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PROGRESS RELATING TO CIVILIAN APPLICATIONS DURING AUGUST, 1961

by

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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 under way at Battelle.

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 temperature under normal and ultrahigh pressures in sealed platinum capsules. The major emphasis at present is on the study of reactions between U_3O_8 and Al_2O_3, CaO, CdO, MgO, and ZnO. The reacted samples are examined visually, and selected regions are studied by X-ray diffraction to determine phase relationships.

Progress Reported Previously

Some of the above mixtures were prepared and runs were made at elevated temperature and pressure. Samples were sectioned and examined in the microscope. There was separation into black and colored areas, which are believed to be due to nonuniform temperature along the sample axis.

Work This Period

Runs were made at 1200 C at both 60,000 atm and at ambient pressure with mixtures of U_3O_8-Al_2O_3, U_3O_8-CaO, U_3O_8-CdO, U_3O_8-MgO, and U_3O_8-ZnO, in which the ratio of uranium to other metal ion was 1 to 2 for the Al_2O_3 additions and 1 to 1 for the other additions.

At ambient pressure and 1200 C, there was no detectable reaction between U_3O_8 and Al_2O_3 or U_3O_8 and ZnO. In the U_3O_8-CaO mixture, the sample was converted to calcium uranate, $CaUO_4$, at 1200 C and ambient pressure. The U_3O_8-CdO sample reacted. The X-ray pattern of the reacted material was very similar to the calcium uranate pattern. Calcium and cadmium are divalent ions with nearly the same ion radius and form many analogous compounds. In the U_3O_8-MgO mixture, a reaction also occurred. The sintered sample was a mixture of an unidentified phase and unconverted U_3O_8.

In no case were the products obtained from mixtures subjected to 60,000 atm and 1200 C completely similar to those run at ambient pressure. In the U_3O_8-CaO, U_3O_8-CdO, U_3O_8-MgO, and U_3O_8-ZnO mixtures, face-centered-cubic type phases, with lattice parameters ranging from 5.23 to 5.38 A were found in the high-pressure samples. In samples of U_3O_8 alone sintered at high pressure and temperature, the smallest observed lattice parameter of a face-centered-cubic type structure has been 5.42 A. The low lattice constants in the mixtures subjected to high pressure suggest that the additive ions enter the cubic U_3O_8-type lattice. The presence of the cubic phases in the pressure samples is in accord with the theory that pressure will favor formation of a denser phase.

The cubic phases in all cases were black crystalline products which were extracted mechanically. Possibly because of nonuniformity of temperature, some portions of the high-pressure samples were not all black. X-ray diffraction analysis of specimens extracted from nonblack regions generally disclosed additional phases. In the U_3O_8-MgO high-pressure sample, the additional phase was the same as that seen in the ambient-pressure sample. In the U_3O_8-CaO high-pressure sample, microscopic evidence of another phase was seen, but it has not as yet been observed by X-ray diffraction. In the U_3O_8-CdO and U_3O_8-ZnO high-pressure samples, at least one unidentified additional phase has been observed.

No cubic phase has been seen in U_3O_8-Al_2O_3 high-pressure samples. There is evidence of reaction, the products of which have not been identified.

Plans for Future Work

There is definite evidence of pressure effects on reactions between U_3O_8 and Al_2O_3 and between U_3O_8 and the divalent oxides CaO, CdO, MgO, and ZnO. A better understanding of these effects will be obtained by systematic changes in U_3O_8-additive ratios and/or in pressure and temperature conditions.

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 properties of AISI Type 347 stainless steel.

Program Approach

Subsize tensile, cyclic-strain fatigue, and impact specimens of 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 total accumulated fast-flux exposure for capsules currently operating in or removed from the ETR prior to the end of Cycle 33 (December 14, 1960) was given in a previous progress report, BMI-1489. The results from Charpy and subsize Izod specimens irradiated to exposures of about 5.7×10^{21} nvt and tested at room temperature and -300 F were reported in BMI-1448 (Rev.). A definite embrittlement was noted in the irradiated specimens, particularly those tested at lower temperature.

Work This Period

Ten capsules are currently being exposed in the I-13 position of the ETR. The ETR is presently operating in Cycle 39, which is scheduled to terminate on or about September 5, 1961. The last two cycles, 38 and 39, have been of short duration with a total accumulation of approximately 2200 megawatt-days of full-power operation. To date, the capsules exposed in maximum-fast-flux positions have accumulated fast-flux exposures of about 9.0×10^{21} nvt.

Plans For Future Work

The capsules will continue to be irradiated in the I-13 position of the ETR. The capsules will be periodically discharged from the reactor for testing at exposures ranging from about 1.0 to greater than 3.0×10^{22} nvt.

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

F. R. Shober, P. B. Shumaker, and A. P. Young

Objective

To compare the creep resistance of Zircaloy-2 at 650 F in a fast flux of 1 x 10^{14} nv with its creep resistance 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. Total deformation will also be indicated during the test by a differential transformer, the output of which will be recorded. 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 in-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.

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. The magnitude of the yield point was found to be influenced by the apparent amount of solute elements retained in solution. Water quenching after heat treatment increased the magnitude of the yield considerably.

Work This Period

Fabrication of all components for a second in-reactor creep capsule, with the exception of final machining of the auxiliary heater blocks, was completed. A supplemental engineering analysis covering several minor design modifications of the capsule was prepared for the WTR Safeguards Committee review.

Additional short-time tensile tests were made at 650 F to study the influence of solid-solution elements and grain size on the yield-point phenomenon. The effects of solid-solution elements were again noted by a marked yield point. Grain-size determinations and second-phase particle-size and -distribution analyses of these samples have not been completed.

Plans for Future Work

The in-reactor creep capsule will be assembled during the early part of September. It is expected to begin irradiation in October. More tensile tests will be made with Zircaloy-2 at 650 F to continue the study of the influence of grain size, of solute-element concentration, and of second-phase particle size and distribution upon the yield-point phenomenon.

Friction and Wear of Sliding Surfaces in Sodium Environments

W. A. Glaeser and J. W. Kissel

Objective

To develop general principles applicable to selection of materials and 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 measuring the stick-slip friction behavior of a ball sliding on a flat in liquid sodium and under a carefully controlled environment. Interpretation of stick-slip traces is supplemented by analysis of surface films formed during exposure to sliding in sodium. Sliding experiments are also planned for high-purity flowing-sodium systems and between specimens having artificially applied lubricant layers. These studies are designed to investigate, on a semiquantitative basis, the effect of oxygen content on film formation, and the durability of pre-formed films of known composition and thickness.

Progress Reported Previously

The friction and wear behavior between slowly rubbing molybdenum surfaces in sodium environments are described in detail in a topical report, BMI-1405, and in paper No. 61AM2B-1 at the 16th Annual Meeting of the American Society of Lubrication Engineers (Philadelphia, April, 1961).

Friction and surface damage experiments involving specimens of tungsten, Inconel X, tungsten carbide, and titanium carbide (the latter two materials being cermets each with 20 w/o cobalt binder) are described in Paper No. 61-LUBS-16 presented at the ASME Lubrication Symposium at Miami in May, 1961.

In general, improved performance has been observed for the metal or 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 the cemented carbide surfaces in the presence of sodium. In this case, however, thermodynamic considerations do not favor the formation of reaction products.

Experiments with stainless steels, Types 347 and 304, exemplify the lowered friction condition noted above for metal and alloy combinations. The drop in friction coefficient for Type 304 stainless is from around 2 to about 0.2. This lowered friction persists after removal of sodium – by heating – from the specimen surfaces. This is further evidence that the frictional behavior observed is caused by a surface film rather than the presence of liquid sodium. Other features of the friction record suggest that the physical and chemical behavior of the surface films are altered by changes in environmental conditions.

Work This Period

Friction records for dry and sodium-lubricated conditions were obtained for specimens of 2-1/4 Croloy and of Stellite No. 6. Croloy is an iron-chromium-molybdenum alloy, 2-1/4 being the per cent of chromium. Dry friction between specimens of this material varied from about $\mu = 0.7$ to $\mu = 1.4$ over the temperature range explored. Following introduction of sodium, the friction remained in the region between about $\mu = 0.3$ to 0.4. Stellite No. 6 demonstrated a quite different behavior after the addition of sodium. In this case, dry friction varied between $\mu = 0.3$ and 0.55, and with sodium lubrication the friction was between $\mu = 0.2$ and 0.7. Friction with the sodium lubrication also was very erratic, thereby suggesting unstable conditions at the surfaces.

Plans for Future Work

Specimens of Chromallized 52-100 steel and of nitrided steel have been fabricated and metallographically prepared. These will be friction tested dry and with molten-sodium lubrication.

STUDIES OF FUELS (AEC-DRD)

R. F. Dickerson

This research is supported by the AEC, Division of Reactor Development, for the purpose of contributing to the technology of both existing and new materials.

