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PROGRESS RELATING TO CIVILIAN APPLICATIONS
DURING NOVEMBER, 1961

by

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REPORTS RELATING TO CIVILIAN APPLICATIONS
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- BMI-1548 "A Method for the Study and Correlation of Fission-Gas-Release Behavior of Fuel Materials During Irradiation", by Gilbert E. Raines, Charles W. Townley, Stephen D. Beck, and William H. Goldthwaite.
- BMI-1549 "Progress Relating to Civilian Applications During October, 1961", by Russell W. Dayton and Clyde R. Tipton, Jr.
- BMI-1551 "Diffusion Coefficients of Uranium and Carbon in Uranium Monocarbide", by Walston Chubb, Charles W. Townley, and Ralph W. Getz.

REACTOR MATERIALS AND COMPONENTS (AEC-DRD)

R. F. Dickerson

Research having the general objective of providing basic structural material and component support to the over-all Atomic Energy Commission reactor-development program is reported in the following section.

High-Pressure High-Temperature Solid-State Studies

A. P. Young, G. N. Panagis, D. A. Vaughan, and C. M. Schwartz

Objective

To determine if ultrahigh pressure can be utilized to produce new materials.

Program Approach

Samples of oxide mixtures are reacted at elevated temperatures under normal and ultrahigh pressures in sealed platinum capsules. Reacted samples are examined visually and by X-ray diffraction to determine phase relationships.

Progress Reported Previously

U_3O_8 -MgO, U_3O_8 -CaO, U_3O_8 -ZnO, and U_3O_8 -CdO mixtures run at 60,000 atm and 1200 C exhibit a UO_2 -type structure with reduced lattice parameters. At 1200 C and ambient pressure these mixtures either form uranates or do not react.

U_3O_8 -CaO high-pressure samples with a uranium/calcium ratio of 3 to 1 show diffraction lines of a simple cubic phase with a lattice constant of 5.38 A. U_3O_8 -CdO high-pressure samples with a uranium/cadmium ratio of 3 to 1 gave only the UO_2 face-centered-cubic diffraction pattern.

U_3O_8 - Al_2O_3 samples do not react at normal pressure and elevated temperatures while at 60,000 atm and 1200 C they react to form an unidentified phase. When seen at 90,000 atm and elevated temperature U_3O_8 - Al_2O_3 samples exhibit a UO_2 -type structure by X-ray diffraction analysis.

Work This Period

The divalent oxides CaO, CdO, MgO, and ZnO all react with U_3O_8 under pressure to form UO_2 -type structures. NiO is also a divalent oxide. Since MgO and NiO both have the sodium chloride structure with nearly the same lattice parameter ($a = 4.20$ A), in which Mg^{++} and Ni^{++} ions have the same Goldschmidt ion radius, 0.78 A, it was of interest to determine whether NiO and U_3O_8 would react. U_3O_8 and NiO were run at 1200 C at normal pressure and at 60,000 atm. There was no reaction at normal pressure. In samples run at 1200 C and 60,000 atm, an unidentified phase and NiO were detected by X-ray diffraction.

Pressure samples of U_3O_8 -CaO with a uranium/calcium ratio of 3 to 1 were heated at 300 and 600 C in air to determine whether the structure with a simple cubic lattice might be disordered to random fluorite structure in this manner. In this case the diffraction lines which indicate the simple-cubic-type lattice would disappear. In the sample heated at 300 C, there was little change in the pattern. At 600 C, there was some evidence of decomposition with faint U_3O_8 lines. However, the simple-cubic-type pattern was also present. If the simple-cubic pattern is due to an ordered fluorite structure with calcium at the corners and uranium at the face centers, it apparently cannot be made random by heating in air since decomposition occurs first.

ZrO₂ normally has a monoclinic structure at ordinary temperatures. At elevated temperatures, it has the cubic fluorite or tetragonal structure. X-ray data indicate that the high-temperature phases are denser. Therefore, pressure should decrease the transformation temperature. A sample of monoclinic ZrO₂ was run at 60,000 atm and 1150 C. The sample after pressing was still monoclinic. Either the sample did not transform or the quenching rate in the high-pressure cell was not rapid enough to preserve the high-temperature structure.

Plans for Future Work

Mixtures of U₃O₈-CaO with uranium/calcium ratios between 1 to 1 and 2 to 1 will be run to try to determine a phase boundary between the material with fluorite structure, which is seen in the 1-to-1 mixture, and the material with simple-cubic structure, which has been seen in both 3-to-1 and 2-to-1 mixtures. A 3-to-1 mixture will be run at much lower pressure than previously studied to try to determine the pressure limit for the formation of the "simple-cubic" phase.

U₃O₈-NiO mixtures with lower NiO content will be run to try to obtain single-phase reaction product with this mixture.

A U₃O₈-Al₂O₃ mixture will be run in the explosive-forming apparatus to determine whether material with the fluorite structure, which was formed statically, can be formed by high-pressure shock.

Irradiation-Surveillance Program on Type 347 Stainless Steel

W. E. Murr, R. Leiberman, F. R. Shober, and R. F. Dickerson

Objective

To determine the effects of fast-neutron irradiation (neutrons having energies greater than 1 Mev) on the mechanical properties of AISI Type 347 stainless steel.

Program Approach

Subsize tensile, cyclic-strain fatigue, and impact specimens of AISI Type 347 stainless steel are being irradiated in capsules in core positions of the ETR. Evaluation of irradiation-induced changes will be based on results of mechanical tests made after exposures of about 0.5 to greater than 3.0×10^{22} nvt (fast). The information thus obtained will be used to predict the safe operating life for in-pile loops constructed of this material.

Progress Reported Previously

The results of tests upon Charpy and Izod specimens irradiated to exposures of about 5.7×10^{21} nvt and tested at room temperature and -300 F were reported previously in BMI-1489 (Rev.). A definite embrittlement was noted in the irradiated specimens, particularly those tested at the lower temperature.

Work This Period

Several dosimeter wires that were exposed in the I-13 position of the ETR for Cycle 38 have been analyzed. This position was occupied by the surveillance capsules during ETR Cycles 34 through 41. The dosimeters included both pure nickel and pure iron wires for comparison of the instantaneous fast flux by the two techniques. Results of the analysis of the dosimeter wires are given in Table A-1. It can be seen that results obtained by both techniques for the I-13 West position are in reasonably good agreement, although the nickel-wire dosimeter consistently gives the lower of the two values. The magnitudes of difference between the two types of dosimeters is greater for the I-13 Center position. This larger difference is unexplainable by present data,

although it is known that the thermal flux is high (7.0 to 8.0×10^{14} nv) in both the Center and West positions. The iron-wire dosimeter data shown in Table A-1 continue to support the previous decision to use this technique for determining fast flux.

TABLE A-1. RESULTS OF DOSIMETRY FOR THE I-13 POSITION OF THE ETR FOR CYCLE 38

Location of Dosimeter Wire in Core-Filler Piece	Fast Neutron Flux, nv	
	As Determined by Nickel Dosimetry ^(a)	As Determined by Iron Dosimetry ^(b)
<u>I-13 West</u>		
12 in. above midplane	6.8×10^{13}	7.9×10^{13}
6 in. above midplane	1.4×10^{14}	1.9×10^{14}
Midplane	2.0×10^{14}	2.4×10^{14}
6 in. below midplane	2.4×10^{14}	2.8×10^{14}
12 in. above midplane	2.1×10^{14}	2.1×10^{14}
<u>I-13 Center</u>		
12 in. above midplane	4.4×10^{13}	7.2×10^{13}
6 in. above midplane	1.2×10^{14}	1.7×10^{14}
Midplane	1.8×10^{14}	2.6×10^{14}
6 in. below midplane	1.9×10^{14}	2.8×10^{14}
12 in. below midplane	1.8×10^{13}	2.6×10^{14}

(a) Determination of fast flux by counting Co^{58} isotope produced by the nuclear reaction, $\text{Ni}^{58} (n, p) \text{Co}^{58}$, with correction of the Co^{58} for thermal burnup, based on a $\text{Co}^{58} (n, \gamma) \text{Co}^{59}$ cross section of 1650 barns.

(b) Fast flux determined by assaying Mn^{54} isotope produced in iron wire by the nuclear reaction $\text{Fe}^{54} (n, p) \text{Mn}^{54}$.

The individual exposures of the stainless steel surveillance capsules as of the end of Cycle 38 (July 6, 1961) are reported in Table A-2. The ETR has completed three additional cycles since this time, but fast flux data are not yet available for determining additional capsule exposures.

TABLE A-2. STATUS OF CAPSULES IN TYPE 347 STAINLESS STEEL IRRADIATION-SURVEILLANCE PROGRAM

Capsule	Type of Specimen in Capsule	Total Exposure as of July 6, 1961, 10^{21}	
		Top	Bottom
BMI-24-2	Tensile and fatigue	8.495	9.226
BMI-24-4	Tensile and fatigue	7.226	8.91
BMI-24-6	Tensile and fatigue	9.589	7.93
BMI-24-8	Tensile and fatigue	7.910	8.696
BMI-24-10	Tensile and fatigue	8.569	7.065
BMI-24-12	Tensile and fatigue	9.709	9.313
BMI-24-16	Impact	8.895	8.082
BMI-24-18	Tensile and fatigue	4.072	5.586
BMI-24-20	Tensile and fatigue	3.786	6.054
BMI-24-22	Tensile and fatigue	4.130	6.006

The surveillance capsules were removed prior to ETR Cycle 42 from the I-13 to the G-13 position. It was erroneously reported in the previous progress report (BMI-1549) that the new capsule position was G-12.

A cycle-by-cycle revision of the estimated instantaneous fast flux to which all stainless steel surveillance capsules have been exposed is nearly completed. This revision is based upon corrected nickel dosimetry for early cycles, as well as the more recent iron-wire dosimeter data that have been accumulated for the appropriate core positions occupied by the surveillance capsules.

Plans for Future Work

The capsules will continue to be irradiated in the ETR core. As soon as the cycle-by-cycle flux revision is completed, exposures based upon the best available flux data will be used for calculating new accumulated exposures. At this time, one or more capsules will be discharged from the reactor and returned to Battelle for examination.

A topical report, summarizing the surveillance program from its inception to the present time, is being prepared.

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

F. R. Shoher and R. W. Getz

Objective

To compare the creep properties of Zircaloy-2 at 650 F in a fast flux of 1×10^{14} nv with those at 650 F in the absence of a fast flux and to study strain aging of Zircaloy-2 to elucidate the extent to which it may influence creep strength at 650 F.

Program Approach

In-reactor and out-of-reactor creep experiments are planned in which total deformations of sheet specimens in 1000-hr creep tests will be compared. The effects of stress under these creep conditions will be investigated by incorporating three different widths in the gage section of the test specimens. The reactor portion of these tests is to be conducted at the WTR and the out-of-reactor part at Battelle. Evidences of strain aging are being investigated by means of short-time tensile tests and deformation studies performed at room and elevated temperatures.

Progress Reported Previously

An instrumented capsule, described in an earlier progress report (BMI-1430), was designed, constructed, inserted in a core position of the WTR, and irradiated for one reactor cycle. The test temperature, 650 F, was maintained by a combination of electrical and gamma heating. As the result of heater failures, excessively high gamma heating in the specimen-grip section, and premature failure of the test specimen in the grip section, the influence of irradiation on the creep behavior of Zircaloy-2 could not be evaluated. The results were described in an earlier report (BMI-1524). A second in-reactor capsule was designed and included modifications to improve the heat-transfer characteristics of the grip sections of the test specimen. Bench tests were made on the second in-reactor creep capsule prior to insertion into the WTR which showed all components to be functioning properly. The capsule was transported to the WTR for irradiation. Failures of the bellows and some auxiliary heaters early in Cycle 18 precluded the possibility of gaining useful information from the experiment. Hence, the experiment was discontinued, and the capsule was discharged at the end of the Cycle 18.

Tensile tests made at 75, 350, 400, 450, 550, 650, and 750 F as part of strain-aging studies have shown a yield point after straining and aging at temperatures of 450 F and above. The presence of the yield point appeared to be related to the following: (1) grain size, (2) prior heat treatment, and (3) strain rate.

Additional tensile tests were made at 650 F to study the effect of solute concentration, grain size, and hydrogen concentration on the magnitude of the yield point. The results of the study of the effect of solute concentrations were inconclusive. Increasing the hydrogen content from 14 to 100 ppm did not appear to influence the yield-point behavior in any respect. However, an investigation of various strain rates from 0.0033 to 0.066 in. per in. per min showed a maximum drop in load at 0.0066 in. per in. per min, indicating some sensitivity of the yield phenomenon to the rate of deformation.

Work This Period

No new work was started relating to in-reactor creep testing of Zircaloy-2. Several additional short-time tensile tests were made at elevated temperatures on Zircaloy-2 and on sponge zirconium. A study was made of the magnitude of the drop in load at the yield point in Zircaloy-2 as function of the test temperatures from 450 through 750 F. It was found that the maximum drop in load occurred at 650 F. A yield point was also observed when unalloyed zirconium was tested at 650 F. Test conditions were the same as for Zircaloy-2, but the tests showed a somewhat smaller drop in load in sponge zirconium than in Zircaloy-2.

Metallographic examination has been completed on test samples representative of the heat treatments and test conditions studied. An attempt was made to relate drop in load on yielding with grain size and/or with the distribution of second phase in Zircaloy-2. It was found that drop in load decreased with increasing grain size to about 0.09 mm in diameter after which there appeared to be a slight increase in load drop. It is thought that this increase can be attributed to twinning of Zircaloy-2. If sufficient twinning occurred then in essence the effect of grain size is reduced. No major effect of the size and distribution of the second phase on the drop in load was noted.

Plans for Future Work

A topical report will be prepared.

Development of a Nondestructive Test for an Aluminum-Clad Uranium Fuel Element

V. W. Storhok, J. C. Smith, A. A. Bauer, and R. F. Dickerson

Objective

To develop a nondestructive test for the detection of uranium and nickel penetration into the aluminum cladding of Sylcor-produced uranium fuel slugs.

Program Approach

Attempts to use the more usual nondestructive test techniques such as ultrasonic energy and eddy currents in the detection of uranium and nickel penetration into the aluminum cladding of uranium fuel slugs have proven unsuccessful. However, beta-ray back-scattering technique appears to provide a reasonable method of detection. The technique requires a beta source and a detector to measure the number of beta particles from the source which are reflected back from the cladding. Since the number of beta particles reflected will vary with the atomic weight of the element, the

presence of either nickel or uranium in aluminum will produce an increase in back-scattered beta activity from a given source.

It is intended to investigate the feasibility of this technique and to secure performance data which will permit design of a suitable test unit. Two of the major requirements are speed of testing and minimization of human judgment in the evaluation of test results. To achieve the latter requirement, the test is being designed on the basis that 20 mils of unpenetrated aluminum cladding is acceptable. For this purpose, strontium-90 with a thin window shield to limit beta penetration to 20 mils of aluminum is being employed as the source.

Fuel slugs, supplied by Sylcor, will be used as test specimens.

Progress Reported Previously

Satisfactory test sensitivity could not be attained using 5 to 250 microcurie strontium-90 beta sources, necessitating fabrication of a stronger source. A lathe has been adapted for use as a test jig to move the fuel slug past the source-detector so that the entire slug surface can be scanned (BMI-1549).

Work This Period

On the basis of measurements made using flat aluminum and nickel sheet over a natural uranium plate as a stand-in for the fuel slug, it appears that a 2-millicurie strontium-90 beta source will give a satisfactory degree of test sensitivity. However, it is not certain whether the desired test speed can be attained. Cerium-144 was also considered for use as a beta source, but the high gamma background was found to interfere excessively with beta counting.

Plans for Future Work

Surfaces of fuel slugs known to be of sound quality will be surveyed to provide baseline back-reflection patterns. Slugs suspected of containing areas of excessive uranium and nickel penetration will be surveyed and areas where flaws are indicated will be sectioned and examined metallographically to determine the type of flaw capable of detection. Also, a slug in which defects are intentionally introduced will be similarly surveyed and examined. Variables to be investigated include test speed and distance of the source-detector from the slug surface.

Friction and Wear of Sliding Surfaces in Sodium Environments

W. A. Glaeser and J. W. Kissel

Objective

To develop general principles applicable to the design of mechanical parts involving sliding surfaces in high-temperature molten sodium.

Program Approach

The friction and wear between various materials resistant to corrosion in liquid sodium are being studied by recording the stick-slip friction and wear behavior of a ball sliding on a flat in liquid sodium. Interpretation of stick-slip traces is supplemented by analysis of surface films formed during sliding in sodium.

Progress Reported Previously

Improved friction and wear performance has generally been observed for metal and metal-alloy materials following the addition of molten sodium to the rubbing surfaces. Studies of the composition of surface films found before and after sodium

additions proved that reaction products were largely responsible for this behavior. On the other hand, friction did not decrease between cemented carbide surfaces in the presence of sodium. In this case, however, thermodynamic considerations do not favor the formation of reaction products.

Recent experiments have involved stainless steels, 2-1/2 Croloy, and nitrided steel. Results from these experiments indicated that improved friction and wear characteristics result from the use of molten sodium between the rubbing surfaces. Chromalized 52-100 steel demonstrated unusually low friction in the stick-slip apparatus. Kinetic dry friction was always below 0.4 throughout the entire temperature range; and, when sodium lubricated, coefficients between 0.1 and 0.25 were recorded for temperatures between 650 and 1200 F.

Work This Period

Results from friction experiments with dry and sodium-lubricated specimens of 52-100 steel were compared with those of the Chromalized specimens referred to above. These indicate increasing friction coefficients from room temperature to 1200 F - from around 0.7 to 1.3. When sodium was added, no such increase with temperature was noted - the coefficients ranged between 0.1 and 0.4.

Analysis of the friction records for stick-slip characteristics and for electrical resistance between specimens was completed for each of the alloy materials mentioned above.

Plans for Future Work

Efforts to utilize the damped-oscillation technique for measuring changes in the surface viscosity will be continued; and techniques for estimating rates of formation of oxide and molybdate films will be considered.

Reactor Flow Studies

L. J. Flanigan and H. R. Hazard

Objective

To study coolant flow and mixing in the PWR Core 2 reactor.

