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PROGRESS RELATING TO CIVILIAN
APPLICATIONS DURING
NOVEMBER, 1960

by

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REPORTS RELATING TO CIVILIAN APPLICATIONS
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- BMI-1379 "Addendum to Hazards Summary Report for the GCRE Critical-Assembly
(Rev.) Experiments", by Joel W. Chastain, Harold M. Epstein, William S.
Hogan, and David A. Dingee.
- BMI-1459 "Development of Corrosion-Resistant Alloys for Use as Container
Materials for Decladding Solutions or as Welding Alloys", by Charles L.
Peterson, David C. Drennen, Merritt E. Langston, A. M. Hall, and
Walter K. Boyd.
- BMI-1471 "Alumina Coating of UO_2 Shot by Hydrolysis of Aluminum Chloride Vapor",
by Melvin F. Browning, Neil D. Veigel, Thomas E. Cook, Ward S.
Diethorn, and John M. Blocher, Jr.
- BMI-1472 "Determination of Interstitial Solid-Solubility Limit in Tantalum and
Identification of the Precipitate Phases", by Dale A. Vaughan, Oliver M.
Stewart, and Charles M. Schwartz.
- BMI-1473 "Progress Relating to Civilian Applications During October, 1960", by
Russell W. Dayton and Clyde R. Tipton, Jr.
- BMI-1475 "Progress on the Use of Gas-Pressure Bonding for Fabricating Low-Cost
Ceramic, Cermet, and Dispersion Fuels", by Stan J. Paprocki.

REACTOR MATERIALS AND COMPONENTS (AEC-DRD)

R. F. Dickerson

The investigation of high temperature-high pressure reactions is currently being concentrated on the study of the $U_3O_8-Al_2O_3$ system. The work is being directed toward verification of the possible compound which was reported in BMI-1473.

The irradiation of ten capsules containing Type 347 stainless steel is continuing in the ETR. Since the recent move to the G-13 position, the lack of accurate flux information has made it impossible to predict the exposures being received by the specimens. These data will be available soon. The analysis of irradiated iron for manganese-54 as a means of determining the fast flux exposure is being studied, and initial results are quite promising; therefore, the effort will be continued. According to present plans, one of the capsules, exposed to over 5.0×10^{21} nvt, will be discharged from the reactor for postirradiation testing early in 1961.

Niobium-base binary alloys containing 2.37 w/o chromium, 3.34 w/o zirconium, 5.21 w/o vanadium and a ternary alloy containing 1.56 w/o zirconium and 1.09 w/o chromium are being tested in creep at 650 and 1000 C. The results to date corroborate the statement that on a weight percentage basis chromium is the most potent strengthener of the additions examined. Creep studies will be continued as will the recrystallization studies. It should be remembered that these alloys are of interest as potential cladding for the EBR.

Because of changes in the predicated gamma-heat production at the WTR, it was necessary to make some minor changes in the capsule to be used for the in-reactor creep test on Zircaloy-2. These modifications have been accomplished; however, loading cannot be carried out until there is assurance that the required neutrons will be available to the program. Electron microscopy work as a part of the strain-aging study is continuing as are additional mechanical tests.

The particles of hafnium diboride which were irradiated to relatively high burnup have been examined by X-ray diffraction techniques. These studies show that both the natural and enriched materials suffered complete loss of crystallinity and were degraded to amorphous structures. The lithium analyses have been completed, but the necessary calculations to complete the burnup analyses are not finished. With the determination of burnup, experimental work will be complete.

The study of principles which are applicable to selection of materials and design of mechanical parts for use in high-temperature sodium has continued. It has been shown that the formation of a surface film improves friction and wear behavior in high-temperature sodium. Actual experiments with various materials and thermodynamic predictions as to probability of surface-film formation are in good agreement. Those materials in which film formation was predicted reacted well in test. Those predicted to be unfavorable toward surface-film formation did not react well.

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 uranium-oxygen system and on the reactions of uranium oxide with various mixed oxides. Previous work on this program has been directed toward the development of equipment and techniques and their application to study of the uranium-oxygen system under pressure. *

Current work is continuing on the study of a possible reaction between U_3O_8 and Al_2O_3 to form a compound. This work is directed toward verification of the compound and the conditions of temperature, pressure, and composition requisite for its formation. Results will be reported when available.

Irradiation-Surveillance Program on Type 347 Stainless Steel

W. E. Murr, R. L. Ritzman, F. R. Shober, and R. F. Dickerson

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 mechanical testing of specimens contained in approximately 14 capsules which are being irradiated in core positions of the ETR and WTR test reactors. The specimens, which include subsize tensile, cyclic-strain fatigue, and impact 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 F (process-water temperature) and 600 F. The information obtained from postirradiation mechanical tests will be used for predicting the in-reactor life for operating ETR loops constructed of this material.

At the present time, ten capsules are undergoing irradiation in the G-13 position of the ETR; and two capsules are being irradiated inside tubular fuel elements in the WTR, with a third capsule scheduled for WTR insertion at a later date. The remaining capsule of the series, BMI-24-14, was discharged from the ETR at the end of Cycle 28 at an estimated fast-flux exposure at time of discharge of approximately 5.7×10^{21} nvt. Results of postirradiation impact tests performed on part of the specimens from this capsule were reported previously in BMI-1448.

Attempts to improve techniques for fast-flux measurements in the ETR are continuing. Stainless steel filings from specimens irradiated for one and for two cycles in the ETR were analyzed to determine the amount of manganese-54 produced in the $Fe^{54}(n, p)Mn^{54}$ reaction. The manganese-54 was separated from the cobalt-60 present by passing an acidified solution of the sample through an ion-exchange column and then measuring the amount of manganese-54 by means of a gamma-ray spectrometer. The calculated fast-flux values (corrected to include only the neutrons having energies greater than 1 Mev) are compared in Table A-1 with extrapolated low-power nickel dosimetry

*Wilson, W. B., "Device for Ultra-High-Pressure High-Temperature Research", Rev. Sci. Instr., 31, 3, 331-333 (1960).

Wilson, W. B., "High-Pressure High-Temperature Investigation of the Uranium-Oxygen System", J. Inorg. & Nuclear Chem., to be published.

Wilson, W. B., "Application of Ultrahigh-Pressure High-Temperature Equipment to Study of UO_2 Reactions", BMI-1328 (March 18, 1959).

measurements. The two values are in good agreement, indicating that the measurement of manganese-54 as determined from filings of irradiated samples is a feasible technique for determining total fast-neutron exposure. Further studies are being conducted to substantiate these initial findings.

TABLE A-1. COMPARISON OF FAST-NEUTRON-FLUX DOSIMETRY TECHNIQUES

Capsule	ETR Cycle	Irradiation Time, days	Estimated Fast-Flux Exposure, 10^{14} nv	
			Extrapolated ^(a) From Low-Power Nickel Dosimetry	Dosimetry From Manganese-54 Production ^(b)
BMI-24-1	11	7.52	2.38	2.10
BMI-24-1	11	7.52	2.38	2.22
BMI-24-17	6 and 7	18.8	2.00	2.15
BMI-24-17	6 and 7		2.00	2.15

(a) The flux values were obtained by extrapolation of N_L nickel-wire results to full-power operation.

(b) This technique involved analysis of filings from irradiated specimens of Type 347 stainless steel.

The ETR operated a total of 1360 megawatt-days during Cycle 31 and 5320 megawatt-days during Cycle 32. The reactor is now operating in Cycle 33, which is scheduled to terminate on December 12. The exposure received by the capsules during Cycles 31 and 32 will be determined after an analysis of dosimeter wires exposed during these cycles. The total estimated neutron exposures for both 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 tensile and fatigue specimens of one of the capsules exposed to about 5.0×10^{21} nvt early in 1961.

Development of High-Strength Niobium-Base Alloys

J. A. De Mastry, F. R. Shober, and R. F. Dickerson

Anticipated operation of the sodium-cooled Experimental Breeder Reactor, a fast reactor, above the iron-uranium eutectic (725 C) prevents utilization of AISI Type 347 stainless steel as a cladding material. Niobium-base alloys are being considered for EBR cladding applications since no eutectic reaction occurs between uranium and niobium, and alloying of niobium will produce material with high elevated-temperature strength. It has been shown (BMI-1417) that additions of chromium, vanadium, and zirconium improve the elevated temperature strength of niobium. Even though the high melting points of these alloys suggest that they should retain their strengths well above 1000 C, an experimental study was needed to determine the magnitude of strengthening.

Alloys of niobium containing 2.37 w/o chromium, 3.34 w/o zirconium, and 5.21 w/o vanadium and 1.56 w/o zirconium-1.09 w/o chromium were double melted in a consumable-electrode arc-melting furnace, forged to 1/2-in.-thick slabs at 540 C, and rolled to 0.040-in. sheet at the same temperature. Specimens for tensile and creep tests were given a 1-hr anneal at 1650 C prior to testing.

Tensile tests have been completed at 650, 800, and 1000 C, and the data are reported in BMI-1469. All alloys showed very little change in yield strength between 650 and 1000 C. On a weight percentage basis, chromium appears to be the most potent strengthener of niobium of the alloying additions being investigated.

On the basis of incomplete data, the stresses to produce deformation at rates of 0.001, 0.01, and 0.1 per cent per hr at 650 and 800 C are shown in Table A-2. Data are insufficient to establish rates at 1000 C. The final results may be refined somewhat as tests at other stresses are completed.

TABLE A-2. RESULTS OF ELEVATED-TEMPERATURE CREEP TESTS ON THREE BINARY NIOBIUM ALLOYS

Alloy	Test Temperature, C	Stress to Produce Deformation at Indicated Rate, psi		
		0.001 Per Cent Per Hr	0.01 Per Cent Per Hr	0.1 Per Cent Per Hr
Nb-2.37 w/o Cr	650	47,500	53,000	57,500
Nb-5.21 w/o V	650	52,000	58,000	64,000
Nb-2.37 w/o Cr	1000	11,000	17,500	24,000
Nb-3.34 w/o Zr	1000	7,000	13,500	20,000
Nb-5.21 w/o V	1000	15,000	20,000	25,000

The recrystallization temperature of materials cold worked 95 per cent and given a 1-hr anneal is being determined. Specimens of each alloy being studied have been annealed at 982 and 1038 C. These specimens are being prepared for hardness and metallographic examination. Annealing of other specimens at 1093, 1149, 1204, 1260, and 1316 C has not been completed. The recrystallization temperature will be estimated by hardness measurements and then confirmed by metallographic examination.

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 is being conducted to study the creep properties of Zircaloy-2 during irradiation at elevated temperatures. The total creep deformation as obtained under a high fast flux (1.0×10^{14} nv) at 650 F will be compared with that deformation obtained under similar stress and temperature conditions out of the reactor. Since strain aging appears to play a major role in determining the creep characteristics of unirradiated Zircaloy-2, a study of this phenomenon is being made.

Changes have been made in the design of the instrumented capsule to increase its heat-transfer properties to insure attainment of 650 F under maximum gamma-heating influence. It is necessary to combine and balance both gamma-energy heat and auxiliary electrical heat to attain and maintain a specimen temperature of 650 F during test. A major portion of this energy requirement will be produced by gamma heating with a

controlled auxiliary electrical circuit to supply the remainder. Control of the electrical power will compensate for any irregularities in the gamma-heat production. The original design based on the use of a 0.070-in. -thick MgO powder annulus as a principal barrier to heat flow proved inadequate. As pointed out in BMI-1473, it has been necessary to alter the heat-transfer characteristics of the original capsule design to compensate for reactor overpower situations. Several methods of achieving metal-fin contact across the 0.070-in. annulus between the capsule outer shell and the heater body have been investigated in thermal mock-up tests. At the present time, the most promising of these involves the use of 1/8-in. -OD by 0.030-in. -wall copper tubing rolled to 0.069 in. Approximately 15 such tubes equally spaced vertically around the heater body appear to provide the appropriate annulus thermal conductance.

Assembly of the in-pile capsule will commence after confirmatory thermal tests with copper tubes are run. The earliest reactor insertion date now under consideration is January, 1961. By that time WTR flux data being gathered during current operation should be processed.

An earlier study of thin sections of cold-rolled Zircaloy-2 using the electron microscope had shown striations which were believed to be related to deformation during rolling. Further studies of room-temperature deformation were made by straining a prepolished thin tensile sample and taking surface replicas after approximately 15 per cent strain. Examination of the replica showed slip to have occurred on more than one slip system within a single grain. However, where intersection of slip lines occurred, there was no evidence of an offset of either slip line. Thin sections for electron microscopy were prepared from the deformed specimen and viewed in the microscope. In grains with basal planes nearly parallel to the sample, surface striations were seen which were probably bases of slip on the prismatic planes. In some grains slip on two sets of prismatic planes were seen. These striations were segmented and did not appear to cross, indicating that slip on one set of prismatic planes did not penetrate active slip planes of a second set.

It is planned to strain a similar tensile sample at 350 C. Thin sections will be examined for changes in mode of deformation which may alter tensile and creep characteristics at this temperature.

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. X-ray diffraction studies of two high-burnup irradiated specimens show that both the natural and the enriched HfB_2 suffered complete loss of crystallinity and were degraded to amorphous structures. Lithium analyses of these specimens, for use in calculation of burnup, have also been completed.

Friction and Wear of Sliding Surfaces in Sodium Environments

W. A. Glaeser and J. W. Kissel

Research is being carried out to develop general principles which would be applicable to selection of materials and design of mechanical parts involving sliding surfaces in high-temperature molten sodium.

The friction and wear behavior of various materials resistant to corrosion in liquid sodium is being studied by measuring the stick-slip friction characteristics of a ball sliding on a flat in liquid sodium and under a carefully controlled gaseous environment. Interpretation of stick-slip traces is supplemented by analysis of surface films formed during exposure to sliding in sodium.

The friction and wear behavior of molybdenum, tungsten, Inconel X, Stellite 3, tungsten carbide, and titanium carbide has been investigated, and in each case, except for the two carbides, improved operation resulted when sodium was present between the rubbing surfaces. In the case of molybdenum sliding against itself, the improved behavior has been definitely correlated with the presence of a Na_2MoO_4 film. It was subsequently shown that formation of this film is thermodynamically feasible under any of the variety of temperature-environment conditions which might prevail during friction experiments.

Recently, thermodynamic analyses were applied to possible reactions involved between materials other than molybdenum and sodium. These indicate a favorable tendency toward formation for Na_2WO_4 and Na_2CrO_4 but a generally unfavorable tendency toward formation for Na_2CoO_3 and for Na_2C_2 . These results suggest logical explanations for the observed frictional behavior.

Techniques of applying some recently published theoretical analyses of stick-slip sliding to friction records are being developed. This effort is expected to provide information concerning the role of environmental factors with respect to changes in specimen-surface phenomena, especially films. A topical report will be prepared and will include the information obtained from the analysis of stick-slip behavior.

STUDIES OF ALLOY FUELS (AEC-DRD)

R. F. Dickerson

In an attempt to prevent the microsegregation which is thought to be the cause for the unfabricability of niobium binary alloys containing 30 to 60 w/o uranium, skull-melting studies have been initiated. The first ingot prepared cast well but reacted in one spot with the copper mold. The mold will be water cooled and casting of the final ingots will proceed. Two capsules are ready for irradiation.

The evaluation of the compounds ThB_4 , ThC , ThC_2 , ThN , Th_3P_4 , ThS , Th_3Si_2 , and ThSi , and the same compounds with the addition of 10 a/o of uranium, is progressing. Oxidation tests in air at 1200 F show that the beryllides and borides are comparatively oxidation resistant. The addition of uranium to the beryllide and monosilicide decreases oxidation resistance, while a similar addition to the boride and sesquisilicide improves oxidation resistance. Similar oxidation tests in 1200 F CO_2 also show that beryllides and borides are the best performers.

In Santowax at 350 C the aluminides and beryllides are the best performers of the thorium and thorium uranium compounds. The addition of small amounts of metal carbides to the thorium uranium dicarbides did not change their behavior, while similar additions to the monocarbides were of no help as the specimens decomposed in the desiccator.

Thorium-base binary alloys containing 5 and 10 w/o plutonium have been studied by metallographic techniques and appear to be single-phase alloys. The cast structure of the thorium-30 w/o plutonium alloy is, on the other hand, two phase. Hardness measurements indicate increasing hardness with increasing plutonium content. Heat-treatment studies are being initiated. Niobium-plutonium-silicon and niobium-plutonium-zirconium have been "drop" cast and are ready for further study. All alloys exhibiting strength and alloys exhibiting microstructures consisting of solid solutions or fine dispersions will be studied in more detail.

The compatibility study of PuO_2 with various metallic and ceramic materials has been continued. Ten volume per cent of an extraneous Pu_2O_3 was tentatively identified in the as-received material. Sintering tests of PuO_2 pellets in hydrogen and argon atmospheres at 1700 C indicate that either a chemical reduction or a thermal decomposition is occurring. Work has been initiated to clarify the situation.

Development of Niobium-Uranium Alloys

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

Niobium-uranium alloys are being developed as reactor fuels for use at elevated temperatures.

In an attempt to produce niobium alloys containing 30 w/o or more of uranium with structures amenable to fabrication, a program to investigate skull-melting

techniques has been initiated. Six ingots of compositions varying from pure niobium to niobium-50 w/o uranium will be prepared by means of consumable-electrode arc melting in a skull prior to pouring into a copper mold.

One ingot has been prepared from low-purity stock for the purpose of establishing melting procedures. An ingot of niobium was cast during preparation of the skull. The molten metal available filled the mold cavity satisfactorily, indicating that the skull had been built up to sufficient thickness. A cursory examination of this ingot revealed a very good surface with the exception of an area at the top or thickest section of the ingot. Apparently the heat capacity of the copper mold was not sufficient and overheating and alloying of the ingot with the mold resulted. In view of these findings, the mold will be water cooled, thereby reducing the possibility of a mold-metal reaction. Casting of the final ingots is ready to proceed as soon as the adequacy of the water cooling of the mold has been demonstrated.