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, 2 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 are presented in two topical reports, BMI-1400 and BMI-1536.

The planning of and construction of capsules for irradiation tests were reported in an earlier progress report, BMI-1464.

Annealing studies of irradiation control samples at 1600 and 1800 F were completed (2400 hr).

The center-line temperature of specimens in Capsule WR-20 (inserted in the WTR during the shutdown for Cycle 14) was raised from 1100 F to 1600 F by adding neon (45 volume per cent) to the helium in the capsule annulus. Auxiliary heat for control purposes is being supplied by resistance heaters.

Work This Period

The enriched control samples which were heat treated at 1600 and 1800 F were examined. There was no evidence of attack by sodium at either temperature. Table B-1 summarizes the slight weight changes observed in both sodium and in vacuum. Microstructural examination has not been completed.

Plans for Future Work

A second capsule containing 15 fuel pins for irradiation at 1800 F will be inserted into the WTR during the shutdown for Cycle 17 (October 5 to 7, 1961). 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.

TABLE B-1. RESULTS OF 2400-HR CORROSION TESTING OF ENRICHED NIOBIUM-URANIUM SPECIMENS

Composition (Balance Niobium), w/o	Test Conditions: Temperature, F	Test Conditions: Testing Environment	Weight Changes, mg per cm^2
10 U	1600	Sodium	0.02
	1800	Sodium	0.10
	1600	Vacuum	0.007
	1800	Vacuum	0.00
20 U	1600	Sodium	0.07
	1800	Sodium	0.23
	1600	Vacuum	0.01
	1800	Vacuum	0.00
30 U	1600	Sodium	0.13
	1800	Sodium	0.23
	1600	Vacuum	0.04
	1800	Vacuum	-0.006
20 U-10 Zr	1600	Sodium	0.08
	1800	Sodium	0.40
	1800	Vacuum	0.04
20 U-20 Zr	1600	Sodium	0.12
	1800	Sodium	0.28
	1800	Vacuum	0.03

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. Within limitations dictated by funding, the four most promising compounds have been selected for initial 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 the preparation and corrosion resistance in various media of thorium- and thorium-uranium-compounds of beryllium, boron, carbon, aluminum, and silicon. The results of these studies are presented in the following progress reports: BMI-1480, BMI-1489, BMI-1496, BMI-1504, BMI-1509, and BMI-1514. Three cylindrical castings (3/8 in. in diameter by 2-1/2 in. long) of each of the four compounds selected for more detailed study were cast for use in property determinations.

Work This Period

The castings were radiographed to determine their soundness. All three castings of the $(Th_9U)C_2$ compound were sound. Two each of the $(Th_9U)C$, $(ThU_9)B_4$, and $(Th_9U)Be_{13}$ castings showed some cracks. These are being recast in an effort to produce crack-free material.

The sound castings are being prepared for use in property determinations.

Plans for Future Work

As soon as specimens are prepared the following property studies will be initiated.

(1) Thermal-conductivity and thermal-expansion measurements from room temperature to 1800 F
(2) Compression-strength measurements at room temperature and at 1200 and 1800 F
(3) Compatibility studies at 1100, 1500, 2000, and 2500 F with potential cladding materials (molybdenum, niobium, zirconium, stainless steel, etc.)
(4) Oxidation tests at 900, and 1300 F of the $(Th_9U)B_4$ and $(Th_9U)Be_{13}$ to supplement data obtained at 1100 F
(5) Hot-hardness measurement to determine the softening temperature of each compound.

A Study of Plutonium-Containing Fuel Alloys

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

Objective

To investigate those plutonium alloys which may have potential application as fuels for reactors operating at relatively high temperatures or as fuels for possible use as "spike" elements in power reactors.

Program Approach

Plutonium-containing alloy systems have been selected for study on the basis of the behavior of similar uranium-alloy systems. The compositions selected are based on the niobium, throium, uranium-molybdenum, and uranium-niobium alloy systems with plutonium. 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

The results of previous studies, which are summarized in a recently issued topical report (BMI-1532), indicate that thorium-plutonium-base alloys and alloys consisting of plutonium silicides dispersed in niobium and of plutonium-zirconium alloy phases dispersed in niobium-zirconium matrices show sufficient promise to warrant further study.

Studies of uranium-niobium-base alloys with plutonium additions have begun; and cast uranium-10 w/o niobium-5, -10, -15, and -20 w/o plutonium alloys were found to increase in hardness with increasing plutonium content, as reported last month in BMI-1534.

Work This Period

Cast uranium-10 w/o niobium-5, -10, -15, and -20 w/o plutonium alloys were found to exhibit cored microstructures. At this point, it is uncertain whether a second phase is associated with these cored structures. Microstructures of cast uranium-10 w/o niobium-10 w/o plutonium-5 and -10 w/o zirconium, uranium-15 w/o niobium-15 w/o plutonium-15 w/o zirconium, and uranium-20 w/o niobium-10 w/o plutonium-10 w/o zirconium alloys were similarly cored. Hardness values for these quaternary alloys were 420, 356, 343, and 285 DPHN, respectively. Specimens of these alloys are being heat treated for homogenization, and it is expected that the structure of these alloys will be more clearly discernible and the reasons for hardness variations amenable to interpretation.

A button theoretically composed of $PuSi_2$ has been prepared by arc melting stoichiometric proportions of plutonium and silicon. Metallographic examination indicates that the button consists essentially of a single phase, identified by X-ray diffraction analysis as $PuSi_2$. A very small amount of precipitate, noted in the grains, is considered insignificant. This $PuSi_2$ button was prepared as a preliminary step in preparing an alloy consisting of 30 volume per cent $PuSi_2$ dispersed in a niobium matrix. This two-step procedure was adopted since previous attempts to prepare such an alloy by simultaneous melting of all three components were unsuccessful, with plutonium apparently being lost.

A heat treatment of 4 hr at 1000 C followed by furnace cooling resulted in severe cracking of previously prepared niobium-plutonium-silicon alloys. Reasons for this cracking are not certain, but could be indicative of transformation in the alloys.

Plans for Future Work

The effects of heat treatment on the structures of the uranium-niobium-plutonium and uranium-niobium-plutonium-zirconium alloys will be investigated.

Recrystallization characteristics of thorium-plutonium alloys at a temperature of 600 C will be studied by hardness measurements and metallography.

An attempt will be made to prepare a niobium-30 volume per cent $PuSi_2$ alloy by arc melting master-alloyed $PuSi_2$ with a proper proportion of niobium.

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 of 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 provide data to establish the temperature and time dependence of fission-gas-release rates.

Progress Reported Previously

In the past several months, modifications were made in the equipment to increase its over-all reliability and to extend the operating-temperature range of the specimen furnace. Extensive out-of-pile tests of furnace designs were conducted. An electrical-resistance furnace with a tantalum-wire resistor was selected for use in future experiments.

Work This Period

The program of UO_2 irradiations and fission-gas analysis has been resumed. At present, a sintered UO_2 sample of 93 per cent of theoretical density is being irradiated at 500 F. Analyses of released fission xenon and krypton are continuing, but evaluation of the data is not yet complete.

Plans for Future Work

Upon completion of the experiment now in progress at 500 F, the same UO_2 sample will be irradiated at a series of temperatures up to 3000 F. Additional UO_2 samples having densities between 90 and 100 per cent of theoretical will be irradiated in future experiments.

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 are described in a previous progress report, BMI-1509. Examination of three other specimens exposed to NaK at 150 C for 834 hr indicated that under these conditions the uranium-glass-aluminum material was not subject to significant corrosive action by the NaK. The irradiation capsules were designed and constructed of aluminum. However, fabrication difficulties encountered during the assembly of the capsule necessitated replacing some of the aluminum parts with nickel components. The lower thermal conductivity of the nickel, as compared to aluminum, has required that the capsule be irradiated in a neutron flux about 20 per cent lower than that permissible with aluminum in order not to exceed specimen temperatures of 100 C. This change introduces an increased irradiation time to reach the desired 25 per cent burnup of the uranium-235. Present estimates indicate that the modified capsule can be irradiated in a thermal-neutron flux of 2 x 10^{14} nv, and that the desired burnup can be reached in 50 days (three MTR cycles).

Work This Period

The new nickel capsule components were fabricated, and the capsule was assembled and loaded. The welds were inspected by helium leak detection and X-ray examination and found to be satisfactory. The capsule was sent to the MTR for insertion during the shutdown for MTR Cycle 161. However, space in the core at the desired flux was not available, and the insertion was delayed. It is expected that the capsule can be charged into the reactor during the shutdown for MTR Cycle 162 during the week of September 4, 1962.