Program Approach

A 0.4-scale model of the reactor, using air as the working fluid, is being used to obtain data for the design of the prototype.

Progress Reported Previously

The model core was disassembled and the Tygon flexible instrumentation tubing, which reacted with model materials, was replaced with vinyl tubing which exposure tests showed does not react with model materials.

A plug-in panel board was constructed and mounted on the model stand. The panel board will provide a means for directly measuring small pressure differentials in the model with high-precision differential manometers.

Work This Period

In November, the model core was reassembled and installed in the model. The external instrumentation was modified to include the plug-in panel. The core orifices were resized and reinstalled in the model core.

An in-place calibration of the core orifice in each simulated fuel assembly is in progress.

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Plans for Future Work

In December, core-orifice calibration will be finished, and flow studies will be started.

STUDIES OF FUELS (AEC-DRD)

R. F. Dickerson

Research concerned with providing fuel-material support for the over-all reactor-development program of the Atomic Energy Commission is reported in this section.

Irradiation Studies of Niobium-Uranium Alloys

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

Objective

To investigate the irradiation stability of high-strength niobium-uranium alloys.

Program Approach

A program has been in progress to investigate and determine those physical, mechanical, and corrosion properties of niobium-uranium alloys which are of interest in evaluating their usefulness as reactor fuels. These studies have been completed, and the irradiation behavior of these alloys is now being investigated. For this purpose, two capsules containing 15 specimens each are to be irradiated. The specimens are to be irradiated to a burnup of 1 total a/o at temperatures of 1600 and 1800 F. The specimens include binary niobium alloys containing 10, 20, and 30 w/o uranium and ternary alloys containing 20 w/o uranium with 10 and 20 w/o zirconium.

Progress Reported Previously

Property determinations related to this program have been completed and were discussed in two topical reports, BMI-1400 and BMI-1536. The planning of and construction of capsules for irradiation tests were discussed in an earlier progress report, BMI-1464.

The first capsule, WR-20, was inserted in the WTR during the shutdown for Cycle 14. During the first cycle the capsule operated with a specimen center-line temperature of 1100 F. The temperature increased to 1300 F at the start of the next cycle and then a helium-45 volume per cent neon mixture was introduced into the capsule annulus to raise the specimen center-line temperature to 1600 F. A failure in the inlet valves of this capsule toward the end of the cycle prevented further mixing of neon and helium with a resultant lowering of the center-line specimen temperature to about 1200 F. The valves on this capsule could not be reactivated and the capsule was moved to a position expected to have a higher flux in an attempt to increase the specimen temperature. The expected increase in temperature did not occur, indicating a lower flux than for the previous position. Capsule WR-20 was subsequently removed from the reactor for further repositioning during the shutdown for Cycle 19.

Capsule WR-21 was inserted during the shutdown for WTR Cycle 18. Temperature measurements indicated that desired specimen center-line temperatures of about 1800 F were obtained.

Work This Period

Capsule WR-20 was repositioned in the WTR during a midcycle shutdown and a center-line temperature of approximately 1200 F was obtained. Only one thermocouple remains operative and no further moves are planned.

Capsule WR-21 is currently operating with a maximum specimen center-line temperature of about 1820 F.

Plans for Future Work

Specimens in the two capsules will be irradiated to a total of 1 a/o burnup after which they will be returned to the Battelle Hot-Cell Facility for postirradiation examination.

Development of Uranium-Containing Thorium Compounds

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

Objective

To investigate thorium-uranium compounds with the aim of developing high-temperature fuels for thermal breeder reactors and of determining those properties which will indicate the suitability of such compounds for various reactor applications.

Program Approach

Thorium-uranium aluminides, beryllides, borides, carbides, nitrides, silicides, and sulfides were selected as promising materials for study using high melting point, low thermal-neutron cross section, high thorium (and uranium) density, and crystal structure as the criteria for selection. These compounds are to be evaluated as to preparation, corrosion resistance in various media, and mechanical and physical properties. The four most promising compounds have been selected for more detailed study, primarily on the basis of ease of preparation by casting as determined during the first year of study. These compounds and compositions are $(Th_9U)B_4$, $(Th_9U)Be_{13}$, $(Th_9U)C$, and $(Th_9U)C_2$.

Progress Reported Previously

During the first phase of this program, data were obtained on thorium and thorium-uranium compounds of aluminum, beryllium, boron, carbon, and silicon. The results of these studies are reported in the following progress reports: BMI-1480, BMI-1489, BMI-1496, BMI-1504, BMI-1509, and BMI-1514.

Three cylindrical castings (3/8 in. diameter by 2-1/2 in. long) of each of the four compounds selected for study have been cast for use in more extensive property determinations. Radiographs of these castings indicated that the $(Th_9U)C_2$ castings were sound; however, two each of the other three compositions showed cracks. Additional castings of the $(Th_9U)C$ compound were prepared and were found to be crack-free.

Work This Period

High-purity boron has been obtained and casting of the $(Th_9U)B_4$ compound is in progress. High-purity beryllium for use in preparing the $(Th_9U)Be_{13}$ compound is still on order.

Thermal-conductivity, thermal-expansion, and compressive-strength measurements at temperatures up to 1800 F are currently being conducted on the $(Th_9U)C$ and $(Th_9U)C_2$ compounds. Some difficulties were encountered in the initial compression tests because of misalignment of the specimen ends. This misalignment produced a shear stress in the specimens and led to premature failure. This is being corrected by more rigid specimen machining tolerances which will produce more exactly parallel ends.

Specimens for testing the compatibility of two carbides with potential cladding materials (molybdenum, niobium, vanadium, zirconium, stainless steel) at 1000 and 1500 F are being prepared. Disks of the cladding materials and the carbides are to be sandwiched into a tantalum tube. The metal and carbide disks will be brought into intimate contact by pressure exerted by a push rod of high-thermal-expansion material (such as stainless steel) and an outer shell of a low-thermal-expansion material (such

as Invar). As the assembly is heated the differences in thermal expansion will force the metal and carbide disks together.

Plans for Future Work

Testing of the $(\text{Th}_9\text{U})\text{C}$ and $(\text{Th}_9\text{U})\text{C}_2$ compounds will be completed.

Casting and radiographic examination of the $(\text{Th}_9\text{U})\text{B}_4$ compound will be completed, and specimens will be prepared for property evaluation.

As sound casting of the borides and beryllides are produced specimens will be prepared for various physical- and mechanical-property studies. These include thermal-conductivity, thermal-expansion, hot-hardness, and compression-strength measurements, and oxidation and compatibility tests.

A Study of Plutonium-Containing Fuel Alloys

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

Objective

To investigate those plutonium alloys which appear to have potential as fuels for relatively high-temperature propulsion applications or for possible use as "spike" fuel elements in power reactors.

Program Approach

Plutonium-containing alloy systems have been selected for study mainly on the basis of the behavior of similar uranium-alloy systems. The compositions selected are based on the niobium, thorium, iron, uranium-molybdenum, and uranium-niobium systems. These systems are being studied at the plutonium-lean end. Initial evaluation is based on metallographic studies and hardness measurements. Those alloys exhibiting desirable phase distribution and evidence of strength on the basis of hardness measurements will be subjected to more detailed property studies.

Progress Reported Previously

A niobium-25.6 w/o plutonium-6.1 w/o silicon alloy prepared using master-alloyed PuSi_2 was found to consist of approximately 30 volume per cent PuSi_2 in a niobium matrix (BMI-1549).

On the basis of hardness measurements, 90 per cent cold reduced thorium-5 and -10 w/o plutonium alloys exhibit no tendency to recrystallize within 10 min at either 500 or 550 C (BMI-1549). At 600 C, the alloys appear to recrystallize within 3 to 5 min (BMI-1546).

Uranium-niobium-plutonium and uranium-niobium-zirconium-plutonium alloys continued to exhibit cored microstructures after being heat treated for 5 hr at 800 C (BMI-1549).

Work This Period

Uranium-10 w/o niobium-5, -10, -15, and -20 w/o plutonium alloys were heat treated 6 hr at 900 C and furnace cooled. This resulted in improved homogeneity, although some coring was still evident. The coring appeared most severe in the alloys containing 15 and 20 w/o plutonium. In addition to the cored dendritic structure, the microstructures appeared to consist of gamma uranium-niobium grains surrounded by a transformation product at the grain boundaries. Apparently the cooling rate through the transformation range was rapid enough for partial retention of the gamma phase. Hardnesses of 431, 494, 506, and 509 DPH were measured for the heat-treated alloys containing 5, 10, 15, and 20 w/o plutonium, respectively.

Iron-27.6 w/o plutonium and iron-27.6 w/o plutonium -11.9 w/o chromium alloys have been prepared by arc-melting techniques. These alloys are expected to consist of approximately 30 volume per cent PuFe_2 in iron and iron -20 w/o chromium matrices. The chromium addition should strengthen the iron matrix, improve oxidation resistance, and stabilize ferritic iron up to the liquidus temperature, while probably not affecting the PuFe_2 .

Plans for Future Work

Studies to determine the effects of heat treatment on uranium-niobium-plutonium and uranium-niobium-zirconium-plutonium alloys will continue.

Attempts will be made to coalesce the PuSi_2 phase in the niobium-25.6 w/o plutonium-6.1 w/o silicon alloy by heat treatment.

The effects of heat treatment on the distribution of the PuFe_2 phase in the iron and iron-chromium-base alloys and the fabricability and melting temperature of these materials are to be investigated.

Measurements of the Volatility of PuO_2

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

Objective

To determine the volatility of PuO_2 in the 1400 to 1750 C temperature range.

Program Approach

The volatility of PuO_2 will be determined in various atmospheres. Transpiration vapor-pressure techniques will be utilized with analysis of the condensate accomplished by absolute alpha-radiation-counting methods. On the basis of an abbreviated prior study, it is predicted that PuO_2 will be the congruently vaporizing plutonium-oxygen compound in nonreducing atmospheres. The vapor pressures are expected to be several orders of magnitude lower than those for uranium oxides under oxidizing conditions. X-ray diffraction studies will be performed subsequent to some of the transpiration runs in an attempt to identify the elevated-temperature structure of the oxides. Such interpretation of room-temperature studies may be justified on the basis of the relatively sluggish phase transformations generally associated with oxide systems.

Progress Reported Previously

Transport rates have been measured at room temperature and determined to be temperature independent. The mobility is attributed to alpha self-propulsion and is sufficiently high to require compensation during vapor-pressure calculations. Gas flow is conducive to the particulate mobility.

The variation in vapor pressure as a function of temperature has been determined in air at 1500, 1555, 1600, 1650, and 1700 C with an essentially linear relationship between temperature and vapor pressure resulting. The range of vapor pressures was between 3.17×10^{-9} atm at 1500 C and 2.03×10^{-8} atm at 1700 C.

Work This Period

The heating element in the transpiration apparatus failed early in the month, and replacement was delayed by the order time for the platinum-rhodium alloy. Only one volatility determination was performed. At 1450 C in air the measured vapor pressure was 1.41×10^{-9} atm. This value is slightly higher than would be predicted; however, the condenser was left in the apparatus for an unknown period of time. Alpha contamination was probable.

Plans for Future Work

The replacement heating-element material has been received and is being fabricated. Experimental work will begin very quickly and will be first directed toward expanding the range of temperatures investigated in air atmospheres and in verifying earlier work. When air has been evaluated as an atmosphere, determinations will be made in other gaseous media.

Fission-Product Release From Refractory Fuels

J. B. Melehan and F. A. Rough

Objective

To establish an understanding of the important causes of fission-product release during irradiation of refractory fuels, starting with uranium dioxide, and to obtain engineering information which will aid in the design of fuel elements utilizing these fuels.

Program Approach

The present program consists of experiments to study the fission-gas-release behavior of UO_2 during irradiation. UO_2 samples having densities between 90 and 100 per cent of theoretical will be irradiated in the BRR at temperatures to 3000 F. The atmosphere surrounding the irradiation sample will be a streaming mixture of purified helium and hydrogen. Released fission gases will be continuously transported in the gas stream to a single-channel gamma-ray spectrometer for determination of fission-gas concentrations in the streaming atmosphere. Following this continuous activity monitoring, the fission gases will be extracted from the atmosphere stream and concentrated by adsorption on refrigerated charcoal. Gamma-ray spectrometric analysis of the concentrated fission gases will establish the temperature and time dependence of fission-gas release rates.

Progress Reported Previously

A series of UO_2 irradiations spanning the temperature range of 700 to 3000 F was completed. Release-rate data were obtained for the isotopes krypton-85m, krypton-87, xenon-133, and xenon-135. The release rates for xenon-135 were utilized in the calculation of xenon-135 diffusion coefficients. At 2700 and 3000 F, adjustments had to be made in the diffusion-coefficient solutions to take into account sintering effects that occurred in the UO_2 specimen and corresponding changes in the surface area. It was found that below 1900 F there was no significant increase of release rates or of apparent diffusion coefficients with increasing temperature. Above 1900 F, the diffusion coefficients reflected a thermally activated fission-gas-release mechanism with an activation energy of 95 kcal per mole.

Work This Period

A sample of natural uranium-5 w/o carbon alloy was irradiated in the temperature range 700 to 2600 F. Released fission gas was periodically sampled during irradiation and analyzed by gamma spectrometry for concentrations of several krypton and xenon isotopes. From the information on fission-gas abundances in the gas samples, calculations were made of the fractional release rates (R/B , where R is the instantaneous isotope release rate, atoms per sec, and B is the steady-state isotope production rate, atoms per sec). Irradiations of several days at 700, 1650, 1900, and 2300 F were completed. While evaluation of the results is continuing, it appears that for xenon-135 R/B remains in the region 5×10^{-4} to 1×10^{-3} throughout the temperature range from 700 to 2300 F. Failure of the vacuum system at 2600 F with possible damage to the

carbide specimen limited the experimental results obtained at that temperature. A similar treatment of experimental results for other xenon and krypton isotopes is continuing.

Plans for Future Work

The irradiation of UO_2 and UC with measurements of fission-gas release will continue with a view to increasing the understanding of the mechanisms of fission-gas release. Data obtained to date will be examined further to develop a more detailed understanding of the processes going on.

Radiation Stability of Aluminum-Uranium Fueled Glass Fiber Fuel Plates

R. J. Burian and J. E. Gates

Objective

To determine the effects of reactor irradiation on the mechanical properties of a material composed of aluminum-50 volume per cent uranium-fueled glass fiber.

Program Approach

Tensile and bend-test specimens will be prepared from fuel plates fabricated by the Clevite Corporation from aluminum-50 volume per cent glass fibers. The glass fibers contain about 50 w/o fully enriched U_3O_8 . The specimens will be irradiated to a burnup of about 25 per cent of the uranium-235 at temperatures below 100 C. This material was developed by the Clevite Corporation as a part of the AEC Fuel-Cycle Development Program. The radiation stability of the material will be evaluated by a comparison of mechanical properties before and after irradiation.

Progress Reported Previously

Six clad and three unclad specimens were fabricated from aluminum-uranium-fueled glass-fiber compacts. The specimen design and materials were described in BMI-1509, and the capsule design was described in BMI-1518 and BMI-1534. The capsule, BMI-41-1, containing the nine specimens was inserted into an MTR core position during the week of September 4, 1961 (shutdown for MTR Cycle 162).

Work This Period

Capsule BMI-41-1 was discharged from the MTR on November 6, 1961, after irradiation for 50.2 days to an estimated burnup of about 25 per cent of the uranium-235. The fast-neutron flux (>1 Mev) during the irradiation is estimated to have been 2.5×10^{13} nv. The maximum specimen temperature during irradiation was less than 100 C.

After irradiation, the capsule was clean and bright and evidence of thermal or mechanical damage was not visible on the exterior surface of the capsule body. The recovery of the specimens from the capsule was initiated.

Plans for Future Work

The postirradiation examination of the specimens will begin as soon as they are recovered from the capsule. These examinations will include visual inspection, measurements of dimensional distortion, and a study of the changes in selected structural properties.

Effects of Fabrication Variables on the Radiation
Stability of Uranium Carbide

D. G. Freas, E. O. Fromm, J. F. Lagedrost,
J. H. Stang, J. E. Gates, and R. F. Dickerson

Objective

To investigate the effects of compositional and fabrication variables on the radiation stability of uranium monocarbide at high irradiation temperatures and high fission-heat production rates.

Program Approach

Five uranium carbide specimens will be irradiated in each of three instrumented capsules in the ETR as part of the United Nuclear Corporation fuel-cycle program. These specimens will be fabricated by United Nuclear by both arc-casting and powder-metallurgy techniques. The carbon contents of the specimens will range from approximately 4.4 w/o to 5.2 w/o and the uranium will be of 12 per cent enrichment. Each fuel specimen will be 3/8 in. in diameter by 3/4 in. long and will be immersed in liquid sodium inside a 30-mil niobium-1 w/o zirconium can.

The anticipated irradiation conditions for these specimens will include surface temperatures of 750 C and center-line temperatures of 1250 C. The target irradiation exposure is a 15,000-MWD/T burnup of the uranium. To evaluate the effects of the compositional and fabrication variables, the postirradiation examinations will include measurements of physical properties and fission-gas release and studies of microstructure.

Progress Reported Previously

The design and fabrication of the irradiation capsules described in BMI-1534 have been completed. The niobium cans in which the fuel specimens will be immersed in sodium have been fabricated, and the final purification of the sodium has been performed. Uranium-4.4 w/o carbon alloy and uranium-5.2 w/o carbon-alloy specimens are being separately exposed to sodium in niobium-1 w/o zirconium cans at 1500 F (the surface temperature of the irradiation specimens) for 200 hr as a compatibility test.

Work This Period

Experiments to study the compatibility of uranium-4.4 w/o carbon alloy and uranium-5.2 w/o carbon alloy in sodium at 1500 F for 200 hr are complete. A comparative microstructural study of these alloys both before and after heat treatment did not reveal any structural changes. Also, no changes in physical dimensions or density were detected. Although the results of these studies do not suggest any reaction between the uranium carbides and sodium, a further microstructural study of the niobium-1 w/o zirconium cans in which the specimens were contained will be performed to determine if the sodium may have acted as a carbon-transfer medium. The irradiation specimens representing two types and compositions of uranium carbide have been received from the United Nuclear Corporation. Microstructural and physical-property studies of these specimens are presently being conducted.

