiation of core assembly.

The model core assembly and installation in the model were completed on October 8, and flow studies were started immediately.

The following flow studies have been completed:

- Core flow-measuring-orifice calibration
- Core-flow distribution, three- and four-loop
- Detailed pressure losses, three- and four-loop
- Interpass mixing, three- and four-loop
- Lower plenum mixing, four-loop.

The model operated very satisfactorily, and no unexpected flow phenomenon occurred.

Work on the project was terminated on October 31.

DEVELOPMENT OF MATERIALS FOR THE MGCR AND HTGR (GA)

W. H. Goldthwaite

Research on core materials for the MGCR and HTGR is in progress. Radiation-effects studies of fuel specimens of UO_2 dispersed in BeO and UC and UC_2 dispersed in graphite are reported. Techniques for coating uranium and thorium carbide powder with carbon by vapor deposition are being investigated.

DEVELOPMENT AND EVALUATION OF FUEL
ELEMENTS FOR THE MGCRRadiation-Effects Study of Potential Fuels

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

A study of the radiation stability of ceramic-type fuels under conditions simulating those of the MGCR concept is in progress. The types of fuels under study are uranium dioxide dispersed in beryllium oxide, uranium monocarbide dispersed in graphite, and uranium dicarbide dispersed in graphite. The loading in each type is equivalent to 20 volume per cent of UO_2 . The specimens will be irradiated over a range of about 1 to 12 a/o uranium at a specimen-surface temperature of about 1500 F. Four capsules, equipped with thermocouples and electric heaters, containing two specimens of each fuel composition make up the program.

One of the four capsules, GA-BRR-1, was opened and the specimens examined at Battelle. After irradiation in the Battelle Research Reactor to specimen burnups of about 1.0 to 1.7 uranium a/o, all of the specimens were in good condition. Two of the remaining three capsules, BMI-31-2 and BMI-31-3, were irradiated in the MTR and were shipped to the General Atomic hot cells for the postirradiation examination of the specimens. This examination is now in progress. The final capsule, BMI-31-1, is continuing to operate satisfactorily at about 1500 F. This capsule is scheduled to be discharged from the MTR in March, 1961, and will be sent to GA for examination.

Experimental Capsule Studies

N. E. Miller, P. M. Steinback, and J. H. Stang

Fuel pellets of UO_2 (30 volume per cent, 12 per cent enriched) and BeO will be contained in Hastelloy X cans and irradiated at 1700 F for 18 months at the MTR in a NaK-filled double-wall capsule equipped with thermocouples and auxiliary electric heaters. The fuel pellets will be clad and encapsulated as soon as they become available from General Atomic.

COATING OF URANIUM AND THORIUM CARBIDE
POWDERS FOR THE HTGR

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

Solid-solution thorium-uranium dicarbide fuel particles are being coated with pyrolytic carbon to provide air-stable fuel particles for fuel-compact development studies. Besides protecting the carbide from moisture, the coating may also help retain fission products during reactor operation. The carbon was deposited by the pyrolysis of acetylene in a fluidized bed of the fuel particles.

Nine lots of mixed carbide were coated during October at temperatures ranging from 1650 to 2750 F and with coatings of 2 to 55 μ of carbon. Two of these lots were enriched material and seven were depleted material. The enriched material has been shipped to General Atomic for compact fabrication and evaluation.

A variation in apparent coating porosity was found to result from variations in the partial pressure of acetylene in the feed gas. If the presence of this apparent porosity can be confirmed by physical tests, it may be possible to provide a carbon coating with improved fission-product-retention properties.

Every lot of the coated particles was tested for porosity by screening it into fractions and exposing the weighed fractions to laboratory air at ambient conditions until there was no further weight gain. Analysis of the data from the hydrolysis test indicates that the smaller the particle size, the larger the weight gain. To help verify the apparent effect of particle size on percentage weight gain, duplicate runs have just been completed on narrowly screened samples of thorium-uranium dicarbide powders. Evaluation of the results of these experiments will be completed shortly.

Bench-scale coating studies are to be recessed and consideration is being given to an investigation of the feasibility of coating the mixed carbide particles on a larger scale of operation.