Plans for Future Work

The capsule, BMI-41-1, will be inserted in the MTR during the shutdown for Cycle 162, if possible. The irradiation will be continued for three MTR cycles with an expected discharge date of November 6, 1961. The specimens will then be examined and evaluated.

Irradiation Study of Metastable Beta-Phase Uranium Alloys

M. S. Farkas, B. D. 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, are to be irradiated in two temperature-controlled capsules. The specimen compositions and heat-treatment conditions were reported in BMI-1489.

Capsule design is intended 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 are to be irradiated in the BRR to a burnup of 0.01 to 0.05 a/o over a 2 to 3-day period. These specimens are to be examined after irradiation, primarily to determine if the beta phase has been retained during irradiation. The specimens in the second capsule are being irradiated under similar conditions to a burnup of approximately 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 discussed in two recent progress reports, BMI-1518 and BMI-1524.

The irradiation of the second capsule was begun during MTR Cycle 156 in Position A-26-NE, and the capsule remained in this position through Cycle 158. During this period the monitored temperatures ranged from about 425 F at the top specimen zone to about 625 F at the bottom specimen zone, with specimen center-line temperatures being about 200 F higher. More uniform specimen temperatures could not be achieved in

A-26-NE because, with the specimen array located across the peak flux, temperatures were excessive. During Cycle 159 the capsule was located in Position A-20-SE, and monitored temperatures ranging between about 510 F and 690 F were achieved. Late in July the capsule was moved into Position A-20-NE in an attempt to increase the general temperature level.

Work This Period

During most of Cycles 160 and 161, the monitored temperatures have remained fairly steady in the neighborhood of 560 F at the top zone to about 625 F at the bottom zone. Consequently, it appears that during this two-cycle period the estimated center-line temperatures of all specimens were close to or within the 800 to 850 F range.

Plans for Future Work

Additional X-ray diffraction studies are being performed on several of the alloys irradiated in the BRR.

The MTR capsule will be discharged from the reactor at the end of Cycle 161 and shipped to the Battelle Hot-Cell Facility for postirradiation examination. Present plans call for examination to begin during the first week in October.

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 fabricational variables on the radiation stability of uranium monocarbide at high-irradiation temperatures and high fission-heat production rates.

Program Approach

Five uranium monocarbide (UC) specimens will be irradiated in each of three instrumented capsules as part of the United Nuclear Corporation fuel-cycle program. These specimens will be fabricated at United Nuclear by both arc-casting and powder-metallurgy techniques. The carbon content of the specimens will include materials of both above and below the stoichiometric composition as well as stoichiometric material. The specimens in all cases will contain uranium of 12 per cent enrichment and will be 3/8 in. in diameter by 3/4 in. long.

The anticipated irradiation conditions for these specimens will include surface temperatures of 750 C with a center-line temperature of 1250 C. The target irradiation exposure is 15,000 MWD/T of the total uranium.

Progress Reported Previously

The basic design of the irradiation capsules has been established and was described in a recent progress report, BMI-1524. The design was based on irradiation in the ETR. Experimental heat-transfer studies of the capsules were planned which would supply information necessary to establish dimensional tolerances in the finned region between the inner and outer shells of the capsule.

Work This Period

Design studies of the irradiation capsule were essentially completed, and procurement of materials and fabrication of components were initiated. Assembly of a mock-up capsule for heat-transfer studies was started.

Plans for Future Work

Experimental heat-transfer studies pertinent to the capsule design will be completed. As soon as specimens are received, the capsules will be loaded and irradiated.

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 uranium oxides under oxidizing conditions. X-ray diffraction studies will be performed subsequent to some of the transpiration runs in an attempt to determine 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

The program was initiated during this month; however, research will be strongly influenced by results obtained from volatility experiments performed during the past year. The basic experimental apparatus is available immediately and radiochemical analysis techniques are well established.

Work This Period

No results have been obtained during this month.

Plans for Future Work

Slight modifications pertaining to temperature control and heat losses will be undertaken. It is expected that vapor-pressure determinations will commence during the early part of the next month.

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

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

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 are being 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 will contain six specimens, two each of the three materials indicated above, and will be 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 2350 F for the UO_2-chromium. The two capsules will 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 design and fabrication of the capsule components were completed; thermal parameters for the irradiations were summarized in a recent progress report, BMI-1514. The design engineering analysis for the proposed irradiations was completed and sent to the Materials Testing Reactor. Assembly of the two capsules, identified as BMI-39-1 and BMI-39-2, was begun.

Work This Period

Assembly of the second capsule, BMI-39-2, was completed. Capsule BMI-39-1 was scheduled into the MTR for Cycle 162 on September 4, 1961.

Plans for Future Work

Capsule BMI-39-2 will be sent to the MTR early in September.

Fabrication of Cermet Fuel Elements

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

Objective

To evaluate compatibility of UC with promising matrix materials and to develop fabrication techniques for producing UC cermets containing a minimum fuel loading of 60 volume per cent to densities of 90 per cent of theoretical or greater.

Program Approach

UC metal couples are being fabricated by hot isostatic pressing to insure intimate contact for heat treatments for determining metal ceramic compatibility. In addition, physical-property data are being obtained on UN cermets for a direct comparison with UO_2 cermets.

Progress Reported Previously

Emphasis last fiscal year was directed toward the development of fabrication techniques for preparing dense UN cermets and obtaining metal-UN and -UC compatibility. Compatibility data for UN and UC with molybdenum, niobium, rhenium, tantalum, and tungsten as well as the thermal conductivity of an 80 volume per cent UN-molybdenum cermet have been presented in previous progress reports.

Work This Period

Thermal-conductivity values on an 80 volume per cent UN-chromium cermet have been measured to 470 C. The cermet was prepared by blending minus 100-mesh UN with minus 325-mesh chromium powder, cold compacting the blend at 30 tsi, and isostatically hot pressing the cold-compacted cores at 10,000 psi for 3 hr at 2500 F. The fabricated specimen was 0.4195 in. in diameter by 1.46 in. in length, having a theoretical density of 89.5 per cent. Interpolated thermal-conductivity values were 0.163, 0.169, 0.176, and 0.182 w/(cm)(C) at 200, 300, 400, and 500 C, respectively. Interpolated electrical resistivity measured concurrently with thermal-conductivity measurements were, respectively, 149, 161, 172, and 183 microhm-cm. Since the interface bonding the sample to the thermal conductivity standard completely fractured at 470 C, the values reported are questionable and should only be considered as tentative data.

Plans for Future Work

An additional 80 volume per cent UN-chromium cermet is being prepared for thermal-conductivity measurement. Also, 80 volume per cent UN-molybdenum and UN-chromium cermets are being fabricated for room-temperature bend and compressive testing. In addition, UC-chromium, UC-molybdenum, and UC-vanadium couples are being prepared for compatibility testing at tentative temperatures of 2200, 2400, 2600, 2800, and 3000 F. UC containing 4.8 and 5.2 w/o carbon is being used for comparison.

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

Two approaches are under current investigation for fabricating low-cost tubular fuel elements. One of these involves the assembly of concentric core and cladding components followed by densification, bonding, and sizing by techniques such as explosive forming and pressure bonding. Various loading techniques have been considered for incorporation of the core powder material into the annulus formed by two concentric stainless steel cladding tubes.

In another approach, multicored fuel plates are fabricated in a conventional manner with each core running the length of the plates. Subsequent to rolling, two such plates are lined up core-for-core and joined together by seam welding in such a manner that the core areas between the welds can be expanded to form tubular shapes. The core area from each plate then makes up half of the circumference of the expanded tube. Expansion of the individual tubes is accomplished by mechanical means in a drawbench operation.

Progress Reported Previously

Several techniques for loading loose powders into the annulus formed by concentric cladding tubes have been investigated. Highest as-loaded densities (60 per cent of theoretical) were obtained by thermal cycling the tubes during loading.

Explosive forming appears to be the most promising consolidation technique to produce tubes which have the most uniform dimensions. A rather elaborate backup die assembly with provision for evacuation was found necessary to eliminate cladding-die welding problems when forming the tubes. Mild-steel sleeves are used on the inside of the fuel tubes to more uniformly size the inside surface of the fuel tubes and produce smooth inside surfaces. The midl-steel sleeve adheres to the fuel tube after detonation and is removed by pickling in a solution of 1:1 nitric acid and water. The fuel tubes are then gas-pressure bonded to enhance core-cladding bonding and to sinter the compacted fuel annulus.

Initial seam-welding studies using stainless steel flat plates indicated the need for a more rigid fixture to clamp the plates during welding. Such clamping is necessary to prevent warpage and to accurately position the seam welds along the edge of the fuel cores. Expansion of the individual tubular sections for some of the welded assemblies indicated that the spacings between welds were not uniform. Larger mandrels than were anticipated were required to fully expand some of the tube sections.