The creep strength of the niobium-10, -20, and -30 w/o uranium and niobium-20 w/o uranium-20 w/o zirconium alloys will be determined at 1600 and 2000 F. Testing will be initiated as soon as specimens have been fabricated.

An irradiation program is under way. Two capsules have been assembled and are ready for irradiation. Each capsule contains 15 fuel pins which will be irradiated to a target 1 total a/o burnup at temperatures of 1600 and 1800 F.

Development of Uranium-Containing Thorium Compounds

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

Compounds containing thorium and thorium-uranium are being studied with the aim of developing a high-temperature nuclear fuel suitable for use in a thermal breeder reactor. Low thermal-neutron cross section, high melting point, and high thorium (uranium) density were properties considered in choosing the compounds to be studied. These are: ThB_4 , ThC , ThC_2 , ThN , Th_3P_4 , ThS , Th_3Si_2 , and ThSi , and these same compounds with 10 a/o additions of uranium.

Oxidation tests in dry air at 1200 F have been completed, and the results are presented in Table B-1. It is readily apparent that the beryllides and borides are comparatively oxidation resistant, while the high rates at which the other compounds oxidize preclude their exposure to high-temperature air. It is interesting to note that in the case of the beryllide and the monosilicide the addition of uranium to the thorium compound apparently decreases oxidation resistance, while in the case of the boride and the sesquisilicide the addition of uranium appears to improve the oxidation resistance. However, the presence of uranium may or may not be the cause of changes in oxidation resistance, as slight deviations from stoichiometry of a compound may produce the same effect. Preliminary results of corrosion tests in 1200 F CO_2 indicate that the beryllides and borides are again the best-performing compounds by a large margin.

The corrosion resistance of thorium and thorium-uranium compounds in 350 C Santowax has been determined. The results are reported in Table B-2. The aluminides and beryllides are the best performers, while the silicides are less corrosion resistant than the carbides tested. The addition of small amounts of low-cross-section metal

TABLE B-1. OXIDATION OF THORIUM AND THORIUM-URANIUM COMPOUNDS
IN DRY AIR AT 1200 F

Compound	Time of Test, hr	Average Weight Gain During Test Period, $\text{mg}/(\text{cm}^2)(\text{hr})$
$\text{Th}_2\text{Al}^{(a)}$	0.66	144
ThBe_{13}	16.5	0.02
$(\text{Th}_9\text{U})\text{Be}_{13}$	5.0	1.0
ThB_4	18.75	0.22
$(\text{Th}_9\text{U})\text{B}_4$	6.75	0.07
ThSi	0.33	268
$(\text{Th}_9\text{U})\text{Si}^{(a)}$	0.33	339
$\text{Th}_3\text{Si}_2^{(a)}$	0.16	597
$(\text{Th}_9\text{U})_3\text{Si}_2$	0.16	202

(a) Average of two tests.

TABLE B-2. THE CORROSION RATES OF THORIUM AND THORIUM-
URANIUM COMPOUNDS IN 350 C SANTOWAX

Compound	Corrosion Rate, $\text{mg}/(\text{cm}^2)(\text{day})$
$(\text{Th}_9\text{U})_2\text{Al}$	0.04
ThBe_{13}	0.3
$(\text{Th}_9\text{U})\text{Be}_{13}$	0.07
ThSi	54
$(\text{Th}_9\text{U})\text{Si}$	39
Th_3Si_2	9
$(\text{Th}_9\text{U})_3\text{Si}_2$	15
ThC-10 w/o UC	2
$\text{ThC-10 w/o UC-5 w/o SiC}$	3
$\text{ThC}_2\text{-10 w/o UC}_2$	0.3
$\text{ThC}_2\text{-10 w/o UC}_2\text{-5 w/o ZrC}$	0.2
$\text{ThC}_2\text{-10 w/o UC}_2\text{-2.5 w/o Mo}_2\text{C}$	0.4
$\text{ThC}_2\text{-10 w/o UC}_2\text{-5 w/o NbC}$	0.2
$\text{ThC}_2\text{-10 w/o UC}_2\text{-5 w/o SiC}$	0.7

carbides to the thorium-uranium dicarbides did not significantly affect their corrosion resistance. Thorium-uranium monocarbides with additions of zirconium carbide, niobium carbide, or molybdenum carbide were not tested since they decomposed while stored in a vacuum desiccator.

Presently corrosion resistance in boiling water and CO_2 are being determined. The thermal-expansion, thermal-conductivity, and compressive-strength characteristics of the most promising compounds will be investigated.

A Study of Plutonium-Containing Fuel Alloys

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

A program to develop irradiation-resistant plutonium-bearing alloys is being conducted. Alloy systems being studied include niobium-base, thorium-base, uranium-molybdenum base, and uranium-niobium-base alloys. Selection of these systems for study is based on the irradiation behavior of similar uranium-containing alloys. Initial studies of these systems are being conducted mainly by metallographic examination and hardness testing.

Metallographic examination of thorium-base alloys show the as-cast structures of thorium-5 and -10 w/o plutonium alloys to be essentially one phase. The as-cast structure of a thorium-30 w/o plutonium alloy is two phase, probably alpha-thorium solid solution and $\text{Th}_6\text{Pu}_{13}$. An as-cast thorium-10 w/o plutonium-10 w/o zirconium alloy contained a small amount of second phase. The effect of heat treatment on the microstructures of these alloys will be investigated. Hardness measurements indicate increasing hardness with increasing plutonium content and a substantial hardening effect from the ternary zirconium addition. Diamond-pyramid hardness numbers for the as-cast iodide thorium-5, -10, and -30 w/o plutonium and the thorium-10 w/o plutonium-10 w/o zirconium alloys are 48, 53, 74, and 89, respectively.

Niobium-25.6 w/o plutonium-6.1 w/o silicon, niobium-10 w/o plutonium-10 w/o zirconium, and niobium-10 w/o plutonium-30 w/o zirconium alloys have been prepared by arc-melting and drop-casting techniques. It is hoped that the silicon-containing alloy will consist of a dispersion of PuSi_2 in a niobium matrix and that the zirconium-containing compositions will be solid-solution alloys. The investigation of binary niobium-plutonium alloys has been dropped as the microstructures, which consist of niobium matrices with plutonium in the grain boundaries, suggest poor irradiation resistance.

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 alloys will be prepared by arc-melting techniques. The effect of ternary additions of zirconium on the uranium-niobium-base alloys will also be investigated. It is hoped that these alloys will behave as similar alloys containing no plutonium.

Alloys exhibiting strength (as evaluated by hardness measurements) and alloys exhibiting microstructures consisting of solid solutions or fine dispersions of compound in a nonfissile metal matrix will be studied in greater detail.

Compatibility of PuO₂

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

The purpose of this program is to study the compatibility of PuO₂ with various metallic and ceramic materials. Heat treatments of powder mixtures and abutted surfaces of PuO₂ with the material in question will be conducted. The appearance of new phases will be followed by X-ray diffraction measurements, metallographic examinations, and observation of changes in the physical condition of the compacts.

The results of a preliminary screening procedure using calculations of standard free energies to indicate metal-PuO₂ reactions are summarized in BMI-1455.

Using petrographic techniques, the as-received PuO₂ particles were determined to average 9.4 μ in diameter with generally submicroscopic grain size and to be spherical in shape. Ten volume per cent of an extraneous phase was identified tentatively as Pu₂O₃.

Sinterability studies showed that as-pressed green density increased with compacting pressure and reached a maximum of 60 per cent of theoretical. Sintering in argon for 1 hr at a temperature in excess of 1600 C increased the density to 90 per cent of theoretical.

During the past month, a series of pellets was sintered in argon and a duplicate series in hydrogen for 1-1/2 hr at 1700 C. In each case, individual pellets were sintered on trays of molybdenum, tantalum, tungsten, Al₂O₃, BeO, MgO, and ZrO₂. In both atmospheres, melting and alloying with the trays occurred. The argon atmosphere gave slightly less drastic reactions. Comparison of the degree of reaction between PuO₂ and various refractory trays indicated that MgO is the most compatible material with PuO₂ at this temperature.

It appears that either a chemical reduction or thermal decomposition from PuO₂ to Pu₂O₃ is occurring. The Pu₂O₃ melts at 1630 C or below the sintering temperature involved. Reactions at the PuO₂-sintering tray interface may also be involved or the results may indicate melting of the Pu₂O₃ present in the as-received powder. Work to clarify the situation is to be initiated. Sintering and compatibility studies will be conducted at temperatures below the melting point of Pu₂O₃ in air, argon, and hydrogen atmospheres. Attempts to remove the Pu₂O₃ from the powder will involve air oxidation of loose powder at 800 C for several hours prior to pressing.

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

J. B. Melehan, S. G. Epstein,
M. Kangilaski, and F. A. Rough

The objective of this program is to understand the important causes of fission-product release in UO_2 and other important refractory reactor fuels and to collect data needed for design of fuel elements. Studies have been in progress on diffusion of xenon-133 during postirradiation heating, on characterization of sintered materials, and on the continuous release of fission products during irradiation of sintered materials. The research on postirradiation diffusion of xenon-133 has shown that diffusion in single crystalline UO_2 is much lower than in sintered materials, being about $2 \times 10^{-17} \text{ cm}^2$ per sec at 1400 C.

Characterization of UO_2 and Model of Gas Release

In this research, quantitative metallographic measurements have been made to determine the surface area of the porosity for several specimens. In a 90 per cent dense specimen, excellent correlation was obtained between the value obtained, corrected for the amount of closed porosity, with the surface area determined by gas adsorption, indicating that at least in this case gas adsorption is a satisfactory process for determining the open surface area before irradiation. So far, there is little information available in the literature concerning the effect of irradiation upon the open surface area.

Several heat treatments have been made upon sintered material in dry hydrogen to provide control of the oxygen/uranium ratio and eliminate oxygen absorbed on the surfaces during storage. It was found that the oxygen/uranium ratio could be reduced to 2.002 ± 0.003 to 2.004 ± 0.003 readily. It was also found that for oxygen/uranium ratios of less than about 2.02 the lattice parameter determined by X-ray diffraction varied linearly with the oxygen/uranium ratio. Presumably, however, the correlation between lattice parameter and oxygen/uranium ratio as determined by ignition or polarographic techniques will vary slightly, depending on the impurities and history of the material being used.

In-Pile Studies

In order to better establish the meaning of earlier experiments on sintered material, a single-crystal plate specimen of UO_2 was irradiated at 500, 1000, and 1500 F for periods of 48 hr; and the fission gases krypton-85m, krypton-87, xenon-133, and xenon-135 were analyzed. These experiments were conducted so that effectively all of the surface recoils would lodge in a surrounding holder of molybdenum. Since the specimen had only about 2 cm^2 of geometric surface area as compared to open surface areas of about 1000 cm^2 or more in the earlier sintered specimens, any surface releases should be much lower for the single crystal. The calculated surface recoil fraction $(R/B)^*$ which would lodge in the molybdenum for the single-crystal experiments was 4×10^{-3} . This compares with an estimated surface release rate (R/B) of about 5×10^{-6} based on

*Where R = rate of release, atoms per sec.

B = rate of production, atoms per sec.

earlier experiments. The release rates from the single-crystal experiments are given in the following tabulation:

<u>Run</u>	<u>Temperature, F</u>	<u>Release Fraction^(a), R/B</u>
1	500	2×10^{-5} to 5×10^{-5}
2	1000	1×10^{-4} to 3×10^{-4}
3	1500	2×10^{-4} to 6×10^{-4}

(a) Includes data for krypton-85m, krypton-87, and xenon-135 at equilibrium and xenon-133.

These results are interpreted to mean that most of the surface recoils remain in the surrounding molybdenum, the number released being dependent upon the temperature.

While additional experiments are needed to more fully understand the mechanism of releases from UO_2 observed at 500 to 1500 F, the data can best be interpreted on the basis of a vaporization release induced by the passage of recoils through open surfaces.

GENERAL FUEL-ELEMENT DEVELOPMENT (AEC-DRD)

S. J. Paprocki

In previous studies, techniques for the fabrication of cermet fuels containing 80 volume per cent UO_2 were developed, and physical and mechanical properties of these materials were determined. Based on these studies, UO_2 -chromium, UO_2 -molybdenum, and UO_2 -niobium specimens are being prepared for irradiation testing.

Techniques are being developed for the fabrication of cermets embodying approximately 80 volume per cent UN. The current studies are concerned with determination of sintering characteristics and compatibility with various potential metallic matrices.

Various fabrication techniques are being investigated for the preparation of tubular fuel elements. Most of the techniques selected for this study are nonconventional; they were selected on the basis of being capable, if successful, of producing a quality product at a low relative cost.

A study is being made of the mechanism of solid-state bonding of metals by the application of heat and pressure. The initial phase of this work was concerned with the determination of the effects of time, temperature, pressure, grain size, surface roughness, and impurities on the self-bonding of copper. Future work will be concerned with the evaluation of the self-bonding characteristics of metals other than copper.

Fabrication of Cermet Fuel Elements

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

Several methods of producing 60 to 90 volume per cent UN cermets of 90 per cent of theoretical density have been considered. Currently sintering and gas-pressure-bonding techniques are being investigated. These fabrication techniques are also being used to select compatible cermet matrix metals.

Green-pressed cermets containing 60 volume per cent UN dispersed in molybdenum and niobium have been vacuum sintered for 2 hr at 3000 F. The molybdenum-cermet density was unchanged by sintering. The niobium-cermet density was increased to 99.5 per cent of theoretical, a 41.5 per cent increase over green-pressed density. A microscopic examination of the niobium cermet showed that an extensive reaction between the UN and niobium had occurred and that a molten phase had been present in the grain boundaries to promote sintering. The molybdenum-matrix cermet structure indicated slight or no reaction between the UN and molybdenum. This suggested that the reaction with niobium is probably not due to the thermal dissociation of UN into nitrogen and free uranium. Vacuum sintering 60 volume per cent UN-Type 347 stainless steel and chromium cermets for 2 hr at 2400 F resulted in density losses of 5.1 and 3.6 per cent, respectively. Microscopic examinations of the cores indicated little or no reaction had occurred between the matrix metals and the UN.

In addition, 80 volume per cent UN-Type 347 stainless steel, -molybdenum, -niobium, and -chromium green-pressed cermets have been gas-pressure bonded for

3 hr at 2200 F under a helium gas pressure of 10,000 psi. The UN particle size was varied through the range of minus 100 plus 400 mesh to determine the size effect upon gas-pressure-bonded densities and microstructures. Gas-pressure-bonded densities ranged from 82 to 89 per cent of theoretical. Metallographic examinations are being conducted to determine compatibility and structure differences.

Also, UN cermetes are being prepared for gas-pressure bonding to determine pressure-bonding parameters and the effect of fuel loading.

Irradiation of Cermet Fuels

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

This program involves radiation-damage studies of 80 volume per cent UO_2 -refractory metal fuels; the metals of interest are chromium, molybdenum, and niobium. Cylindrical specimens of each cermet are to be irradiated in two capsules under conditions combining high temperature and high fission-heat-generation rates.

During November, preliminary studies of the various specimen parameters as they pertain to capsule design were continued. Tentative values thus far established are: uranium enrichment, 20 per cent; specimen-surface heat flux, approximately 2×10^6 Btu/(hr)(ft²); and approximate specimen center-line temperatures, 2550 F for UO_2 -molybdenum, 2500 F for UO_2 -niobium, and 2250 F for UO_2 -chromium.

According to present plans, design of the two capsules will be identical. Each will contain two specimens of each cermet; they will be irradiated in similar neutron fluxes for different time intervals to achieve two significantly different burnup levels. Since the various cermetes have different thermal conductivities, it will be necessary to vary the specimen diameters and to locate the specimens at vertical positions in the reactor facility selected to achieve desired specimen center-line temperatures and burnup rates.

The study concerning the selection of the cladding material and the fabrication of the specimens is continuing. Specimens are being prepared with ferritic stainless steel and Hastelloy X cladding. Methods of incorporating void space to absorb fission gases at the ends of the fuel and within the cladding are being investigated. Pressed pellets of Al_2O_3 powder and fused alumina containing drilled holes are two materials being tested as gas sinks in this study. The fuel section of these dummy specimens will contain 80 volume per cent UO_2 with molybdenum, since this metal represents the most dissimilar thermal-expansion relationship between core and cladding.

The specimens will be gas-pressure bonded, tested by thermal cycling under simulated reactor conditions, and examined metallographically. The irradiation specimens will then be fabricated on the basis of this evaluation.

Development of Low-Cost Tubular Fuel Elements

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

Techniques are being developed for fabricating tubular fuel elements of consistent quality with a minimum of cost. The excellent thermal and structural properties inherent in the tubular configuration make it an attractive design for use in high-power-density fuel-element applications.

Studies during the past month have been concentrated in two areas. One of these is concerned with determining the burnout characteristics of some of the cold-binder plastics being considered for use in forming metal-powder mixtures. Mixtures of minus 200-mesh Type 316 stainless steel powder with polyvinyl alcohol, methyl cellulose, and Aeryloid B-7 were prepared and rolled into thin sheet. Heating these mixtures at 2300 F for 1 hr in hydrogen resulted in considerable sintering of the sample containing the Aeryloid mixture. Some sintering also took place in the samples using the polyvinyl alcohol and methyl cellulose, but not to the same extent. It was found that 75 w/o of the Aeryloid binder was removed during the sintering operation, leaving 3 to 4 w/o binder residue in the sintered sample. Attempts to remove the binders by first heating at 1000 F in air resulted in complete binder removal, and the sample returned to its original powder form. Future studies will be concerned with reducing the weight percentage of the plastic binder in the mixture by diluting the plastic while maintaining its forming characteristics. Optimum powder-binder ratios will be determined for both extruding and rolling operations, and the effect of various heat treatments on the amount of binder residue and degree of sintering produced will be studied.