Plans for Future Work

Studies of the physical properties of the irradiation specimens will be conducted as they are received from the United Nuclear Corporation. As these studies are completed, the specimens will immediately be encapsulated in the niobium-1 w/o zirconium cans in sodium. After the specimens have been encapsulated, the three capsules will be shipped to the ETR for irradiation.

Irradiation Study of Metastable Beta-Phase Uranium Alloys

M. S. Farkas, D. B. Hamilton, A. A. Bauer, and
R. F. Dickerson

Objective

To study the irradiation behavior and phase stability during irradiation of metastable beta-phase uranium alloys.

Program Approach

Uranium alloys with additions of 0.3 w/o chromium and 0.3 w/o chromium-0.3 w/o molybdenum designed to retain the metastable beta phase have been developed by Nuclear Metals, Inc. Eighteen specimens of these alloys, prepared from uranium of 10 per cent enrichment, in a variety of conditions of heat treatment, have been irradiated in two temperature-controlled capsules. The specimen compositions and heat-treatment conditions were reported in a previous progress report, BMI-1489.

The capsules were designed to maintain the specimen center-line temperature at 850 F at an effective flux of about 10^{13} nv. The specimens in the first capsule were irradiated in the BRR to an estimated burnup of 0.008 a/o over a 3-day period to determine the stability of the beta phase. The specimens in the second capsule were irradiated under similar conditions in the MTR to an estimated burnup of about 0.3 a/o to investigate the irradiation stability of the alloys.

Progress Reported Previously

Results of the postirradiation examination of the beta-phase uranium alloys irradiated in the BRR were presented in two earlier progress reports, BMI-1518 and BMI-1524.

Results of the MTR irradiation were reported last month in BMI-1549.

Work This Period

Cesium-137 analyses of one specimen from each capsule were performed to determine burnup. The total uranium burnup for specimens from the BRR and MTR capsules was found to be 0.0051 and 0.226 a/o, respectively.

X-ray diffraction analyses of specimens from the BRR capsules are nearly complete. The results thus far are given in Table B-1.

Plans for Future Work

Dosimeter wires from the MTR capsule will be analyzed to provide flux-profile data which will be used to approximate burnup of all the specimens in the capsule.

TABLE B-1. X-RAY DIFFRACTION RESULTS FROM METASTABLE BETA-PHASE URANIUM-
ALLOY SPECIMENS IRRADIATED IN THE BRR

Specimen Composition (Balance Uranium), w/o	Heat Treatment	Observed Peaks, A	
		Alpha	Beta
0.3 chromium-0.3 molybdenum	As extruded (unirradiated control)	2.542	2.700
		2.292	2.585
			2.508
			2.430
			2.350
0.3 chromium-0.3 molybdenum	As extruded	2.270?	2.592
			2.550
			2.515
			2.435
			2.355
			2.304
0.3 chromium-0.3 molybdenum	Beta quenched and alpha transformed	2.595	2.366
		2.542	
		2.494	
		2.292	
	Gamma quenched	To be run	

GENERAL FUEL-ELEMENT DEVELOPMENT (AEC-DRD)

S. J. Paprocki

These studies are being performed for the AEC Division of Reactor Development.

Irradiation of Cermet Fuels

E. Fromm, J. H. Stang, G. W. Cunningham, and D. E. Lozier

Objective

To investigate the irradiation stability of UO_2 -refractory metal (chromium, molybdenum, and niobium) cermet fuels.

Program Approach

Two capsules containing clad (with 35 mils of Type 430 stainless steel or niobium) cylindrical specimens of UO_2 -refractory metal fuels have been prepared for high-temperature high-heat-flux irradiation at the Materials Testing Reactor. Three refractory-metal matrices, chromium, molybdenum, and niobium are involved; each is combined with 80 volume per cent minus 100 plus 200-mesh spherical UO_2 enriched to 20 per cent. Each capsule contains six specimens, two each of the three materials indicated above, and is equipped with thermocouples for specimen-temperature monitoring. Target center-line temperatures for the three cermets are 2800 F for the UO_2 molybdenum and the UO_2 -niobium, and 2400 F for the UO_2 -chromium. The two capsules are to be irradiated in similar thermal-neutron fluxes but for different periods of time to achieve two levels of specimen burnup (approximately 2 per cent of the total uranium atoms present during three normal MTR cycles in the one case, and approximately 5 a/o during eight normal cycles in the other).

Progress Reported Previously

The irradiations of the two capsules, BMI-39-1 and BMI-39-2, which were inserted into the MTR at the beginning of Cycles 162 (September 4, 1961) and 163 (September 29, 1961), respectively, were continued. Temperature data recorded during the early part of the irradiations, as reported in detail in BMI-1546, indicated that the specimen center-line temperatures were fairly close to those desired, the largest estimated deviations being approximately 300 F.

The thermocouples in BMI-39-1 became erratic early in the irradiation. It is probable, however, that operating temperatures did not vary significantly from initial satisfactory levels. Temperature performance of BMI-39-2 was satisfactory.

Work This Period

The irradiation of BMI-39-1 was terminated on November 6, 1961, as scheduled after a 50-day exposure. A specimen burnup level of approximately 2 per cent of the total uranium atoms present is estimated for this irradiation. The capsule was shipped to the Battelle Hot-Cell Facility for postirradiation examination.

The irradiation of BMI-39-2 was continued. Although some of the thermocouples have become erratic, performance is still satisfactory.

Plans for Future Work

The irradiation of BMI-39-2 will be continued. The experiment is scheduled for discharge in March, 1962.

Fabrication of Cermet Fuel Elements

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

Work on this program was recessed during November.

Development of Low-Cost Tubular Fuel Elements

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

Objective

To develop techniques for fabricating low-cost tubular fuel elements of consistent quality.

Program Approach

Several techniques for fabricating tubular fuel elements have been considered in this program. One approach has been selected for further development as it appears to offer the best possibility of meeting the two basic requirements of this program; low relative cost with consistent quality. The process selected involves the assembly of concentric cladding tubes to form an annulus. The annulus is then loaded with dry-blended fuel and matrix powders, and the tube assembly is evacuated and sealed. Compaction and sizing of this assembly are accomplished through explosive forming in a die. Sintering of the compacted annulus and core-cladding bonding are achieved by gas-pressure bonding.

Progress Reported Previously

Studies to date in the concentric-component approach have resulted in a reference procedure being established for loading of blended core powders, compaction and sizing, and pressure bonding to produce a tubular fuel element. A combination of thermal cycling and loose-powder loading of the annulus formed by concentric cladding tubes provided the most uniform and highest as-loaded densities. Stainless steel powder end plugs are incorporated during the loading cycle at both ends of the tubes.

Compaction and sizing using explosives resulted in tubes which had uniform overall dimensions as well as smooth inside and outside surfaces. A special two-part split die is used to shape outside of the tubes. The explosive is placed inside a mild steel sleeve that is inserted in the fuel tubes prior to detonation. Use of this sleeve helps to densify the powder core more uniformly and to produce a smooth inside surface. It is subsequently removed by acid pickling. The compacted and sized fuel tube is then loaded into an autoclave and gas-pressure bonded at 2100 F and 10,000 psi for 3 hr. After bonding, the wrought end plugs are removed and the bonded tubular fuel element is complete. Tubes approximately 6 in. in length have been produced to date, but a larger explosive die is being constructed to allow tubes 2 ft in length to be made.

Work This Period

Construction of an explosive-forming backup die for compacting and sizing tubular fuel elements 27 in. long has been completed. The die consists of a tapered split-die insert which fits into an outside housing divided into four parts. Heat-treated Vega tool steel was used for both the die insert and housing. Close tolerances were maintained between the insert and housing to prevent the insert from becoming enlarged from repeated use. A means for evacuating a portion of the die was provided to prevent inadvertent welding between the specimen cladding and die wall resulting from entrapped

air. The sections of housing are provided with grooves and O-ring washers on their mating surfaces to effect a vacuum seal at these areas. The top and bottom housing sections contain annular washers for sealing the top and bottom of the assembly. This method of sealing permits both ends of the assembly to remain open for relief of the pressures resulting from the explosives. The explosive charge is placed inside the tube specimen to be compacted after it is positioned in the split die. Detonation takes place with the entire die assembly submerged in water.

Plans for Future Work

Additional tube specimens are planned for electron-beam welding studies to alleviate the problems that have been met in sealing the as-loaded tubes. Fuel tubes 27 in. long will be loaded, compacted, and pressure bonded for evaluation of finished dimensions, fuel-dispersion characteristics, and core-cladding bonding. The results of these tests will determine what changes might be necessary in the reference process to obtain the desired results.

An analysis of the fabrication costs involved for this approach will be made for comparison with those for other ways for producing dispersion fuel elements.

Gas-Pressure Bonding of Ceramic Fuel Elements

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

Objective

To develop and evaluate the gas-pressure-bonding process as applied to vibratory-compacted fuel rods supplied by GE-Hanford and rolled UO₂ sheet materials as supplied by Gladding, McBean & Co.

Program Approach

This study concerns two areas of research of interest in cooperative programs with Hanford and Gladding, McBean & Co. In these programs, the gas-pressure-bonding process as combined with vibration powder-packing techniques and UO₂ sheet-rolling techniques is being investigated. The vibration packing-pressure bonding studies are directed toward the definition of practical vibration-packed UO₂ materials which will provide the initial density and ceramic activity required for successful pressure bonding. Flat-plate designs consisting of pressure-bonded sheet-rolled UO₂ will serve to demonstrate unique low-cost fuel-element designs.

Progress Reported Previously

In previous studies, the evaluation of various types of uranium dioxide powders has been carried out on the basis of their initial compacting and pressure-bonding characteristics. The results of this work and the development of pressure bonding techniques for various fuel geometries were discussed in a phase report, BMI-1475. More recently, preliminary process specifications for pressure bonding several basic Type 304 stainless steel-clad UO₂ fuel-element shapes were partially developed. A report of the fabrication of fuel rods for irradiation testing in the VBWR has been drafted.

Initial tests with pressure-bonded sheet-rolled UO₂ platelets have demonstrated behavior similar to that expected from pressed and sintered cores. A slight tendency toward cracking during bonding has been reduced by altering the rate of pressure application during initial heating.

Work This Period

Compacted and assembled fuel rods for pressure bonding have been received from Hanford. The cladding materials represented in these tests include Inconel X, Type 304L stainless steel, Type 406 stainless steel, Zircaloy-2, and Zircaloy-4. The uranium dioxide in these samples, which consist of individual types and mixtures, has been compacted by either vibration or swaging. Presently, these fuel rods are being gas-pressure bonded at conditions normally required for self-bonding of the respective cladding materials.

A brief comparison of the pressure-bonding behavior of ceramic uranium dioxide powders as obtained from different suppliers has been initiated. It is intended that this series of tests will define a reproducible commercially available oxide most suitable for pressure bonding subsequent to vibration packing. Initial samples have been pressure bonded for 3 hr at 2100 F and 10,000 psi. In general, structures of uniform high density were obtained; however, the oxygen/uranium ratio and density determinations did not appear consistent with these structures. Further tests are being conducted to resolve these differences.

Plans for Future Work

Future tests will include evaluations of the pressure-bonded Hanford assemblies and additional tests of Gladding, McBean & Co. cores as contained in pressure-bonded flat-plate elements.

Factors Affecting Pressure Bonding

D. C. Carmichael and J. W. Spretnak

Objective

To determine the mechanism of solid-phase bonding of metal components under the application of heat and pressure.

Program Approach

Hot pressing or gas-pressure bonding under various conditions of temperature, pressure, and time are being used to bond specimens for study. The specimens are of different designs and preparations and are composed of two components of the same or different metals. Evaluation of the bonds is based chiefly on metallographic examination, using special techniques. Observations are made of the extent of contact between mating surfaces and of the behavior of grain boundaries, dislocations, and interfacial voids which affect the elimination of the interface during bonding.

Progress Reported Previously

The mechanism for the self-bonding of copper has been established and the effects of pressure, temperature, grain growth, impurities, and surface roughness have been investigated. It was found that the first basic requirement for producing a metallurgically perfect bond is the application of sufficient pressure to place the surfaces in intimate contact, which for copper could be related to Meyer hot-hardness values. The second requirement was found to be the minimization of voids in the bond interface which prevent grain growth across the interface. These results are discussed in detail in a topical report, BMI-1512.

The pressure-bonding mechanism is now being studied in more detail and in other metals than copper. Results of recent studies of the deformation behavior during formation of bonds in nickel-copper couples as compared to copper-copper couples have been discussed in a previous progress report, BMI-1549.

Work This Period

The deformation mechanism for bringing the surfaces into intimate contact for the self-bonding of aluminum is being studied. Relationships are being determined between the applied load and temperature of hot pressing and the resulting area of contact between the mating surfaces. For this study, platelets of pure, annealed aluminum with surfaces shaped to a roughness of 500 μ in. rms are being pressed into similar platelets with smoothly ground surfaces. The rough surfaces could not be shaped to the uniform geometry desired, however, which prevents accurate mathematical treatments of the results but still permits general comparison and analysis.

Aluminum-aluminum couples of this type have been vacuum hot pressed at 620 F and 2000 psi for 5 min and at 230 F and 10,000 psi for 5 min and have been evaluated. For comparison with the previously bonded and reported copper-copper couples, the aluminum bonding temperature of 620 F represents the same fraction (about 0.5) of the melting point as did the 1000 F bonding temperature used for the copper couples. At the temperature of 230 F, the strength of pure aluminum is comparable to the strength of pure copper at 1000 F. The fractional area of contact measured for the 620 F and 2000-psi specimen was 0.51 and for the 230 F and 10,000-psi specimen it was 0.47.

Thus, the area of contact obtained at 2000 psi in the aluminum couple at 620 F was considerably greater than that produced at the same pressure in a copper couple at 1000 F, as was expected since the strength of aluminum is less than copper at the respective temperatures. On the other hand, the aluminum couple bonded at 10,000 psi and 230 F showed much less contact area than a copper couple pressed at 10,000 psi and 1000 F, although their strengths are comparable at their respective bonding temperatures. This difference is apparently due to a work-hardening effect in the aluminum as related to the mechanism of flow in bringing the rough surface into contact with the smooth surface. Comparison of the mechanism of flow in the two aluminum specimens based on metallographic examinations shows that the asperities of the rough aluminum surface were flattened more during bonding at 620 F than at 230 F. It was not possible in these experiments to eliminate a thin oxide film on the bonding surfaces, but it is not expected to have affected the mechanism of flow.

Additional aluminum couples of the same type have been vacuum hot pressed at 230 or 620 F using various pressures in order to permit further observations of the flow behavior for aluminum self-bonding.

Plans for Future Work

The study of the flow mechanism during self-bonding of aluminum will be continued. Metallographic evaluation of the specimens which have been hot pressed will be completed and additional couples will be prepared.

GROWTH OF UO_2 SINGLE CRYSTALS (AEC-DR)

J. D. Jackson, C. A. Alexander, and H. L. Goering

Objective

To prepare large single crystals of UO_2 .

Program Approach

This program is being supported by the AEC Division of Research. An apparatus to allow crystal growth from the melt by the Bridgman technique is being developed.

Progress Reported Previously

Several high-temperature sources have been investigated, and a tungsten-resistance furnace has been selected for use in the program. Effort has been expended in improving shielding, electrode, and resistance-element design so that the 2800 C temperature necessary to melt UO_2 may be achieved. Temperatures in excess of 2800 C have been reached for periods of 1 hr, and UO_2 has been successfully melted.

Work This Period

During November, Bridgman drop experiments were made at low drop rates at a temperature just above the melting point of UO_2 (about 2760 C), where a furnace life of several hours may be expected. In two experiments, drop rates of about 0.35 in. per hr were employed for 1/2 and 2 hr with a temperature of 2780 to 2790 C in the hottest zone of the furnace. Examination of the charges from these runs shows that fusion had occurred, but considerable porosity existed in the center portion of each charge. In both experiments, considerable loss of UO_2 occurred, about 9 and 16 per cent, respectively. It is interesting that considerable grain growth, crystals up to 1/16 in., occurred at the top of the crucible, which was probably below the melting point of UO_2 .

In these, as in previous experiments, porosity of the charge has been associated with evaporation losses of UO_2 , whether at high temperature and high drop rate or low temperature and low drop rate. In one experiment performed previously, a good ingot was formed and no loss of UO_2 occurred. It is believed that the porosity results indirectly from loss of UO_2 vapor from the system. Such loss would prevent establishment of equilibrium between vapor and condensed phases and allow significant transfer of material to the vapor phase in the course of the rather protracted experiments. If this is true, control of the UO_2 losses from the crucible may produce UO_2 with little or no porosity.

Plans for Future Work

Experiments are planned in which the loss of UO_2 from the crucible will be held to a minimum. An inert atmosphere (reduced pressure) can be used in the furnace to lower the evaporation rate of the charge. Although sufficiently high temperatures could not be reached in previous experiments, in which an atmosphere was used, recent improvements in furnace design and power supply may permit attaining temperatures near 2800 C.

Consideration is also being given to other methods of sealing the tungsten crucible.

RADIOISOTOPE AND RADIATION APPLICATIONS (AEC-DID)

D. N. Sunderman

These studies are in support of the program of the AEC Division of Isotopes Development, Isotopes Technology and Production and Radiation Branches.

Use of Intrinsic Radioactive Tracers for Process Control

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

Objective

To investigate the feasibility of using in-process isotope neutron sources to produce short-lived intrinsic radiotracers useful for stream analysis and process control.

Program Approach

It is planned to determine first the optimum practical conditions for in-process activation of stream constituents to form short-lived intrinsic radiotracers. This will be accomplished by activating a standard solution of manganous sulfate, along with indium and gold foils, while varying such parameters as activation volume, geometry, holding time, etc. Experiments will then be performed under the optimum conditions to determine general stream activation efficiencies for different process streams as a function of neutron-source strength. Finally, the concept of in-process neutron activation will be tested on several processes using simulated plant conditions.

Progress Reported Previously

The present phase of the intrinsic radiotracer program began July 1, 1961. After an initial survey of neutron sources and source technology, a 10-curie polonium-beryllium source was obtained from Mound Laboratory. A combination neutron-source storage shield and activation-cell holder has been constructed and is being used for optimization studies and neutron-flux measurements.