Work This Period

Two sets of fuel plates were fabricated at 2200 F out of a hydrogen atmosphere to a thickness of 0.035 in. The plates were then annealed at 2200 F and air cooled. Surface oxidation was removed by pickling in a bright acid solution. The plates were then cold rolled to 0.027 in. to flatten them and straighten the cores. The cores in each plate were located and two plates for each set were aligned so that the cores in one were matched to the cores in the other. The two-plate composites were first tacked together to maintain their alignment and them seam welded with the edge of the welds placed as close to the core edges as possible. Some difficulty was found in positioning the welds and it is believed that the plastic jig used to clamp the plates may not be sufficiently rigid to properly guide the seam rollers. Very little warpage was noticed in the composites after welding.

Consideration has been given to a larger backup die to be used in explosive-forming studies for producing tubular fuel elements 2 ft long for subsequent irradiation tests. Although the general design will be similar to that of the split die already in use, the larger die assembly can be more readily evacuated through the use of rubber O-ring seals. This will eliminate the need for gooch tubing for evacuation purposes.

An annularly loaded fuel tube that had been explosively compacted was gas-pressure bonded at 2100 F and 10,000 psi for 3 hr. No change in dimensions was noted for the tube after bonding. Some oxide formation was present on the stainless steel cladding tubes, but the surfaces were free from wrinkling.

Plans for Future Work

Expansion of the two sets of seam-welded fuel-plate composites will be accomplished to evaluate more closely the placement of the welds with respect to the fuel cores.

No additional fuel-loading studies are planned as it is believed that satisfactory loadings can be made by pouring loose powders and using thermal cycling to displace entrapped air. Explosive forming appears to offer the most promising means of densifying the fuel annulus and maintaining dimensional control over a long length of tube. Several problems must be worked out in the larger backup die design which will allow for improved surfaces on the compacted fuel tubes. It is believed that the larger backup die assembly now being designed should be utilized in further consolidation studies.

Factors Affecting Pressure Bonding

D. C. Carmichael, G. W. Cunningham, 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 prepare 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. In order to produce a metallurgically sound bond, i.e., no discontinuities or changes in the structure in the bond region, the first requirement is the application of sufficient pressure to place the surfaces in intimate contact. For copper, it was found that the pressure required could be related directly to Meyer hot-hardness values.

Grain growth across the bond interface and thus elimination of the interface has been found to be dependent upon the presence or absence of microvoids in the bond region. Microvoids can be created during bonding by insufficient flow of metal or by condensation of excess vacancies at impurity nuclei. Decreasing the number and size of microvoids to a level permitting grain growth depends upon the availability of vacancy sinks in the metal. In copper, the most effective sinks were found to be grain boundaries.

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.

Work This Period

Experiments are being conducted to study further the relationships determined using copper among the hot hardness at a given temperature, the applied load in hot pressing, and the resulting area of contact between mating surfaces. For this study, platelets of pure nickel which have surfaces shaped to a roughness of 500 μ in. rms are being hot pressed into annealed copper platelets ground to a surface roughness of 8 μ in. rms. As discussed in BMI-1518, the results obtained by pressing the hard nickel asperities into soft copper are necessary to confirm the explanation which has been made of the results obtained by pressing copper into copper. Nickel-copper specimens were vacuum hot pressed at 1000 F using pressures of 2,000, 4,000, 8,000, and 12,000 psi applied for

5 min. Metallographic examination of these couples has now been completed, and the fractional area of contact obtained in each couple has been measured using a Hurlbut counter. The results obtained are given below:

Applied Pressure, psi	Resulting Fractional Area of Contact
2,000	0.18
4,000	0.30
8,000	0.55
12,000	0.82

These results at 2,000, 4,000, and 8,000 psi are in close agreement with those previously obtained at these pressures using copper-to-copper couples. These values show a straight-line relationship on a graph of applied pressure versus area of contact. The contact area observed at 12,000 psi in the copper-nickel couple falls on the same straight line, whereas the contact area produced in a copper-copper couple at the same pressure deviated from the straight-line relationship on the graph. Another copper-nickel couple is currently being hot pressed at these same conditions (12,000 psi) to check this result, however, because some possible contamination was observed in the bond interface which could have affected the area-of-contact measurement.

Metallographic examination of the mode deformation in the copper-nickel specimens indicates that the deformation occurred by penetration of the wedge-shaped nickel asperities into the nickel. It appears that very little, if any, deformation of the nickel asperities occurred which is as expected since the nickel is much harder than the copper. In the copper-copper couples, it had been observed that considerable deformation of the asperities had occurred, which complicated the analytical comparison to classical pressure-contact area relationships.

The data obtained for the copper-nickel specimens will not be examined in terms of these relationships, especially the Hill-Lee-Tupper* treatment. To derive their relationship, they assumed that the material is not work hardened, that the included angle at the apex of the wedge remains constant, and that there is no friction or adhesion between the wedge and the metal surface being penetrated. For this purpose, the included angle of the wedge-shaped asperities in the copper-nickel couples bonded at each of the four pressures is being measured to determine if any change occurred during the deformation process. It may also be found that it will be useful to bond additional copper-nickel specimens in which the nickel asperities are more perfectly wedge-shaped than they are in the shaped surfaces.

Plans for Future Work

The study of the bonding of the copper-nickel specimens will be continued to investigate the deformation relationships involved. The investigation of the formation and subsequent elimination of the bond interface in aluminum couples will be continued later.

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* Hill, R., Lee, E. H., and Tupper, S. J., "The Theory of Wedge Indentation of Ductile Materials", Proc. Roy. Soc. (Londo A188, 273 (1947).

GAS-PRESSURE BONDING OF CERAMIC FUEL ELEMENTS (AEC-FUEL CYCLE)

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 UO_2 sheet material as supplied by Gladding, McBean & Co.

Program Approach

This program is being carried out in support of the AEC Fuel-Cycle Development Program. The present program will concern three areas of research. The first area involves the fabrication of UO_2 fuel rods by direct pressure bonding for irradiation testing in the VBWR. The remaining two efforts, which will be carried out simultaneously, represent cooperative programs with Hanford and Gladding, McBean & Co. In these programs, the gas-pressure-bonding process as combined with the vibratory packing techniques and the UO_2 sheet-rolling techniques will be evaluated.

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 of work on 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 UO_2 fuel-element shapes were partially developed. A report of the fabrication of fuel rods for irradiation testing in the VBWR is being drafted.

Work This Period

Four full-scale pressure-bonded VBWR fuel rods have been completed. These rods will be sent to the Atomic Products Division of the General Electric Company for irradiation testing in the VBWR. These bonded assemblies, using the alternate ceramic grade UO_2, demonstrated a greater degree of surface roughness and axial misalignment than experienced previously. A swage sizing operation followed by a second pressure-bonding cycle was found to improve the surface and straightness of these rods.

Gladding, McBean & Co. is presently encountering difficulties with rolling and platelet-punching techniques. As a result, it is estimated that additional UO_2 platelets will not be available for Battelle study until November, 1961. Specimens have been prepared from the 17 UO_2 platelets previously supplied by Gladding, McBean & Co. for tentative evaluation. Tests are being performed upon these specimens to establish the effects of the gas-pressure-bonding operation on the density, stoichiometry, and general structure of the UO_2 platelets.

A cooperative vibration packing-pressure bonding program has been established by Battelle and Hanford. Agreement as to the type, size, and estimated number of specimens to be processed was made. It is expected that the initial packed assemblies will be available for pressure bonding in approximately 1 month.

Plans for Future Work

The evaluation of the initial Gladding, McBean & Co. UO_2 platelets will be completed.

DEVELOPMENT OF URANIUM CARBIDE (AEC-FUEL CYCLE)

F. A. Rough and W. Chubb

Uranium carbides and their alloys are being developed for the AEC Division of Reactor Development as a part of the Fuel-Cycle Development Program.

Melting and Casting Techniques for Uranium-Carbon Alloys

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

Objective

To develop and evaluate techniques for the large-scale production of high-quality cast shapes of uranium carbide.

Program Approach

Skull-type arc-melting techniques are being applied to the melting and casting of uranium carbide. The effects of process variables such as the quality of the charge material, furnace atmosphere, arc current and voltage, and the grades of graphite used for the electrode tip are being investigated in the interest of producing high-quality castings with good compositional control. Evaluation of composition and homogeneity are based upon chemical analyses supplemented by metallographic examinations. Casting quality is determined by radiography and macroscopic examination of the casting surfaces.

Progress Reported Previously

Casting research covering the first phases of this program is summarized in two phase reports, BMI-1370 and BMI-1488. Research in the several months since the issuance of BMI-1488 has dealt with the effect of vacuum-outgassed commercial-grade graphite (used both as a charge addition and as an electrode tip) upon the control of the composition of the product. It was found that five castings produced using outgassed graphite varied less than ±0.1 w/o carbon while castings produced from the same grade of graphite that had not been outgassed varied by ±0.3 w/o carbon.