Another technique being investigated for producing low-cost tubular fuel elements is based on a commercial process for producing thin-walled tubing economically. The annulus between two concentric cladding tubes is filled with a UO_2 -stainless steel core powder mixture. After evacuating and sealing the sample element is press flattened and rolled. Rolling schedules and equipment are chosen with the aim of imparting a maximum of the total work and reduction into densification and bonding of the core material. Subsequent to rolling the flattened tube is opened by mechanical, pneumatic, or hydraulic means into the original tubular shape. The results of rolling a few of these tubes indicate that considerable care is needed to insure a uniform loading of the powder. Non-uniform loadings cause local thinning and wrinkling of the tube walls during rolling. Additional work is planned using this concept with concentration on better loading techniques and varying fabrication parameters to obtain more uniform core dimensions.

Factors Affecting Pressure Bonding

G. W. Cunningham, D. C. Carmichael, and J. W. Spretnak

An investigation of solid-phase bonding of metals under application of heat and pressure is being conducted in an effort to establish the mechanism of the process.

has been completed on a broad study of the mechanism of pressure bonding utilizing copper, and a topical report is being prepared on this phase. The program is directed toward examining the mechanism in more detail and in metals other than

The effects of pressure, temperature, time, grain size, grain growth, impurities, and surface roughness on the self-bonding of copper have been investigated and the mechanism of bonding established. In order to produce a metallurgically sound bond, i. e. , no discontinuities or changes in the structure in the bond region, the first requirement is the application of sufficient pressure to place the surfaces in intimate contact. In copper, the pressure required is equivalent to the Meyer hot hardness at which the penetration of the indenter is equal to the average depth of the surface asperities.

At the time intimate contact is attained, the bond interface may be described as an irregular jogged boundary similar in appearance to a high-angle grain boundary. Plastic deformation occurs in the region adjacent to the interface, and thus the interface is surrounded by stressed metal. The interface may also contain impurity particles and microvoids.

These microvoids are created during the bonding operation by insufficient flow of metal into regions at the base of very small asperities or by the condensation of excess vacancies at impurity nuclei. The total void volume is a maximum within a few minutes after obtaining intimate contact. After this time, the total void volume may increase, and the number of voids may decrease with the larger voids growing by the elimination of smaller voids. Whether or not the void volume decreases depends upon the availability of vacancy sinks, and in copper the most effective sinks have been found to be grain boundaries.

Grain growth across the bond region and thus the elimination of the interface has been found to be dependent upon the presence or absence of microvoids. At temperatures high enough for grain growth to proceed at a reasonable rate, the driving force to straighten the irregular-shaped interface is sufficient to provide grain growth across the bond region if there are only a few microvoids. If the number of microvoids is large (10^6 voids per cm^2), grain growth can occur only if the grain size is much smaller than the size usually existing at that temperature or if recrystallization or an allotropic transformation occurs during bonding. The latter has been investigated for zirconium and iron.

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

S. W. Porembka

The objective of this program is to refine and further develop the gas-pressure-bonding process as a method for fabricating low-cost ceramic, cermet, and dispersion fuel elements. Since it is possible to achieve both fuel densification and cladding in a single bonding operation, the technique appears to be a promising method for producing quality fuel elements at a reduced cost. Of the fuel systems considered in this program, the uranium dioxide fuel as clad with Type 304 stainless steel has received the major emphasis. The approach utilized in this investigation has involved studies of initial oxide compacting techniques, characteristics of pressure-bonded oxides, and possible fuel geometries.

Various types of uranium dioxide powders have been evaluated through studies of their initial compacting and pressure-bonded characteristics. Of the oxides studied, mixtures of ceramic grade and fused oxides offer the best characteristics for successful pressure bonding. The effects of pressure bonding on the properties of Type 304 stainless steel have also been defined. Current efforts are directed toward preliminary process specifications for the most promising fuel elements.

Uranium Dioxide Compaction Studies

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

Studies of uranium dioxide compacting characteristics have concerned the development of high-density green fuel cores which will provide good dimensional control and high final densities upon pressure bonding. Present studies involving ceramic and fused uranium dioxide mixtures are aimed at optimizing the compacting and pressure-bonding behavior for process specifications. A brief comparison of the various binders utilized in the program has also been included with a similar objective.

Additional flat platelets of fused and ceramic oxide mixtures were compacted for use in powder-component compartmented flat plates. Mixtures containing from 60 to 75 w/o fused oxide yielded densities from 72 to 85 per cent of theoretical. To achieve the higher densities, it was necessary to use a Ceremul "C" binder with mixtures containing 70 to 75 w/o fused oxide. In all cases, compacts with good handling characteristics were achieved.

Gas-Pressure-Bonding Studies

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

The gas-pressure-bonding technique has been used to fabricate Type 304 stainless steel-clad UO_2 rod and flat-plate elements and flat-plate assemblies. Excellent dimensional tolerances have been achieved when high-density cores have been used, and, in

general, dimensional control has been found to be dependent on the initial oxide and method of compaction. Investigations are being continued to develop process specifications for the fabrication by gas-pressure bonding of several basic fuel-element shapes utilizing high-density green-pressed oxide cores.

To help establish final process specifications on the green-pressed oxide cores, the study of the effect of different binders and methods of binder removal upon the final stoichiometry and density of oxide mixtures is being continued. The results obtained thus far are given in Table E-1. From these results and other data compiled throughout this program, it has become apparent that polyvinyl alcohol cannot be considered as a possible binder for this process.

TABLE E-1. EFFECTS OF BINDERS AND METHODS OF BINDER REMOVAL UPON THE STOICHIOMETRY AND DENSITY OF UO_2 MIXTURES^(a)

Tube	Binder	Method of Binder Removal	Oxygen-to-Uranium Ratio	Density ^(b) , g per cm
106	2 w/o polyvinyl alcohol semislurry	(c)	2.09	10.5
108	Ceremul "C" (1 drop per 3 g of UO_2 powder)	(d)	2.017	10.7
109	1/2 w/o of saturated solution of Carbowax 6000 and methyl alcohol	(d)	2.013	10.3
110	2 w/o of saturated solution of Carbowax 6000 and methyl alcohol	(d)	2.014	10.5
111	2 w/o polyvinyl alcohol semislurry	(d)	2.065	10.6

(a) The UO_2 used in these studies was a 60 w/o fused-40 w/o ceramic mixture pressed at 50 tsi. All tubes were pressure bonded at 2100 F for 3 hr at 10,000 psi.

(b) Densities by pycnometer measurements.

(c) Specimen was evacuated to 5 μ prior to heating. Successive evacuations to 5 μ were performed at temperatures of 400 to 1200 F with temperature intervals of 100 F. At 1200 F, the sample was held under vacuum for 6 hr. Throughout the heating, the vacuum did not exceed 70 μ .

(d) Specimen was evacuated to 5 μ prior to heating. Sample was then heated to 1200 F and held for 1-1/2 hr under vacuum.

A ninth subassembly measuring 1 by 2-1/4 by 6 in. and containing five compartmented fuel plates has been pressure bonded. In this case, each fuel plate contained nine sintered uranium dioxide fuel cores. This assembly is being sectioned for a complete dimensional study in order to define the degree of dimensional control that can be achieved with compartmented flat-plate assemblies.

Four additional flat-plate specimens containing pressed and sintered stainless steel powder frames and green-pressed UO_2 cores have been pressure bonded. These elements contained cores of a 75 w/o fused and 25 w/o ceramic (as received) UO_2 mixture cold pressed at 50 tsi to densities of 74.1, 74.5, and two at 77.0 per cent of theoretical. These cores were loaded into powdered frames with densities of 74.0, 77.0, 77.0, and 82.0 per cent of theoretical, respectively. The assemblies were pressure bonded at 2100 F for 3 hr at 10,000 psi. Simultaneous densification of the core and frame components was achieved; however, all surfaces over the core area were rough and only one element measured the same thickness over the core and frame, within 5 mils. Other specimens are being planned in which pressure plates will be inserted next to the cladding to help overcome the difficulties described.

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. The objective of this program is to lower the cost of power produced from nuclear energy by lowering the costs of manufacturing, using, and reprocessing reactor fuels. Foremost among the various ways in which these costs can be reduced are: (1) lowering the cost of fabricating the fuel, (2) increasing the temperature at which the reactor coolant and fuel operate, (3) increasing the life of the fuel in the reactor, and (4) decreasing the cost of reprocessing the fuel. Uranium carbides seem capable of producing dramatic savings in connection with the second and third of these items, and the costs of fabricating and reprocessing of fuel elements using uranium carbides will probably be quite similar to the costs of fabricating and reprocessing fuel elements containing uranium oxide or uranium metal.

To assist in the accomplishment of the aforementioned objective, an integrated program of research on the preparation and properties of bulk uranium carbides is in progress at Battelle. This 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, physical, and chemical 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) irradiations designed to produce basic information on the nature of radiation-damage processes in the structure and upon the integrity of uranium carbides. These studies have now been in progress for over a year and a half. During this time the emphasis in the research effort has been in the first four areas mentioned above. During the next year, emphasis will be shifted gradually toward property studies and toward analysis of the nature of radiation-damage processes in uranium carbides.

Significant developments during the last month include a finding to the effect that the use of outgassed commercial graphite in the skull-melting and casting operation may result in better control of the carbon content of the castings. It has also been found that uranium carbide is wet by aluminum-12 w/o silicon alloy and copper-20 w/o silicon alloy at 2000 F (1090 C).

Alternate Fabrication Methods for UC

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

A study of solid-phase fabrication methods for uranium carbide is being made to devise economical fabrication methods.

Six uranium carbide powder compacts have been isostatically hot pressed at 1430 C under a 10,000-psi pressure for 6 hr. All compacts were sealed in niobium capsules under high vacuum by means of an electron-beam welder before hot pressing. Two of the compacts used had been presintered under an argon atmosphere at 1700 C prior to hot pressing. The powder used for these compacts contained 4.64 w/o carbon. It was obtained by crushing buttons arc melted at Battelle. Microscopic examination of

duplicate, presintered specimens showed the structure to contain the UC phase only, with no grain-boundary uranium visible. The densities of these presintered compacts were about 70 per cent of theoretical.

The other compacts used were prepared from crushed commercial shot and Battelle buttons, both of stoichiometric carbon content. The shot was obtained from Vitro Laboratories. The minus 100-mesh fraction of both materials was used in this series. The powder was cold pressed, vacuum degassed to remove the Carbowax binder, and then sealed and hot pressed as already described.

Density measurements and microscopic examinations of these fabricated specimens are in progress.

Another series of capsules containing cold-pressed compacts prepared from the stoichiometric Battelle and Vitro materials has been prepared for hot pressing at 1480 C. The variable in this series is mesh size with the following fractions selected: minus 80, minus 100, minus 200 plus 325, and minus 325.

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. Ten melts of uranium carbide were made using commercial electrode-grade graphite as both electrode tip and alloy addition. Although melting conditions were constant, the analyzed composition of the castings produced varied as much as ± 0.3 w/o carbon from the desired composition.

This month two of a series of ten melts were made using vacuum-outgassed AGX commercial grade graphite. The melting conditions were the same as used for the series of melts employing untreated AGX commercial-grade graphite, and the analytical results, although this series of melts is incomplete, indicate that compositional control using vacuum-outgassed AGX commercial-grade graphite is better than compositional control using untreated AGX commercial-grade graphite. The carbon contents of the two melts were within 0.1 w/o.

Next month AGOT and TSF reactor-grade graphite will be studied by observing carbon-content fluctuations of the cast UC under the same melting conditions as used in runs involving the commercial-grade material. The series of melts using outgassed AGX graphite will be completed. Studies of graphite-tip erosion will also be made.

Mechanical and Physical Properties of Uranium Monocarbide

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

The potential of uranium monocarbide as a nuclear fuel is dependent upon its high-temperature properties. Obtaining reliable measurements of mechanical and physical properties, such as strength, hardness, and thermal conductivity, at elevated temperatures is one of the prime objectives of this program. In addition, the effect of alloy additions, protective coatings, and heat treatment on properties must be evaluated.

While specimens for property measurements are being prepared, attention during this period centered upon various coatings and techniques for applying them to UC. Several metals and alloys were investigated in terms of providing an adherent coating for UC specimens by hot dipping. The metals and alloys examined as dip coatings for uranium carbide are as follows: Aluminum-12 w/o silicon, copper, copper-20 w/o silicon, nickel, lead, lead-62 w/o tin, tin, and zinc. Specimens were immersed in molten baths of each of these alloys for 1/2 min. At temperatures near the melting points of the materials tested no wetting of UC was observed. However, in two cases, aluminum-12 w/o silicon alloy and copper-20 w/o silicon alloy, the temperature was raised to 2000 F. At this bath temperature wetting was observed in both instances by metallographic inspection. Although the coatings were not continuous in either case, they were adherent at those points where wetting had occurred.

Vapor deposition was also attempted as a means of applying coatings. The alloy, chromium-50 w/o nickel, was heated in a crucible to 3000 F. Vapor passed through an orifice in a plate which covered the crucible and coated a UC specimen which was held above the orifice for 1/2 hr. The coating produced was roughly 1/32 in. thick, and, although the majority of the coating was porous, the coating exhibited a thin, even layer next to the UC which appeared to be mechanically bonded to the UC surface. In a second method, aluminum and copper were each vaporized by a tungsten resistance coil heated to about 3000 F. The vapor was condensed on a UC specimen held near the hot filament containing the metal. Copper appeared to offer a more adherent coating than aluminum, although in both cases the bond was only mechanical in nature. From a handling standpoint, coatings produced in this manner appeared too fragile to be given further consideration.

Alloys of UC which have recently been prepared and which are to be evaluated in terms of electrical resistivity, corrosion resistance, and metallographic analysis are tabulated below with their room-temperature hardnesses.

<u>Alloy</u>	<u>Hardness, KHN</u>
UC-10 w/o NbC-5 w/o vanadium	1370
UC-5 w/o NbC-5 w/o vanadium	1240
UC-10 w/o VC-5 w/o niobium	1660
UC-10 w/o NbC-5 w/o molybdenum	1540
UC-5 w/o NbC-5 w/o molybdenum	1280

Further attempts to coat UC specimens by varying the immersion period in the metal bath, the bath temperature, and the surface finish of the uncoated UC specimen are planned. Specimens for measuring thermal conductivity, high-temperature resistivity, and high-temperature hardness of UC are being prepared.

Uranium Monocarbide Diffusion Studies

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

The rate of self-diffusion of uranium in uranium monocarbide is being investigated using a tracer technique. Diffusion rates in uranium monocarbide are useful in understanding the atomic structure of this material and in predicting the elevated-temperature behavior of this material. Previously in this study, the rate of interdiffusion of carbon and uranium was determined between 1600 and 2000 C by holding liquid uranium saturated with carbon in contact with graphite and metallographically measuring the thickness of the interdiffusion zone. The rate of self-diffusion of uranium monocarbide is now being studied using a diffusion couple consisting of a foil of enriched uranium sandwiched between two lengths of depleted uranium monocarbide rod; and it is planned to study the rate of self-diffusion of carbon in uranium monocarbide by bonding a length of monocarbide containing carbon-14 to a length of monocarbide made with normal carbon.

In the study of the rate of self-diffusion of uranium, a couple was bonded and then annealed for 480 min at 2000 C. The couple was sampled, and these samples have been submitted for analyses for enrichment using the 0.184-Mev gamma-ray activity of uranium-235. A preliminary calculation of the activation energy for the self-diffusion of uranium in uranium monocarbide was made previously. (The results of this calculation were reported in BMI-1464.) It is planned to complete this study after evaluation of the couple annealed for 480 min at 2000 C.

A technique has been devised for studying the rate of self-diffusion of carbon in uranium monocarbide. The carbon-14 for this study has been received and uranium monocarbide having a specific activity of 80 to 100 mc per g is being prepared. It is planned to bond the monocarbide rod containing the carbon-14 to a rod containing carbon-12 using a 0.001-in. foil of normal uranium. After appropriate heating the couple will be ground in 0.0005 to 0.0009-in. increments starting with the carbon-12 end of the couple. The beta activity of the ground face will be measured after each slice is removed in order to determine the concentration-penetration curve for the couple. It is planned to determine the rate of self-diffusion of carbon in the temperature range from 1600 to 2000 C.

Future work will include the analysis of uranium diffusion for the couple annealed for 480 min at 2000 C and the preparation and analysis of couples for the study of the rate of self-diffusion of carbon in uranium monocarbide.

Effects of Radiation on UC

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

The 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 are presently being irradiated in four NaK-filled capsules in the BRR. During the initial reactor cycle, the neutron flux was

found to be approximately 20 per cent lower than had been anticipated, causing lower temperatures than expected. However, by incorporating inert gas mixtures in the outer gas annuli of the capsules, temperatures nearer to the design range (1400 to 1600 F) have been attained. It has been decided that the burnups of the specimens in two of these capsules should be approximately 0.15 a/o of the uranium while those of the remaining two capsules should be approximately 0.7 a/o of the uranium.

PHYSICAL RESEARCH (AEC-DR)

F. A. Rough

A program supported by the AEC Division of Research and directed toward development of high-quality UO_2 single crystals is continuing.

Growth of UO_2 Single Crystals

J. D. Jackson and C. A. Alexander

During the past month, effort was continued on modifying the resistance furnace, described in previous monthly reports, so that it would be possible to maintain a temperature of 2800 C for an extended period of time. Modification of this apparatus was completed late in this month, and preliminary heatings indicate that the upper electrode will not fail as had previously occurred. In tests completed to date a temperature of 2500 C was maintained with relative ease.