Work This Period

Emphasis this month has been on the optimization experiments using the 10-curie polonium-beryllium neutron source.

Manganous sulfate solutions are being irradiated in the activation cell to determine the effective neutron flux as a function of distance from the source. Table H-1 shows the results obtained in a solution containing 100 ppm manganese. The detection cell used for these measurements was calibrated against a manganese-54 standard source, by comparison of the photopeak areas of the manganese-54 0.84-Mev γ and the manganese-56 0.85-Mev γ using a single-channel analyzer.

Some indium-foil activations were also carried out this month. However, these measurements are not yet standardized. Relative counting data agree quite well with the manganous sulfate solution work.

Plans for Future Work

It is anticipated that the optimization phase of the program will be completed next month. Design work on equipment modification for the simulated process studies will begin.

TABLE H-1. EFFECTIVE NEUTRON FLUX IN MnSO_4 SOLUTION
CONTAINING 100 PPM MANGANESE

Average Distance From Center of Source, cm	1.9	2.5	3.1	3.9	5.1	6.5	8.3	10.9	13.8
Average Neutron Flux, 10^4 nv	8.1 ± 0.4	9.1 ± 0.2	9.1 ± 0.2	7.9 ± 0.2	7.1 ± 0.1	5.1 ± 0.1	3.5 ± 0.1	1.8 ± 0.2	0.9 ± 0.2

Graft-Polymerization Studies

F. A. Sliemers, M. Luttinger, E. Gulbaran*, W. B. Gager,
R. Lieberman, J. F. Kircher, and R. I. Leininger

Objective

To determine the influence of structural and compositional factors on the behavior of polymers subjected to gamma irradiation and to correlate the radiation-induced changes which occur with the ability of the polymer to form graft copolymers.

Program Approach

It is anticipated that a mechanism of radiation attack on the various polymer systems can be established. Polymers are irradiated in vacuo to various total doses. Free-radical site concentrations are determined both as a function of dose and as a function of time, using electron paramagnetic resonance (EPR) measurements. Vapor-phase chromatography and mass spectrometry are used to determine quantitatively the volatile fragments of the irradiation. Grafting techniques are employed to correlate free-radical site concentrations and grafted copolymer.

Progress Reported Previously

A number of polyalkylmethacrylates have been examined extensively. The data suggest that: (1) free-radical formation is accomplished by means of a whole or partial ester scission, (2) the size and configuration of the hydrocarbon tail of the ester group influence the efficiency of site formation, and (3) site formation is accompanied, in most cases, by a scission on the polymer backbone. More recently the work has been extended to other polymer systems. In addition, several preliminary grafting studies were initiated.

Work This Period

During November, experimental work included: (1) continuation of the grafting studies, (2) measurement of free-radical concentration as a function of dose for poly-methyl- α -cyanoacrylate, (3) measurement of the types and concentrations of volatile products formed during the irradiation of poly-2-hydroxypropyl methacrylate, (4) continuation of the study of the chemical measurement of free-radical sites, and (5) preparation of the methyl- α -bromoacrylate monomer.

*I. A. E. A. Fellow.

The results from a new series of graft-polymerization experiments are listed in Table H-2. In this study, PMMA was again used as base polymer and acrylonitrile was used as graft monomer. The water-insoluble acetone-soluble fraction of the copolymer was collected and analyzed for nitrogen. Approximately the same degree of grafting was noted in these experiments as when vinylpyrrolidone was used as graft monomer. Also, as was the case with the vinylpyrrolidone work, the solubility behavior of the copolymer resulted in its incomplete recovery. The acetone-insoluble material collected from one grafting contained 17.7 per cent nitrogen (acrylonitrile nitrogen content is 26.4 per cent).

TABLE H-2. RESULTS OF GRAFT-POLYMERIZATION STUDIES ON PMMA^(a) AND ACRYLONITRILE^(b)

Run	Weights of Base Polymer, g	Total Dose, rads x 10 ⁻⁷	Sites Per Monomer Unit x 10 ³ (c)	Nitrogen(d), per cent
G-50	5.03	0.02	0.4	0.87
G-52	5.00	0.10	1.2	0.49
G-53	5.01	0.34	2.5	0.58
G-54	5.01	0.50	3.3	0.51
G-55	5.02	0.96	5.5	0.23
G-57 ^(e)	5.01	2.90	8.0	1.49

(a) The weight-average molecular weight of the base polymer is 24,000.

(b) Grafting was carried out at 25 C for 1.25 hr using 25 cm³ of acrylonitrile.

(c) These values were determined by EPR measurements.

(d) These nitrogen values are for the water-insoluble acetone-soluble copolymers. Multiplication of these data by 2.64 gives the percentage of acrylonitrile in the copolymers.

(e) The water-insoluble acetone-insoluble material from this run contained 17.7 per cent nitrogen. This material is probably a mixture of homopolymer of acrylonitrile and copolymer high in acrylonitrile content.

Possibly, a more straightforward way in which the correlation between concentration of grafted polymer and total free-radical sites can be established is to measure weight changes following grafting. This method will be evaluated during December.

Site concentration has been determined as a function of dose for polymethyl- α -cyanoacrylate using EPR measurements (Table H-3). The results indicate that this material is less radiation susceptible than either PMMA or the α -chloro derivative.

In last month's report (BMI-1549) it was noted that poly-2-hydroxypropyl methacrylate had given a three-line EPR spectrum, whereas all of the unsubstituted esters had given five-four spectra. Using this observed spectrum as a basis, it appears that the mechanism of free-radical formation in this polymer differs from that obtained for the unsubstituted polymethacrylates. Since the five-four spectrum is generally

attributed to the backbone free radical, $\bullet\text{C} \begin{array}{c} \text{CH}_3 \\ | \\ \text{C} - \text{C} - \text{C} \sim \\ | \quad | \quad | \\ \text{COOR} \end{array}$, it was hypothesized that the

three-line spectrum might indicate that the stable free radicals are located at some other position, perhaps on the ester side chain. It was mentioned that the measurement

of M_N following irradiation as well as analyses for CO and CO₂ might establish if the predominant stable free radical is actually on the side chain.

The insolubility of the polymer, probably the result of cross linking initiated by dimethacrylate impurities, has discouraged attempts to measure M_N . However, the concentration of CO and CO₂ have been determined quantitatively by mass spectrometry following irradiation of the polymer to a dose of 3×10^7 rads. These data, presented in Table H-4, indicate that considerable ester-chain scission is taking place during irradiation. Consequently, the supposed differences in mechanism between the unsubstituted and hydroxy-substituted polymethacrylates, if present, are not easily resolvable. More work is planned in this area.

TABLE H-3. SITES-VERSUS-DOSE MEASUREMENTS FOR
POLYMETHYL- α -CYANOACRYLATE

Dose, rads	Sites Per Monomer Unit $\times 10^3$ (a)
5×10^4	N.D.(b)
1×10^5	N.D.(b)
2×10^5	0.055
5×10^5	0.26
1×10^6	0.39
4.8×10^6	1.07
9.6×10^6	1.23
1.9×10^7	1.92
3.4×10^7	1.98

(a) These values were determined by EPR measurements.

(b) Not detectable. The total number of sites in the samples fell below the sensitivity of the instrument.

TABLE H-4. COMPOUNDS FORMED DURING IRRADIATION OF
POLY-2-HYDROXYPROPYL METHACRYLATE TO A
DOSE OF 3×10^7 RADS(a)

Compound	Amount Found(b), microliters
Hydrogen	1715
Carbon monoxide	4050
Carbon dioxide	2540
Methane	100
Ethane	1320

(a) These compounds were identified quantitatively by mass spectrometry. Evidence for several organics was also found but analyses were not carried out.

(b) Based upon 5 g of polymer.

The work designed to investigate chemical methods for measuring free-radical concentrations has been continued during November. This study was undertaken primarily to establish if such a method might be applicable to the measurement of total site concentrations in the very low site-concentration range where sample size limits the sensitivity of the EPR instrument.

The hydroquinone-DPPH method, although initially showing some promise, has been proven ineffective. Recent results have shown that the scavenger system reacts with unirradiated polymer to the same extent as it does with the "active" polymer. Pursuing this unidentified reaction appears to be beyond the scope of the program. Consequently, other scavenger systems, including iodine and tetranitromethane, are now being investigated.

During November, in line with the proposed study of the effect on site formation of the group attached to the carbon alpha with respect to the ester group, methyl- α -bromacrylate was synthesized according to the procedure of Marvel, et al.* This monomer will be polymerized during December. Further, it will be used as the starting material for the preparation of the α -amino polymer.

Plans for Future Work

During December, work will be continued in the several areas described above.

* Marvel, C. S., Dec, J., Cooke, H. G., Jr., and Cowan, J. C., JACS 62, 3495 (1940).

MATERIALS DEVELOPMENT AND EVALUATION (HAPO)

F. R. Shober

Research and development work being conducted on materials to be used in conjunction with construction of the NPR (New Production Reactor) and operation of the PRTR (Plutonium Recycle Test Reactor) at Hanford Atomic Products Operation is reported in this section.

Mechanical Properties of Zirconium Alloys

J. A. VanEcho

Objective

To obtain creep and creep-rupture data, especially long-term creep, on annealed and cold-worked Zircaloy-2 sheet and to evaluate the effect of stress and/or temperature cycle on the creep properties of this alloy.

Program Approach

The tests are being conducted in standard Battelle creep machines and furnaces. In order to prevent excessive contamination of the specimens, all tests are made in vacuum of the order of 0.5 to 0.1μ of pressure. Test temperatures are 550, 650, and 750 F. Creep measurements are taken approximately daily by means of an optical system. In all respects, the tests meet or exceed the requirements for creep testing specified in ASTM Standard E-139-58T.

In the stress- and temperature-cycle tests, the stress is removed first and then the temperature is lowered to room temperature 1 day per week. The stress is re-applied after the specimen has been returned to test temperature. No temperature-cycle tests are being conducted at present, but these will be resumed in the near future.

Progress Reported Previously

Nearly all tests now in progress have been in progress for times ranging from about 5,000 to 35,000 hr. Therefore, data have been accumulating for some time and they have been reported in progress reports from time to time. The latest summary was presented in the September progress report (BMI-1546). Since all tests are long-term, there is little change in test results, other than accumulated time, from one month to the next.

Work This Period

All 20 tests reported in progress as of the end of October were continued through this period. These tests consist of the following:

- (1) Two creep tests on 15 per cent cold-worked material at 650 and 750 F
- (2) Nine creep tests on annealed sheet at 550, 650, and 750 F
- (3) Five temperature- and stress-cycled tests on annealed material at 550 and 650 F
- (4) Four creep tests on annealed PRTR pressure-tube material at 450 and 550 F.

Plans for Future Work

The 20 tests in progress are scheduled to continue. Creep data will be accumulated until a failure has occurred or it is believed that continuing a test will no longer

J-2

contribute useful data. As tests are discontinued, new tests will be started on specimens taken from the PRTR or the NPR pressure tubes. In addition, more tests are scheduled to be made on specimens which are stress and/or temperature cycled.

COATED-PARTICLE FUEL MATERIALS (AEC-DRD)

R. W. Dayton and J. H. Oxley

This program is being carried out for the AEC Division of Reactor Development to determine the characteristics of various coated-particle fuels and to develop optimum materials for specific reactor applications.

MATERIALS-DEVELOPMENT PROGRAMUncoated Fuel-Particle Studies

G. W. Cunningham, J. M. Fackelmann, and S. J. Paprocki

Objective

To characterize and evaluate fuel particles of UO_2 , UC, and UC_2 for use in coating studies and in subsequent coated-fuel irradiation programs.

Program Approach

Fuel materials are purchased as required from commercial vendors and are characterized with respect to enrichment, composition, particle size, porosity, shape factor (sphericity, roundness, and surface roughness), hardness, crushing strength, phase structure, grain size, fraction of particle agglomerates, surface-activity ratio, xenon-release ratio, and chemical impurities. As the relative importance of the various factors becomes known, emphasis will be increased or decreased on these factors.

Progress Reported Previously

Uranium carbide powder prepared by 3M was found to contain UC_2 as the major phase with varying amounts of UC, graphite, and inclusions present. Ninety per cent of the most recently received material was in the shape classification corresponding most closely to perfect spheres, a significant difference over previous lots.

The pore size distribution in natural ANL UO_2 was determined using a mercury-intrusion technique. The largest pore volumes were associated with 0.15 to 0.5- μ -diameter pores.

Postirradiation heat treatments of UC_2 Batch E2 resulted in equal amounts of Xe^{133} release at temperatures of 815, 1090, and 1320 C. Similar treatment to UO_2 Batch E3 resulted in 60 per cent of the Xe^{133} being released at 1320 C.

Work This Period

Surface-area measurements were completed on five lots of fully enriched UO_2 powder. These data were obtained by means of measurement of the amount of nitrogen adsorbed by a particular quantity of powder. The results have been summarized in Table L-1 and show that surface areas range from 0.02 to 0.08 m^2 per g. If these areas are converted to the diameter of fully dense spheres having the given surface areas, these diameters range from 7 to 28 μ , compared with screen sizes of 105 to 240 μ . Thus, by this measurement, Batch UO_2 -E5 is most porous and UO_2 -E8 most dense of the five lots evaluated.

TABLE L-1. SUMMARY OF SURFACE-AREA MEASUREMENTS ON ENRICHED UO_2 POWDER

Powder	Average Screened Particle Diameter, μ	Surface Area, m^2 per g	Diameter Corresponding to Surface Area ^(a) , μ
UO_2 -E4	127	0.07	8
UO_2 -E5	127	0.08	7
UO_2 -E6	127	0.03	18
UO_2 -E7	178	0.04	14
UO_2 -E8	127	0.02	28

(a) Assuming fully dense spheres.

Plans for Future Work

Quantitative particle-shape studies are being made on the five lots of UO_2 . In addition, mercury-intrusion measurements of surface-connected porosity will be made.

Alumina-Coating Studies

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

Objective

To establish conditions and equipment design for coating fuel particles with Al_2O_3 of optimum quality.

Program Approach

Alumina is being deposited on UO_2 particles by hydrolysis of Al_2Cl_6 vapor in a fluidized bed. The coatings are being evaluated by microscopic examination, measurement of physical properties, alpha counting, and measurement of fission-gas release on postirradiation heating. Promising materials will be evaluated by in-pile tests at elevated temperatures.

Progress Reported Previously

Coatings of near theoretical density deposited at a temperature of 1000 C consist of small crystallites of alpha alumina dispersed in an amorphous matrix. Coatings deposited at temperatures from 500 to 700 C are porous, of lower density, and completely amorphous, but become crystalline on heat treating at temperatures about 1000 C and above. Heat treatment of the dense coatings deposited at a temperature of 1000 C results in increased average crystalline size.

All coatings prepared at 1400 C even though relatively free of impurities exhibited a coating integrity inferior to that of the better 1000 C coatings.

Surface protrusions of 1000 C Al_2O_3 coatings can be minimized by interrupting the growth pattern with a thin layer of Al_2O_3 deposited at a lower temperature. However, the coating integrity is adversely affected.

The following specific types of coated fuel particles have been prepared for in-pile evaluation:

Lot 715A, dense Al_2O_3 on dense UO_2 -E2; coating temperature: 1000 C
(Xe^{133} release: 3.2 ppm at 1300 C)

Lot 721C, dense Al_2O_3 over porous Al_2O_3 on dense UO_2 -E3; coating temperatures: 100 C, 700 C (Xe^{133} release: 1.3 ppm at 1300 C)
 Lot 725D, dense Al_2O_3 on dense UO_2 -E3; coating temperature: 1225 C (Xe^{133} release: 0.33 ppm at 1300 C).

Work This Period

The major effort during this period has been the preparation of coated-particle specimens with composite coatings tailored to resist fission-recoil damage, permit substrate swelling, and limit crack propagation. These materials have been prepared with enriched UO_2 so as to be immediately available for in-pile evaluation. The following specimens have been prepared or are in process:

Lot	UO_2	Coating Type	Estimated Date of Completion ^(a)
731D	Dense	5 μ of dense Al_2O_3 + 15 μ of porous Al_2O_3 + 5 μ of carbon + 25 μ of dense Al_2O_3	Completed
732D	Porous	45 μ of dense Al_2O_3	Completed
733B	Dense	45 μ of dense Al_2O_3	December 1
735	Dense	15 μ of dense Al_2O_3 + 30 μ of carbon	December 5
736	Dense	15 μ of carbon + 30 μ of Al_2O_3	December 8
737	Dense	15 μ of Nb + 30 μ of Al_2O_3	December 8

(a) Except for postirradiation fission-gas release evaluation.

The coating of Lot 731D was adversely affected by heat treatment at 1250 C for 48 hr in hydrogen, as indicated by the 660 ppm of UO_2 removed in a subsequent acid-leach evaluation. An effort is being made to upgrade this material by chemically culling with alternate heat treatment, oxidation, and acid leaching. Lot 732D is being evaluated by postirradiation fission-gas release.

An effort to upgrade by both hand and chemical culling of the 1400 C coatings (Lots 727C, 728C, and 729C) described in the last monthly report (BMI-1549) was relatively successful. It was only necessary with Lot 729C to remove by hand culling the particles with irregular protrusions on their surface to obtain a product which satisfactorily resisted heat treatment at 1400 C for 48 hr in hydrogen, oxidation at 1100 C for 24 hr in oxygen, and acid leaching. A sample of Lot 729C will be prepared for post-irradiation fission-gas-release evaluation.

A 45- μ dense Al_2O_3 coating was deposited on a dense enriched UO_2 substrate at 1300 C (Lot 734C). The evaluation, except for the postirradiation fission-gas-release determination, should be completed early in December.

The effort to improve the stability and general properties of the Al_2O_3 coatings by heat treatment was continued. The heat treating of Lot 715A and Lot 725D as described in BMI-1549 resulted in a sample (Lot 715AHT) with a postirradiation fission-gas release

of 0.17 ppm at 1300 C. Lot 725AHT showed an excessive alpha count. Attempts to upgrade this material by oxidation and acid-leach treatment were not successful. The two samples (Lot 901) annealed at 1800 C after an overcoat of PyC as described in BMI-1549 were found to be 100 per cent cracked after the carbon was burned off. Evidently the thermal stresses which developed at 1800 C were sufficient to crack the $53\text{-}\mu\text{ Al}_2\text{O}_3$. The $120\text{-}\mu\text{ PyC}$ outer coating did not appear to offer effective support.