Variations in carbon content in successive melts have been attributed to decreasing contributions of carbon from the starting skull due to segregation within the skull. The variations within the skull can be minimized through the use of prealloyed UC in the preparation of the skull. Variations in carbon content of successive melts from this skull can then be held within very narrow limits through the use of prealloyed UC plus an adequate quantity of elemental uranium to compensate for carbon pickup by electrode-tip erosion.

Work This Period

The emphasis during the past month has been directed toward completing certain skull-melting-furnace modifications necessary for the continuation of the program. These alterations will permit the furnace operator to employ various charging and melting procedures during melting to gain better compositional control of the cast product. One of the important variables that will be investigated is the uranium charging rate, since previous experience has shown that unalloyed uranium tends to segregate in the bottom of the crucible and results in carbon contents higher than anticipated in the castings. It is believed that this can be alleviated to a great extent by charging the uranium in small increments during melting, thus permitting each increment of uranium to be alloyed before additional uranium is introduced.

These alterations have been completed and the equipment is being evaluated in regards to the reliability of the components and the leak rate of the system. Minor changes in the equipment are now in progress.

Plans for Future Work

The modified skull-melting facility will be employed to investigate various charging and melting procedures for preparing the skull and for the production of castings. The optimum graphite electrode diameter will be determined prior to the production of cast shapes. The electrode performance will be evaluated on the basis of arc stability, electrode erosion, and the control of the molten pool attainable. After optimizing the electrode diameter, a series of melts will be made for the purposes of establishing melting parameters and establishing the electrode consumption rate. Of particular importance will be the ability of the operator to compensate for electrode erosion by the periodic addition of elemental uranium. A final series of melts will then be made employing the optimized electrode and uranium feed rate to ascertain the homogeneity and control attainable when building a new skull and in producing castings. The deposited skulls and the castings will be evaluated in terms of homogeneity and soundness as determined by metallographic techniques, radiographic examinations, and chemical analyses.

Effects of Radiation on UC

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

Objective

To study and understand the important mechanisms of damage occurring in uranium monocarbide as a result of irradiation.

Program Approach

Specimens of uranium monocarbide (UC), uranium sesquicarbide (U_2C_3), and uranium dicarbide (UC_2) have been irradiated in the Battelle Research Reactor at surface temperatures ranging from 500 F or less to 1600 F and to target burnups ranging from 0.01 a/o to 0.7 a/o of the uranium. The specimens were contained in six NaK-filled capsules. Evaluation of these irradiation specimens is based on their dimensional stability and microstructural stability and on measurements of their electrical resistivities and lattice parameters.

Progress Reported Previously

The irradiation of six capsules has been completed. The postirradiation examinations of the specimens recovered from four of these capsules have been completed, and the results were presented in a previous progress report, BMI-1524. In general, it was found that a saturation of lattice strain occurred during irradiation exposures to 0.01 a/o burnup of the uranium at temperatures of less than 500 F. This saturation of lattice strain was accompanied by initial large increases in the electrical resistivity which reached a limit after a burnup of 0.01 a/o of the uranium. The one high-temperature capsule in this series of four failed during irradiation.

Examination of specimens irradiated in Capsules 5 and 6 has been initiated. These specimens were exposed to target uranium burnups of about 0.7 a/o and experienced average surface temperatures ranging from 1100 to 1400 F. Two centrally positioned specimens of uranium-8.5 w/o carbon alloy, essentially UC_2, in Capsule 5 were completely disintegrated, and one of two specimens of uranium-6.7 w/o carbon alloy, essentially U_2C_3, was partially broken. The remaining specimens, all uranium-5.0 w/o carbon alloy, from both Capsules 5 and 6 were in excellent condition with no apparent evidence of cracking or deterioration.

The physical measurements of the unaffected uranium-5.0 w/o carbon specimens of Capsules 5 and 6 showed negligible diametral changes. Density decreases in these specimens were in the order of 1 per cent or less. The one specimen of uranium-6.7 w/o carbon, U_2C_3 + UC, which appeared unaffected showed a diametral decrease of about 1 per cent and a density increase of 1.34 per cent.

Work This Period

The postirradiation examinations of the specimens from Capsules 5 and 6 are in progress. The isotopic burnup analyses of representative specimens from both capsules have shown that the specimens sustained a uranium burnup of about 0.5 a/o. Analysis of the gases collected from Capsule 5 indicated that about 2 per cent of the total fission gas produced during irradiation was released from the specimens. However, two specimens of this capsule were totally disintegrated. Analysis of the gases collected from Capsule 6 indicated a release of only 0.32 per cent of the fission gas, or about 2.5 times that which would be expected from recoil.

The microstructural studies of the uranium-5.0 w/o carbon specimens from these capsules revealed several types of behavior during irradiation. In most instances a depletion of the UC_2 second phase was noted in a band around the outer circumference of each specimen. This condition may have been caused by diffusion of carbon into the niobium tube which contained them. In one specimen which had been heat treated prior to irradiation for stress relief, a second phase of U_2C_3 was noted at the grain boundaries and the grains were nearly equiaxed rather than columnar, typical of cast specimens. The preirradiation microstructure of control specimens which had been identically heat treated showed columnar grains and UC_2 second phase; it is suspected that the observed microstructure may have resulted from the irradiation. Another specimen of uranium-5.0 w/o carbon alloy showed a complete disappearance of the UC_2 second phase. Although this condition may represent an irradiation effect, it might also have been a result of insufficient grinding during the metallographic preparation which exposed only a section across the carbon-depleted band at the surface of the specimens, as noted earlier.

In attempting to replicate the metallographic surfaces of these specimens for electron microscopic studies, it was found that extreme pullout resulted. Since the replicas must not be contaminated, this phase of the structural studies has been temporarily recessed.

Plans for Future Work

The X-ray diffraction studies will be completed, and the electron microscopic studies reattempted. The compatibility of UC_2 in NaK at temperatures near 1600 F is to be studied briefly in an effort to isolate the source of the failure during irradiation of the uranium-8.5 w/o carbon specimens. Final evaluation of the effects of irradiation on the carbide specimens of this program will then be completed.

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. Various high-temperature sources are being evaluated to melt UO_2 (melting point about 2760 C) and to control its solidification to form large single crystals.

Progress Reported Previously

The following fusion methods have been investigated:

(1) Electron-beam melting
(2) Resistance heating
(3) Arc melting
(4) Plasma-jet impingement
(5) Induction heating
(6) Radiation heating from resistance element.

Most of the experimental program has employed the last method. The resistance furnace developed, using a tungsten heating element, can now reach and maintain a temperature of 2800 C. This temperature is thought sufficient to melt UO_2 in the capped tungsten crucible employed for Bridgman experiments in this furnace.

Work This Period

During August, Bridgman drop experiments were continued. In one run, good operating conditions were maintained throughout the experiment. The charge was initially held at 2500 C for 30 min to reach steady-state temperature. The temperature was then raised to about 2820 C and maintained for 1 hr. During this time, the charge was dropped through the furnace at 2 in. per hr. After the drop, the charge was cooled in a controlled manner, over a 2-hr period, to prevent thermal shock which might cause fracture of the crystals.

Observation of the charge showed that melting had not occurred throughout, and only the outer 1/8 in. showed fusion. In addition, a few cracks were found running transgranularly through the crystals. The crystals were about 1/16 to 1/8 in. in size. The cracks probably resulted from uneven cooling between the fused outer layer and the sintered inner core.

It is believed that the entire charge did not melt because either the drop was too fast or the temperature too low. In another experiment, a lower drop rate, 1 in. per hr, was employed at about the same temperature, 2830 C. Incomplete melting was also obtained in this case.

Plans for Future Work

A higher temperature will be employed in future Bridgman experiments. In addition, calibration of sight ports will be employed prior to each experiment to obtain more accurate temperature readings. Precautions have been taken to avoid clouding of the sight port by lengthening its distance from the hot zone, and by cooling its perimeter to condense any vapors present before they can reach the glass surface.

Work will be continued in an effort to improve the temperature capability of the furnace. Heavier and stronger heating elements are being constructed to replace the thin fragile ones now being used.

RADIOISOTOPE AND RADIATION APPLICATIONS (AEC-OID)

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 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 some chemical element (e.g., manganese) while varying such parameters as activation volume, geometry, holding time, etc. After determining optimum conditions, experiments will be performed to determine general stream activation efficiencies for different process streams as a function of neutron-source strength. The concept of in-process neutron activation will then be tested on several processes using simulated plant conditions.

Progress Reported Previously

The first phase of the over-all intrinsic radiotracer program demonstrated the technical feasibility of using intrinsic radiotracers as process-control tools. This phase was completed in June.