While the furnace modifications were being made, additional experiments on growth of single crystals of UO_2 were made employing the thermal decomposition of uranyl chloride, UO_2Cl_2 . In these experiments UO_2 was chlorinated to the uranyl chloride at about 700 C, and the vapor was carried by a carrier gas to a region of higher temperatures where decomposition back to UO_2 and chlorine would occur. Attempts to carry this reaction out with HCl(g) as the chlorinating compound were unsuccessful; hence, the reaction: $\text{UO}_2 + 2\text{HCl} \rightarrow \text{UO}_2\text{Cl}_2 + \text{H}_2$ was not found to proceed at a rate suitable for crystal growth at 700 C. (With U_3O_8 in place of UO_2 , the reaction proceeds, since in this case water is a product and its free energy of formation is enough to make this reaction thermodynamically more favorable.)

Use of chlorine rather than HCl also produces UO_2Cl_2 , but in this case the single crystals are not stoichiometric UO_2 , but U_4O_9 or U_3O_8 single crystals. Had it been possible to cause the reaction to proceed with HCl , the hydrogen would have allowed the crystal growth to proceed under reducing conditions, thus producing UO_2 . It would appear that thermal decomposition of UO_2Cl_2 is a convenient means of growing crystals of higher oxides but cannot be used, without modification, for UO_2 because of the thermodynamics of the reaction. It may be possible to develop a two-stage process in which the last stage, decomposition of UO_2Cl_2 , takes place in presence of hydrogen, depositing UO_2 .

During the coming month it is planned to continue work with the furnace to establish conditions under which 2800 C may be maintained for periods of time sufficient to ensure melting 20-g charges of UO_2 . It is considered that the furnace is capable of this at present, although it may be necessary to improve the thermal shielding because of the extreme radiative losses occurring at the temperatures involved.

RADIOISOTOPE AND RADIATION APPLICATIONS (AEC-OID)

D. N. Sunderman

Research to establish the applicability of intrinsic tracers to process control and to study the effect of structural factors in graft polymerization was continued this month.

The process-model system has been cleaned and made ready for further studies of the use of intrinsic tracers in precipitation processes. The next process type will be chosen soon. A general evaluation of the hazards to the populace of large-scale intrinsic tracer applications is under way with initial emphasis upon a definition of degree of association between the individual and the product containing the tracer.

The extension of total dose to over 10^8 rads has confirmed earlier data which indicated a reduction in total active sites with increased dose beyond a peak value. Mass spectrometric and vapor chromatographic analyses are under way, and results will be compared with those obtained at lower doses. Future work is outlined through January, 1961.

Use of Intrinsic Radioactive Tracers for Process Control

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

During the month of November the process-model system used in the iron-removal study was cleaned and reconditioned for experiments involving a different purification process. Preliminary work has begun to choose the next process for experimental study, and the hazards evaluation of an intrinsic radiotracer application is under way.

For the experimental study two purification processes are currently under consideration: (1) the removal of manganese from industrial process water using manganese-54 as a tracer, and (2) the removal of impurities such as cobalt and arsenic from the electrolyte used in the electrolytic refining of manganese.

Information is being gathered for the evaluation of hazards associated with the application of intrinsic radiotracers to the production of steel and steel products. This study will be continued next month.

Also during the month a draft of a topical report was prepared describing the results of the iron-removal study.

Next month, in addition to continuation of the hazards evaluation, a choice will be made of the purification process for laboratory study, and experimental work will be initiated.

Graft-Polymerization Studies

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

Earlier in the program, curves of total free-radical sites versus dose were obtained for the methyl-, ethyl-, n-propyl-, isopropyl-, n-butyl-, sec-butyl-, isobutyl-, and n-amylmethacrylate polymers. It had been observed, without exception, that the free-radical-site concentration began to decrease quite rapidly at the higher doses. During November, these data were extended to include a total dose of approximately 1.4×10^8 rads.

At this dosage, the n-butyl- and n-amylmethacrylate polymers degraded to wax-like materials, and, consequently, EPR measurements could not be used to determine total site concentrations. Moreover, nearly all of the other polymers contained less total measurable sites than had been observed at very low doses ($\sim 5 \times 10^5$ rads) using the same dose rate. Apparently, beyond the maximum of the curve of sites versus dose, the production of free radicals by gamma radiation can no longer compete with the destruction of sites by the scavenger by-products of the reaction.

The volatile products from the irradiation of polymethyl- α -chloroacrylate were quantitatively measured by means of mass spectroscopy and vapor-phase chromatography. The results of this work will be available early in December. Comparison of this fragment analysis with similar analyses for polymethylmethacrylate (PMMA) and polymethylacrylate (PMA) should give considerable additional insight into the mechanism of free-radical formation.

Because of the difficulty of accurately scheduling radiation in subsequent analytical procedures, the ground to be covered within a period as short as 1 month cannot be readily estimated. For this reason, the anticipated program for the 2-month period ending January 31, 1961, is outlined below:

- (1) Completion of the fragment analysis from the irradiation of polymethyl- α -chloroacrylate
- (2) Preparation of poly-t-butylmethacrylate and comparison of it with the other butyl isomers with respect to: (a) efficiency of site formation and (b) identity and concentration of volatile irradiation fragments
- (3) Extension of the investigation now in progress of free-radical site formation as a function of dose for polymethylacrylate to higher doses
- (4) Determination of the volatile fragments of irradiation for PMMA and PMA
- (5) Extension of the experiments designed to establish the influence on the efficiency of site formation of polymer molecular weight
- (6) Continuation of the experiments, also now in progress, in which iodine is 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 STUDIESR. O. Wooton, F. G. Peter, J. Foglia,
H. M. Epstein, and J. W. Chastain

The objective of this program is to investigate axial void distribution and heat transfer for rectangular coolant passages over a range of experimental conditions of interest in light-water reactors.

In the experimental procedure, a positive-displacement pump is used to circulate distilled water in a closed, pressurized loop consisting of piping, test section, pre-heater, condenser, sump, and accumulator. The void distribution in the test section is measured using a beta-ray attenuation technique. The experimental void fractions are correlated in terms of the significant heat-transfer parameters, such as mass flow rate, fluid temperature, heat flux, slip ratio, and quality.

During the past months, a new method has been developed for attaching the end pieces to the test section, and the new test section has been used to investigate special experimental conditions. Several runs were taken at pressures around 900 psi to re-examine the variation in the slip ratio, and the results indicated that the slip ratio exhibits an instability of ± 25 per cent.

A correlation of the Bankoff flow parameter, K , in terms of the Reynolds number was developed, and plans were made to test the behavior of the correlation at low Reynolds numbers (45,000).

During November experimental data obtained to examine the behavior of the Bankoff flow parameter at low Reynolds numbers have confirmed the general validity of the previous correlation.

The flow parameter appears to have a parabolic shape, approaching 1.0 for $N_{Re} < 45,000$ and for $N_{Re} > 100,000$ with a minimum of 0.7 around $N_{Re} = 75,000$.

It has been postulated that the instability in the slip ratio at pressures around 900 psi may be caused by a resonance phenomenon existing between the bubble frequency and the alternating-current power supplied to the test section. A 40-kw d-c motor-generator has been obtained to examine this hypothesis and to further investigate the influence of a-c power.

During the coming month the installation of d-c power to the test section will be completed, and test runs will be started to investigate the effects of a-c and d-c power.

DEVELOPMENT OF URANIUM MONONITRIDE

D. L. Keller

This program is concerned with an evaluation of uranium mononitride as a fuel material for power reactors. This evaluation will be based on cursory fabrication studies and property measurements, with the major emphasis on determining irradiation resistance of uranium mononitride. Results of fabrication studies by both powder-metallurgy and melting and casting techniques are reported in the following sections. Capsule-design studies for irradiation experiments were initiated this month and will be reported in December.

Powder-Metallurgy Studies

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

The method of preparing minus 100-mesh UN powder with an average particle size of $66\ \mu$ and a nitrogen content of 5.47 ± 0.03 w/o is outlined in BMI-1365.

A binder composed of a saturated solution of Carbowax 6000 in methyl alcohol has given the best green-pressing results.

A problem in vacuum sintering, that of decomposition of the UN resulting in a liquid uranium phase, was encountered at temperatures of 2000 to 2100 C. Efforts to renitride the resultant uranium matrix at 1300 C under pressures of 1 atm of nitrogen resulted in formation of higher nitrides and loss of compact structural integrity.

Sintering of stoichiometric mixtures of U_2N_3 and uranium at lower temperatures failed to produce dense compacts, but compacting of higher nitride powder and decomposition in situ during sintering gave indications of a possible slight increase in sinterability.

With the isostatic hot-pressing technique, a series of cold-compacted cores of somewhat inferior UN powder pressed at 1290 C under a 5-tsi pressure for 3 hr yielded compacts of 84 to 91 per cent of theoretical density.

During the past month, results on the vacuum sintering of a series of specimens indicated that UN decomposition occurs at 1700 to 1800 C under measured system pressures of 10^{-4} to 10^{-3} mm mercury. Efforts to sinter the relatively coarse powder for long times at lower temperatures resulted in adhesion of particles but no significant densification. With minor modifications of existing equipment, attempts have been made to retard this decomposition by sintering under partial pressures of nitrogen, but difficulties such as leak rates and the nitriding of furnace radiation shields have been

encountered. Further studies would require major modifications and development work on existing equipment and are beyond the scope of this program. There were indications, however, that temperatures of 1900 C for up to 3 hr did not result in significant densification unless the liquid uranium phase was present.

A finer UN powder has been produced by hydriding the uranium before nitrogenation, but no significant increase in the sinterability of this powder has been observed. Further evaluation of the compacting of higher nitride specimens with decomposition to UN in situ during sintering has shown that no significant increase in the sinterability of the resultant UN occurs.

In experiments with the isostatic hot-pressing technique, uranium nitride powder containing 5.40 w/o nitrogen has been cold compacted into pellets about 70 per cent dense, sealed under vacuum in hollow thin-walled stainless steel cylinders, and densified into bodies of up to 95 per cent of theoretical density by means of isostatic hot pressing at 1290 C under a pressure of 5 tsi. Uranium nitride cores 1/2 in. in diameter and up to 2 in. in length, clad with 20 mils of stainless steel, have been produced by this method. An excellent metallurgical bond has been consistently attained with the stainless steel, although 30- μ diffusion zones were also observed. Cores stacked atop one another have bonded together, as evidenced by structural integrity after removing the cladding by machining. Pertinent characteristics of the powder used in these studies along with density measurements before and after hot pressing are summarized in Table I-1. Quantitative microscopic porosity measurements have tended to corroborate the higher density values measured by the pycnometer method rather than by the X-ray method. It is noteworthy to observe that the highest density value obtained was measured on a core that had been implosively compacted to 80 per cent of theoretical density before hot pressing, as indicated in the table.

TABLE I-1. RESULTS OF ISOSTATIC HOT PRESSING COLD-COMPACTED DEGASSED URANIUM NITRIDE POWDER COMPACTS 3 HR AT 1290 C AND 10,000 PSI

Powder Characteristics					Density, g per cm ³		
Oxygen Content ^(a) , w/o	Nitrogen Content ^(a) , w/o	Uranium Content, w/o	Mesh Size	Binder	Green	After Hot Pressing	
						X-Ray	Pycnometer
--	5.40	94.6	-80	None	11.4 ^(c)	12	13.6
--	5.40	94.6	-80	Zinc stearate	10.6	12	12.8
--	5.40	94.6	-80	Zinc stearate	10.5	12	13.2
0.0290	5.47	--	-80	Carbowax 6000	10.5	10 ^(d)	--
0.0290	5.47	--	-200+400	Carbowax 6000	10.3	12 ^(d)	--
0.0290	5.47	--	-200+400	Carbowax 6000	10.5	10 ^(d)	--
0.0290	5.47	--	-400	Carbowax 6000	10.3	10 ^(d)	--
0.0290	5.47	--	-400	Carbowax 6000	10.3	11 ^(d)	--

(a) Vacuum-fusion analysis.

(b) Kjeldahl analysis.

(c) Implosively compacted.

(d) 10-mil-wall stainless steel container leaked during hot-pressing cycle.

Metallographic examination of these specimens has shown a structure containing at least 95 volume per cent of the UN phase with no minor phase present in the grain

boundaries. Up to 5 volume per cent of an unidentified phase, probably a higher nitride, was evenly dispersed throughout the core. The grain size of the UN was extremely fine, averaging about $10\ \mu$. This indicates that the powder particles were multigrained and offers another variable in the solid-phase fabrication of these bodies.

In another series of hot-pressing experiments, it was found that a 10-mil wall thickness of the stainless steel container was not thick enough to insure a leakproof capsule. All five such capsules, containing the 5.47 w/o nitrogen powder, leaked during hot pressing, resulting in a loss of pressure on the compacts and a lack of appreciable densification as shown in Table I-1. It is theorized that the leaks were caused either by a mechanical puncturing of the stainless steel by the UN from the inside or by a chemical reaction between the stainless steel and the silica sand environment used to minimize temperature differentials.

Melting and Casting Studies

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

Uranium mononitride has considerable potential as a high-integrity nuclear fuel and is being studied in connection with the European power-reactor program. However, an investigation of feasible methods of preparation of UN and of its behavior in a reactor environment must precede any comprehensive development program. Several methods of preparation are being considered; in this portion of the research, melting and casting procedures are being investigated. The ultimate goal is the production of a limited number of dense UN bodies to be used for the determination of physical and mechanical properties and for irradiation tests.

Preliminary studies of UN have indicated that both pressure and temperature control the stoichiometry of UN prepared by direct melting techniques. Pressure-temperature relationships at or above the melting point have been extrapolated from existing thermochemical data. These calculations bear out the expected sensitivity of the UN system to slight changes in temperature and nitrogen pressure. As part of an investigation of the preparation of UN by arc melting of uranium in a nitrogen atmosphere, two groups of experiments were recently completed. In one case, an attempt was made to establish a reaction time at an arbitrarily chosen pressure of 3 atm (absolute), while in the other case the nitrogen pressure over the melt was varied for a series of uranium and uranium nitride melts.

In the first series of melts, 100-g buttons of premelted center-cut biscuit uranium were reacted with nitrogen by arc melting for 4, 6, 8, 10, and 12 min. Purified nitrogen, containing less than 50 ppm oxygen was used, and the pressure was maintained at 3 atm during the melting operation and during subsequent cooling. Each melt was turned over midway through the allotted reaction time. Arc conditions of amperage (400 amp) and voltage (40 v) were approximately the same for each melt. The resultant materials were found to vary in nitrogen content from 3.97 w/o for the 4-min melt to 4.46 w/o for the 12-min melt as shown in Table I-2. For the conditions of the experiment, the reaction was 99 per cent complete after 10 min. Of particular interest was the structure of the castings near the edges. This area, farthest from the arc, was the coolest and the first to solidify. The edges were found by metallographic techniques to be essentially stoichiometric UN, again emphasizing the importance of the temperature-pressure relationship in the preparation of UN.

TABLE I-2. ANALYSES OF MATERIAL REACTED UNDER 3 ATM OF NITROGEN BY ARC MELTING FOR VARIOUS TIMES

Reaction Time, min	Nitrogen Content, w/o
4	3.97
6	4.20
10	4.41
12	4.46

(a) Charge weight was 100 g of uranium and arc current was 400 amp with a potential of 40 v.

In a study of the effect of nitrogen pressure as a variable, pressures of 1/4, 1/2, 1, 2, and 3 atm were chosen for investigation using the same arc conditions as imposed in the reaction-time series. Reactions were timed for 10 min, and the 100-g melts were again turned over at midreaction time. As a further check on the pressure effect, 50-g compacts of UN powder containing initially 5.47 w/o nitrogen (98.4 per cent of stoichiometry) were melted using identical pressures and times as for the 100-g uranium melts, but under conditions of lower arc amperage and voltage to compensate for the reduced charge weight. Materials generated by melting uranium metal varied from 3.85 w/o nitrogen at 1/4 atm pressure to 4.53 w/o nitrogen at 3 atm; materials generated by melting the UN compacts varied from 3.15 w/o at 1/4 atm to 4.82 w/o at 3 atm. Analyses are given in Table I-3. The higher nitrogen values ultimately obtained by melting the UN compacts is attributed to the lower temperature of the melt and implies that pressures of less than 10 atm can produce stoichiometric UN only if the temperature is kept low by using a relatively low arc amperage.

TABLE I-3. ANALYSES OF MATERIALS GENERATED DURING NITROGEN-PRESSURE EXPERIMENTS^(a)

Starting Material	Charge, g	Nitrogen Pressure, atm	Nitrogen Content, w/o
Uranium	100	1/4	3.85
		1/2	3.97
		1	4.51
		2	4.32
		3	4.53
UN	50	1/4	3.15
		1/2	3.66
		1	4.23
		2	4.79
		3	4.82

(a) Reaction time in all cases was 10 min total or 5 min per side.

Metallographic inspection of sections of castings indicated greater homogeneity of the material produced by melting UN compacts. However, this inspection also revealed that the casting produced by reacting uranium metal under 1/2 atm of nitrogen contained a central, lens-shaped core that contained stoichiometric UN, distinguishable even macroscopically. Although the mechanism by which this core was produced is open to conjecture, the occurrence of the core was an encouraging demonstration that, under certain conditions, dense UN castings probably can be produced even at relatively low nitrogen pressures provided the temperature is properly controlled. A further demonstration of the effect of temperature and arc amperage was accomplished by preparing a 100-g button under a 3-atm nitrogen pressure. Instead of extinguishing the arc at 400 amp, as before, the arc amperage was gradually reduced to 200 amp (on each side of the casting) before extinguishing the arc. In this case, the average nitrogen content was found to be 5.15 w/o or 92.6 per cent of stoichiometry.

During the course of the various experiments, little or no erosion of untreated tungsten tips was evidenced by the appearance of the tip, by weight change, or by analysis of the melt. On the other hand, in trial experiments with yttrium-treated tungsten tips, considerable erosion was noted despite a significant increase in tip weight. Tungsten pickup by the melt was as high as 0.3 w/o.