DEVELOPMENTAL STUDIES FOR THE SM-1 AND SM-2 (ALCO)

S. J. Paprocki

Instrumented fuel plates of the blocked-channel design incorporating SM-1 fuel plates have been fabricated. The plates contain a Zircaloy filler clad with niobium to prevent reaction with the stainless steel during the bonding treatment.

Six capsules containing SM-2 reference and alternate fuel specimens are being irradiated in the ETR. It is planned to irradiate the specimens to a burnup of 70 per cent of the contained uranium-235. Work has been concluded on the materials-development and fuel-element thermodynamics studies.

SM-1 PROGRAMProcess Development for the Preparation of Instrumented
Fuel Plates of the SM-1 Type

S. J. Paprocki, E. S. Hodge, G. W. Cunningham,
C. B. Boyer, and E. G. Smith, Jr.

A process has been developed through the use of the gas-pressure-bonding technique for producing SM-1 instrumented fuel plates of the blocked-channel design. Plates of both the stationary and control design have been produced for in-pile test evaluation in the SM-1 core.

The instrumented fuel plates consist of a massive low-cross-section filler material (Zircaloy-2) which will contain the thermocouples. This filler plate is enclosed in a Type 304-L stainless steel frame which is bonded between two SM-1 fuel sheets of one-half the standard thickness. These fuel sheets were roll bonded with niobium clad to one face to prevent reaction between the filler and the core during the gas-pressure-bonding operation. Earlier development studies indicated that 3 mils of niobium was sufficient to prevent any reaction. Strips of niobium 20 and 25 mils thick were also used between the edges of the filler and stainless frame.

The entire plate assemblies were gas-pressure bonded at a temperature of 2100 F for 3 hr at 10,000 psi to obtain good stainless steel self-bonds.

Three full-size stationary instrumented fuel plates have been fabricated and two control and one stationary plate are being processed. The shipment of these six plates to Alco will complete all of the work planned for this program.

SM-2 PROGRAMRadiation-Stability Studies of Reference and Alternate SM-2 Fuels

J. H. Stang, L. K. Seymour, J. F. Lagedrost,
and W. H. Goldthwaite

Capsule-irradiation studies of SM-2 reference and alternate fuel materials are in progress at the ETR. Six instrumented capsules, each containing six clad UO₂-stainless steel specimens, are being exposed under conditions designed to achieve specimen-surface temperatures of 650 F and fuel burnups up to 70 a/o of the uranium.

The status of each of the in-pile capsules as of the end of ETR Cycle 32 (October 31, 1960) is summarized below.

BMI-32-5. This capsule completed approximately 30 full-power days in the reactor. An estimated time of 140 days is needed to achieve 70 a/o total uranium burnup. Surface temperatures of the individual specimens range from 625 to 775 F. All thermocouples are operating satisfactorily; no electric heat has been required.

BMI-32-6. This capsule completed approximately 90 of the estimated 160 days at full power required to achieve 70 a/o burnup. Specimen-surface temperatures during the first 55 days ranged from 500 to 650 F with the addition of supplementary electric heat. At the start of ETR Cycle 31 (September 8, 1960), all thermocouples became erratic simultaneously. Therefore, for Cycles 31 and 32, no electric heating was possible. Specimen-surface temperatures for this period are estimated to be 100 to 150 F below design. Circuit checkouts are scheduled for the current shutdown.

BMI-32-7. This capsule, which is being irradiated at a flux level somewhat higher than the above two, completed approximately 55 days at full power; an estimated time of 125 days is required for 70 a/o burnup. Specimen-surface temperatures have ranged from 520 to 750 F with no electric heating. Of the three operating thermocouples, one appeared to be failing during Cycle 32. It will be checked during the current shutdown.

BMI-32-8. This capsule completed approximately 90 of the estimated 150 days at full power required to reach 70 a/o burnup. Individual specimen-surface temperatures have ranged from 530 to 650 F with the aid of electric heat. One thermocouple experienced a sudden 100 F decrease during Cycle 32, while the others remained steady. Its circuit will be checked during the current shutdown.