The present phase of the intrinsic radiotracer program began July 1. The initial survey of neutron sources and source technology indicated that a polonium-beryllium α-n source would best meet the program requirements. It was decided, therefore, to obtain a 10-curie polonium-beryllium source for preliminary testing and optimization experiments, and to obtain a higher intensity polonium-beryllium source later in the program for the process-simulation experiments.

Work This Period

The principal effort this month has centered on the design and construction of experimental equipment for the neutron-activation studies. Design of a versatile source storage shield and holder for the neutron-activation cell has been completed. This equipment is presently under construction.

A 10-curie polonium-beryllium α-n source has been ordered from Mound Laboratory, Miamisburg, Ohio, with delivery expected in September.

Some time has been spent this month on the survey of efficient beta-counting techniques. On the basis of this study it was decided to construct a simple multiprobed Geiger counter for use in the experimental program. The components for this detection system will be assembled next month.

Plans for Future Work

Work next month will include completion of the neutron-source storage and activation equipment, and a series of cold experimental runs to develop operating techniques.

Preliminary experimental work will begin when the source arrives at Battelle.

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 resulting data suggests that (1) free-radical formation is accomplished by means of hydrogen scission or complete or partial ester scission, (2) the size and configuration of the hydrocarbon tail of the ester group influence the efficiency of site formation, (3) site formation is accompanied, in most cases, by a scission on the polymer backbone, and (4) the amount of graft copolymer is influenced by polymer structure as well as by the concentration of available free-radical sites. More recently, the work has been extended to other polymer systems.

Work This Period

During August, experimental work included: (1) continuation of the grafting studies, (2) measurement of free-radical concentration as a function of dose for poly-n-hexylmethacrylate, (3) initiation of a study of the chemical measurement of free-radical sites using the diphenyl picrylhydrazil (DPPH)-hydroquinone method, (4) investigation of procedural techniques employed in the analytical methods (mass spectrometry and gas chromatography) used for the determination of volatile product concentrations from irradiated polymethacrylates, and (5) preparation of several additional vinyl polymers.

In all grafting experiments completed to date, polymethylmethacrylate has been used as base polymer and vinyl pyrrolidone as graft monomer. Additional grafts have been performed for the purpose of establishing the correlation between concentration of grafted polymer and total free-radical sites. Further, experiments designed to establish the effect on total grafting of contact time between polymer and graft monomer have been completed. In this work, the water-insoluble copolymers were collected and washed with methanol. Residual monomer was removed from each copolymer by extraction with water for 24 hr in a Sohxlet. Following extraction, the material was dried in vacuo at 70 C. Nitrogen concentrations and number-average molecular weights will be determined for each copolymer. These data should be available for inclusion in the September report.

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*I.A.E.A. Fellow.

Site concentration has been determined as a function of dose for poly-n-hexylmethacrylate using EPR measurements (Table H-1). The results are in agreement with those which would be predicted from an examination of the sites-versus-dose curves for the C_1 through C_5 unbranched methacrylate polymers. Comparison of the EPR data obtained for the n-hexylmethacrylate polymer with those obtained for the cyclohexyl- and phenylmethacrylate polymers indicates, interestingly, that fewer stable sites exist in the former polymer. This is probably due to increased molecular mobility, which is suggested by the rubberlike nature of this material.

Several preliminary experiments have been completed in which free-radical concentrations have been determined chemically using the DPPH-hydroquinone method. For PMMA irradiated to a dose of 6×10^6 rads, EPR measurements indicate a site concentration of approximately 4×10^{-3} site per monomer unit. Values of 1.5×10^{-3} and 0.5×10^{-3} site per monomer unit have been obtained from duplicate chemical measurements on the same material. The comparative measurements will be repeated at several lower doses in order to: (1) establish the sensitivity and reproducibility of the chemical method, and (2) determine if concentrations measured by the two methods are, perhaps, related by some factor which might provide insight into polymer conformation.

During the month, the investigation of procedural techniques for determining volatile product concentrations from irradiated polymethacrylates was continued. Absorption and/or solubilization of the low-molecular-weight fragments by the polymer has rendered questionable some of the data obtained from mass spectrometry and gas chromatography, particularly in the case of the terbutylmethacrylate polymer. It is anticipated that the necessary procedural changes can be completed early in September.

Several additional polymers have been prepared by irradiation methods. These include, in addition to the n-hexylmethacrylate polymer described above, polyethyl dimethylaminomethacrylate and polyacrylonitrile. These polymers will be examined in a later phase of the program.

Plans for Future Work

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

TABLE H-1. SITES VERSUS DOSE FOR POLY-N-HEXYLMETHACRYLATE

Dose, rads	Sites Per Monomer Unit x 10^3(a)
2×10^5	(b)
5×10^5	(b)
1×10^6	0.006
3.2×10^6	0.054
4.8×10^6	0.016
9.6×10^6	(b)

(a) By EPR measurements.

(b) Total site concentration below sensitivity of instrument.

MATERIALS DEVELOPMENT AND EVALUATION (HAPO)

F. R. Shober

Research and development work are 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.

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 0.1 μ 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 temperature-cycle tests, the temperature is allowed to drop normally to room temperature 1 day per week and then increased back to test temperature in about 2 hr. In the tests involving stress and temperature cycles, the stress is removed first and then the temperature is lowered to room temperature 1 day per week. The stress is reapplied after the specimen has been returned to test temperature.

Progress Reported Previously

All 20 tests now in progress have been in progress for times varying from about 3000 to 33,000 hr; therefore, accumulated data have been reported from time to time. The latest summary was presented in the April progress report (BMI-1514). 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

The 20 tests reported in progress as of the end of July were continued through this period. No additional tests have started or discontinued. Creep data were accumulated for later interpretation and analysis.

All temperature-cycle tests have been concluded. These tests were made on annealed and 15 per cent cold-rolled Zircaloy-2 sheet at 550 and 650 F. Indications are that temperature cycling accelerates creep as compared with static (no cycle) tests and also with the tests involving combined stress and temperature cycles.

Plans for Future Work

The 20 tests in progress are scheduled to continue. Creep data will be accumulated until it is evident that continuing a test will no longer contribute useful data. As tests are discontinued, new tests will be started on specimens taken from the PRTR or the NPR pressure tubes.

Thermal Conductivity and Electrical Conductivity of UO_2

H. W. Deem and C. F. Lucks

There has been no new experimental work undertaken this month. A summary of the previous work is being prepared.

Materials of Construction for the Fused Chloride-Electrolytic UO_2 Process

E. L. White, P. D. Miller, D. C. Drennan, C. H. Lund,
H. J. Wagner, A. M. Hall, and W. K. Boyd

Objective

To develop a metallic container material for the HAPO fused chloride-electrolytic fuel-recovery process.

Program Approach

Several commercially available metals and alloys were screened as candidate materials of construction by using a bench-scale apparatus employing chlorine to produce uranyl chloride from uranium oxide. Experimental iron- and nickel-base alloys, designed for this application on the basis of the results of the screening program, were evaluated by weight changes during exposure to a 750 to 800 C equimolar solution of sodium and potassium chlorides containing U_3O_8 and sparged with chlorine.

Progress Reported Previously

Results in the program have been summarized in several past progress reports, BMI-1504, BMI-1514, BMI-1518, and BMI-1524. The work has shown that experimental alloys based on the nickel-aluminum system and the nickel-aluminum-silicon system have superior corrosion resistance for the environment contemplated. Corrosion rates in the liquid were less than 100 mils per month. Attack at the gas-liquid interface was somewhat greater.

Work This Period

A topical report covering the preparation, corrosion properties, and metallographic studies of the alloys is being prepared.

Plans for Future Work

The topical report will be completed.

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 PROGRAM

Uncoated 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

A lot of fully enriched UC_2 powder was found to contain 9.45 w/o total carbon and 1.62 w/o free carbon. In appearance, the particles were found to be spherical with a 4-mil rim of graphite and some graphite in the grain boundaries. The UC_2 was the major phase, and there were small amounts of UC present. Results of X-ray diffraction verified the presence of graphite, UC_2 (as the major phase), and UC. No agglomeration or sticking of particles was detected.

Work This Period

A quantitative estimate of the deviation of particle shapes from perfectly round spheres has been obtained by examining the shape of individual particles and assigning the appropriate classification to each individual particle. The results, which were obtained on a 5500-particle sample, are presented in Table L-1. A total of 90.4 per cent of the particles studied was in the classifications corresponding most closely to perfectly round spheres. This compares with 35 per cent of enriched MCW UO_2, batch UO_2-E3, as was reported in BMI-1518. The amount of agglomeration of small segments to normal particles in UC_2-E2 was negligible, 0.1 per cent.