Carbon-Coating Studies

A. C. Secrest, V. M. Secrest, M. F. Browning,
and J. M. Blocher, Jr.

Objective

To establish conditions for coating fuel particles with pyrolytic carbon of optimum quality.

Program Approach

Pyrolytic-carbon coatings are being applied by the decomposition of hydrocarbons in a fluidized bed of particles at temperatures of 1000 to 2000 C. The coatings are being evaluated by means of microscopic examination, measurement of physical properties, direct alpha counting, and by measurement of fission-gas release on postirradiation heating. Promising materials will be evaluated by in-pile tests at elevated temperatures.

Progress Reported Previously

Pyrolytic carbon coatings have been applied to fuel particles at temperatures of 950 to 1800 C.

The following lots of carbon-coated enriched fuel particles have been submitted for in-pile evaluation:

Lot 918A, columnar coating on UC-E1; coating temperature: 1400 C
(Xe¹³³ release: 2.7 ppm at 1300 C)

Lot 919A, laminar coating on UC₂-E1; coating temperature: 1400 C
(Xe¹³³ release: <0.5 ppm at 1300 C)

Lot 920A, laminar coating on UO₂-E3; coating temperature: 1400 C
(Xe¹³³ release: 0.9 ppm at 1300 C)

Lot 921, laminar-columnar mixture PyC coating on dense Al₂O₃-coated UO₂-E2; coating temperature: 1400 C (Xe¹³³ release: 0.5 ppm at 1300 C)

Lot 922, laminar-columnar mixture PyC coating on dense;porous Al₂O₃-coated UO₂-E3; coating temperature: 1400 C (Xe¹³³ release: 3.1 ppm at 1300 C)

Lot 1010A, faintly laminar coating on enriched UO₂-E3; coating temperature: 1600 C (Xe¹³³ release: 1.3 ppm at 1300 C).

The density of PyC coatings prepared by the fluidized-bed technique was found to decrease from 1.91 g per cm³ at 950 C to a minimum value of approximately 1.50 g per cm³ at 1500 C, and then increase to a density of 1.94 g per cm³ at a temperature of 1800 C. The density of PyC coatings increased slightly with deposition rate or hydrocarbon partial pressure over the limited range of conditions studied.

Work This Period

Work was limited to the preparation of multilayer coatings of alumina and carbon for in-pile evaluation. These composite coatings are to be evaluated for the purpose of limiting crack propagation, permitting stress-free swelling of the substrate, and minimizing fission-recoil damage.

Plans for Future Work

Two enriched UO_2 samples will be coated with high-density pyrolytic carbon for in-pile evaluation. One coating will be deposited at 950 C, the other at 1800 C.

In an effort to form a fuel particle with voidage between the substrate and the coating, work will be initiated on the deposition of carbon on uranium shot to be later reacted in place.

Continued assistance will be given in preparing composite coatings which include pyrolytic carbon.

Chemical-Fabrication Studies

A. C. Secrest, M. F. Browning,
V. M. Secrest, and J. M. Blocher, Jr.

Objective

To establish conditions for chemically sintering or infiltrating a packed bed of coated fuel particles with the purpose of forming a strong dense matrix in situ.

Program Approach

Fuel particles will be coated as required to give the desired fuel loading when packed in a static bed having dimensions of the finished fuel specimen. The matrix will be formed by chemical vapor deposition from appropriate reactants passed through the interstitial structure. A carbon matrix could be formed by thermal decomposition of a hydrocarbon, a metal oxide matrix by hydrolysis of an appropriate halide, and a metal matrix by hydrogen reduction of the corresponding halide or by decomposition or reduction of other vapor compounds.

Progress Reported Previously

Packed beds of PyC:Al₂O₃-coated UO₂ particles were infiltrated with pyrolytic carbon by thermal decomposition of methane at temperatures of 1150 to 1175 C. A residual porosity of less than 10 per cent was obtained at a loading of 1.4 volume per cent UO₂.

A fueled pellet, 0.275 in. in diameter by 0.165 in. high, prepared as described above, was subjected to crushing-strength tests. The pellet crushed at a load of 9300 psi, as compared with crushing strengths of 6900, 6500, and 6300 psi for unfueled pellets machined from ATJ graphite in three perpendicular orientations with respect to the block.

In fracture, most of the chemically fabricated pellet was reduced to fine powder. However, a few fragments survived. Faces of these fragments showed examples of all types of cleavage, with that between the Al₂O₃ and the initial pyrolytic-carbon coating predominating. The bond between the outer pyrolytic-carbon coating applied in the fluidized bed and the matrix applied by infiltration of a static bed was particularly good, as indicated by the over-all strength and the infrequent bond cleavage. Acid leaching of the fractured pellet revealed that about 0.2 per cent of the contained UO₂ was exposed in the fracture.

Work This Period

While preparations were being made to demonstrate the feasibility of producing prototype fuel elements by this technique, pellets of the size used for irradiation testing were prepared. Because of the small quantity of fuel involved, it was decided to prepare these specimens with alumina-coated enriched fuel from an earlier lot (715A). In this way, evaluation of the small pellets can be carried out during the feasibility tests, and irradiation testing can be initiated immediately upon demonstration of the feasibility of prototype element preparation.

Fueled pellets approximately 0.276 in. in diameter and 0.17 in. long with a UO₂ loading of approximately 1.72 volume per cent were prepared by the procedure used in the second run described in BMI-1518. In the present work, the gas flow was maintained until the pressure drop across the pellet assembly reached approximately 600 mm of mercury. The terminal pressure was increased in an effort to better infiltrate the voids with matrix material. Lot 715A (Al₂O₃-coated enriched spherical UO₂) was used as the basic fuel element and was overcoated with PyC to a total diameter of approximately 375 μ to give the desired loading. These pellets are undergoing out-of-pile evaluation as possible candidates for in-pile study.

Plans for Future Work

Out-of-pile evaluation of the chemically sintered specimens described above will be continued along with demonstrations of the feasibility of preparing prototype elements (GA or ORNL) by present techniques.

Graphite-Matrix Studies

M. C. Brockway, D. J. Bowers, and W. H. Duckworth

Objective

To prepare and characterize graphite-matrix bodies containing coated fuel particles.

Program Approach

Bodies which are representative of current graphite technology are being studied. Specimens are to be prepared with the minimum of development studies. The

development work is to be carried out only to the extent needed to achieve reasonable physical properties, reproducibility, and fuel dispersion. Later, improvements in these qualities will be sought.

Progress Reported Previously

Processing conditions were selected for unfueled specimens made from AGOT graphite flour and medium-hard coal-tar pitch. Those processing conditions which yielded reasonably dense (1.65 g per cm^3) and sound unfueled specimens were then used as a basis to prepare specimens in which Al_2O_3 -clad UO_2 particles were dispersed. Randomness of fuel dispersion was evaluated by radiographs and metallography. Uniformity of fuel loading was nondestructively determined from gamma emission. Cladding damage during processing was evaluated by neutron-activation tests.

A satisfactory batch method was developed for dispersing Al_2O_3 -clad UO_2 in graphite by conventional processing, except for the use of solvent mixing. Use of this method to disperse dense Al_2O_3 -clad UO_2 (Lot 715A) in a carbon matrix and porous-dense Al_2O_3 -clad enriched UO_2 (Lot 721C) in both carbon and carbon-bonded graphite matrices produced specimens having low neutron-activation fission-gas release (see BMI-1534). Three specimen sets were selected for static capsule irradiation. Summary characterization data for these sets were given last month in BMI-1549.

Specimen or small-quantity dispersal methods have been developed and used in combination with pressure-baking procedures similar to those outlined in GAMD-2221. Six types of clad enriched fuel (Lots 4E, 918A, 919A, 920A, 725D, and 1010A) have been dispersed by these methods. Fueled-matrix specimens containing these fuels had neutron-activated fission-gas release at 1150°C of the same order as, or lower than, those of the respective undispersed fuels at temperatures of 1300°C .

Work is in progress on methods to achieve equidistant particle spacing of fuel dispersions in graphite matrices. The approach being explored is to individually pelletize clad fuel particles with matrix and then form these pellets into fueled-matrix specimens.

Work This Period

Pelletizing techniques developed last month for coating $400\text{-}\mu$ -diameter, or larger, PyC-clad fuel were modified to achieve pelletizing of 210 to $250\text{-}\mu$ -diameter Al_2O_3 -clad depleted UO_2 . (This is the size range of most clad particles of interest.) The pellets were then pressure baked to yield a very close approach to equidistant fuel dispersion. Except for "chemically sintered" matrix specimens, this is the first true approach to an ideal fuel dispersion in graphite.

While the objective of this exploratory pelletizing work is to prepare ideal, equidistant fuel dispersion, it is of practical importance that it be accomplished with matrix material which will produce good graphite properties. Therefore, limited property data have been obtained on pressure-baked specimens prepared from matrix materials used successfully in the pelletizing experiments. It is emphasized that these data are limited and are not presented as average or typical, but they do indicate good matrix properties. Table L-2 presents data for both fueled (pelletized followed by pressure baking) and unfueled pressure-baked specimens.

Heat-treated Lot 715A fuel (dense Al_2O_3 -clad enriched UO_2) is being incorporated into graphite-matrix specimens for neutron activation. The usual random-dispersion pressure-baking procedures are being used.

TABLE L-2. PRELIMINARY PROPERTY DATA FOR PRESSURE-BAKED GRAPHITE-MATRIX SPECIMENS

	Specimen Type	
	Fueled	Unfueled
Fuel Loading,		
UO ₂ , w/o	11.4	--
Al ₂ O ₃ -Clad UO ₂ , volume per cent	8.8	--
Density, g per cm ³	2.07	1.75-1.78
Tensile Strength, psi	1,670	2,300
Crushing Strength, psi	No data	10,000
Electrical Resistivity, 10 ⁻³ ohm-cm	--	4.9

Plans for Future Work

Neutron-activation tests will be performed on the heat-treated Lot 715A fuel dispersed in graphite. New fuel types will be dispersed by the random-dispersion procedures.

The use of pelletizing to prepare "ideal" graphite-matrix dispersions will be continued. In particular, attention will be given to determining to what degree these dispersions will permit use of higher forming pressures (or cold forming) without cladding damage, and what improvement in matrix density and strength can be achieved by the increased forming pressures.

Explosive Compaction Studies

S. J. Paprocki, R. J. Carlson, and P. H. Bonnell

Objective

To determine the applicability of explosive compaction techniques for consolidating powder dispersions of pyrolytic-carbon-coated particles in a graphite matrix.

Program Approach

Power dispersions of pyrolytic-carbon-coated particles in graphite will be prepared and explosively compacted by established techniques to relatively high density. Evaluation of the applicability of these techniques will be on the basis of the as-compacted integrity of the particle coatings, which will be determined by microstructural and acid-leaching studies of the consolidated material.

Progress Reported Previously

Specimens of pyrolytic-carbon coatings with both columnar and laminar structures were blended with natural graphite in the ratio of 20 volume per cent coated particles to 80 volume per cent graphite. Each structure type was blended separately and the dispersions were loaded by tamp packing into mild steel tubes for compacting. Leach tests of the blended powders showed no damage caused during blending operations.

Two specimens were explosively compacted in a large-diameter housing. These did not have a cohesive density. One specimen was explosively compacted in a small-diameter mild steel housing. This housing ruptured but the specimen appeared to have attained a cohesive density. However, when the cladding was removed the compacted dispersion fell apart.

Work This Period

A small-diameter stainless steel housing was obtained and four specimens were explosively compacted. Two of these contained natural graphite and two contained reactor-grade graphite in the same ratio of coated particle to reactor grade graphite. The cladding on one specimen containing natural graphite ruptured and the other specimen was not compacted to a cohesive density. Both specimens containing reactor-grade graphite visually appeared to have compacted to a greater density than those containing natural graphite.

Plans for Future Work

The cladding of both specimens containing reactor-grade graphite will be removed by pickling. Additional specimens containing reactor-grade graphite will be explosively compacted. All specimens compacted to a cohesive density will be subjected to micro-structural studies. Specimens containing each of the two types of pyrolytic-carbon-coated particles will be subjected to acid-leaching studies in hot nitric acid to determine if the coatings remained intact during explosive compaction.

Pressure-Bonding Studies

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

This program has been recessed temporarily. Work will be initiated when suitable materials have been prepared.

FUEL-IRRADIATION PROGRAM

W. A. Goldthwaite, D. N. Sunderman, and R. F. Dickerson

Sweep Capsule SP-5

G. E. Raines, R. H. Barnes, and R. J. Burian

Work on this program was deferred during November.

Static Capsules CFP-1A, -1B, -1C, and -1D

R. J. Burian, N. E. Miller, W. J. Zielenbach, and R. H. Barnes

Objective

To determine the effects of reactor radiation on alumina-coated uranium dioxide particles as a function of irradiation temperature, flux, matrix support, and particle porosity.

Program Approach

The fission-product-retention properties of UO_2 fuel particles coated with Al_2O_3 may be improved by using various combinations of high- and low-density UO_2 and Al_2O_3 . It is planned to irradiate three types of fuel particles, as described below. The combinations of coated fuel particles contemplated for study in this set of experiments are:

- (1) UO_2 particles coated with dense Al_2O_3 (Type 715A) dispersed in a graphite matrix and unsupported
- (2) UO_2 particles coated first with low-density Al_2O_3 and then with dense Al_2O_3 (Lot 721C) dispersed in both graphite and carbon matrices and also unsupported
- (3) UO_2 particles coated first with dense Al_2O_3 and then pyrolytic carbon (Lot 921) and irradiated unsupported.

Four static capsule experiments are planned for irradiation in core positions in the BRR. Each type of particle will be irradiated in each capsule experiment. The specimens in the four capsules will be irradiated at temperatures of about 150 and 980 C to burnups of approximately 2 and 6 a/o of the uranium. Evaluation of the specimen performance will be achieved by pre- and postirradiation examinations, which will include measurements of physical dimensions and weights, visual examinations, metallographic examinations, and strength tests. In addition, the quantity of fission gases released from each specimen will be determined. Dosimeter wires within the capsule will be analyzed to obtain flux data, and selected specimens will be analyzed for uranium burnup.

Progress Reported Previously

The capsule design has been completed and is described in two previous progress reports, BMI-1489 and BMI-1496. The six types of specimens are fully described in BMI-1546. The specimens were fitted into individual sealed cans for irradiation. One specimen of each type was loaded into each of four capsules designated as Capsules CFP-1A, CFP-1B, CFP-1C, and CFP-1D. The capsules were positioned in the BRR during the shutdown during the week of August 7, 1961. The intended burnup for specimens in Capsules CFP-1A and CFP-1B was 6 a/o of the uranium at about 980 C and 60 C,

respectively. Capsules CFP-1C and CFP-1D were discharged from the BRR on October 2, 1961, after irradiation at 980 C and 60 C, respectively, to a burnup of about 3 a/o of the uranium. The specimens were recovered from the capsule and the gas within the specimen cans was sampled. Analyses of the gas and the dosimeter wires recovered from the two capsules were initiated.

Work This Period

Analyses of the dosimeter wires and gas samples from specimens irradiated in Capsules CFP-1C and CFP-1D were completed. The results of dosimeter-wire analyses appear inconsistent with normal data obtained by this method. The analytical procedures are being reviewed to discover the source of this apparent inconsistency. The results of the gas analyses indicate that alumina-coated particles supported in a matrix tend to retain fission gas better than unsupported particles and also that the gas-retention properties of the particles are probably better when irradiated at elevated temperatures. Alumina-coated particles with a porous layer in the alumina coating tend to retain fission gas better than particles with completely dense alumina coatings at low temperatures.

Capsules CFP-1A and CFP-1B were discharged from the BRR on November 13, 1961, after an estimated specimen fuel burnup of 6 a/o. The specimens in these capsules were irradiated at about 980 C and 60 C, respectively. The specimens were recovered from the capsules, and sampling of the gases in the specimen cans was initiated. The dosimeter wires in the capsules were recovered and prepared for analysis.

Plans for Future Work

The analysis of the dosimeter wires recovered from Capsules CFP-1C and CFP-1D will be rechecked. The specimens will be recovered from the capsules and examined. Additional studies planned for these specimens include uranium leach analyses to determine the degree of coating failure, metallographic examination, and heat treatment to obtain data concerning gas diffusion through the coatings.

The gases recovered from specimen cans irradiated in Capsule CFP-1A and CFP-1B will be analyzed for fission-gas content. The dosimeter wires recovered from these capsules will also be analyzed. The specimens will be recovered and subjected to uranium leach analyses, metallographic examinations, and fission-gas diffusion examinations during postirradiation heat treatment.

Sweep Capsules CFP-S-2 and -2R

N. E. Miller, G. E. Raines, T. S. Elleman, and R. J. Burian

Objective

To obtain preliminary fission-gas-release information on pyrolytic-carbon-coated uranium dicarbide particles supported in a graphite matrix.

Program Approach

The fuel specimen is contained in a sweep-gas capsule, and the fission gas released by the specimen is continually removed from the capsule during the irradiation by a helium sweep gas. Periodic analysis of the sweep gas is made to determine the fission-gas-release characteristics of the fuel as a function of burnup.

Progress Reported Previously

In BMI-1524, it was reported that in June, 1961, after 36 hr of in-pile operation, the CFP-S-2 experiment was terminated when a gas leak occurred between the argon-filled capsule annulus and the sweep-gas system. As reported in BMI-1541, a

replacement capsule, CFP-S-2R, was operated for 2 weeks during August until a restriction in the gas passages occurred which prevented the flow of sweep gas. The irradiation of the capsule was continued under static conditions, with the fuel-specimen temperature maintained at 1070 C, until an estimated 25,000-MWD/Tonne burnup was accumulated early in October when the test was terminated. The fission-gas-release data were given in BMI-1546. The findings of a preliminary hot-cell examination of these two capsules were reported last month in BMI-1549.