BMI-32-9. This capsule completed approximately 75 full-power days as of October 31; it will require an estimated total of 90 days to reach the 70 a/o burnup level. Since the start of this irradiation, specimen-surface temperatures have dropped gradually; levels during Cycle 32 ranged from 570 to 1090 F, compared with 800 to 1250 F during Cycle 29. As reported previously, this capsule was inserted into a

reactor position having neutron fluxes much greater than those designed for. Three of its thermocouples remain operative; the electric heaters have not been required.

BMI-32-10. This capsule completed approximately 30 full-power days; it will require an estimated 125-day total to achieve 70 a/o burnup. Specimen-surface temperatures range from 550 to 790 F by fission and gamma heating only. Three thermocouples are operative.

GAS-COOLED REACTOR PROGRAM (AGN)

D. L. Keller

Studies for Aerojet-General Nucleonics (AGN) and the Army Reactor Branch (ARM) are reported in this section. A large part of these studies are directed to support the ML-1 reactor fuel-element development program in which the reference fuel consists of both solid UO_2 and BeO-UO_2 pellets clad with Hastelloy X tubing.

During October a program was initiated to evaluate the possible advantage of using Al_2O_3 -coated UO_2 as a fuel material for advanced ML-1 cores. Spherical UO_2 particles will be coated with Al_2O_3 by a vapor-deposition process. This material will then be used in sintering and hot-pressing studies to determine to what extent it might be densified into sound bodies. These bodies will contain 60 volume per cent UO_2 or greater. If sound bodies can be obtained they will be corrosion tested to determine their oxidation resistance.

Study of Processing Variables in the
Fabrication of BeO-UO_2 Ceramics

H. D. Sheets, A. K. Smalley, and W. H. Duckworth

The objective of this program is to develop optimum techniques for fabricating BeO-UO_2 fuel-element pellets for use in the ML-1.

In earlier work (BMI-1442) it was found that sintered pellets containing 60 w/o UO_2 and 40 w/o Brush LOH BeO could be disintegrated by oxidation in air at 1000 F. Then up to 10 w/o of disintegrated material could be added to a virgin batch without decreasing the sinterability of the batch. Since LOH BeO no longer is available, recent work has been concerned with assessing the sinterability of UO_2 - BeO ceramics made from other BeO powders. With the powders evaluated, sintering at 3200 F for 1 hr was required to obtain densities in excess of 95 per cent of theoretical. Comparable pellets containing LOH BeO sintered to high density in 1 hr at 2700 F.

During October, the effects of utilizing fired and unfired scrap on the sintered density of pellets containing Brush UOX BeO were investigated. Fired scrap was prepared by disintegrating sintered pellets by heating in air. Unfired scrap was prepared by either crushing pressed, dried pellets to pass through a 12-mesh screen, or by oxidizing pressed pellets in air. Batches containing these two scrap materials and virgin materials were prepared as shown in Table U-1.

The addition of sintered scrap reduced sintered density slightly, the decrease being greater as the scrap content increased. The effect was less marked at the higher forming pressure. Within the range studied, fuel content did not affect the magnitude of density decrease as fired scrap was increased. Use of green scrap, whether crushed or oxidized, had little or no effect on sintered density.