Plans for Future Work

Evaluation of pycnometer, sedimentation, petrographic, and Coulter counting devices will continue in an effort to obtain particle-density values to a high degree of accuracy for both porous and dense particles. Fuel materials of UC, UC_2, and UO_2 have been ordered and will be evaluated as received.

TABLE L-1. QUANTITATIVE DESCRIPTION OF PARTICLES OF FULLY ENRICHED 3M URANIUM CARBIDE, BATCH UC_2-E2

Type of Defect	Occurrence, per cent
Agglomeration of segments(a)	0.1
Deviation from sphericity, 2:1 or more(b)	0.1
Deviation from sphericity, 1.2:1 to 2:1	0.7
Deviation from sphericity, 1.2:1 or less	5.7
Deviation from roundness, $R<0.3$(c)	0.3
Deviation from roundness, R 0.3-0.7	8.4
Deviation from roundness, $R>0.7$	84.7
	100.0

(a) A segment is attached to a normal particle.

(b) Ratio refers to ratio of major to minor axis of ellipsoidlike particles.

(c) Refers to egg-shaped particles where R = ratio of radius at highly curved end to that at less curved end.

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

A general summary of the experimental work on Al_2O_3-coated particles prepared prior to this program was given in a topical report, BMI-1471. Modifications of the fluidized-bed reactor design and water-vaporization system have improved process control and have extended the operating time to permit deposition of the desired coating thickness without interruption. 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 (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 (Xe^{133} release: 1.3 ppm at 1300 C).

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.

Work This Period

All effort this period has been devoted to development of the technique and equipment for preparing coatings at temperatures above 1100 C and to the evaluation of these coatings. The purpose of studying higher coating temperatures is to increase the useful temperature range for Al_2O_3-coated fuel particles by minimizing the effects of differential thermal expansion. In addition, there is some evidence that higher coating temperatures result in smoother coatings.

Two lots of Al_2O_3-coated depleted UO_2 were prepared at 1400 C in a Mullite reactor. The first, Lot 723B, was coated with approximately 27 μ of Al_2O_3 by steam hydrolysis of Al_2Cl_6, and the second, Lot 724B, was coated with approximately 33 μ of Al_2O_3 by the reaction of Al_2Cl_6 with CO_2 and hydrogen. Both coatings appeared to be essentially transparent, dense, and free of voids. Lot 724B was slightly more transparent. The crushing strength of these coatings was approximately 50 per cent of the crushing strength of a typical 1000 C coating of the same thickness. However, the hardness was about equal to the hardness of the 1000 C coatings and of sapphire. Neither coating had surface protrusions.

In the sectioned and as-polished condition, both 1400 C coatings appeared to consist of columnar grains with the grain boundaries extending from the substrate to the coating surface. Most of the particles in Lot 723B failed when heated at 1400 C for 48 hr in an oxygen atmosphere, but were essentially unaffected by heating at 1400 C for 48 hr in hydrogen as determined by a subsequent 48-hr oxidation exposure at 1000 C and an acid leach test. It therefore appears that excessive oxygen diffusion at 1400 C caused the failure. Lot 724B was unaffected by oxidation testing at 1400 C for 48 hr. The evaluation of Lot 724B has not been completed. However, if the evaluation results continue to be favorable, similar coatings on enriched UO_2 could be prepared for in-pile study by the end of September.

Coating contamination could be a significant factor in the 1400 C coated material. It was found that the coating of Lot 723B contained 6000 ppm silicon while the coating of Lot 724B contained only 1000 ppm. This variation in coating contamination could have resulted from the variation in coating procedure, or from the use of a new Mullite reactor for preparing Lot 723B. Lot 724B was prepared in a reactor which had been used previously and hence the reactor may have already been at least partially leached free of impurities.

In exploring the preparation of coatings at intermediate temperatures, Lot 725D was prepared with enriched UO_2-E3 as a substrate at a coating temperature of 1225 C in a quartz reactor. The coating was intentionally deposited in five relatively short runs in an effort to decrease the nonuniformity of coating thickness encountered in the two-step process used previously. Extensive adherence of particles to the reactor walls was observed at every step and it is questionable whether coating uniformity was improved. It is suspected that operating a quartz reactor near its upper temperature limit may have contributed to the problem. When the coated particles were sectioned all layers of the coating were discernible, apparently transparent, dense, and free of voids. The particles were free of large surface protrusions. The sample should be ready for in-pile study in September, provided that evaluation tests now in progress continue to be satisfactory.

Thin-section mounts for petrographic examination of 500, 700, 900, 1000, and 1100 C coatings, as well as various heat-treated samples and one highly contaminated with silicon, have been procured and are now being examined in an effort to better understand the crystal morphology of the Al_2O_3 coatings.

Plans for Future Work

Depending on the results of evaluations now in progress, 1400 and 1225 C coatings will be prepared for in-pile study. In addition, a 1000 C coating will be made in multiple steps as a reference material for the in-pile study of samples prepared at various coating temperatures.

Further studies will be made of factors relating to the incidence of surface protrusions. In this connection, the petrographic examination of thin sections of various coatings will be continued.

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 tumbling-bed reactor or 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

In a previous program, pyrolytic-carbon coatings were applied to UC, UC_2 and UO_2 by the pyrolysis of hydrocarbons in a fluidized bed of the powders at temperatures up to 1500 C. Promising coatings were obtained, but there was some indication of mechanical failure of the coatings at temperatures above the coating temperature.

In the present program, the fluidized-bed reactor design for coating temperatures under 1500 C was altered to eliminate the occurrence of poorly coated particles due to inadequate fluidization. Carbon-coated particles were prepared which exhibited good fission-gas-retention characteristics above the coating temperature.

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

Lot 918A, columnar coating on enriched UC_2-El (Xe^{133} release: 2. 7 ppm at 1300 C)

Lot 919A, laminar coating on enriched UC_2-El (Xe^{133} release: <0. 5 ppm at 1300 C)

Lot 920A, laminar coating on enriched UO_2-E3 (Xe^{133} release: 0. 9 ppm at 1300 C)

Lot 921, laminar-columnar mixture PyC coating on dense Al_2O_3-coated UO_2-E2 (Xe^{133} release: 0. 5 ppm at 1300 C)

Lot 922, laminar-columnar mixture PyC coating on dense:porous Al_2O_3-coated UO_2-E3 (Xe^{133} release: 3. 1 ppm at 1300 C).

Work This Period

An experimental program to relate the coating conditions, temperature, reactant composition, gas velocity, and reactant preheat, with coating properties such as density, structure, and strength is in progress.

During the current month, a high-temperature fluidized-bed reactor developed for another program was made available for the preparation of specimens of PyC-coated powder at temperatures above 1500 C. This reactor consists of a cone-bottomed 3/4-in. -ID graphite tube inductively heated. It is insulated with graphite powder and enclosed in a Vycor envelope purged with inert gas to prevent oxidation of the carbon. Methane was used in these experiments instead of acetylene because of its higher temperature coefficient of reaction. It should have less tendency to decompose at intermediate temperatures and plug the gas inlet to the reactor.

The following lots of high-temperature pyrolytic-carbon coatings were prepared for general evaluation and/or possible in-pile study:

	Lot	Fuel	Average Coating Thickness, μ	Coating Temperature, C
(1)	1001A	Depleted UO_2	65-70	1700
(2)	1004A	UC_2-El	57-65	1800
(3)	1006A	UC_2-El	35-38	1700

These materials were all made in two steps to minimize contamination of the coating of uranium. Coatings of the three lots had columnar structure which showed orientation under polarized light. The coating thickness was more nonuniform than that normally obtained in the low-temperature deposition unit. However, this condition was improved in the later runs by reducing the temperature of the gas-inlet line and increasing the fluidizing gas flow. The crushing strengths of the carbon coatings prepared above 1600 C were approximately the same as those deposited a temperature below 1500 C, and the maximum average coating density determined was 1. 94 g per cm^3 for the 1800 C coating, which is only slightly higher then the 1. 90 g per cm^3 obtained for at least one coating prepared at 1400 C. Sectioned coatings examined microscopically appeared to have carbon soot inclusions which were more numerous the higher the coating temperature. On the basis of limited acid-leach data, the high-temperature carbon coatings appear to be nonporous.

Lots 1004A and 1006A showed surface contaminations of 550 and 1600 ppm, respectively, in alpha assays. To determine whether the contamination was due to exposed uranium or inclusions in the coating, fragments of coating, leached free of core particles, were subjected to alpha assays with similar results. An effort is also being made to determine by autoradiography techniques whether these high alpha counts are the result of a few bad particles or a general coating contamination. It is suspected that diffusion of uranium at the high coating temperatures may be causing contamination of the coatings.

Lot 1001A (65 to 70 μ of 1700 C PyC on UO_2) was found to have a surface contamination of 7.1 ppm by alpha assay. This would suggest that pyrolytic-carbon-coated UO_2 has some promise as a high-temperature fuel particle as long as the carbon shell remains intact.