The occurrence of the lens-shaped core of UN in a button casting produced at a nitrogen pressure of 1/2 atm has suggested a method by which dense UN bodies of the required shape may be produced. It appears that the UN core was produced by first being formed on the hearth side of the melt by a diffusion process at or just below the melting point of UN. As melting proceeded the UN probably floated upward toward the center of the melt. In order to verify the diffusion method as a means of preparation of dense UN, a substoichiometric drop casting of UN is to be produced by arc melting uranium under a 1/2-atm or greater pressure of nitrogen. It is expected that such a casting would contain at least 3 w/o nitrogen and would be composed of a structural network of UN sufficient to provide strength for subsequent processing by induction heating of the casting just below the melting temperature (about 2500 C) under about 1 atm of nitrogen pressure. The cylinder produced in this manner is to be analyzed chemically and metallographically.

MATERIALS DEVELOPMENT AND EVALUATION (HAPO)

F. R. Shober

An investigation of the electrical- and thermal-conductivity properties of un-irradiated and irradiated UO_2 is being conducted. The long-time creep properties of annealed and of cold-worked Zircaloy-2 are being studied over the 290 to 400 C temperature range.

A number of available materials are being tested in corrosion and are being evaluated as possible container materials for a fused chloride-electrolytic fuel-recovery process.

Thermal Conductivity and Electrical Conductivity of UO_2

C. F. Lucks and H. W. Deem

Measurements have been completed on the first UO_2 disk specimen supplied by HAPO. The data presented in Table J-1 are considered tentative until such time as a possible apparatus malfunction is checked. The specimen was prepared by cold pressing micronized PWR-grade UO_2 , mixed with Carbowax binder, in a 300-ton press and sintering the resultant compact in hydrogen at 1650 C. The measured density for this specimen is 95.3 per cent of theoretical.

TABLE J-1. TENTATIVE INTERPOLATED THERMAL CONDUCTIVITY OF UO_2 SPECIMEN 1(a)

Temperature, C	Thermal Conductivity, w/(cm)(C)	
100	0.076 ^(b)	0.080 ^(c)
200	0.064	0.067
400	0.049	0.051
600	0.039	0.041
800	0.032	0.034
1000	0.029	0.030
1200	0.026	0.027

(a) Sintered UO_2 designated by HAPO as Fabrication A. The specimen is 95.3 per cent of theoretical density.

(b) Values read from a smooth curve drawn through experimental data.

(c) Values adjusted linearly with density to 100 per cent theoretical density.

A furnace has been constructed for use in the measurement of the electrical resistivity of the irradiated UO_2 specimens. The furnace fits existing vacuum equipment already at the hot cells.

No experimental work was done on irradiated specimens or the vibrationally compacted UO_2 specimens this month.

Mechanical Properties of Zirconium Alloys

J. A. VanECHO

The objective of this research program is to determine the long-time creep properties of Zircaloy-2 sheet in the temperature range of 290 to 400 C.

The program is divided into four phases:

- (1) Creep of 15 per cent cold-worked sheet at 290, 345, and 400 C for times extending to 25,000 hr
- (2) Creep of annealed material at 290, 345, and 400 C for times approaching 15,000 hr
- (3) Effect of temperature cycle on both cold-worked and annealed materials at 290 and 345 C
- (4) Effect of temperature and stress cycle on annealed Zircaloy-2 sheet.

All tests are made in vacuum of the order of $1/2 \mu$ in standard Battelle creep machines. Twenty machines are being used on the program.

Since this is a long-time creep program, the 20 tests now in progress have been running for times ranging from about 2,500 to 25,000 hr. A summary of test results was given in BMI-1469. Very little significant change occurs in such long-time creep tests from one month to the next. Therefore, the results shown in BMI-1469 report can be considered as reasonably current.

With the exception of the one temperature-cycle test, all the tests now in progress will probably continue through December.

Materials of Construction for the Fused Chloride-Electrolytic UO₂ Process

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

Research has continued on the program for developing container materials for the HAPO fused chloride-electrolytic fuel-recovery process. In the first phase of the study, the durability of available alloys is being measured under conditions simulating the actual process. The alloys were chosen so that trends could be established and the influence of various elements could be learned.

Coupons of the materials being evaluated are suspended in an equimolar melt of sodium and potassium chlorides containing U₃O₈. The molten salt is maintained at 750 to 800 C. The melt is sparged with a mixture of chlorine, oxygen, and argon gases for 4 hr, then with chlorine and argon for 4 hr, for a total exposure time of 8 hr.

Three specimens are exposed in each experiment. These are located (1) in the melt below the sparge tube, (2) in the melt adjacent to the sparge tube, and (3) at the vapor-liquid interface of the melt above the sparge tube.

To date, about 60 metals and alloys have been evaluated. Alloys based on cobalt, iron, nickel, niobium, and tantalum have been studied, together with a number of pure metals. None of the metals or alloys investigated, except silicon, has shown rates of attack of less than several hundred mils per month in all three locations in the test chambers. Table J-2 shows the corrosion rates determined for some of the more promising materials.

TABLE J-2. THE CORROSION RATES OF VARIOUS MATERIALS IN FUSED CHLORIDE MELTS SPARGED WITH CHLORINE AT 750 TO 800 C

Material	Corrosion Rate, mils per month			
	Below Sparge	Near Sparge	Interface	Average
Silicon	2	7	3	4
Alnico 3B	210	290	370	290
Hastelloy B	170	490	230	297
INOR-1	220	180	560	320
Inconel 702	300	340	450	363
Unitemp 600	240	270	600	370
Tantalum	330	330	520	394
Molybdenum	410	360	430	400
Alnico 5	260	370	710	447
Inconel 713	320	290	830	480
Duriron	120	790	690	500
Astrolloy	330	470	720	507
Alumel	190	300	1070	520
Alnico 2	190	610	820	540

The results to date have indicated the following general conclusions: (1) In the nickel-alloy system, additions of aluminum, molybdenum, and silicon increase resistance, while chromium, niobium, and tungsten reduce resistance. Cobalt and iron appear to have little effect. Of the nickel alloys investigated, Hastelloy B seems to be the best. (2) In the iron-alloy system, aluminum, molybdenum, and silicon tend to improve the resistance, while chromium reduces resistance. Of the iron alloys investigated, Alnico 3B is least rapidly attacked. (3) In the tantalum-alloy system, titanium lowers the resistance. (4) All of the alloys of the niobium system were attacked at excessive rates. Only limited data are available at present on cobalt-base alloys.

Considering the data now available, the most promising alloy systems are nickel-base or iron-base compositions with additions of aluminum, molybdenum, and silicon. Tantalum-, cobalt-, and niobium-base alloys also should be investigated further.

In the future, work will continue on the evaluation of the effects of alloy additions using available alloys and will be expanded to include the making up and testing of nickel- and iron-base experimental alloys.

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

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

Vibrational energy is capable of accelerating or impeding certain solid-state reactions that occur in metals and will cause some reactions to occur at temperatures other than those at which they occur when exposed to thermal energy alone. Among metals that exhibit a relatively large contraction upon solidification, the application of vibrational energy to the solidifying melt will often result in a refined, more uniform, and more randomly orientated cast structure.

In applications where it is desirable to use coextruded uranium a relatively smooth core-to-cladding interface and a uniform cladding thickness is necessary. However, these are not always attained due to the large grain size or the preferred orientation of the uranium casting or extrusion blank. It was believed that the use of vibrational energy in conjunction with the normal heat treatments might result in a randomly oriented refined grain size which will be more amenable to the coextrusion process. Therefore, a program to investigate the feasibility of utilizing vibratory energy as a means of attaining the desired structure in uranium coextrusion blanks has been in progress at Battelle.

In the course of the program, specimens have been utilized in the hot-alpha-rolled and the gamma-annealed conditions. The gamma-annealed specimens were obtained by heat treating uranium extrusion blanks at 800 C for 1 hr and furnace cooling. Specimens in both conditions were heat treated at temperatures from 30 C above to 10 C below the alpha-to-beta transformation temperature for periods of time up to 20 min. Specimens in the alpha-rolled condition were also heat treated at 550 C for as much as 20 min. All these heat treatments were repeated using both a 20-kc, 2.5-kw and a 10-kc, 2.5-kw energy source to supply vibratory energy to the uranium specimens.

The metallographic examination consisted of macroetching and comparing the specimens heat treated and vibrated with those heat treated without being vibrated plus a microscopic examination of the areas adjacent to the control thermocouples in all specimens.

The examination of the macroetched extrusion blanks revealed what appeared to be a significant effect upon the structure of the uranium that could be attributed to the vibrational treatments. For instance, in those cases where the gamma-annealed specimens had been heated to approximately 650 C for 20 min and subjected to vibratory energy, the structure appeared to be greatly refined. The width of the refined zone appeared to be a function of the frequency of the vibrations. The specimens subjected to the 10-kc vibrational frequency exhibited a refined zone almost twice as wide as the zone exhibited by the material vibrated at 20-kc. In all cases, there was sufficient evidence of structural changes to warrant microscopic examination of the specimens to validate the macroscopic evaluation.

The microscopic examinations were made primarily on transverse sections of each heat-treated specimen. These specimens were prepared in the usual manner and electropolished in 4:1 chromic acid and water. The electropolished specimens were

examined at 100X under polarized light. From the microscopic examination it was impossible to denote any effects upon the material that could be attributed to the vibrational treatment alone. Those specimens which exhibited apparent grain refinement due to vibrational treatment when examined by macroetching exhibited structures under the microscope that did not differ greatly from the specimens heat treated without the vibrational treatment.

Therefore, it can be stated that with the techniques employed during this study and the power levels utilized for vibrating the material there is no discernible effect upon the microstructure of unalloyed uranium which can be detected by conventional micrographic techniques.

COATED-PARTICLE FUEL MATERIALS (AEC-DRD)

R. W. Dayton and J. H. Oxley

An irradiation program for alumina-coated uranium dioxide particles has been planned. Coatings approximately 40 μ thick on 100- μ uranium dioxide particles were selected as the basis for this study. Porosity of both the fuel particle and of an inner layer of the coating material and the effect of a graphite-matrix support will be treated as variables during the in-pile evaluation program. Some studies will be begun on in-pile evaluation of beryllia- and carbon-coated particles during Fiscal Year 1961, using sweep capsules for the preliminary evaluations.

A comprehensive program for testing and out-of-pile evaluation of coated particles purchased from vendors was prepared. Particle-coating studies have been initiated aimed at preparing Al_2O_3 , BeO , and carbon coatings under reproducible conditions. Studies of the effect of fabrication variables on the properties of graphite bodies containing coated particles and fission-gas-release studies to help elucidate the mechanisms of coated-particle failure have commenced. The latest information on Capsule SP-5 is reported and interpreted in terms of plausible failure mechanisms. Studies of fuel compatibilities will be started next month.

Evaluation of Commercial Products

T. S. Elleman and D. N. Sunderman

Tests for the evaluation of commercially prepared coated fuel particles have been selected. The tests are designed to measure surface uranium contamination, coating strength, and permeability of coatings to fission gases for submitted coated-particle specimens. A general letter and enclosure describing the coated-particle requirements, tests, and test acceptability limits has been prepared for use by interested companies.

Particle-Coating StudiesM. F. Browning, H. Carlton, A. C. Secrest,
R. B. Landrigan, and J. M. Blocher, Jr. Al_2O_3 Coating of Fuel Particles

Investigation of the coating of UO_2 powders with Al_2O_3 by hydrolysis of Al_2Cl_6 vapor in a fluidized bed of the powder has been resumed. The initial objective of this work is to develop reproducible coating methods and determine the effects of process variables on the coating properties. It is intended that this program be well advanced in time to prepare "best" specimens for in-pile evaluation scheduled for early 1961.

During November, exploratory coating runs were made in fluidized-bed reactors of modified design in an effort to prevent undesirable settling of the Al_2O_3 dust formed as a by-product of the coating process. This situation can lead to

- (1) Plugging of the reactor, necessitating shutdown
- (2) Feedback of dust to form islands of porosity in the coating
- (3) Deposition of coated particles on deposits of dust on the walls of the reactor above the fluidized bed.

Two design modifications have shown considerable promise. In one, the entire overhead section of the fluidized bed leading to a horizontal disengagement chamber is maintained at 1000 C, which effectively prevents the deposition of dust particles in the vertical reactor. In the other, a conical orifice above the bed prevents return of agglomerates of dust particles.

Improvements have also been made in preheating the reactant vapors and in controlling the $\text{Al}_2\text{Cl}_6/\text{H}_2\text{O}$ feed ratio.

BeO Coating of Fuel Particles

Investigation of the conditions for depositing BeO on fuel particles by hydrolysis of $\text{BeCl}_2(\text{g})$ in a fluidized bed has been resumed. Although thin coatings were obtained in three experiments, minor equipment difficulties limited the length of the runs, and the products were not satisfactory for evaluation. It is expected that these problems will be solved and that suitable conditions for depositing useful BeO coatings can be established shortly.

Pyrolytic Carbon Coating of Fuel Particles

Investigation of the coating of fuel particles with pyrolytic carbon by the decomposition of hydrocarbon in a fluidized bed of the particles was also resumed. The primary purpose for this program is to develop conditions for depositing dense uniform coatings reproducibly so that specimens can be prepared for an in-pile evaluation scheduled for early 1961.

A sample of 100 to 140-mesh UO_2 shot previously clad with 53 μ of Al_2O_3 was coated with approximately 120 μ of pyrolytic carbon by decomposition of acetylene at 1400 C in a fluidized bed. Metallographic examination revealed that a dense, uniform, homogeneous coating has been deposited. This composite specimen will be evaluated for high-temperature compatibility of the Al_2O_3 - UO_2 and Al_2O_3 -carbon couples.

To extend the temperature capability of the pyrolytic-carbon-deposition equipment to at approximately 2000 C, two new reactors are under construction. One is a resistance-heated graphite fluidized-bed reactor. The other is an induction-heated rotating-tube reactor. Preliminary tests are under way with the latter apparatus.

When these reactors are in operation, the effect of coating temperature on properties of carbon-coated UC_2 and UO_2 will be studied.

Ceramic-Matrix Studies

M. C. Brockway and W. H. Duckworth

The objective of this phase of the program is to prepare and characterize graphite-matrix bodies containing coated fuel particles.

Experimental investigation of irradiation effects on coated fuel particles dispersed in graphite will be most meaningful if the graphite used in the matrix is representative in qualities to that likely to be used in fuel-element production. Therefore, the literature has been reviewed and graphite manufacturers consulted to determine the current state of the art of graphites for fuel-element matrices. On this basis, a reference graphite has been selected.

Raw materials for the preparation of the reference graphite body are being obtained, and processing equipment is being made ready. Binder pitch is now on hand. If the selected graphite filler, finely ground, well-graphitized needle coke, is not received soon, initial specimens will be made from AGOT graphite flour in order to meet the program schedule.

Effort next month will be on laboratory preparation and characterization of fueled and unfueled graphite bodies.

Fuel-Irradiation Studies

W. J. Zielenbach, N. E. Miller, G. E. Raines,
and W. H. Goldthwaite

The fuel-irradiation studies involve the high-flux irradiation phase of the coated-particle development program. Irradiation capsules containing various coated-particle fuels of interest will be designed, fabricated, and operated at various conditions of specimen temperature, flux, and burnup level. Two general types of capsules will be used. The first type will be used for the irradiation of canned graphite matrices containing selected quantities of coated particles. Postirradiation examination of these specimens will include fission-gas-release determinations by cladding piercing, gas collection, and analysis techniques and visual and metallographic examinations of the fueled body to establish radiation-induced changes in the particles. The second type of capsule will be used for the irradiation of unclad specimens containing coated particles. These capsules will be swept with helium continuously during irradiation so that the fission-gas constituents of the helium can be collected in out-of-pile traps. Trap radioassays will provide data that can be interpreted in terms of gas-release rates and probable mechanisms.

During November the program activity was generally along two lines as follows: (1) formulation of plans for capsules to be built and irradiated during the next few months and initial design of the first series of capsules and (2) final operation of the Battelle Research Reactor of SP-5. Capsule SP-5 is part of the high-flux sweep experiment operated previously in conjunction with Battelle's fission-gas-release studies for the Pebble-Bed Reactor under Sanderson & Porter auspices.

During the final irradiation cycle of SP-5, a 29-hr daughter-trap run was made. In such a run, the helium sweep stream is diverted into a stainless steel-mesh-filled trap located very close to the test capsule. Short-lived fission products decay in the trap to give long-lived solid daughters which deposit on the mesh at the point of decay. The subsequent trap assays provide information directly concerning the release of short-lived gases. A detailed summary of this technique is presented in BMI-1466.

The following paragraphs summarize the significant details relative to the two November activities outlined above.

Capsule Activity

Plans are under way for the irradiation of four capsules which will contain Al_2O_3 -coated UO_2 particles (40- μ coating and 100 to 200- μ highly enriched UO_2). Half of the particles will be supported in a graphite matrix and the other half will be unsupported. These capsules will not involve sweep systems but rather the fueled specimens will be sealed into individual cans for postirradiation fission-gas-release examinations. The release of fission gas, together with the other postirradiation data, will establish the fission-product-retention capabilities of the Al_2O_3 coatings that are now being produced. As outlined in BMI-1473, the experimental parameters of major interest include porosity of the UO_2 , effect of matrix support, in-pile temperature (300 F and 1800 to 2000 F), and burnup (in the 20,000 to 50,000-MWD/TONNE range). The specimens showing maximum resistance to radiation damage will be studied further in high-flux high-temperature sweep experiments.

In the capsule design for these static irradiations the canned specimens will be supported by a graphite-block framework. The blocks will be housed in a stainless steel shell which will provide capsule containment. Two of the four static capsules will be operated with the specimens at a low temperature (nominally 300 F), the other two capsules will be designed for elevated-temperature exposures (1800 to 2000 F). The high-temperature systems will be provided with thermocouples and auxiliary electrical heaters for specimen temperature monitoring and control.