TABLE U-1. EFFECT OF SCRAP ADDITIONS ON SINTERED DENSITY AND DIAMETRAL SHRINKAGE OF BeO-UO₂ CERAMICS

Batch	Batch Composition, w/o			Pressed Scrap		Total UO ₂ , w/o	Forming Pressure, psi	As-Formed Density, g per cm ³	Sintered Density ^(c)		Diametral Shrinkage, per cent
	Virgin BeO ^(a)	Virgin UO ₂	Sintered Scrap ^(b)	Treatment	Amount, w/o				G per Cm ³	Per Cent of Theoretical	
74-2 ^(d)	40	60	--	--	--	60	20,000	2.74	5.10	96.4	18.8
74-2 ^(d)	40	60	--	--	--	60	40,000	2.97	5.16	97.5	16.9
74-3 ^(d)	30	70	--	--	--	70	20,000	3.12	5.82	92.4	18.9
74-3 ^(d)	30	70	--	--	--	70	40,000	3.38	5.90	93.7	17.1
124-1	38	57	5	--	--	60	20,000	2.72	4.98	94.1	18.2
124-1	38	57	5	--	--	60	40,000	2.94	5.12	96.8	16.6
124-2	36	54	10	--	--	60	20,000	2.74	4.89	92.4	17.4
124-2	36	54	10	--	--	60	40,000	2.99	5.06	95.6	16.0
124-3	32	48	20	--	--	60	20,000	2.82	4.76	90.0	16.2
124-3	32	48	20	--	--	60	40,000	3.06	4.90	92.6	14.8
125-1	28.5	66.5	5	--	--	70	20,000	3.05	5.60	88.9	18.4
125-1	28.5	66.5	5	--	--	70	40,000	3.30	5.73	91.0	16.8
125-2	27	63	10	--	--	70	20,000	3.08	5.52	87.7	17.8
125-2	27	63	10	--	--	70	40,000	3.36	5.70	90.4	16.4
125-3	24	56	20	--	--	70	20,000	3.18	5.47	86.8	16.8
125-3	24	56	20	--	---	70	40,000	3.46	5.64	89.5	15.6
126-1	36	54	5	Crushed ^(e)	5	60	20,000	2.74	4.98	94.1	18.0
126-1	36	54	5	Crushed	5	60	40,000	2.98	5.12	96.8	16.5
126-2	34	51	5	Crushed	10	60	20,000	2.74	4.98	94.1	17.9
126-2	34	51	5	Crushed	10	60	40,000	2.96	5.11	96.6	16.5
126-3	30	45	5	Crushed	20	60	20,000	2.75	4.96	93.8	17.6
126-3	30	45	5	Crushed	20	60	40,000	2.99	5.12	96.8	16.3
127-1	36	54	5	Oxidized ^(f)	5	60	20,000	2.72	5.10	96.4	18.2
127-1	36	54	5	Crushed	5	60	40,000	2.96	5.16	97.5	16.6
127-2	34	51	5	Crushed	10	60	20,000	2.70	5.10	96.4	18.1
127-2	34	51	5	Crushed	10	60	40,000	2.93	5.12	96.8	16.0
127-3	30	45	5	Crushed	20	60	20,000	2.67	5.06	95.6	18.0
127-3	30	45	5	Crushed	20	60	40,000	2.88	5.05	95.4	16.8

(a) Lot 100-W-24, Brush Beryllium Company, Cleveland, Ohio.

(b) Prepared by heating sintered pellets to 1000 F in air, and holding at temperature for 2 hr.

(c) Theoretical densities of 60 w/o UO₂-40 w/o BeO and 70 w/o UO₂-30 w/o BeO were taken as 5.29 g per cm³ and 6.30 g per cm³, respectively. Pellets were sintered for 1 hr at 3200 F in hydrogen.

(d) Previously reported data, repeated for clarity.

(e) Prepared by crushing pressed pellets to pass through a 12-mesh sieve, using a mortar and pestle.

(f) Prepared by heating pressed pellets to 1000 F in air, and holding at temperature for 2 hr.

Future work will include studies of the effects of argon-hydrogen or forming-gas sintering atmospheres on the sinterability of BeO-UO₂ compositions.

Advanced ML-1 Fuel Pellets

J. W. Lennon and A. K. Smalley

The objective of this program is to develop ceramic fuel pellets of UO₂ and Al₂O₃. The pellets are to be resistant to oxidation by the oxygen-containing nitrogen coolant to be used in the ML-1. The pellets are to contain at least 60 volume per cent of UO₂, a minimum volume of voids, and the balance Al₂O₃. The experimental bodies will measure about 1/4 in. in diameter by about 1/4 in. long.