Work This Period

In the postirradiation examination of the CFP-S-2R capsule it was desired to examine both the fuel specimen, for particle damage, and the sweep-gas passages within the fuel-specimen can, for restrictions to gas flow. To do this, the specimen can was filled by a vacuum technique with epon resin and sectioned axially on a plane through the gas passages and specimen chamber. The resin did not provide sufficient support for the fuel specimen and it was completely abraded away by rubbing on the face of the cutting wheel. The walls of the inlet gas passage were coated with thick layers of a light-colored material, tentatively identified as Nb_2O_5 , in the peak temperature zone adjacent to the specimen chamber. An accumulation of loose pieces of this material was found which appeared to completely block the inlet passage where it turned to enter the specimen chamber. These deposits are believed to be the major cause of flow restrictions which caused the termination of dynamic operation of this experiment. The walls of the specimen chamber and outlet passage were clean and free of deposits, indicating the gas-metal reaction was completed in the inlet passage.

No further examination will be made of the CFP-S-2 capsule, beyond that reported last month, because of its short total irradiation.

Plans for Future Work

Further work on this experiment will be limited. Attempts will be made to identify the material deposited in the gas passage, and the microstructure of the specimen can will be compared with that of a similar unirradiated heat-treated niobium can.

Sweep Capsule CFP-S-3

N. E. Miller, G. E. Raines, T. S. Elleman,
and R. J. Burian

This work is currently recessed.

Static Capsule CFP-4

R. J. Burian, N. E. Miller, W. J. Zielenbach,
and R. H. Barnes

Objective

To evaluate the performance of unsupported fuel particles of uranium compounds coated with various ceramic materials during low-temperature irradiation.

Program Approach

Fuel particles of UO_2 and UC_2 coated with Al_2O_3 and pyrolytic carbon of various types and porosities will be irradiated without support from a matrix material in the BRR. The coatings will be characterized and studied before and after irradiation to evaluate the stability of the coating.

Progress Reported Previously

Six types of coated fuel particles were selected for irradiation. The particles were characterized before irradiation, and random samples of the particles, each containing about 0.06 to 0.07 g of uranium, were prepared for irradiation. Each group of particles was sealed in a helium-filled annulus in a Hastelloy X can. The six cans were placed end to end in a NaK-filled aluminum capsule. The specimen cans were centered by three longitudinal dosimeter wires spaced 120 deg apart.

The capsule was discharged from the BRR after irradiation for five reactor cycles. The temperature of the specimens during irradiation was between 65 and 120 C, and the specimens experienced a fission burnup of 2.9 to 3.4 a/o of the uranium-235. After irradiation the Hastelloy X specimen cans appeared clean and in good condition. The gases in the specimen cans were sampled and analyzed for fission-gas content. The results of these analyses, presented last month in BMI-1549, indicated that for these irradiation conditions, unsupported alumina-coated particles with a porous layer in the alumina coating retained fission gas better than unsupported particles with an alumina coating which was dense throughout. Also, pyrolytic carbon coatings on unsupported particles appear to have about the same gas retention potential as the alumina coatings under these irradiation conditions.

Work This Period

The particles were recovered from the specimen cans and examined at magnifications of 1 to 30 diameters. They appeared generally clean and unaffected, although some fragmented coatings were observed. The degree of coating failure could not be determined by these observations. Uranium-leaching techniques are planned for coating-failure evaluations.

The particles have been subdivided into smaller groups and prepared for further examinations.

Plans for Future Work

A group of each type of particle irradiated in Capsule CFP-4 will be leached and the leach solution analyzed to determine the degree of coating failure. Some of these particles will then be heat treated to obtain gas diffusion data on the coating materials. Selected types of particles will be examined metallographically.

Sweep Capsule CFP-S-5

N. E. Miller, G. E. Raines, T. S. Elleman,
and R. J. Burian

Objective

To obtain high-temperature high-burnup fission-gas-release information on alumina-coated uranium dioxide particles supported in a graphite matrix.

Program Approach

A small cylindrical specimen similar to the sphere used in the SP-5 experiment is to be contained in the sweep section of a sweep capsule and irradiated in the core of the BRR. Periodic analysis of the helium sweep gas, which is continuously passed through the capsule, will be used to determine the fission-gas-release characteristics of the fuel specimens. In addition, coated-fuel-particle specimens will be irradiated within static compartments in the capsule to obtain preliminary irradiation-damage information on other materials by postirradiation examinations.

Progress Reported Previously

The description of the sweep specimen and the four static specimens was given in BMI-1541. This sweep-gas experiment was begun in September, 1961. In October, a leak developed between the argon-filled capsule annulus and the sweep-gas system, causing the termination of dynamic operation. The irradiation was continued under static conditions. The fission-gas-release data were reported last month in BMI-1549.

Work This Period

The static irradiation of the capsule has continued. Variations in neutron flux have caused a greater than normal variation in the specimen temperatures, ranging from approximately 1050 to 1250 C.

Plans for Future Work

The static irradiation of the capsule will be continued with a tentative termination date of January 8, 1961.

Sweep Capsule CFP-S-6

N. E. Miller, G. E. Raines, T. S. Elleman,
and R. J. Burian

Objective

To evaluate the fission-gas-release characteristics at elevated temperature of alumina-coated uranium dioxide particles in a graphite matrix.

Program Approach

The fuel specimen is incorporated in a sweep capsule, and the fission gas released by the specimen is continually removed from the capsule during the irradiation by a helium sweep gas. Periodic analysis of the sweep gas is made to determine the fission-gas-release characteristics of the fuel as a function of burnup. During the irradiation the fuel specimen is maintained at a nominal temperature of 1050 C.

Progress Reported Previously

The description of the four static specimens which will be irradiated in this capsule, and the capsule design modifications which have been made to overcome those problems encountered in previous sweep capsules, were presented last month in BMI-1549.

Work This Period

The assembly of the irradiation capsule, which is presently near completion, has been held up by a delay in the procurement of some materials required by the change in the capsule design. Because of this delay, it has been decided to defer the incorporation of the laminar-pyrolytic carbon-coated uranium dioxide particles (Specimen G-20) as a sweep specimen in favor of obtaining fission-gas release information on the alumina-coated uranium dioxide particle specimen (G-21). No change has been made in the static specimens.

Plans for Future Work

This experiment is expected to begin about mid-December.

Intermediate-Temperature Design Studies

N. E. Miller and W. J. Zielenbach

Objective

To develop an irradiation-capsule design for operation in the temperature range of 1100 to 1700 C.

Program Approach

An attempt will be made to extend the maximum temperature of the present capsule design upward by creating a greater barrier to the loss of fission heat, by reducing the requirements for auxiliary heating, and by replacement of the present stainless steel construction materials with materials which are serviceable at higher temperature, meeting both strength and anticorrosion requirements. This program is primarily directed toward sweep capsules for intermediate temperature range, but will also be applicable to static capsules.

Progress Reported Previously

An electrical heater has been fabricated which is designed for use in an irradiation capsule to provide fuel-specimen temperatures up to 1650 C. The heater is essentially a tantalum resistor wire and a high-purity alumina insulation on a 3/4-in. -diameter tantalum tube. The heater shell is contained in a 1-3/8-in. -diameter argon-filled stainless steel shell and is being evaluated by operating the heater at temperatures up to 1500 C with the outer shell immersed in water.

Work This Period

The laboratory operation of the mock-up capsule to determine the potential life of the tantalum-resistor heater at 1450 C is still in progress. In addition preliminary tests of carbon-fiber resistor elements have begun with operating temperatures up to 1800 C. Capsule designs for carbon-resistor heaters are in progress.

Plans for Future Work

The work will be continued on the present design concept, and, in addition, new heater designs will be investigated.

Fission-Gas Analytical Studies

R. L. Ritzman and T. S. Elleman

Work on this program was recessed during November.

SUPPORTING RESEARCH PROGRAM

Physics and Engineering Guidance

D. E. Wetzel, R. O. Wooton, H. M. Epstein, and J. W. Chastain

Objective

To investigate the importance of effects of coated fuel particles on the physics and engineering characteristics of reactor systems which may utilize these coated particles.

Program Approach

Reactor systems previously designed by other laboratories are selected and examined to determine the effects of substituting coated fuel particles for the given fuel. Parameters investigated are coating material, coating thickness, particle size, fuel and moderator loading, and reactor size. The calculation techniques are based on the simplest analysis available consistent with the program objectives. Other miscellaneous problems for which solutions will assist the total program are also considered.

Progress Reported Previously

Results of an investigation to examine the nuclear-criticality problems involved in designing a chemical reactor of cylindrical geometry which could be used to coat 125- μ -diameter fuel particles with Al_2O_3 or carbon (and BeO) were presented in an earlier progress report, BMI-1524. A later expansion of this investigation defined the dependence of maximum "safe" reactor diameters upon particle coating thickness. A preliminary investigation was begun to determine the range of fuel loadings of interest in the development of reactor fuel elements containing coated particles. The feasibility of employing coated particles in current superheater reactor design was studied, the initial results suggest that several advantages, including economics and safety, might be realized by a direct substitution of low-enrichment fuel particles, coated with a low-absorption-cross-section steam-corrosion-resistant material (MgO , ZrO_2 , or Al_2O_3), into the high-enrichment cell configuration currently being employed in the Pathfinder superheater concept.

Work This Period

The nuclear analysis of the Pathfinder superheater reactor employing 17 per cent enriched fuel coated with 20 μ of magnesia (MgO) has been completed. Whereas the system has several advantages (economics and safety) from the nuclear standpoint, there is doubt about the structural integrity of an annular ring of particles coated with 20 μ of material due to the extreme conditions imposed upon a superheater fuel element.

Plans for Future Work

Further work will be performed as necessary and desirable.

Radiation-Damage Studies

T. G. Knorr and C. M. Schwartz

Objective

To study fission-recoil damage in nonfissionable ceramic materials.

Program Approach

Decoration and replica procedures are being investigated to allow observation of the fission tracks with optical and electron microscopical techniques. Alpha Al_2O_3 and other ceramics will ultimately be the materials studied, but the alkali halides will be investigated initially because of ease of handling and the information already available.

Progress Reported Previously

Direct carbon-replica techniques with the electron microscope have revealed fission damage on the surfaces of NaCl, KBr, and LiF. The damage region is usually the defected cross-sectioned area resulting from the entry of fission products into the crystals almost normal to the surface. The size ranges from a few hundred angstroms to about $2\ \mu$. Longer, thinner areas have also been observed which are due to the fission recoils which enter the surface at lower angles. Pit damage is also seen but is much less frequent. Decoration by precipitated silver in AgCl has revealed dislocations, grain boundaries, and fission damage. In the NaCl crystals, a considerable change in the registered damage has been observed which must apparently be related to a humidity or temperature effect.

Work This Period

Further experimental studies have been deferred temporarily, while more detailed analytical studies are being made to obtain a better understanding of the mechanisms by which the observed damage patterns may be produced. The types of features which would be expected from melting, strain, surface migration which may be enhanced by moisture, the effects of dislocations, and the effects of interstitials and vacancies are being examined in an attempt to decide which of these is the likely cause of damage. No conclusions from this study have been reached.

Plans for Future Work

The analysis of the data will be concluded, and it is hoped that the analysis will indicate critical experiments which should be performed to further elucidate the fission-recoil damage mechanism in nonmetallic materials.

Fission-Gas-Diffusion Studies

R. H. Barnes, D. L. Morrison, T. S. Elleman,
and D. N. Sunderman

Objective

To determine diffusion coefficients in the absence of radiation for rare-gas fission products in alumina, pyrolytic carbon, and other potential coating materials for nuclear fuel particles.

Program Approach

Small particles of the coating material of interest are packed in uranium dioxide powder and irradiated at low temperatures in the Battelle Research Reactor. These particles, which contain fission products recoiled into them from the surrounding uranium dioxide powder during irradiation, are separated from the powder and heated at constant temperature in a flowing helium stream. The fission gases released from the particles during heating are collected from the helium on activated charcoal at liquid-nitrogen temperature and radioassayed. The results of the measurements are then used to determine fission-gas diffusion coefficients.

Progress Reported Previously

Diffusion coefficients for xenon-133 in Linde Company 1000- μ -diameter alpha-alumina spheres at temperatures ranging from 800 to 1500 C were discussed in previous progress reports, BMI-1504 and BMI-1546. Xenon-133 diffusion coefficients for pyrolytic carbon at temperatures ranging from 700 to 1300 C were also presented in previous progress reports, BMI-1514 and BMI-1518.

Work This Period

Diffusion measurements were made on Linde single-crystal alpha-alumina spheres which had been irradiated in contact with fully enriched UO_2 for a period of about 5 weeks in a thermal-neutron flux of about 1.6×10^{13} nv. Preliminary analysis of the data from these measurements indicated that the diffusion coefficients for xenon-133 are from ten to a hundred times lower than those reported for alpha-alumina spheres which contained lower fission-fragment concentrations. The fission-fragment concentration in the present experiment is 5000 times that used in prior xenon diffusion studies. The activation energy over the temperature range from 800 to 1500 C appears to be constant at a value lower than the 70 kcal per g mole obtained in prior experiments. Similar measurements are being made on pyrolytic-carbon spheres irradiated under the same conditions.

Plans for Future Work

Diffusion measurements will be performed using alpha-alumina and pyrolytic-carbon spheres irradiated approximately 8 weeks in a thermal-neutron flux of 1×10^{13} nv to demonstrate the effect of prolonged irradiation on observed fission-gas release. An experiment to further explore the effect of low xenon-133 concentration on diffusion in alpha alumina is to be conducted using single-crystal alpha-alumina plates with fission-product concentrations about 100 times lower than that used in the low-concentration diffusion experiment described in last month's report (BMI-1549).

Techniques for measuring solid fission-product diffusion in alumina and pyrolytic carbon are to be developed, and diffusion studies involving other potential coating materials for fuel particles are planned.

Fuel-Compatibility Studies

G. W. Cunningham, D. C. Carmichael, R. H. Barnes,
and S. J. Paprocki

Objective

To investigate reactions which may affect the maximum operating time or temperature of coated fuel particles.

Program Approach

The stability of oxide-coated UO_2 particles in an oxidizing atmosphere will be studied by measuring weight changes and by determination of changes in oxygen content of the UO_2 in the temperature range of 1000 to 1800 C. Specimens will be examined by metallographic and X-ray diffraction techniques, and the results will be compared with weight-change data obtained on a recording microbalance. Initial studies on the oxide-carbon reactions will involve the use of gravimetric methods to determine reaction kinetics of the oxides with pyrolytic graphite. Later studies will require the measurement of gas pressures. Diffusion of uranium through pyrolytic carbon coatings on UC, UC_2 , and UO_2 particles will be investigated by annealing coated particles of fully

enriched uranium compounds at temperatures of 1000 to 1800 C for various times. The distribution of uranium in the carbon coating will then be determined by a combination of metallographic, autoradiographic, and alpha-counting techniques. The redistribution of uranium due to a thermal gradient will be studied by annealing specimens of carbon-coated UC₂ in a temperature gradient for specified periods of time.

Progress Reported Previously

Particles of enriched uranium carbide coated with pyrolytic carbon were heat treated at 1400, 1600, and 1800 C for varying time periods to study uranium diffusion through the coatings. Results of X-ray diffraction analyses on the particles were presented in a recent progress report, BMI-1524. At temperatures of 1600 C or above, the UC is converted to UC₂ in a few hours. At lower temperatures, UC and U₂C₃ also exist. Alpha-counting and autoradiographic measurements were performed on these specimens to obtain data to be used for calculating the rate of diffusion of uranium through the coatings. Studies of the oxidation of Al₂O₃-coated UO₂ particles indicated that the observed failure of particles at a critical temperature is due to cracking of the coatings by thermal stresses resulting from the difference in thermal expansion of the Al₂O₃ and the UO₂. This temperature of rapid failure was found to depend on the coating thickness, UO₂ particle density, and the original temperature of application of the coating. It was also found that oxidation of the UO₂ by diffusion of oxygen through the Al₂O₃ coating can occur at temperatures below that at which the coatings crack due to thermal stress. Thermogravimetric methods were used to determine permeability rates.

Work This Period

The studies involving the use of autoradiographic and alpha-counting techniques to measure uranium diffusion through pyrolytic-carbon coatings on UC particles are continuing. Derivations of the equations for interpreting the results of the measurements in terms of diffusion coefficients have been completed.* The concentration of a diffusing species in a uniform annular coating on a spherical fuel core for an initial uniform concentration of the diffusing species in the fuel core and an initial concentration of zero in the coating can be approximated by the equation:

$$c = \frac{2\nu a c_0 \sqrt{D_2 t}}{r \sqrt{\pi} (1+\nu)} \left\{ \left[\frac{1}{(r-a)} - \frac{2D_2 t r}{a(r-a)^3} \right] \exp \left[-\frac{(r-a)^2}{4D_2 t} \right] - \left[\frac{1}{(2b-r-a)} - \frac{2D_2 t (2b-r)}{a(2b-r-a)^3} \right] \exp \left[-\frac{(2b-r-a)^2}{4D_2 t} \right] \right\}$$

for $\frac{D_2 t}{(b-a)^2}$ less than 0.2 where,

c = concentration in coating, atoms per cm³

c_0 = initial concentration in fuel core, atoms per cm³

D_1 = diffusion coefficient for core region, cm² per sec

* The mathematical derivations were performed by L. E. Hulbert, BMI.

D_2 = diffusion coefficient for coating region, cm^2 per sec

a = radius of fuel core, cm

b = outside radius of coating, cm

t = diffusion time, sec

r = radial coordinate between a and b , cm

$$\nu = \frac{D_1}{D_2} .$$

The rate at which alpha particles are emitted from a composite sphere with the distribution of the source of alpha radiation represented by the preceding equation is approximated by the equation below for the case where the coating thickness is greater than the

alpha-particle range in the coating and $\frac{D_2 t}{(b-a)^2}$ is less than 0.2.

$$R = \frac{2\lambda \nu c_o (D_2 t)^{3/2}}{\delta \sqrt{\pi} (1+\nu)} \left\{ - \left[\frac{(b+\delta)^2 - b^2}{b^2 (b-a)^2} \right] \left[1 - \frac{4D_2 t (b+a)}{a (b-a)^2} + \dots \right] \exp \left[- \frac{(b-a)^2}{4D_2 t} \right] + \right. \\ \left. \frac{2D_2 t b}{(b-\delta)^2 (b-\delta-a)^3} \exp \left[- \frac{(b-\delta-a)^2}{4D_2 t} \right] - \frac{2D_2 t b}{(b-\delta)^2 (b+\delta-a)^3} \exp \left[- \frac{(b+\delta-a)^2}{4D_2 t} \right] + \dots \right\} ,$$

where R = alpha-particle emission rate, particles per sec

δ = alpha-particle range in coating, cm

λ = radioactive-decay constant for alpha emitter, sec^{-1} .