These four static irradiation experiments are scheduled to start in February. It is presently planned to irradiate all four systems simultaneously in a 3 by 3-in. core position at the Battelle Research Reactor. In-pile periods of three and six 2-week reactor cycles are contemplated.

Two sweep-gas systems are also presently planned for irradiation at the BRR early in 1961. The irradiation temperatures in these systems will be in the 1800 to 2000 F range. One capsule will contain specimens based on BeO-coated UO_2 particles while the other will contain specimens based on carbon-coated UC or UC_2 particles. These experiments are designed to provide preliminary information about coatings and coating techniques not hitherto evaluated in a high-flux high-temperature environment. The gas-release behavior of the specimens will serve to guide the coating-development activity for the materials involved.

As an adjunct to the present encapsulation activity, effort is being devoted to capsule design for future coated-particle irradiations in the 2000 to 3000 F range. A major phase of this program is the appraisal by laboratory proof tests of heaters and thermocouples for elevated-temperature service. Commercially available heaters and

thermocouples as well as noncommercial heaters based on refractory-metal wire elements will be investigated in some detail. Elevated-temperature heat-transfer problems will also receive a considerable amount of attention.

Operation of SP-5 Experiment

An evaluation of the fission-gas-release behavior during irradiation of an element fueled with alumina-coated UO_2 particles has been in progress since April. Previous reports of progress have appeared in Section P of this report. The experiment is described in detail in BMI-1468.* Samples of the long-half-life gases are obtained by adsorbing xenons and kryptons from the sweep helium stream on liquid nitrogen-cooled activated charcoal or by sampling the flowing gas.

The irradiation experiment was terminated during November and the capsule was delivered to the hot cell laboratory for postirradiation examination of the specimens. Additional release measurements of both relatively long- and short-half-life gases were obtained. The results of these measurements are shown in Table L-1 along with previous data for comparison. Some general comments concerning these measurements are in order, although a complete appraisal of this information is not yet available. First, the lack of a strong half-life dependency in the earlier data suggest that for the rather small amount of materials being released no appreciable diffusion resistance was present. This lack of half-life dependency can be explained by the fact that a small amount of fuel was found external to the particle coatings by an alpha survey prior to the irradiation. The activity being generated by this small fraction of the material would find very little diffusional resistance to release and would show very little half-life dependency. Secondly, an apparent diffusion release occurs as burnup increases, indicated by the significant half-life dependency from xenon-133 through krypton-87 that subsequently develops. These data can probably be explained on the basis of degradation of the coating due to irradiation damage of some nature. They cannot be explained by fracturing of particle coatings, since a large half-life dependency would not be expected. Finally, the increase in the release of the short-half-life species is thought to be a result of cracking or fracturing of a certain number of the particle coatings. However, the most significant feature of these results is that the release of the short-half-life species is quite low after about 6 per cent uranium-235 fission burnup or 50,000MWD/TONNE total uranium content, even though the release of the long-half-life species has become relatively large. The daughter products of these short-half-life species are a potential source of trouble insofar as maintenance of a reactor during shutdown is concerned. Further discussion of these results will be presented when a more complete correlation is available.

It is of interest to compare the release behavior of the sphere (E8-7) in another sweep compartment with that of the sphere containing coated particles. Sphere E8-7 has a thin siliconized SiC coating which failed early in the irradiation. This sphere is fueled with bare UC_2 particles. The failed coating on Sphere E8-7 is not expected to provide significant resistance to release. Hence the behavior of this specimen, at a burnup comparable to that of the coated-particles specimen, should be representative of that expected from uncoated fueled graphite. Table L-2 illustrates the results of the last several samples of long-half-life gases that were obtained from this sphere. From

*"Progress on Ceramic Coated Fuel Particles at Battelle", BMI-1468 (September 16, 1960).

TABLE L-1. SUMMARY OF RESULTS OF FISSION-GAS-RELEASE MEASUREMENTS

Sample Date	Sample Type ^(a)	Estimated Uranium-235 Fission ^(b) Burnup, per cent	Estimated Specimen-Surface Temperature, F ^(c)	5.27-Day	9.13-Hr	4.36-Hr
				Xenon-133	Xenon-135	Krypton-85m
4-11-60	A	0.1	1150	1.8×10^{-7}	1.9×10^{-8}	1.6×10^{-8}
4-15-60	A	0.2	1150	1.0×10^{-7}	4.0×10^{-8}	5.9×10^{-8}
4-28-60	A	0.6	1350	1.9×10^{-6}	4.9×10^{-7}	4.7×10^{-7}
5-5-60	A	0.9	1350	5.8×10^{-6}	2.1×10^{-6}	7.3×10^{-6}
5-6-60	A	0.9	1350	1.3×10^{-7}	8.2×10^{-9}	2.6×10^{-8}
5-10-60	A	1.0	1300	4.8×10^{-8}	4.2×10^{-9}	7.0×10^{-9}
5-11-60	A	1.0	1300	4.0×10^{-8}	1.6×10^{-9}	3.1×10^{-9}
5-13/15-60	D	1.1	1350	--	--	--
5-19-60	A	1.3	1350	9.1×10^{-7}	1.2×10^{-8}	1.8×10^{-8}
5-25-60	A	1.4	1150	5.0×10^{-8}	4.0×10^{-9}	7.6×10^{-9}
7-8-60	A	2.6	1350	2.8×10^{-4}	7.8×10^{-7}	4.0×10^{-6}
7-22-60	A	2.9	1350	8.0×10^{-4}	7.9×10^{-7}	5.9×10^{-6}
7-22-60	A	2.9	1350	2.1×10^{-4}	5.3×10^{-7}	2.1×10^{-6}
8-3-60	A	3.2	1300	1.2×10^{-3}	1.5×10^{-6}	6.1×10^{-6}
8-18-60	A	3.7	1200	1.1×10^{-3}	4.2×10^{-6}	1.2×10^{-5}
9-14-60	A	4.4	1300	4.1×10^{-3}	1.4×10^{-4}	4.7×10^{-4}
9-28-60	A	4.9	(1150)	4.7×10^{-3}	1.4×10^{-4}	2.5×10^{-4}
9-30-60	V	4.9	(1150)	8.1×10^{-3}	2.4×10^{-4}	4.8×10^{-4}
10-1/3-60	D	5.0	(1150)	--	--	--
10-3-60	V	5.0	(1150)	1.7×10^{-2}	1.9×10^{-4}	2.5×10^{-4}
10-12-60	V	5.3	(1150)	9.7×10^{-3}	2.3×10^{-4}	4.6×10^{-4}
10-20-60	V	5.5	(950)	6.9×10^{-4}	2.2×10^{-5}	3.0×10^{-5}
10-21-60	V	5.5	(950)	1.5×10^{-4}	1.8×10^{-5}	3.7×10^{-5}
10-26-60	V	5.6	(950)	1.9×10^{-4}	3.0×10^{-5}	5.2×10^{-5}
10-28-60	V	5.7	(950)	2.0×10^{-4}	1.9×10^{-5}	4.4×10^{-5}
11-4-60	V	5.8	(950)	3.2×10^{-4}	3.5×10^{-5}	5.9×10^{-5}
11-12-60	V	5.9	(950)	3.5×10^{-4}	3.0×10^{-5}	9.5×10^{-5}
11-12/14-60	D	6.0	(950)	--	--	--
11-14-60	V	6.0	(950)	2.4×10^{-4}	3.9×10^{-5}	6.8×10^{-5}

(a) A denotes samples adsorbed on charcoal, D denotes daughter-trap samples, and V denotes volumetric samples.

(b) To obtain MWD/TONNE of total uranium, multiply per cent fission burnup by 8650.

(c) Temperature estimates through the sample of 9-14-60 were based on thermocouple readings; those after 9-14-60 were based on the performance of other experiments in the BRR as all thermocouples in this capsule had failed.

(d) R/B is the ratio of atoms being released per second to atoms being generated per second.

FOR A GRAPHITE SPHERE (NO. 471) FUELED WITH Al_2O_3 -CLAD UO_2 PARTICLES

R/B Ratio ^(d)						
2.77-Hr Krypton-88	78-Min Krypton-87	3.9-Min Xenon-137	3.18-Min Krypton-89	16-Sec Xenon-140	9.8-Sec Krypton-91	1.7-Sec Xenon-141
2.2×10^{-9}	--					
2.7×10^{-8}	1.8×10^{-8}					
2.9×10^{-7}	1.8×10^{-7}					
--	--					
1.6×10^{-8}	--					
7.1×10^{-9}	--					
4.7×10^{-9}	--					
--	--	--	3.0×10^{-7}	1.1×10^{-7}	--	1.3×10^{-7}
7.7×10^{-9}	--					
1.9×10^{-9}	--					
--	1.6×10^{-7}					
--	1.3×10^{-7}					
6.2×10^{-7}	2.0×10^{-7}					
1.0×10^{-6}	2.9×10^{-7}					
3.2×10^{-6}	1.1×10^{-6}					
--	--					
4.8×10^{-5}	7.6×10^{-6}					
1.4×10^{-4}	3.3×10^{-5}					
--	--	7.3×10^{-6}	8.6×10^{-6}	1.9×10^{-6}	3.6×10^{-6}	1.0×10^{-6}
7.4×10^{-5}	1.9×10^{-5}					
1.1×10^{-4}	2.7×10^{-5}					
1.2×10^{-5}	5.9×10^{-6}					
1.6×10^{-5}	6.3×10^{-6}					
1.9×10^{-5}	1.1×10^{-5}					
2.0×10^{-5}	3.4×10^{-6}					
1.8×10^{-5}	1.8×10^{-5}					
3.5×10^{-5}	1.9×10^{-5}					
--	--	1×10^{-5}	1.1×10^{-5}	5.4×10^{-6}	5.9×10^{-6}	6.3×10^{-6}
3.0×10^{-5}	1.2×10^{-5}					

TABLE L-2. RESULTS OF FISSION-GAS-RELEASE MEASUREMENTS FOR A GRAPHITE SPHERE (NO. E8-7) FUELED BARE UC₂ PARTICLES

Sample Date	Estimated Specimen Surface Temperature ^(a) , F	R/B Ratio ^(b)				
		5.27-Day Xenon-133	9.13-Hr Xenon-135	4.38-Hr Krypton-85m	2.77-Hr Krypton-88	78-Min Krypton-87
10-27-60	1100	4.5×10^{-2}	8.0×10^{-3}	1.3×10^{-2}	5.5×10^{-3}	3.1×10^{-3}
11-4-60	1100	2.3×10^{-2}	4.2×10^{-3}	9.2×10^{-3}	--	1.9×10^{-3}
11-12-60	1100	5.9×10^{-2}	5.2×10^{-3}	1.2×10^{-2}	3.8×10^{-3}	2.5×10^{-3}
11-14-60	1100	2.6×10^{-2}	6.7×10^{-3}	1.3×10^{-2}	4.6×10^{-3}	3.5×10^{-3}

(a) Estimated by performance of other experiments as all thermocouples in this capsule had failed.

(b) R/B is the ratio of atoms being released per second to atoms being generated per second.

this comparison, an idea of the worth of individual particle coatings is obtained. A daughter sample for this specimen was also obtained in November but its analysis is not complete.

It is anticipated that during the next month the analysis of the daughter sample from Sphere E8-7 and a final correlation of the fission-gas-release data will be completed.

Fission-Gas-Release Studies

T. S. Elleman and D. N. Sunderman

Fission-Gas Analytical Techniques

A program has been undertaken to improve gamma-ray spectrometer techniques for the analysis of fission-gas mixtures. The program will involve the determination of spectrometer detection efficiencies for representative fission gases, the measurement of fission-gas breakthrough on charcoal collection traps, and the investigation of techniques for the analysis of gamma-ray-emitting fission-gas mixtures for individual radioisotopes. The initial experiment will concern the standardization of laboratory spectrometers for fission-product xenon-133 and xenon-133m. Parent iodine-133 will be standardized by 4 π beta counting, and known quantities of xenon-133 and xenon-133m daughter products will be transferred to charcoal collection traps for standardization of the gamma-ray spectrometers. Charcoal traps and the gas-transfer manifold are presently being constructed for the gas standardization.

Fission-Gas-Diffusion Studies

Fission-gas release from coated fuel particles during postirradiation heating will be studied. The purpose of this program will be to elucidate the mechanism of fission-gas release from coated particles and to obtain information on diffusion coefficients and

release rates of the gas. The release of recoil fission products from alumina will be investigated in the initial phases of the program. Subsequent phases will involve similar studies with other coating materials as well as investigations on the effect of burnup and high temperatures on fission-gas release.

The initial experiment in this program will be the measurement of fission-gas release from solid alumina spheres containing recoiled fission fragments. These data on gas release will permit an estimation of fission-gas release from cracked fuel particles during reactor operation. The details of this experiment are presently being planned, and results will be discussed in future progress reports.

Matrix-Separation Techniques

Techniques for the separation of coated particles from a graphite matrix are being developed for use in hot-cell postirradiation studies and burnup analyses. The method adopted should not crack particles during the matrix separation and should involve only simple physical manipulations. Disintegration of the matrix with fuming nitric acid and by electrolysis are two possibilities being considered. Preliminary experiments on trial specimens with fuming nitric acid yielded decomposition of the graphite without apparent damage to the coated particles.

Physics and Engineering Guidance

R. G. Wooton, H. M. Epstein, and J. W. Chastain

For proper guidance of the experimental program on coated-particle fuels, it is necessary to know the characteristics which coated particle fuels must have to be usable in specific reactors. Information on fuel loading, tolerable coating thickness, and degree of fission-product containment is especially important. The purpose of this program is to supply such information from an analysis of data for reactors of interest to the AEC.

Information is currently being gathered on two reactor systems: the Experiment Gas-Cooled Reactor (EGCR) and the Pebble-Bed Reactor (PBR). Both reactors use UO_2 as fuel and graphite as moderator. Analyses are being made on a modified EGCR fueled with $125 \mu \text{UO}_2$ particles with a $40\text{-}\mu \text{Al}_2\text{O}_3$ coating but with the same EGCR core size and element geometry. The fuel enrichment and matrix composition are parameters of the analysis.

A program has been started to examine and summarize available information on the amount of fission-product release which can be tolerated without interfering with normal maintenance of the system. This study will establish permissible release of volatile fission products from coated-particle fuels, and provide guidance for future experimental work and development.

Fuel-Compatibility Studies

S. J. Paprocki, G. W. Cunningham, D. C. Carmichael, and J. J. Ward

A study has been initiated to investigate various chemical reactions which may affect the maximum operating conditions for either the fuel particles or their coatings. In the first phase of the program, emphasis will be placed upon an investigation of oxygen diffusion through Al_2O_3 coatings, reaction of Al_2O_3 with carbon, and diffusion of uranium through pyrolytic carbon coatings. Thermodynamic calculations, a literature survey, and a limited number of tests will also be run to obtain preliminary information on the compatibility of UO_2 , UC, and UC_2 with Al_2O_3 , ZrO_2 , BeO, MgO, and pyrolytic carbon.

The diffusion of oxygen through unirradiated Al_2O_3 coatings is expected to be very slow unless the coatings are cracked or are porous. Therefore, tests will initially be conducted at temperatures on the order of 1700 C. Coated particles will be visually inspected and radiographed before use and only particles which are estimated to be crack-free and dense will be tested. It is anticipated that the rate of oxygen diffusion can be determined on the basis of weight change as measured on a recording microbalance and the change in the oxygen/uranium ratio of the coated UO_2 . Thermodynamic calculations indicate that no chemical reaction will occur between UO_2 and Al_2O_3 at temperatures below 2000 C, and existing phase diagrams indicate that there is little mutual solid solubility of these oxides and that a eutectic exists at about 1850 C. Therefore, diffusion of oxygen through the coating and reaction to form U_3O_8 would appear to be the most probable mechanism of possible failure due to chemical reactions.

As indicated in Table L-3, for Al_2O_3 -coated UO_2 in a graphite matrix there is a possibility that Al_4C_3 will be formed at the higher temperatures, particularly in the presence of a flowing atmosphere or a partial vacuum. This reaction will be investigated with UO_2 coated with a layer of Al_2O_3 coated with pyrolytic graphite and with Al_2O_3 -graphite couples. Several grades of graphite will be utilized to obtain information on their relative reactivity.

TABLE L-3. THERMODYNAMICS OF THE REACTION

Temperature, K	Free Energy, ΔF , cal	Equilibrium Constant, $\log_{10} K$	Logarithm of Equilibrium Pressure of CO, $\log_{10} P_{\text{CO}}$, atm
1000	+164,880	-36.031	-12.009
1200	+137,440	-25.029	-8.342
1500	+96,860	-14.111	-4.703
1800	+56,820	-6.898	-2.299
2000	+30,320	-3.313	-1.104
2200	+2,260	-0.224	-0.075
2500	-38,380	+3.355	+1.118

Techniques involving the use of uranium-235 will be used to study the diffusion of uranium through pyrolytic carbon coatings. Initial studies may involve massive couples made with enriched UC or UC_2 and pyrolytic carbon. However, emphasis will be placed upon uranium diffusion from enriched UC or UC_2 powder through pyrolytic carbon

coatings vapor deposited on the particles. Preliminary tests will be run at temperatures of 1700 C or higher. Materials are being prepared and equipment is being modified for this study.

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

R. A. Ewing and C. L. Peterson

Chemical and radiochemical analyses of solutions obtained from the dissolution of two high-burnup Consolidated Edison prototype fuel pins are continuing. Radiochemical analysis of offgas scrubber solutions indicates a greater carry-over of radioniobium with Darex decladding than with Sulfex decladding. Activity of Darex decladding solution was found to be greater than that of the Sulfex decladding solution, in line with the larger losses of fuel materials with Darex decladding.