Effort during October consisted of planning the laboratory program and procuring necessary materials. Briefly, the approach to the problem will be to form sound bodies from fuel particles which have been previously coated by a vapor-deposition method with an impermeable shell of Al₂O₃. Various size fractions of coated UO₂ particles will be mixed with Al₂O₃ powder and a suitable binder, cold pressed, and sintered to densify the Al₂O₃ matrix. The oxidation resistance of the sintered pellets will be measured by heating at temperatures in the range of the anticipated operating temperature of ML-1 in the ML-1 coolant or in other oxidizing atmospheres.

Laboratory work has been started.

Encapsulation Studies

W. J. Zielenbach, J. P. Logedrost, and J. H. Stang

Irradiation studies of Hastelloy X-clad compacts of highly enriched UO₂ and UO₂-BeO are in progress. Specimens are contained in capsules designed to maintain cladding-surface temperatures at 1725 ± 25 F for periods of time of interest to the ML-1 core-development program. At present, two capsules are being irradiated, one at the Materials Testing Reactor and one at the Battelle Research Reactor.

Capsule BMI-38-1, containing specimens composed of 70 w/o and 80 w/o UO₂ in a BeO matrix, completed its second cycle in MTR Position A-27-NE on October 24, 1960. Prior to the start of this cycle (MTR Cycle 146) the capsule was raised so that the specimens were positioned along the peak of the flux profile. During the cycle, the average cladding-surface temperature of the hottest 70 w/o specimen was 1570 F with the addition of approximately 2 kw of auxiliary electric heat. Since this temperature is somewhat lower than that desired, some consideration is being given to the possibility of relocating the capsule into a position having about 15 per cent higher flux than available in A-27-NE.

In future cycles, the irradiation may be faced with a difficult temperature-interpretation situation inasmuch as five of the six thermocouples originally provided now appear to be inoperative. Three of these failed to function properly after Cycle 147

startup; the leadout circuits of these three will be carefully examined during Cycle 148 shutdown in an attempt to restore sensitivity.

Capsule AGN-BRR-III contains six highly enriched UO_2 specimens. During its first two cycles of irradiation specimen-surface temperatures with the addition of up to 2 kw electric heat were about 200 F lower than the desired 1725 F. To raise the specimen temperatures, the capsule body has been provided with additional thermal resistance between the fission heat source and the pool water by surrounding the body with an aluminum "boot" and filling the internal annulus thus formed with helium. The operating characteristics of the capsule with this modification will be assessed during the reactor cycle starting November 2.

It is not fully understood why both of the capsules described above have operated at temperatures lower than the design levels. It is believed, however, that one contributing factor lies in capsule-specimen flux-perturbation factors greater than predicted by the combination of empirical and analytical techniques employed to predict net perturbation. The magnitude of error involved could easily be related to the fact that the specimens contain an unusually large amount of uranium-235.

Alloy Modification of Hastelloy X

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

There were no studies conducted on this program during October.

High-Temperature Fatigue Evaluation of Hastelloy X Tubing

G. McClure, H. Grover, and A. Duffy

The experimental program for the high-temperature evaluation of Hastelloy X tubing was resumed during the latter part of September. An S-N curve will be developed for each of five temperature levels out to 10^6 cycles at a stress ratio of $A = 1$. The five temperature levels are: 1400, 1600, 1735, 1800, and 2000 F. In order to establish Goodman diagrams based on failure at 10^6 cycles, additional tests will be performed at stress ratios of $A = 0$, $A = 0.25$, and $A = \infty$ (approximately). About 120 specimens will be required for the evaluation. Cyclic speed for all the tests will be 1725 cycles per min, which will produce 10^6 cycles in about 10 hr.

Calibration of fatigue machines as adapted for most of this series of tests has been completed. End clamps which hold the tubing sufficiently tight during testing but which do not introduce detrimentally high stress intensification have been satisfactorily developed.

During October, three S-N curves were partially completed. For stress ratios of $A = 1$, the S-N curves for temperatures of 1600, 1735, and 1800 F indicate that stresses of 18,500, 8,700, and 5,300 psi, respectively, are necessary to instigate the fatigue failure at 10^6 cycles. These stated stresses are the result of preliminary reduction of

data and, therefore, are not conclusive. Twenty-one tests have been conducted so far. Each specimen failed near its center in the heat zone of controlled temperature and consequently indicated proper functioning of the end clamps.