Both equations are limited to the condition that the diffusing species does not penetrate to the outside surface of the coating.

The carbon-coated fuel particles studied to date vary considerably in size and deviate from the spherical geometry, making it difficult to obtain good results. A very approximate analysis of alpha-counting measurements indicates a diffusion coefficient of about $1 \times 10^{-10} \text{ cm}^2$ per sec at 1800 C for uranium in laminar-type pyrolytic-carbon coatings on UC particles. This value assumes a carbon-coating density of 1.7 g per cm^3 with a corresponding alpha-particle range of 19μ . Analyses of results of autoradiographic measurements on the same particles show a uranium diffusion gradient in the coating; however, dimensional uncertainties have prevented diffusion coefficients from being obtained.

Additional study has been made of the permeation of oxygen through the Al_2O_3 coatings by UO_2 particles at temperatures below that at which rapid failure occurs by thermal-stress cracking of the coatings. The permeabilities are calculated on the basis of

weight-gain measurements on coated particles heated in an oxidizing atmosphere obtained using a recording microbalance. Table L-3 contains corrected and additional data to those reported last month. The corrections are primarily due to more accurate measurements on the coating thickness.

TABLE L-3. PERMEABILITY OF OXYGEN THROUGH Al_2O_3 COATINGS AT A PRESSURE OF 1 ATM

Specimen	Test Temperature, C	Test Time, hr	UO_2 Particle Diameter, μ	Coating Thickness, μ	Permeability, $\text{g}/(\text{cm}^2 \times \text{sec})$	Oxidized and Quenched Structure
OXT-13	1300	120	68	65	1.09×10^{-9}	UO_{2+x} U_4O_9
OXT-30 ^(b)	1300	96	62	38	9×10^{-10}	--
OXT-14	1200	96	68	65	5.38×10^{-10}	U_3O_8
OXT-15 ^(a)	1200	96	260	158	4.23×10^{-10}	UO_2 , U_3O_8
OXT-16	1200	1000	63	40	3.08×10^{-10}	UO_{2+x} , U_4O_9 , U_3O_8
OXT-27 ^(b)	1200	139	62	38	2.08×10^{-10}	UO_2
OXT-30 ^(b)	1100	100	62	38	1.24×10^{-10}	--

(a) Porous UO_2 .

(b) Coatings deposited at 1400 C. All other specimens coated at 1200 C.

The reactions between pyrolytic graphite and various oxides are also being studied. Preliminary tests are presently being made to determine the reaction temperature in vacuo. X-ray diffraction examination of a sample of Al_2O_3 and pyrolytic-graphite powders heated at 1200 C for 2 hr in vacuum indicated that no reaction occurred. The tests in vacuo will be followed by thermogravimetric tests in a flowing atmosphere of controlled CO partial pressure. Modification of the equipment for these controlled-atmosphere tests will be completed when the auxiliary equipment which has been ordered is received.

The equipment which has been constructed for investigating the migration of UC_2 in carbon coatings has been tested, and a specimen is being run at 1200 C.

Plans for Future Work

The study of the rate of diffusion of uranium in carbon coatings will be continued using spherical UC particles coated with pyrolytic carbon. Reaction temperatures of various oxides with pyrolytic carbon will be determined in vacuo and at CO pressures of 10, 100, 500 mm of mercury. Thermal diffusion in carbon-coated UC_2 particles will be investigated.

Coolant-Compatibility Studies

C. A. Alexander and A. Levy

Objective

To establish experimentally the compatibility of high-pressure steam with Al_2O_3 , MgO , ZrO_2 , and UO_2 .

Program Approach

A transpiration study is being conducted in which the effect of steam on the rate of volatilization is being investigated. Initial investigations are being conducted at low steam partial pressures; under these conditions vaporization mechanisms may be more precisely determined.

Progress Reported Previously

Results of a literature study were presented in a topical report, BMI-1530. Experimental data on the effect of water vapor on UO_2 volatilization as well as preliminary findings on MgO , have been presented in progress reports.

Work This Period

The effect of water vapor on the volatilization of MgO has been determined over the temperature range from 1460 to 1740 C. It has been found that water vapor does not influence the vaporization of MgO . Determinations have been made in wet and dry argon and in oxygen, and all the data may be represented by the same equation. Mass spectro-metric analyses of the argon employed indicated that oxygen was present in the parts-per-million range. This concentration of oxygen is sufficient to suppress the reaction, $\text{MgO(c)} \rightarrow \text{Mg(g)} + 1/2\text{O}_2\text{(g)}$, at the temperatures employed in this investigation.

It is concluded that under oxidizing conditions magnesia vaporizes congruently. Sufficient, reliable data exist in the literature on the vaporization of magnesia under reducing conditions, or under high vacuum, that it is considered unnecessary to perform any experiments under these conditions. The observed vapor pressure of MgO(g) is considerably greater than the value predicted from tabulations of free-energy functions which have been calculated from spectroscopic data. At a temperature of 1400 C the erosion rate of MgO would be of the order of a mil per year, regardless of the nature of the coolant, unless of course the coolants were reducing, in which case the erosion rate would be higher.

Plans for Future Work

Preliminary studies with Al_2O_3 and ZrO_2 are under way, and it is planned to begin more detailed experimental investigations of these oxides in the coming month. Arrangements are also being made to begin experimental determinations at high steam pressures.

Thermal-Conductivity Studies

H. W. Deem and C. F. Lucks

Objective

To select a method and to design and assemble an apparatus for making thermal-conductivity (thermal-diffusivity) measurements on unirradiated and irradiated specimens containing coated fuel particles.

Program Approach

The problem was approached through a literature survey of methods for measuring thermal conductivity at elevated temperatures.

Progress Reported Previously

The apparatus was assembled, and an operational check run of the system was made at about 500 C using a 15-mil-thick graphite specimen.

Work This Period

A high noise level in the output circuit signal as shown on the oscilloscope caused much concern. An improved specimen holder to shield the detectors from the incoming primary cyclic thermal radiation was installed. Temporary vibration damping of the entire system and its components and improved filtering and grounding methods reduced the unwanted noise to a satisfactory level.

Measurements of about 440 C were made on a 1/4-in. -diameter by 80-mil-thick Type 7087 graphite specimen. A thermal-diffusivity value of 0.59 cm^2 per sec at 438 C was obtained for the specimen. The calculated thermal conductivity of $0.36 \text{ cal/ (sec) (cm) (C)}$ is in reasonable agreement with published data for similar material.

Further evaluations of the data and method are in progress.

Plans for Future Work

An evaluation of the method will be made on the basis of the past measurements, and any necessary modifications will be made. The apparatus will be prepared for remote hot-cell operation. This preparation will include completion of the specimen-loading assembly, construction of a new heater for remote operation, installation of lead shielding and water cooling in the vacuum chamber, and any other preparations necessary.

Evaluation of Commercial Products

H. S. Rosenberg, T. S. Elleman, and D. N. Sunderman

Objective

To provide information on the availability and quality of coated-fuel particles from commercial sources.

Program Approach

Interested manufacturers of coated particles who request information on the Coated-Particle Program are sent a standard form letter and summary of the evaluation procedures presently used for coated particles. Battelle is authorized to purchase 100-g lots of coated-fuel particles at a price of \$1 per g and to report the results of an evaluation of the material in this section of the report.

Progress Reported Previously

Requests for information on this program from 17 interested commercial firms have been received and answered.

Work This Period

No requests for information on this program were received during November.

Plans for Future Work

No future work is contemplated until offers of commercial products are received.

CORROSION STUDIES OF THE FLUORIDE-VOLATILITY PROCESS (ORNL)

P. D. Miller, E. F. Stephan, and C. L. Peterson

Objective

To evaluate materials of construction for various components of the Fluoride-Volatility process.

Program Approach

Candidate metals and alloys are being subjected to hydrofluorination and fluorination conditions in bench-scale equipment. Also, components from the ORNL Fluoride-Volatility pilot plant are being evaluated by metallographic techniques. These investigations are under study in support of nuclear fuel-reprocessing research being conducted by the Chemical Technology Division at ORNL.

Progress Reported Previously

Various aspects of the work have been described in previous progress reports. A combined hydrofluorination-corrosion study involving ZrO_2 dissolutions and INOR-8 corrosion specimens was described in BMI-1518. A similar investigation using Hastelloy X and tungsten was described in BMI-1534. The corrosion of INOR-8 and of HyMu-80 during hydrofluorination of Zircaloy-2 was described in BMI-1549. Fluorination-corrosion studies have also been in progress utilizing INOR-8, HyMu-80, and "L"-Nickel corrosion specimens as reported in BMI-1524, BMI-1534, and BMI-1546.

Work This Period

A 200-hr corrosion evaluation of INOR-8 and HyMu-80 was conducted under hydrofluorinator conditions in the presence of dissolving Zircaloy-2 in $LiF-NaF-BeF_2$ melts at 650 C. Apparently, this salt mixture is more corrosive than the $LiF-NaF-ZrF_4$ type studied earlier. The corrosion was most severe at vapor-liquid interface areas where the penetration was as high as 110 mils per month for INOR-8 and 85 mils per month for HyMu-80. The corrosion on specimens in the vapor and liquid was much less. The attack did not seem to be intergranular in nature.

Plans for Future Work

A fluorination-corrosion study with HyMu-80, "L"-Nickel, and INOR-8 specimens will be conducted at 495 C in a $LiF-NaF-BeF_2-ZrF_4$ melt.

PROGRESS ON USAEC/AECL COOPERATIVE PROGRAM (SRO)

S. W. Porembka

Work coordinated by SRO relating to the USAEC/AECL Cooperative Program is discussed in this section.

DEVELOPMENT OF COLD BONDING PROCESSESFriction Bonding

S. J. Paprocki, E. S. Hodge, and J. H. Peterson

Objective

To study the end capping of SAP fuel sheaths by friction techniques.

Program Approach

Joint design, pressure, rotational velocity, time, and surface preparation are being investigated in the friction-bonding end-capping studies. Friction extrusion of tube ends is being investigated as a parallel effort. Metallographic techniques are presently employed as the primary method of evaluation. A burst test is being considered as a means for inspecting the integrity of end caps.

Progress Reported Previously

It was reported in BMI-1541 that end-capping studies using SAP materials were initiated. Attempts to join the SAP material by conventional friction-bonding techniques have not been successful. The lack of plastic deformation during the bonding cycle is the primary problem area. Aluminum specimens were joined without difficulty using the same techniques and equipment.

Work This Period

Friction bonding of SAP material has been evaluated using various bonding parameters. The use of low speeds, less than 100 rpm, has yielded encouraging results. Although shearing occurred in the parent components, the bonds at the original SAP-to-SAP interface look excellent. Joining of SAP to aluminum has also been demonstrated with similar results. However, in these specimens, shear occurred in the aluminum member.

The forming of end caps on the SAP fuel sheaths by friction extrusion is being investigated. Initial tests resulted in forming a major portion of the end-cap configuration. The holes remaining after the friction-extrusion operation measured approximately 1/8 in. in diameter. Use of improved die materials and design should permit more complete closure of the tube. Means for closure of the final opening are being considered.

Plans for Future Work

Friction extrusion of the end cap will be continued to obtain a more complete closure of the tube. Friction bonding of SAP materials will be continued with particular emphasis on the lower speeds and high pressures. The use of aluminum as an intermediate layer at the SAP interface will be evaluated in these tests. External heating of the specimens during the friction-bonding cycle will be investigated as a means for obtaining increased plastic deformation.

Roll Forming

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

Objective

To determine the effects of surface preparation and design on metallurgical bonding in roll forming joints between stainless steel and Zircaloy-2 tubular components.

Program Approach

Roll-forming equipment has been assembled to investigate the joining of stainless steel and Zircaloy components. Variations in surface design and the use of intermediate barrier materials are being considered. Flat-plate composites of the two materials are being used in initial screening studies to determine the most effective combination of surface finish and preparation to join the stainless steel and Zircaloy. The most favorable combinations will then be investigated as full-size joint components in a roll-forming operation.

Progress Reported Previously

Various parameters have been established for the apparent joining of flat-plate composites of Type 410 stainless steel and Zircaloy-2 by cold rolling. Surface finishes, surface preparations, and the incorporation of alumina and gold layers at the stainless-Zircaloy interface have been considered. As a result of this study, composites have been consistently joined at 37 to 42 per cent cold-reduction levels. No difference in the interface appearance between these composites and those previously rolled 75 per cent has been observed.

Roll-forming equipment has been set up, and the capabilities and limitations of the operation have been determined. Single-pass reductions of about 15 per cent represent a maximum possible for this equipment using stainless steel and Zircaloy tubular components. Locally high deformations produced by various joint designs have not been sufficient to result in the joining of these materials.

Work This Period

Additional flat-plate tests were conducted in an effort to more precisely define the range of surface roughness most beneficial to the cold joining of stainless steel to Zircaloy. The composites for these tests measured 2 by 4 by 0.100 in. The added length allowed for a longer time of contact with the rolls. Surface roughnesses of 1500, 1800, and 2100 μ in. rms were machined on the stainless steel members and 300 μ in. rms on the Zircaloy components. Because of machining difficulties, there was considerable variation in roughness for each of the stainless steel plates. Surface preparation consisted of degreasing followed by scratch-brushing. Within 30 sec of the scratch-brushing operation, the composites were rolled. Reductions ranging from 25 to 55 per cent were made on these composites in single passes.

A tendency for these longer composites to adhere to and to follow the top and bottom rolls was noted, and, on occasion, the action caused the members to pull apart on the exit side of the mill. Reductions of 38 to 55 per cent were required to join these composites. In several cases, a considerable variation in reduction was evident within the individual specimens. These tests employed significantly rougher surfaces than were used in previous tests, and no lowering of the reductions required for joining was

noted. These results suggest an optimum roughness value above which no further benefit for joining of flat-plate composites is gained. In previous tests increasing the roughness of the stainless steel member from 300 to about 500 μ in. rms lowered the reductions necessary to join the components from 50 to 40 per cent. It now appears that further surface roughening has little effect.

Metallographic examination of specimens from the series of flat-plate tests employing rough surfaces has been completed. Some small separations were visible in areas of specimens reduced less than about 42 per cent. These separations were located in the grooves on the rougher surfaces, indicating that the flow of metal in these cases was insufficient for complete filling of these grooves. The majority of the joined composites, however, exhibited good contact all along their interfaces.

Plans for Future Work

Further flat-plate tests are planned with specimens having electroplated gold layers on both composite members. An attempt will be made to achieve self-bonding between the gold layers. Consideration will be given to the use of soft metal inlays to achieve a tighter mating of the joint members using reductions within range of the roll-forming equipment. A more quantitative evaluation of flat-plate composites joined under various conditions will be made using thermal-cycling tests.

Explosive Joining

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

Objective

To determine the feasibility of achieving mechanical and metallurgical bonding between Zircaloy-2 and Type 410 stainless steel by explosive techniques.

Program Approach

Explosive techniques are being employed to bond Zircaloy-2 to Type 410 stainless steel. Evaluation is based upon bond strength as determined by metallographic examination and mechanical testing.

Progress Reported Previously

Flat-plate assemblies of the subject materials have been explosively joined preparatory to testing full-scale components. Several mock-up tubular-component specimens have been explosively joined. Charge configurations and joint design have been varied during this testing in an effort to produce surface jetting and thus a sound metallurgical bond.

Work This Period

Additional tubular specimens possessing a 1500- μ in. rms surface on the stainless steel and a 300- μ in. rms surface on the Zircaloy-2 were roll formed preparatory to explosive joining. Stand-off charges of 45.5 g at 1/2 in. and 56 g at 1/4 in. in a water transfer medium were employed. The decreased stand-off distances used in these tests were intended to produce an increase in pressure at the stainless steel-to-Zircaloy-2 interface of sufficient magnitude to produce surface jetting. This additional pressure increase, however, resulted in a rupturing of the specimens. Complete separation of the Zircaloy and stainless steel components was noted in the remaining portions of the assemblies. As a result, no metallographic evaluations will be conducted on these composites.

Plans for Future Work

Preparations have been completed for conducting a test in which a presintered low-density stainless steel disk prepared by powder metallurgical techniques will be placed in the stainless steel-to-Zircaloy-2 interface and the assembly explosively impacted. The object of this test will be to attain high surface deformation as the stainless steel and Zircaloy-2 are driven into the pores of the porous stainless steel disk. This action is expected to rupture the oxide films and expose bare metal for metallurgical bonding. The explosive impulse should also result in a densification of the porous stainless steel disk.

Additional tests will be conducted using a combination of the roll-forming and explosive-joining techniques in which a soft metal liner will be placed in the stainless steel-to-Zircaloy-2 joint. Specimens will also be prepared for direct explosive joining in which a 0.010 to 0.020-in. clearance between the stainless steel and Zircaloy-2 will be incorporated. After impacting, these specimens will be subjected to a thermal-cycling test as a means of determining bond integrity.

LONG-TERM CREEP-RUPTURE PROGRAM ON SAP ALLOYS

J. A. VanEcho

Objective

To determine the effect of long-term exposure (to 20,000 hr) on the creep-rupture strength of three sintered aluminum powder alloys (M-583, SAP-895, and M-257) at 700, 800, and 900 F.

Program Approach

Test specimens are placed in standard Battelle creep frames and furnaces to determine their stress-rupture and creep strengths. Test temperatures are maintained to an accuracy of ± 2 F. Rupture times are recorded to the tenth of an hour by interruption of a timer when failure occurs. Creep measurements are taken either daily or more often by means of an optical system which has a sensitivity of 0.00005 in. All requirements for creep testing specified by ASTM Standard E-139-58T are met or exceeded.