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

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

Previous head-end studies have been concerned with relatively low-burnup prototype Consolidated Edison ($\text{ThO}_2\text{-UO}_2$) fuel pins. Two main radiation effects were observed. Ease of core dissolution increased with increasing irradiation, as did losses of fuel during decladding, when decladding was performed by the Darex process.

Current studies are being performed with high-burnup Consolidated Edison prototype fuel pins (15,000 to 25,000 MWD/T). As noted in the previous report (BMI-1473), the same effects were apparent for the first two pins examined, on a magnified scale. Analysis of the core solutions for cesium, thorium, and uranium are continuing.

Gross beta and gross gamma activities of cladding solutions were substantially higher than in previous experiments on lower burnup pins, and ranged from 10^9 to 10^{10} dpm per ml. Activity of the Darex cladding solution was greater than that of the analogous Sulfex solution, in line with the greater losses of fuel and associated fission products to Darex solutions. Cesium analyses of the cladding solutions are also being made to determine the magnitude of the correction to burnup determinations based on cesium analyses of core solutions only. Preliminary indications are that some correction will be necessary.

In the first two experiments with high-burnup pins, offgases from the dissolver were passed through a sulfuric acid scrubber spiked with niobium to determine losses of radioniobium. Results were not conclusive because of an apparent physical entrainment of traces of cladding solution, especially during the Darex run. However, analyses of the scrub solutions from the two runs with a gamma spectrometer indicated that there may have been substantial carry-over of radioniobium with Darex decladding, but not with Sulfex decladding. This experiment will be repeated in the next series of runs, with better provision for trapping any entrained cladding solution.

Remains of two CETR high-burnup (6,000 and 12,000 MWD/T) fuel pins have just been released from postirradiation studies. It is planned to conduct dissolution tests on these and on two more ORR-irradiated high-burnup pins during the next hot-cell campaign, scheduled for early December.

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

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

The research on this project has been concluded, and a topical report is now being processed.

URANIUM CARBIDE AS A REACTOR FUEL (AI)

E. L. Foster

The interest of Atomics International in the development and evaluation of fuels for high-temperature power reactors led to a research program on uranium carbide prepared by arc melting and casting. This program is now complete. It included the preparation of UC materials and their evaluation after burnups as high as 25,200 MWD/T. The irradiations were conducted under conditions that imposed surface and core temperatures as high as 950 and 1600 F, respectively. Postirradiation examinations showed that only slight surface and volume changes occurred.

Postirradiation Examination of Uranium Monocarbide

A. W. Hare and R. F. Dickerson

The postirradiation examination of uranium monocarbide specimens from seven irradiation capsules has been completed. The program consisted of an irradiation at the Battelle Research Reactor of one noninstrumented capsule which contained 1/2-in.-long by 3/8-in.-diameter UC specimens and the irradiation at the MTR of six instrumented capsules which contained 2-in.-long by 3/8-in.-diameter UC specimens.

The purpose of this investigation was to determine the effects of irradiation on massive uranium carbide of compositions at, above, and below stoichiometric at elevated temperatures and relatively high burnups. To obtain initial irradiation information, the BRR capsule was irradiated at 600 F to a burnup of about 400 MWD/T. The instrumented MTR capsules were irradiated under conditions whereby the specimens experienced surface and core temperatures of from 540 to 950 F and from 880 to 1600 F, respectively, and burnups of from 1,400 to 25,200 MWD/T.

Postirradiation diameter measurements taken on the specimens showed increases of from 0.16 per cent for the BRR specimens to a maximum of 1.4 per cent for the highest burnup specimens. Density showed decreases of from 0.29 per cent for the BRR specimens to a maximum of 4.4 per cent for the highest burnup specimens. Some cracks did occur in the specimens during the irradiations. During the postirradiation examinations, the specimens and spacers fractured into numerous large pieces; however, this was caused by the oxidation of NaK entrapped in the microcracks.

The metallographic examination of the two specimens and the top spacers from Capsule BMI-23-4 (burnup of 25,200 MWD/T) was completed last month. The structure of the spacers, which were composed of the stoichiometric composition, uranium-4.8 w/o carbon, showed no change after irradiation. The microstructure of the two specimens, which contained 5.2 and 5.3 w/o carbon, did indicate a change after irradiation. The UC₂ phase, which takes the form of Widmanstätten-type plates in the UC matrix, appeared to be somewhat diffuse and less distinct as compared with the plates in the preirradiated structure.

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, and promising results have been achieved by coating individual fuel particles with alumina. Current efforts are directed toward evaluation of fuel bodies containing fuel particles of UC_2 individually coated with pyrolytic carbon.

In initial neutron-activation tests, experimental fuel bodies containing carbon-coated UC_2 particles have shown high leak rates, indicative of cracked coatings.

Laboratory Evaluation of Fueled-Graphite Spheres

T. S. Elleman

Fission-gas release during postirradiation heating was measured for Sphere FA-25, No. 1, at temperatures of 1650, 2100, and 2400 F. The sphere contained 4.75 g of natural uranium in the form of carbon-coated UC_2 and was irradiated to an integrated flux of 2.3×10^{15} nvt. A total xenon-133 release of 0.11 per cent was obtained at the three temperatures, which is the same order of magnitude as the release observed with an earlier sphere containing carbon-coated particles. The sphere was heated approximately 1 hr at each temperature and the incremental per cent release was relatively constant at each temperature.

In the coming months additional fueled bodies will be tested in an attempt to develop a body with sound coatings on the fuel particles. When this is accomplished, irradiation tests will be initiated.

DEVELOPMENT OF FABRICATION PROCESSES FOR COLD BONDING OF ZIRCALOY-2 TO TYPE 410 STAINLESS STEEL

S. W. Porembka

This investigation was established to develop techniques for cold joining pressure tubes of Zircaloy-2 and Type 410 stainless steel in support of the Canadian NPD-2 and CANDU reactor programs. It is desirable that such techniques have the potential of being performed in pile in order to facilitate tube replacement. Presently, both friction bonding and a study of the effects of surface preparation on bonding tests to date have yielded encouraging results in that Zircaloy-stainless, stainless-stainless, and Zircaloy-Zircaloy joints have been achieved. Preliminary cold roll-bonding tests did not produce bonding because of the limited reduction rates possible with the initial assemblies.

Friction Bonding

S. J. Paprocki, E. S. Hodge, and H. D. Hanes

Friction bonding of Zircaloy to stainless steel is attractive since the process offers the potential of achieving high surface deformation and localized heating in the joint area, thereby minimizing diffusion and compound formation. In order to provide basic data for possible transition joining of these materials, studies of Zircaloy-Zircaloy and stainless-stainless friction bonding have also been included in this portion of the program. Initial tests have involved friction bonding rod samples of these materials combinations in order to provide a rough definition of the process variables. Upon refining the bonding techniques for such specimens, the joining of pressure tubes is planned.

In preliminary tests involving friction bonding of Zircaloy to stainless steel, good apparent bonding was achieved. In order to overcome excessive upsetting of the Zircaloy, it was necessary to employ a chamfered bonding face. Such joint designs resulted in good general bonding of the specimen with some nonbonding apparent at the extreme periphery. Of the limited bonding conditions studied to date, the best joints were achieved with speeds of 1500 rpm and pressures of 10,000 and 20,000 psi at times to 11 sec. Metallographic examination of a selected sample from this group of tests revealed an extremely fine bond interface which also exhibited an interlocking of the two phases.

The joining of Zircaloy to itself was also readily achieved by friction techniques. To minimize oxidation of the joint area, it was necessary to devise an argon system for covering the work during the bonding operation. Process conditions of 500 rpm at 20,000 psi for 3 sec produced good bonds in the Zircaloy-Zircaloy couples as evidenced by metallographic examination. Tensile tests of samples bonded under these same conditions showed ultimate strengths of 44,300 and 57,000 psi with ductile fractures.

In friction bonding of Type 410 stainless steel to itself it was found that good bonding was more difficult to achieve. Those specimens which were joined exhibited bonding of approximately two-thirds of the interface with nonbonded areas at the periphery. Such joining was attained at 1500 rpm at 10,000 psi with times from 2 to 4 sec. Metallographic

examination of these bond areas showed good metallurgical bonds with evidence of heat-affected zones. Ultimate strengths of 37,800 and 70,500 psi were obtained on similar samples, demonstrating the variation of bonds achieved in tests to date.

Further tests with each materials combination are planned in order to more clearly define the process variables and to refine the joining techniques.

Effects of Surface Preparation in Roll Forming

S. J. Paprocki, R. J. Carlson, and C. C. Simons

The effects of surface preparation in roll-form joining are being investigated to establish the feasibility of achieving cold bonding by this technique. Since both direct and transition joining are of interest in this program, three materials combinations are being considered: Zircaloy-stainless, Zircaloy-Zircaloy, and stainless-stainless. The initial approach to this problem involves establishing the degree of bonding that can be achieved in cold rolling and defining the conditions required. In this regard, a preliminary series of Zircaloy-stainless steel cold-rolling tests at low reduction rates have been completed.

Thirteen tack-welded pack assemblies of Zircaloy-2 and Type 410 stainless steel were cold rolled at single reductions ranging from 2 to 26 per cent. All surfaces in these composites were machined to a 300- μ in. rms finish and subsequently cleaned with either the standard pressure-bonding wash cycle or by scrubbing with MgO and sodium carbonate. In all cases, the original tack welds separated, revealing no bonding in the composites. A similar pack press forged to approximately 25 per cent reduction also did not bond. Presently, assemblies are being prepared for cold rolling at considerably higher reductions.

DEVELOPMENT OF MATERIALS FOR THE MGCR AND HTGR (GA)

W. H. Goldthwaite

Radiation-effects studies of potential MGCR fuel materials are in progress. Capsule irradiations of UO_2 dispersed in BeO and UC and UC_2 dispersed in graphite are nearing completion.

DEVELOPMENT AND EVALUATION OF FUEL ELEMENTS FOR THE MGCR

Radiation-Effects Study of Potential Fuels

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

Ceramic-type fuels composed of about 20 volume per cent uranium dioxide in beryllium oxide, 20 volume per cent uranium monocarbide in graphite, and 20 volume per cent of uranium dicarbide in graphite are being investigated as potential fuels for the MGCR. The uranium used in preparation of the specimens was 30 per cent enriched in the uranium-235 isotope.

The specimens, which are in the form of cylindrical pellets approximately 0.222 in. in diameter by 0.250 in. long, are being irradiated over a range of 1 to 12 uranium a/o burnup at surface temperatures near 1500 F. Four capsules equipped with thermocouples and electrical heaters and containing two specimens of each composition are included in the program.

One of the four capsules was opened, and the specimens were examined after irradiation in the Battelle Research Reactor to specimen burnups of 1.0 to 1.7 uranium a/o. All specimens were in good condition. The irradiation in the MTR of two of the remaining three capsules has been completed, and the capsules have been shipped to the General Atomic hot cells for postirradiation examination of the specimens. This examination is now in progress. The final capsule is continuing to operate satisfactorily in the MTR 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

P. M. Steinback and J. H. Stang

Fuel pellets of UO_2 (30 volume per cent, 12 per cent enriched) in BeO are to be contained in Hastelloy X cans and irradiated at 1700 F for about 18 months at the MTR in a NaK-filled double-wall capsule equipped with thermocouples and auxiliary electric heaters. Except for minor heat-transfer details, the design of the capsule has been completed, and many of the capsule components as well as the specimen containment

cans have been fabricated. Some of the capsule subassembly operations are in progress, but the major ones are awaiting delivery of the fuel pellets from General Atomics.

COATING OF URANIUM AND THORIUM CARBIDE
POWDERS FOR THE HTGR

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

Work on this project was terminated during the last week in October.

DEVELOPMENTAL STUDIES FOR THE SM-2 (ALCO)

S. J. Paprocki

Analysis of temperature data from six capsules containing specimens of SM-2 reference and alternate fuels now being irradiated in the ETR has been initiated. It is planned to irradiate the specimens to burnups of up to 70 per cent of the contained uranium-235.

Radiation-Stability Studies of Reference on Alternate SM-2 Fuels

J. F. Lagedrost, J. H. Stang, and W. H. Goldthwaite

A study of the radiation stability of reference and potential fuels for the SM-2 reactor is in progress. The specimens, contained in instrumented capsules, are being irradiated in the ETR. A thorough analysis of temperature data from the six capsules now in the reactor has been initiated. Preliminary results indicate that significant flux variations have occurred at some of the capsule positions. Attempts will be made to relate specimen burnup with temperature for specific intervals where such information seems desirable.

Cycle 32 of the ETR ended on October 31, 1960, with an accumulation of slightly over 30 full-power days. Cycle 33 started on November 11, and is scheduled to end on December 12, 1960. During the shutdown following Cycle 32, the thermocouples in BMI-32-6 were restored to operating condition, and the heaters were found to be satisfactory. Operating conditions of the six capsules during the early part of Cycle 33 are discussed below.

BMI-32-5. All thermocouples are operating. Surface temperatures of the top four specimens ranges approximately from 550 to 850 F. No electric heating has been required.

BMI-32-6. All thermocouples are operating. With the addition of approximately 5 kw of electric heat, surface temperatures of the top four specimens are in the 450 to 700 F range.

BMI-32-7. The temperature indicated by the thermocouple adjacent to the top specimen in this capsule increased gradually (over a period of 2 days) from an original operating temperature of 330 F (400 F at specimen surface) to 750 F (1300 F at specimen surface) shortly after the start of Cycle 32. During Cycle 33, it has continued to indicate this high temperature. Since the other thermocouples in this capsule have experienced no such change, it appears possible that the top thermocouple is faulty; it is scheduled to be checked during the next shutdown. The second thermocouple failed during Cycle 31; the third and fourth indicate specimen-surface temperatures in the 500 to 600 F range during Cycle 33. No electric heating has been used.

BMI-32-8. All thermocouples are operating, although one (adjacent to the fourth specimen) is becoming erratic and will be checked during the next shutdown. Specimen-surface temperatures range from 475 to 575 F with the addition of slightly over 4 kw of electric heating.

BMI-32-9. The temperature performance of this capsule has been especially noteworthy in that temperature levels have decreased significantly during the first few cycles. During Cycle 33, surface temperatures of the second, third, and fourth specimens are in the 450 and 675 F range. The top thermocouple failed during Cycle 31. No electric heating has been used.

BMI-32-10. Thermocouples at the second, third, and fourth specimens continue to operate satisfactorily, indicating surface temperatures in the 400 to 675 F range. The top thermocouple failed on initial startup. No electric heating has been necessary.

GAS-COOLED REACTOR PROGRAM (AGN)

D. L. Keller

Studies for Aerojet-General Nucleonics (AGN) and the Army Reactors Branch (ARM) in support of the ML-1 reactor development program are reported in this section. This program consists primarily of in-pile-capsule studies of UO_2 and $\text{UO}_2\text{-BeO}$ specimens clad in Hastelloy X, which represent the fuel materials for the ML-1, fatigue studies on Hastelloy X, critical-assembly studies on a mock-up of the ML-1 core, and advanced fuel studies with UO_2 bodies vapor coated with Al_2O_3 .

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

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

In this program, the process for fabricating BeO- UO_2 fuel-element pellets by cold pressing and sintering is being examined. The objective is to develop and optimize techniques which will permit a reduction in pellet fabrication costs.

Previously, it was found necessary to sinter pellets containing 30 or 40 w/o of commercially available BeO powder and ceramic grade, minus 325-mesh UO_2 for 1 hr at 3200 F to obtain bulk densities in excess of 95 per cent of theoretical. Numerous investigators* have reported that UO_2 powders sinter more readily in neutral atmospheres than in hydrogen. It has been suggested that excess oxygen in the UO_2 promotes sintering, and that the hydrogen is detrimental because it removes excess oxygen. Other investigators have shown that hydrogen adsorbed on UO_2 surfaces inhibits sintering. In either case, reduction in the amount of hydrogen in the furnace atmosphere might be expected to permit densification at lower temperature. The use of a non-explosive atmosphere would also be an advantage.

Therefore, during this period, BeO- UO_2 pellets were sintered in a 90 volume per cent argon-10 volume per cent hydrogen atmosphere.** The results are compared with the results obtained in a previously reported hydrogen sintering trial in Table U-1. The densities of specimens sintered in hydrogen-argon were significantly lower than those sintered in hydrogen. Further experimentation does not appear to be justified.

This concludes the planned work on this program.

*See, for example, Williams, J., Baines, E., Scott, R., and Hall, A., "Sintering of Uranium Oxides of Composition UO_2 to U_3O_8 in Various Atmospheres", AERE M/R 2653 (August, 1958).

**The high volatility of uranium oxides in neutral or very slightly oxidizing atmospheres made the use of some hydrogen seem necessary.

TABLE U-1. EFFECT OF ATMOSPHERE ON SINTERABILITY OF BeO-UO₂ CERAMICS

Composition	Batch Composition, parts by weight		Forming Pressure ^(b) , psi	As-Formed Density, g per cm ³	Sintering Atmosphere ^(c) , volume per cent	Sintered Density		Diametral Shrinkage, per cent
	BeO ^(a)	UO ₂				G Per Cm ³	Per Cent of Theoretical	
11-2	40	60	20,000	2.73	Hydrogen	4.94	93.2	18.2
11-3	40	60	40,000	2.98	Hydrogen	5.03	95.0	16.3
11-2	30	70	20,000	3.10	Hydrogen	5.60	88.8	17.8
11-3	30	70	40,000	3.39	Hydrogen	5.72	90.7	16.1
11-2	40	60	20,000	2.67	Argon-10 hydrogen	4.76	90.0	17.8
11-2	40	60	40,000	2.97	Argon-10 hydrogen	4.98	94.2	16.0
11-3	30	70	20,000	3.06	Argon-10 hydrogen	5.44	86.4	17.6
11-3	30	70	40,000	3.37	Argon-10 hydrogen	5.62	89.2	16.0

(a) Grade 1, The Beryllium Corporation, Reading, Pennsylvania.