Plans for November are to continue testing specimens at a stress ratio of $A = 1$ in order to firmly establish the S-N curves for 1600 and 1800 F. In addition, the S-N curves for 1400 and 2000 F will be started for stress ratios of $A = 1$.

ML-1 Critical-Assembly Experiments

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

Experiments relating to the ML-1 flooded-core condition were continued during October. The reactivity worth of a fuel element in three peripheral positions was measured. Experiments also were performed to determine the concentration of boric acid in the moderator necessary to provide a shutdown reactivity worth of 1 per cent $\Delta k/k$. In addition, final analysis of all data obtained to date was started in preparation for reporting.

The reactivity worth of a peripheral fuel element was found to be a strong function of core position. Measurements were made in three positions; all other peripheral positions are nearly symmetric with one of those measured. The results of the measurements are given in Table U-2. Measurements were made by adding the element in the position as noted to a 42-element core.

TABLE U-2. REACTIVITY WORTH OF PERIPHERAL FUEL
ELEMENTS IN ML-1 FLOODED CORE

Core Position	Element Worth, per cent $\Delta k/k$
48	0.60
49	0.51
50	0.35

It was found that 0.324 g of boric acid per liter of moderator water gave a shutdown reactivity worth of 1 per cent $\Delta k/k$. This concentration was determined by interpolating between the measured points, which lay on a straight line. The experimentally measured points are given in Table U-3.

Experiments concerning reactivity worths of production-type fuel elements in the unflooded core will be performed during November. In addition, work will continue on data analysis and preparation of a report.

TABLE U-3. REACTIVITY WORTH VERSUS BORIC ACID
CONCENTRATION IN ML-1 FLOODED
CORE

Concentration of Boric Acid, g/per liter	Reactivity Worth, per cent $\Delta k/k$
0.000	0.000
0.264	0.815
0.504	1.565

BRR Loop Program

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

The fuel-element irradiation program in the recirculating gas loop at the Battelle Research Reactor is a part of the Aerojet-General Nucleonics program for the Army Reactors Branch of the AEC. The 1B-2T-1 specimen was the seventh in a series of experiments for this program. Irradiation of this fuel subassembly, which began in May, was terminated after 1940 hr of in-pile operation. The average specimen power generation was 36 kw during the irradiation. For the final operational period of about 300 hrs, specimen temperatures were as follows:

Outlet gas temperature	1300 F
Inlet gas temperature	760 F
Peak fuel-pin surface temperature	1580 F

These conditions were achieved with a coolant-flow rate of 610 lb per hr, which gave a pressure drop across the specimen of about 2.5 psi. Evaluation of the friction factor of the subassembly revealed that an apparent increase, observed when operation at 1300 F exit-gas conditions began, was not significant. At the low flow rates necessary for the 1300 F exit-gas conditions (about 600 lb per hr or less), operation was in a transition region where accurate determinations of friction factor were difficult. The apparent change was within the range of error at the flow rates used for the 1300 F exit-gas conditions.

Fission-product gases were detected in the system during the latter part of the operation. Activity levels in the loop at the end of the operation were as follows:

Xenon-133	25 mc
Krypton-85m	<0.1 mc
Krypton-88	<0.1 mc
Xenon-135	<0.1 mc

Removal of the specimen from the loop was effected in a routine manner. The specimen was sealed in a cask for transfer to a hot cell for posttest examination.

The loop was secured in the back pool of the reactor since no further irradiations are planned at present. Accordingly, no further reports will be issued until such time as irradiation experiments are resumed in the recirculating gas loop.

CORROSION OF THORIUM AND URANIUM UNDER STORAGE CONDITIONS (LIVERMORE)

P. D. Miller

Improvements in nickel-plating procedures for thorium have been obtained by means of leveling agents.

An extensive corrosion-evaluation program under several storage conditions has been initiated at Columbus, Ohio, and at Daytona Beach, Florida.