Stresses are chosen to cause failure in 1, 10, 100, and 1000 hr on each material at each temperature. These data will be used to estimate stresses to cause failure in 10,000 and 100,000 hr. Actual tests will be made to verify the 10,000-hr stresses but the 100,000-hr tests will be discontinued at 20,000 hr.

Progress Reported Previously

Two of the test materials (M-583 and SAP-895) had been received and machined into test specimens. Because of apparent notch sensitivity, it was necessary to increase the fillet radius from 1/8 to 1/2 in. The gaged section was also tapered slightly to encourage failure near the center. Some satisfactory creep-rupture results had been obtained but none was reported as yet.

Work This Period

Creep-rupture tests were continued on the two materials on hand at all three temperatures. Most of the tests are scheduled for 1000 hr or longer and therefore

only a few ruptures have occurred to date. Some of these failures were noted in the threaded areas. It is not yet established whether these are isolated instances or not. Should such failures continue to occur, it may be necessary to make further changes in the specimen.

All test results, including those of the fillet and thread failures, are shown in Tables Q-1 and Q-2.

Plans for Future Work

Additional tests will be made to determine whether another change in specimen design is needed. Even though the cross-sectional area of the threaded section is at least 50 per cent greater than that of the gaged section, it may be necessary to further reduce the gage diameter to perhaps 0.400 in.

When notch failures no longer occur, tests will be continued to determine the 1-, 10-, 100-, and 1000-hr rupture stresses preliminary to estimating the 10,000-hr and 100,000-hr rupture stresses of all test materials.

TABLE Q-1. CREEP-RUPTURE DATA ON SAP-895 SINTERED ALUMINUM POWDER ALLOYS AT 700, 800, AND 900 F

Specimen	Temperature, F	Stress, psi	Rupture Time, hr	Elongation, per cent	Reduction of Area, per cent	Minimum Creep Rate, per cent per hr
6-4	700	13,000	2.5	2.6	6.3	0.48
6-5	700	12,000	17.5	2.2	3.3	0.058
1-8	700	11,500	37.3	0.9	3.8	0.016
1-9	700	11,500	32.9	2.2	4.0	--
1-6	700	11,000	(In progress 1003 hr)			0.00011
1-1	700	10,000	215.6 ^(a)	--	--	0.00018
6-3	800	11,000	0.9	2.2	8.0	0.47
6-1	800	10,000	12.0 ^(b)	2.2	5.0	0.025
1-2	800	9,000	86.8 ^(b)	0.9	6.9	0.0004
6-7	800	9,000	(In progress 236 hr)			0.0002
6-6	800	8,000	(In progress 1003 hr)			0.00005
6-2	900	10,000	0.1	3.9	9.1	--
1-3	900	9,000	0.9	4.8	15.6	0.22
1-4	900	8,000	5.8 ^(b)	1.3	4.5	0.012
1-10	900	7,400	81.1 ^(a)	--	--	--
1-7	900	7,400	101.5 ^(a)	--	--	--
1-5	900	6,700	(In progress 1050 hr)			0.00004

(a) Failed in thread.

(b) Failed in fillet.

TABLE Q-2. CREEP-RUPTURE DATA ON M-583 SINTERED ALUMINUM POWDER ALLOY AT 700, 800, AND 900 F

Specimen	Temperature, F	Stress, psi	Rupture Time, hr	Elongation, per cent	Reduction of Area, per cent	Minimum Creep Rate, per cent per hr
21-1	700	10,000	0.7 ^(a)	3.1	2.9	2.2
66-1	700	9,000	2.8	2.6	7.8	0.65
66-2	700	8,000	37.9 ^(a)	2.2	3.9	0.01
60-1	700	7,000	(In progress 905 hr)			--
246-1	700	7,000	(In progress 1195 hr)			0.00007
38-1	800	9,000	On loading ^(b)	--	--	--
21-2	800	8,500	0.1	2.6	2.3	--
85-1	800	8,000	0.3	2.6	5.2	2.6
85-2	800	7,000	3.5 ^(a)	2.6	3.6	0.13
44-1	800	6,000	189.9	1.7	3.6	0.0004
44-2	800	5,750	73.7 ^(b)	1.7	3.9	--
60-2	800	5,750	(In progress 713 hr)			0.00006
21-4	900	6,000	0.8	3.5	7.1	1.2
85-3	900	5,400	1.4 ^(a)	3.5	7.1	0.25
246-2	900	5,400	41.7	3.5	11.9	0.004
21-3	900	5,000	194.4	3.9	5.2	0.0004
246-3	900	4,800	819.3	2.2	10.8	0.000055

(a) Failed in fillet.

(b) Failed in threads.

RADIATION-EFFECTS STUDY OF CANDIDATE FUEL
MATERIALS FOR THE MGCR (GA)

P. Steinback, J. E. Gates, and W. H. Goldthwaite

Objective

To determine the irradiation stability of selected ceramic-matrix fuels under conditions of temperature and burnup similar to those anticipated in the MGCR.

Program Approach

Battelle has assisted General Atomic in the development and evaluation of core materials for the Maritime Gas-Cooled Reactor and the High-Temperature Gas Reactor and is now conducting studies of the irradiation damage of BeO-matrix fuels up to relatively high burnups. Clad fuel pellets are irradiated in capsules which are equipped with thermocouples and auxiliary electric heaters and designed to maintain specimen-surface temperatures in the 1500 and 1700 F range, primarily by fission heating. Maximum target burnups are in the range of 40 to 50 per cent of the uranium-235 atoms originally present. Data concerning specimen burnup, swelling, fission-gas release, and microstructural changes are obtained in postirradiation hot-cell examination.

Progress Reported Previously

Capsule BMI-36-1, containing six Hastelloy X-canned specimens of 30 volume per cent UO₂ (12 per cent enriched) in BeO, was prepared for irradiation in the MTR. Design irradiation conditions include fuel-surface temperatures of 1700 F and an in-pile period of 18 months in a specimen-effective peak flux of 2×10^{13} nv. The capsule was delivered to the reactor late in October, 1961.

Work This Period

The irradiation of Capsule BMI-36-1 began with MTR Cycle 165 on November 10, 1961. Although performance during this first cycle was satisfactory, the specimens were not located vertically across the peak of the neutron flux profile in the irradiation facility. Therefore, the capsule was relocated vertically within its facility during the shutdown for Cycle 166. The new location resulted in a more desirable temperature profile along the specimen array. Table S-1 summarizes initial temperature data for this experiment.

TABLE S-1. SUMMARY OF INITIAL TEMPERATURE DATA FOR IRRADIATION CAPSULE
BMI-36-1

Specimen Location (From Top)	Specimen Cladding Temperature ^(a) , F	
	Average During MTR Cycle 165 (November 10-27, 1961) ^(b)	Initial for MTR Cycle 166 (November 30, 1961)
1	1525	1470
2	1490	1520
3	1450	1565
4	1400	1570
5	1250	1530
6	1300	1540

(a) It is estimated that the fuel-surface temperatures are approximately 150 F higher than the cladding temperatures; the latter are approximately the same as temperatures monitored by sheathed Chromel-Alumel thermocouples adjacent to the specimens.

(b) Approximately 1 kw of auxiliary heat was utilized during Cycle 165. None was utilized during Cycle 166.

S-2

Plans for Future Work

The irradiation of Capsule BMI-36-1 will be continued.

RADIATION STUDIES OF SM-2 FUELS (ALCO)

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

Objective

To investigate the effects of irradiation on stainless steel-clad dispersions of UO_2 (nominally 26 w/o, highly enriched) and certain burnable poisons in Type 347 stainless steel.

Program Approach

This study is being conducted in assistance to Alco Products to develop satisfactory fuel materials for Army pressurized-water reactors. Reference and alternate SM-2-type specimens have been fabricated. Capsules containing five to eight specimens each are being irradiated or have been irradiated in the MTR or the ETR. Desired experimental conditions include nominal specimen-surface temperatures of 600 F and burnups of 37 and 70 per cent of the total uranium-235 atoms. The program includes three non-instrumented and seven instrumented (heaters and thermocouples) capsules; the former were designed for irradiation in MTR core-lattice positions, the latter for irradiation in ETR beryllium-reflector positions. This capsule group is identified as the BMI-32 series, and is described in detail in BMI-1516.

Progress Reported Previously

Irradiations of the first four capsules were completed early in 1960; irradiation histories and results of specimen analyses were discussed in BMI-1442, BMI-1448, BMI-1464, and BMI-1516. Capsules BMI-32-6 and BMI-32-8 were discharged on July 10, 1961, and BMI-32-9 on October 2, 1961, all from the ETR. Peak-flux specimen burnups for the former two were estimated to be in the range 65 to 75 a/o (total uranium), and for BMI-32-9, 90 a/o. Specimen-surface temperatures decreased gradually in all three capsules during the irradiations, and were near 400 F just prior to discharge.

Work This Period

The final three capsules of the program, BMI-32-5, BMI-32-7, and BMI-32-10, were discharged from the ETR on November 13, 1961, following Cycle 41. A brief summary of pertinent irradiation data for Capsules BMI-32-5 through BMI-32-10 is given in Table T-1.

TABLE T-1. SUMMARY OF IRRADIATION DATA FOR CAPSULES BMI-32-5 THROUGH BMI-32-10

Capsule	In-Pile Days	Estimated Temperature Range of Peak-Flux Specimen Surface During Irradiation ^(a) , F	Estimated Total Uranium Burnup of Peak-Flux Specimen, a/o
BMI-32-5	183 ^(b)	700-550	70
-6	197	550-350	65
-7	210 ^(b)	650-475	75
-8	197	525-450	67
-9	200	1150-400	90
-10	183 ^(b)	650-500	70

(a) In all cases, temperatures decreased gradually during the irradiations.

(b) Includes estimate for final cycle.

T-2

Plans for Future Work

Capsules BMI-32-5 through BMI-32-10 will be shipped to the ORNL hot cells early in December for postirradiation examination. A brief history of the six irradiations will be prepared.

GAS-COOLED REACTOR PROGRAM (AGN)

D. L. Keller

The Army Reactor Branch (ARB) is developing small portable reactor systems for military applications. Aerojet-General Nucleonics (AGN) is the prime contractor under the Army Gas-Cooled Reactor Systems Program (AGCRSP) and is charged with the design, construction, and operation of the GCRE and ML-1. The studies below are in support of the ML-1 program.

Development of Oxidation-Resistant Fuel Elements

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

Objective

To investigate the simultaneous densification and cladding of Al_2O_3 -coated UO_2 as a method of obtaining a high-density oxidation-resistant fuel.

Program Approach

It has been demonstrated that the oxidation resistance of UO_2 powder can be significantly improved by protecting the powder with a thin coating of Al_2O_3 . In this program, the coated powder will be used as a starting material. Various methods of packing the coated powder into steel tubes will be investigated. The tubes will be evacuated and under isostatic pressure will be collapsed around the ceramic core. The maximum Al_2O_3 diluent is restricted to about 40 volume per cent. The main concern is the retention of coating integrity during densification. This will be evaluated by nitric acid leach tests, oxidation tests, and metallographic studies.

Progress Reported Previously

Spherical UO_2 particles with average diameters of 150 and 50 μ have been coated by a vapor-deposition method with from 5 to 10 μ of Al_2O_3 . Characteristics of the as-received material were described in three previous progress reports: BMI-1489, BMI-1504 (Del.), and BMI-1518. Defective particles were removed by selective leaching in nitric acid followed by heavy-liquid flotation.

Evaluation of fabrication processes utilized prior to isostatic hot pressing, as reported in BMI-1518 and BMI-1524, shows that vibratory compaction eliminates particle breakup associated with other processes such as cold pressing. However, a particle mixture of more ideal packing size distribution is necessary to obtain the desired high densities.

Specimens containing undiluted UO_2 , UO_2 -20 volume per cent chromium cermets, cold-pressed Al_2O_3 -coated UO_2 , vibratory compacted Al_2O_3 -coated UO_2 , or Al_2O_3 -coated UO_2 with chromium diluent were fabricated. The specimens contained a small gas plenum which was defected with a 16-mil hole, after which the specimens were exposed to failure in 1500 F air. The undiluted UO_2 failed in 5 to 10 hr, both specimens containing only Al_2O_3 -coated UO_2 failed after 35 to 42 hr, and the specimen containing coated UO_2 with chromium additive failed after 60 to 72 hr. Comparison with a reference pin of BeO-70 w/o UO_2 showed all specimens with coated particles to be superior, after taking unavoidable variables under consideration.

Factors detrimental to oxidation resistance of cores containing Al_2O_3 -coated UO_2 are discussed in BMI-1546.

Work This Period

Two specimens containing BeO-coated UO_2 cores were fabricated by vibratory compaction followed by gas-pressure bonding. These contained only 30 volume per cent UO_2 since such material was readily available. In addition, one specimen was fabricated by cold pressing and gas-pressure bonding which contained 65 volume per cent UO_2 coated with 18μ of porous Al_2O_3 . It was hoped that the enclosed porosity would "cushion" the compressive stresses during fabrication and prevent particle breakup. Two specimens were also fabricated using Al_2O_3 -coated UO_2 with nearly "idealized" size distribution. All specimens were exposed to 1200 F air after the mild steel cladding was removed by pickling in 50 HNO_3 -50 H_2O at 200 F. The cores containing Al_2O_3 -coated UO_2 showed approximately 90 per cent oxidation of the UO_2 within 15 to 20 min; however, the BeO-coated UO_2 cores showed only approximately 5 per cent oxidation in 30 min. A comparison between the BeO and Al_2O_3 coatings is not completely justified on the basis of this test since the BeO-coated specimen contained approximately twice the amount of diluent that was present in the Al_2O_3 -coated specimen. However, it would appear that an investigation of BeO coatings would be of interest.

Hastelloy X containers have been fabricated for use in preparing a series of tubular pins containing a variety of cores. The cores will be undiluted UO_2 , UO_2 -chromium cermets, various Al_2O_3 -coated UO_2 compacts, and BeO-coated UO_2 . The high oxidation resistance of the Hastelloy X will permit thermogravimetric oxidation testing of the defected pins at 1750 F to obtain quantitative data for comparison.

Radiation Stability of UO_2 -BeO Specimens

D. G. Freas, J. H. Saling, J. E. Gates, and R. F. Dickerson

Work on this program has been recessed.

High-Temperature Fatigue Evaluation of Corroded Hastelloy X Fuel-Pin Tubing

L. G. Beall, A. A. Mittenbergs, and G. M. McClure

Objective

To determine the elevated-temperature fatigue performance of corroded Hastelloy X tubing and to compare the results with existing data for noncorroded tubing.

Program Approach

Fatigue tests and stress rupture tests are being conducted in Krouse axial-load machines at three temperature levels (1400, 1600, and 1800 F) on corroded Hastelloy X tubing to develop a Goodman-type diagram based on 10^6 -cycle lifetimes for stress ratios of $A = 0$, $A = 0.25$, $A = 0.95$, and $A = \infty$ (approximately). Additional tests are being performed to establish S-N curves for $A = 0.95$ at 1600 and 1800 F.

Work Reported Previously

Tests on 15 specimens at 1600 F and 1800 F for A ratios of 0.95 and 0.25 were reported last month in BMI-1549. As reported, the results indicated the 10^6 -cycle fatigue strength of corroded tubing is slightly lower than the noncorroded Type B tubing supply of the previous study but somewhat higher than noncorroded Type A tubing supply.

Work This Period

During November 27 specimens were failed. The majority of the tests were concerned with the various stress ratios for 1600 and 1800 F. The tentative stress values for these temperatures are shown in the following tabulation:

Temperature, F	Stress Ratio, A	Mean Stress, psi	Alternating Stress, psi
1600	0	12,500	0
1600	0.25	11,200	2,800
1600	0.95	10,750	10,250
1600	∞	0	21,000
1800	0	5,000	0
1800	0.25	5,000	1,250
1800	0.95	4,600	4,400
1800	∞	0	15,250

The stress-rupture data ($A = 0$) are for 10-hr lifetimes. This corresponds with the time required to generate 10^6 cycles during a fatigue test.

Plans for Future Work

Plans for December are to conduct tests at 1400 F and a few additional tests at 1600 and 1800 F. These tests will complete the experimental program on corroded Weldrawn tubing.

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

S. J. Paprocki, E. S. Hodge, and J. H. Peterson

Objective

To develop fabrication techniques for cladding UO_2 fuel cores with beryllium.

Program Approach

This work is in support of ORNL's studies on potential materials for gas-cooled reactors. The gas-pressure bonding technique is being used to fabricate specimens. Evaluation is based upon bond and cladding properties after bonding as determined by metallographic examination and mechanical testing.

Progress Reported Previously

Good bonds have been obtained in self-bonding studies of beryllium. Specimens containing UO_2 cores coated with vapor-deposited chromium showed no reaction between core and beryllium cladding. Beryllia (BeO) was shown to be a satisfactory barrier material to prevent reaction between the cladding and spacer materials. A shaper has been set up to prepare surfaces for self-bonding studies.

Work This Period

Self-bonding studies were conducted to evaluate grit-blasted specimens and specimens with abraded surfaces and beryllium powder in the interface. These samples were bonded 4 hr at 1650 and 1850 F and 10,000 psi. The grain growth across the interface of the grit-blasted samples was in excess of 50 per cent in both specimens. The average surface roughness of the beryllium was about $170 \mu\text{in.}$ after blasting with 50-mesh chilled iron grit. The grain growth in the specimens with the powder in the interface compared favorably with the grit-blasted specimens in many areas. However, it was difficult to maintain an even distribution of powder at the interface, and certain portions of the specimens were not well bonded.

Beryllium specimens have been shaper finished to an average surface roughness of $250 \mu\text{in.}$ These specimens have not yet been bonded.

Plans for Future Work

Since the results obtained with the grit-blasted surfaces are very encouraging, the effect of surface roughness on the amount of grain growth will be evaluated. Specimens with powder at the interface will be evaluated only on a limited scale, since powder distribution is difficult to control. Beryllium with various shapered finishes will be bonded and evaluated.

Experimental fuel elements are scheduled for pressure bonding and evaluation. A burst test is being considered as a method of evaluating the strength of the bonds in the elements. Grit blasting will be used as the surface preparation for the initial specimen.

RWD/CRT: all