(b) Specimens about 1/4 in. in diameter by 1/4 in. high were pressed in a tool steel die; 4 w/o Napco CD-108 binder was used.

(c) Specimens were sintered at 300 F for 2 hr.

Advanced ML-1 Fuel Pellets

J. W. Lennon and A. K. Smalley

Laboratory work on an oxidation-resistant ceramic fuel for use in the ML-1 was started during November. The present aim is to produce 1/4-in. -diameter by 1/4-in. -long pellets containing at least 60 volume per cent UO_2 . For the initial approach, UO_2 shot will be coated with Al_2O_3 by vapor deposition, the coated shot will be dispersed in Al_2O_3 powder, and the mixture will be pressed and sintered.

Some UO_2 shot averaging 715 μ (minus 20 plus 30 mesh), 127 μ (minus 100 plus 140 mesh), or 59 μ (minus 200 plus 324 mesh) in diameter was coated with up to about 10 μ of Al_2O_3 by vapor deposition. Oxidation resistance of the coated UO_2 was measured by heating 5-g samples at 1200 F in air for 1 and 5 hr and measuring their weight change. The percentage of UO_2 that was converted to U_3O_8 after 5 hr is shown below:

<u>Average Shot Size, μ</u>	<u>Coating Thickness, μ</u>	<u>UO_2 Converted to U_3O_8, per cent</u>
59	4-7	None
127	4-8	1.4
715	7-10	49

A second sample of the 715- μ shot was coated with from 10 to 13 μ of Al_2O_3 . Oxidation testing gave similar results.

Since from 4 to 8 μ of Al_2O_3 was sufficient to impart oxidation resistance to the two smaller size fractions of UO_2 , it appears that the difficulty in coating the 715- μ particles with a reliable coating of equal or even greater thickness may be inherent in the process. For example, the kinetic energy imparted to the large particles by fluidizing them may be sufficient to crack the coatings when the particles impact each other or the reactor wall.

Attention also was given to the sintering behavior of a series of Al_2O_3 powders of various sizes and degrees of sinterability. This work was aimed at selecting an Al_2O_3 matrix that sinters to a high density with little shrinkage. High density is desired for good thermal conductivity and strength and to aid in protecting the UO_2 against oxidation. Minimum shrinkage of the matrix Al_2O_3 is desired to prevent cracks in pellets from differential shrinkage; the coated UO_2 shot is not expected to shrink during sintering.

Specimens were made of mixtures of 900-mesh and 500-mesh fused Al_2O_3^* and a sinterable form of Al_2O_3 powder developed previously at Battelle. Specimens were pressed using a wax binder and sintered in hydrogen for 1 hr at 2800 F and at 3000 F. Densities of the specimens sintered at 3000 F ranged between 77 and 91 per cent of the theoretical. Shrinkages were between about 4 and 14 per cent. The composition sintering to the highest density also had the highest shrinkage, and conversely. Compositions high in the sinterable Al_2O_3 powder shrank most, and the composition highest in the 500-mesh Al_2O_3 shrank least. There were no indications that the desired combination of high density and low shrinkage could be obtained from mixtures of these Al_2O_3 powders.

*Grade 38, Morton Company, Worcester, Massachusetts.

Planned work includes further investigations on preparing coated UO_2 shot having size ranges suitable for maximum packing density. Experiments will be made to establish the maximum size of UO_2 particles which can be made oxidation-resistant by coating with about $10\ \mu$ of vapor-deposited Al_2O_3 . Preliminary tests will be conducted on incorporating coated UO_2 into several types of Al_2O_3 matrices.

An Alternate Method of Fabricating Advanced ML-1 Fuels

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

The objective of this program is to evaluate the simultaneous densification and cladding of UO_2 bodies diluted with no more than 40 volume per cent Al_2O_3 . In order to insure a continuous matrix of Al_2O_3 , thus providing some oxidation resistance, the UO_2 particles will be coated with Al_2O_3 prior to densification.

Six batches of UO_2 have been coated with Al_2O_3 as shown in Table U-2. Uniform dense coatings of alumina about 5 to $10\ \mu$ thick were applied to UO_2 particles 44 to $150\ \mu$ in diameter by hydrolysis of aluminum chloride vapor in a fluidized bed of the particles at 1830 F.*

TABLE U-2. Al_2O_3 -COATED UO_2

Batch	Type of UO_2	Coating Thickness, μ	Amount, g
A4	Numec natural UO_2 (-100 +140 mesh)	8 to 10	106.9
A5	Numec natural UO_2 (-100 +140 mesh)	5 to 7	101.00
A6	Numec natural UO_2 (-100 +140 mesh)	5 to 8	100.85
A7A	Mallinckrodt natural UO_2 (-270 +325 mesh)	7 to 9	113.45
A8A	Mallinckrodt depleted UO_2 (-270 +325 mesh)	5 to 6	107.0
A9A	50 w/o Mallinckrodt natural UO_2 -50 w/o Mallinckrodt depleted UO_2 (-270 +325 mesh)	5 to 7	107.55

Several compacts were pressed at 50 tsi. These compacts were made of Batch A6, having an average UO_2 diameter of $150\ \mu$. The coating thickness was 5 to $8\ \mu$. These conditions represent 88.90 volume per cent UO_2 or 74.41 w/o UO_2 with a theoretical density of 10.18 g per cm^3 . Compacts with a height-to-diameter ratio of 1.5 to 2.5 exhibited densities of 71.5 per cent of theoretical, and those having a height-to-diameter ratio of 1 or less gave densities of 75 per cent of theoretical. These compacts were loaded into stainless steel tubes and are being isostatically pressed 3 hr at 2300 F and 10,000 psi.

A cursory investigation is planned to check the oxidation resistance of the bare hot-pressed pellets and of clad elements containing intentional defects.

*Browning, M. F., Veigel, N. D., Cook, T. E., Diethorn, W. S., and Blocher, J. M., Jr., "Alumina Coating of UO_2 Shot by Hydrolysis of Aluminum Chloride Vapor", BMI-1471 (October 25, 1960).

Encapsulation Studies

W. J. Zielenbach, J. F. Lagedrost, and J. H. Stang

Two instrumented irradiation capsules are currently being operated to study elevated-temperature irradiation effects on Hastelloy X-clad compacts of highly enriched UO_2 , both alone and dispersed in a BeO matrix.

Capsule BMI-38-1 has operated in Position A-27-NE of the MTR since September 14, 1960; the specimens in this capsule consist of 70 w/o and 80 w/o UO_2 in BeO. Six specimens are stacked vertically in the capsule, the 80 w/o specimens being in the second and fifth positions from the top of the stack. Table U-3 presents a summary of capsule temperature data since startup.

TABLE U-3. SUMMARY OF PEAK TEMPERATURES DURING IRRADIATION OF BMI-38-1

MTR Cycle	UO_2 Content, w/o	Average Surface Temperature of Hottest Specimen, F	Average Heater Input, w
145	70	1450	1550
	80	1530	
146(a)	70	1570(b)	2000
	80	1680(b)	
147(c)	70	1310	--
	80	--	
148(d)	70	1330	740
	80	--	

- (a) The capsule was raised 6 in. before the start of this cycle to bring Specimens 3 and 4 into the peak of the axial flux profile.
- (b) These are estimated temperatures. The reference thermocouples adjacent to the specimens failed during the cycle.
- (c) The heaters were not operated during this cycle owing to thermocouple malfunction.
- (d) During shutdown it was possible to restore two reference thermocouples. However, one of these failed at midcycle. Cycle 148 is still in progress.

The temperature data from BMI-38-1 indicate that a maximum of about 3 a/o total burnup of uranium-235 will have accrued by the end of the present reactor cycle. A total burnup of 9 a/o at the end of the irradiation is desired.

Capsule AGN-BRR-III containing six UO_2 specimens was inserted in CP-12 of the Battelle Research Reactor on September 6, 1960. Since the addition of a heat-transfer barrier in the form of a helium-filled aluminum "boot" around the capsule body, the peak-flux-specimen temperature has been about 1680 F; 1725 ± 25 F was the original specification. The desired burnup of 2 per cent of the original uranium-235 atoms should be reached in about 2 more months of in-pile time.

High-Temperature Fatigue Evaluation of Hastelloy X Tubing

G. M. McClure, H. J. Grover, and A. R. Duffy

The objective of this study is to evaluate the fatigue performance of Hastelloy X tubing under high-temperature environment.

An S-N curve will be developed for each of five temperature levels (1400, 1600, 1735, 1800, and 2000 F) out to 10^6 cycles at a stress ratio of $A = 1$. Goodman diagrams based on failure at 10^6 cycles will also be developed at stress ratios of $A = 0$, $A = 0.25$, $A = 1.0$, and $A = \infty$ (approximately). About 120 specimens are required for the evaluation. All tests are being conducted at a cyclic speed of 1725 cycles per min, which will produce 10^6 cycles in about 10 hr.

End clamps which hold the tubing sufficiently tight during testing but which do not introduce detrimentally high stress intensification have been satisfactorily developed. Partially completed S-N curves for a stress ratio of $A = 1$ and temperature levels of 1600, 1735, and 1800 F indicate a stress of 18,500, 8,700, and 5,300 psi, respectively, is necessary to instigate the fatigue failure at 10^6 cycles. All specimens have failed near the center in the heat zone of controlled temperature.

During the month of November, 17 specimens were failed to develop the S-N curve for a stress ratio of $A = 1$ at the 2000 F temperature level. Preliminary reduction of data indicates that a stress of about 3200 psi is necessary to produce failure in 10^6 cycles. Of interest is the fact that the new supply of tubing used during the development of this S-N curve is slightly different dimensionally than the previous tubing.

Plans for December are to begin to establish the Goodman diagram points for stress ratios of $A = 0.25$ and $A = \infty$ (approximately) for the temperature levels of 1600 and 1800 F. Also, it is planned to calibrate a fatigue machine for use in developing the S-N curve for 1400 F at a stress ratio of $A = 1$. It is necessary to use a heavier beam for the lower temperature tests because of the higher loads involved.

ML-1 Critical-Assembly Experiments

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

Experiments on a mock-up of the ML-1 are being conducted for Aerojet-General Nucleonics in support of the Army Gas-Cooled Reactor Program.

Experiments measuring reactivity worths of two production-type fuel elements and a modified critical-assembly fuel element were performed during November. The production elements are for the ML-1 and 1B-2L reactor cores; the modified critical-assembly element is a mock-up of these production elements. In addition, final analysis of data from previous experiments was continued.

The elements under study were pin-type elements, containing 19 pins each. The center pin in each of the production elements was a solid rod of Hastelloy X. The center pin of the Modification 1' element was an empty tube of Hastelloy X of the same dimensions as the fuel pins in that element. The production elements had a nominal fuel loading of 74.5 g of uranium-235 per inner pin and 29.9 g of uranium-235 per outer pin, for a total loading of 805.8 g of uranium-235 per element. The Modification 1' element had a loading of 77.3 g of uranium-235 per inner pin and 26.9 g of uranium-235 per outer pin for a total loading of 791.1 g of uranium-235.

The reactivity worths of the three elements were measured in a core containing 51 standard critical-assembly elements. Table U-4 lists the reactivity of exchanging each of the experimental elements with six Modification 1' elements for seven standard critical-assembly elements.

TABLE U-4. REACTIVITY WORTHS OF GROUPS OF ELEMENTS

Group of Elements Exchanged	$\Delta\rho$ Over Basic Core (51 Standard Elements), per cent $\Delta k/k$		
	Position 1(a)	Position 13(a)	Position 49(a)
ML-1 production plus six Modification 1'	0.392	0.353	0.200
Seven Modification 1'	0.349	0.218	0.177
1B-2L production plus six Modification 1'	0.322	0.284	0.171
Standard element plus six Modification 1'	0.298	0.253	0.154

(a) Where applicable, the production or standard element was in the indicated core position, and surrounded by the six Modification 1' elements.

Table U-5 lists the reactivity worths associated with the exchange of a single element with the standard core element.

TABLE U-5. REACTIVITY WORTHS OF SINGLE ELEMENTS

Element Exchanged	$\Delta\rho$ Over Standard Element, per cent $\Delta k/k$		
	Position 1	Position 13	Position 49
ML-1 production	0.094	0.103	0.049
Modification 1'	0.055	0.064	0.021
1B-2L production	0.023	0.034	0.019

In December experiments will be performed to evaluate methods of using reactivity differences between elements in a single core position to extrapolate to the reactivity associated with the same element exchange throughout a full core. Experiments toward this goal will use standard core elements with the center pin removed to produce a perturbation in element fuel composition. Final data analysis and report writing will also continue during December.

CORROSION OF THORIUM AND URANIUM UNDER STORAGE CONDITIONS (LIVERMORE)

P. D. Miller

Factors related to blister formation during the corrosion of nickel-plated thorium are being studied.

The Phase III corrosion program in which specimens are being exposed at Columbus, Ohio, and Daytona Beach, Florida, has been under way for about a month. Very little corrosion other than tarnish-film formation has been observed to date.

The Phase II program using exposure to high humidity at 120 F has produced a few failures.

Electrodeposited Coatings for Thorium (SC)

J. G. Beach and C. L. Faust

Nickel-plated (1 to 2 mils) thorium panels (2 by 3 in.) resisted corrosion for more than 500 hr of exposure in water-saturated air at 200 F. After 50 to 100 hr of exposure several small blisters (<1/4 in. in diameter) were evident but did not increase in size during the next 400 hr of exposure. The blisters appeared to be associated with defects, such as nonmetallic inclusions, at the thorium metal surface. There appeared to be local areas of inferior adhesion at which the processing solution was entrapped, or at which there was accumulation of hydrogen. The plate around the blisters could not be peeled. Methods to eliminate these defects in the thorium or to overcome their deleterious effects on the plated coating will be studied further.

The following areas are being investigated:

- (1) Electropolishing of the thorium in a sulfuric-phosphoric acid solution to remove 4+ mils of the surface metal greatly improved the protective quality of the subsequent 2-mil nickel plate. Removal of <2 mils of the surface metal was less effective. Chemical pickling in a nitric-hydrofluosilicic acid solution was less effective than was electropolishing. Electropolishing solutions and conditions will be further appraised toward improving the thorium surface to receive sound protective coatings.
- (2) Activation of the thorium surface for adherent electroplating, using the procedure developed at BMI in 1948 and modified at LRL, results in a roughened or pitted thorium surface (up to 1-mil peak-to-valley distances). Elimination or minimizing this roughness should result in more uniform and smoother plated coatings. Thus, means of activating thorium for adherent electroplating without roughening will be investigated.

- (3) Hydrogen that accumulates in thorium as hydrides (ThH_2 and/or ThH_4) is suspected of being a cause for blistering of the nickel plate. An estimation of hydrides formed during preparation for and during plating will be determined, by analysis for hydrogen content, to help ascertain means of eliminating the detrimental effect of hydrides.
- (4) Joining of thorium to thorium or to other metals by soldering is aided by a plated coating such as nickel. The effects of heating during soldering on the protective quality and the bond strength of the nickel plate on the thorium will be evaluated.
- (5) Chromium plated over nickel-plated thorium to prevent tarnishing and/or corrosion of the nickel is being further studied. Three processes for plating "crack-free" chromium over as-plated or buffed nickel are being compared as to the relative corrosion protection afforded the basis thorium in water-saturated air at 200 F.

Further study of plated metals, other than nickel, directly on thorium is being recessed until the need for properties not supplied by the current nickel plates becomes apparent.

Electropolishing of thorium as a basis for organic coatings has shown promise of improving the protective quality of the organic coating. Improved adhesion and protective quality of the organic coating is attributed to a thorium phosphate film formed on the thorium surface during electropolishing. This film by itself imparts appreciable protection to thorium surfaces. Electropolishing prior to organic coatings will be investigated further.

Two organic materials will be appraised as coatings: (1) epoxy resins and (2) Type-K Tech-Pen ink, marketed by the Mark-Tex Corporation for marking laboratory equipment.

Corrosion Evaluation (UCRL)

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

In a Phase III corrosion-exposure program outlined last month in BMI-1473, specimens were exposed on October 20, 1960, in a warehouse and in an unsealed metal container on a roof at Columbus, Ohio. After about 1 month, none of the specimens have shown any evidence of corrosion other than the normal tarnish colors. In this program, specimens were also exposed in a closed shed at Daytona Beach, Florida, on October 26, 1960.

In a Phase II program, specimens are being exposed to 100 per cent relative humidity at 120 F. After 20 days, two uranium specimens with soldered joints were removed because of separation at the joints. Two uranium specimens coated with aluminum plate 4 and 9 mils thick were removed after 1000 hr of exposure because of severe blistering.

Both programs are being continued.

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

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

Beryllium-clad uranium dioxide fuel elements are being considered by Oak Ridge National Laboratory for use in gas-cooled reactors. The feasibility of fabricating these fuel elements by gas-pressure bonding is being studied.

The material for studying the self-bonding of beryllium as a function of surface preparation and bonding parameters has been received from ORNL. A series of specimens has been bonded for evaluation of bonding parameters for beryllium surface conditioned by pickling in an acid solution that produces a bright, metallic luster. This solution removes approximately 1 mil of beryllium surface metal and contains 450 ml of phosphoric acid, 26.5 ml of sulfuric acid, and 53 g of chromic anhydride. Bonding temperatures studied were 1550 and 1650 F with all specimens held at temperature for 1 hr at 10,000 psi.

In addition to the flat-plate surface-preparation specimens, a series of specimens embodying uranium dioxide cores is being bonded at various temperatures to study the compatibility of uranium dioxide with beryllium at different bonding temperatures. The gas-pressure-bonding process will produce very intimate contact between cladding and core; consequently, reaction is more likely to be observed in these specimens than in conventional diffusion-couple specimens.

Specimens are currently being pressure bonded to study the compatibility of beryllium with other materials. Beryllium was found to react with molybdenum when pressure bonded for 4 hr at 10,000 psi at both 1700 and 1800 F.

RWD:CRT/all