Electrodeposited Coatings for Thorium (SC)

J. G. Beach, E. S. Koglikowski, and C. L. Faust

Thorium specimens were vapor blasted, electropolished, pickled, activated by the procedure developed at LRL, and plated directly with nickel in a Watts' type bath containing coumarin as a leveling agent. Such specimens were smoother than others which were previously nickel plated, and they might be more resistant to corrosion. Metallographic examination of cross sections showed better filling of etch pits caused by anodic etching when the specimens were plated in the leveling nickel baths. Humidity tests were started to compare the leveling nickel deposits with previous nickel plates.

Experimental attempts to deposit nickel adherently on specimens that were not anodically etched have, so far, been unsuccessful. This phase will be continued in the future, however, because etch pits undoubtedly are potential corrosion sites. Complete filling with nickel has not been obtained, up to the present time.

Conventional procedures for chromium plating directly on anodically etched thorium resulted in selective deposition in the etch pits. Future work will be aimed at taking advantage of this unusual selectivity.

Corrosion Evaluation (UCRL)

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

Phase II and Phase III corrosion-exposure programs have been started. In the Phase II program, specimens are being exposed to 100 per cent relative humidity at 120 F. In the Phase III program, specimens are being exposed uncovered in a warehouse at Columbus, Ohio, and in a closed shed at Daytona Beach, Florida. In addition, sets of specimens have been placed in unsealed metal containers on a roof in Columbus and in the shed at Daytona Beach.

The specimens include 2 by 3-in. coupons of bare thorium, uranium, and uranium-10 w/o molybdenum. Thorium coated with an organic-type composition and with electroplated nickel and uranium plated with aluminum are included. Soldered, welded, and screwed joints between thorium and uranium are represented by coupons about 1/8 by 1/2 by 2 in. A variety of metals, including aluminum, beryllium, magnesium, stainless steel, and titanium, have been joined to thorium in this same set.

The specimens will be exposed for a period of 1 year and will be examined visually at 1-, 3-, and 6-month intervals.

GAS-PRESSURE BONDING OF BERYLLIUM-CLAD ELEMENTS (ORNL)

S. J. Paprocki, E. S. Hodge, and J. S. Perrin

The use of beryllium-clad uranium dioxide fuel elements in gas-cooled reactors is being considered by Oak Ridge National Laboratory. The preparation of these fuel elements by gas-pressure bonding is being studied.

Material for use in studying the self-bonding of beryllium as a function of bonding parameters and for the fabrication of beryllium-clad uranium dioxide fuel elements is being machined to final dimensions at ORNL. While waiting for the arrival of this material, a preliminary study using as-rolled beryllium sheet is continuing. Bonds have been achieved between two mating edges of beryllium that were prepared by shearing of sheet stock. In addition, preliminary examination of these specimens indicated that small cracks produced during shearing were healed by the pressure-bonding operation.

The compatibility of various materials at the bonding temperatures to be studied is being determined with specimens prepared from sheet-stock material. These specimens are currently being processed for evaluation.

PLAIN BEARING MATERIALS FOR NITRIC ACID-ALUMINA
SLURRY PUMP (IDO)

H. A. Van Dyke, W. A. Glaeser, and C. M. Allen

This program is directed at the selection and evaluation of plain sleeve-bearing-journal materials for a nitric acid-alumina slurry pump. The slurry is created during cleanup of off-gas from a fluidized-bed calcination of aluminum nitrate liquid wastes to form granular alumina.

A thrust-bearing system consisting of three fixed tapered-land pads and a thrust disk was selected for evaluation. This system was designed to simulate the velocity and lubricant-film thickness anticipated for the guide bearing of a slurry pump.

Duplicate experiments with graphite pads running against a 17-4PH thrust runner indicated a wear of about 2 mils after operating for 24 hr in an aqueous nitric acid slurry containing 40 w/o HNO_3 and 5 w/o Al_2O_3 . These results confirm service results.

Practical considerations indicate that cylindrical boron carbide journal bearings operating with chromium carbide journals should provide adequate life in the nitric acid slurry.

This concludes the present phase of the investigation of circulating pump bearing materials for use in nitric acid slurry systems.

RWD:CRT/all