Considerable effort has been devoted to developing a reliable method of determining the density of coatings on coated particles. This study has been performed with carbon coatings; however, the techniques developed should be applicable to practically any coating which is nonporous relative to the xylene used as pycnometer fluid. The procedures may also be altered to handle more porous coatings by using liquids other than xylene. Of the three density measuring methods which were described in BMI-1534, the system employing a xylene pycnometer measurement of coating fragments prepared by controlled cracking appeared to give the most reliable and reproducible values.

The actual procedure adopted for determining densities consisted of first crushing the coated particles between flat plates separated with shims 50 μ thinner than the diameter of the smallest substrate particle. After acid leaching to remove the substrate, water washing, and drying, flotation in a solution of 2.1 g per cm^3 density was used to separate the clean coating fragments from uncrushed particles. Three tenths of a gram of PyC coating fragments was sufficient for the determination. The substrate-free coating was washed, dried in a vacuum oven, and cooled to room temperature before continuing the determination. The density of the coating was then determined by normal pycnometer techniques except that the xylene was introduced into the pycnometer containing the sample under vacuum. In addition, the various drying, weighing, and handling techniques were evaluated and standardized.

The standard deviation assignable to technique for any series of measurements was demonstrated to be $< \pm 0.003$ g per cm^3. However, variations in characteristics of the same sample from one determination to the next increased the over-all standard deviation of any series of measurements on a given sample to a maximum of ± 0.02 g per cm^3 for determinations made thus far.

The coatings of Lots 918A and 919A which are candidates for in-pile irradiation had densities of 1.73 and 1.54 g per cm^3, respectively. The average densities of the carbon coatings prepared at BMI have been in the range of from 1.46 to 1.94 g per cm^3, depending upon the conditions of preparation.

Densities determined by the above method check within 5 per cent of the results of (1) flotation of coating fragments in solutions of different density and (2) densities calculated from pynometer-measured particle densities and particle analyses. If penetration of connected porosity in the coating by xylene is a problem, it therefore must contribute less than 5 per cent error to the density value.

Plans for Future Work

The experimental program to relate coating conditions and coating properties will be continued. The program should be concluded in the next 2 months.

A lot of enriched UO_2 will be coated with carbon at 1700 C and evaluated as a possible candidate for irradiation evaluation. This should be completed by September 15.

Beryllia-Coating Studies

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

Objective

To develop procedures for coating fuel particles with BeO of optimum quality.

Program Approach

Beryllia is being deposited on UO_2 shot by hydrolysis of $BeCl_2$ in a fluidized bed. The use of other compounds from which BeO can be formed will also be explored. The coatings are being evaluated by microscopic examinations, physical-property determinations, 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

Beryllia coatings have been deposited using essentially the apparatus and techniques previously developed for depositing Al_2O_3 coatings. The BeO coatings (up to 26 μ in thickness) prepared were found to be free of voids and relatively dense, as indicated by acid-leach data. Uniformity of coating thickness from particle to particle was greatly improved over that obtained in exploratory work prior to this program. However, coating quality became inconsistent when an effort was made to alter equipment design and process techniques to permit the deposition of thicker coatings in a continuous operation. Efforts to improve the quality of high-purity BeO coatings by varying the coating conditions have thus far been unsuccessful. In general, only BeO coatings containing impurities appear to be relatively dense and nonporous. All coatings prepared by the oxidation or decomposition of beryllium basic acetate have been contaminated with carbon, contained hydroxy residue, and were soluble in nitric acid.

Work This Period

Efforts to improve the quality of pure BeO coatings prepared by the hydrolysis of $BeCl_2$ were continued. Coatings deposited at 1400 C in a Mullite reactor by steam hydrolysis appear to be translucent, dense, and nonporous. A complete evaluation of this coating is in progress and should be completed in early September.

Exploratory work at 1400 C on the use of a CO_2-hydrogen reaction in place of steam for the hydrolysis of $BeCl_2$ and the use of a method for feeding $BeCl_2$ as a solid directly into the fluidized bed have indicated some promise in improving the quality of the coating. The H_2O:$BeCl_2$ ratio in the coating atmosphere was found to have no significant effect on coating properties within the limits studied (0.5 to 1.5).

Plans for Future Work

Evaluation of the 1400 C coatings will be concluded, and, if the results indicate the coating to have potential as a good fission-gas barrier, a sample for in-pile study will be prepared. The effect on coating properties of the use of the CO_2-hydrogen reaction will be investigated more thoroughly. Decomposition of beryllium basic acetate at 1400 C under mildly oxidizing conditions will also be studied, since the objectionable carbon contamination can thus be minimized and perhaps avoided.

Additional Oxide Coating Studies

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

Objective

To explore briefly the potential of MgO, ZrO_2, Ce_2O_3, and other oxides as coatings for fuel particles.

Program Approach

As the first of a series of oxides to be explored, magnesia is being deposited on UO_2 particles by hydrolysis of MgI_2 vapor in a fluidized bed. The coatings are being evaluated by microscopic examination, measurement of physical properties, alpha count, and measurement of fission-gas release on post-irradiation heating. Promising materials will be evaluated by in-pile irradiation at elevated temperature.

Progress Reported Previously

Experimental work was initiated this period.

Work This Period

A Vycor fluidizing-bed reactor with auxiliary equipment similar to that used for Al_2O_3-coating work was used in the MgO-coating experiments. The coating procedure was similar to that used in depositing Al_2O_3 except for the method of feeding of the metal halide to the system. Since temperatures in excess of 700 C were believed to be required to vaporize and transport the MgI_2 as a vapor in sufficient concentration to give a practical coating rate, the MgI_2 was fed as a solid to be flash vaporized in the bed. The main fluidizing hydrogen gas stream containing the water vapor was introduced into the bottom of the reactor. Magnesium iodide was slowly added as granules (minus 16 plus 20 mesh) from the top of the reactor. As judged from the behavior of particles which became visible when they hit the wall, the MgI_2 particles were mixed well into the lower third of the fluidized bed and evaporated in about 0.5 sec.

This method of feeding the metal halide appeared to work satisfactorily under the particular operating conditions used. When the coating was applied at a rate of 2-1/2 μ per hr at 1000 C, over 70 per cent of the available magnesium was deposited as MgO on the particles. Identification of the coating composition was made by X-ray diffraction.

Two lots of MgO-coated UO_2 were prepared at 1000 C. Lot 1101A was made in two steps with coating thicknesses of 7 and 14 μ, respectively. This coating was translucent and seemed to be dense and free of voids. However, microscopic examination of sectioned particles revealed apparent cracks in the coating. The crushing strength of the MgO-coated particles was found to be low as compared with that of dense Al_2O_3 coatings. The MgO coatings had an average hardness of 720 Knoop, which compares favorably with the 825 Knoop hardness of fused MgO. All particles cracked, and the coatings defoliated when a sample of Lot 1101A was heated at 1000 C in oxygen for 48 hr. It was suspected that the MgO coating had cracked on cooling from the deposition temperature as the result of the difference in thermal-expansion coefficients. To determine whether the failure in oxidation was due to the thermal-expansion cracking or to coating porosity, Lot 1102 of MgO-coated UO_2 (20-μ coating) was prepared. In this experiment the coated particles were subjected to fluidization in oxygen for 2 hr at 1000 C in the coating equipment before being cooled to room temperature in a hydrogen atmosphere. Only about 16 per cent by weight of the coated particles were oxidized sufficiently to be fragmented. Cracking of the coatings during deposition may be explainable as the result of stresses developed in the thermal cycle which each particle experiences when fluidized with a relatively cold gas.

Plans for Future Work

Preparation of Lot 1102 will be repeated except that the fluidizing gas will be preheated to prevent temperature cycling during coating. In this way cracking during the coating operation should be eliminated.

Chemical-Fabrication Studies

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

Experimental work was deferred during August.

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

For initial tests, bodies which are representative of current graphite technology are desired. 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 enriched 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.

Specimen or small-quantity dispersal methods have been developed and used in combination with pressure baking. PyC-clad enriched UC_2 particles dispersed by these methods have had lower fission-gas release than the undispersed fuel in neutron-activation tests.

Work This Period

Two new types of clad fuel particles were dispersed using small-batch pressure-baking methods. The low post-neutron-activation fission-gas release for these specimens is of the same order as for the undispersed fuels. Data for enriched fuel specimens of these types and for the material fueled with UO_2-4E prepared last month are given in Table L-2.

TABLE L-2. FUELED-GRAPHITE-MATRIX SPECIMEN DATA

Specimen	Type of Dispersed Fuel Lot	Fuel	Cladding	Fuel Loading (UO_2 or UC_2), w/o	Composite Density, g per cm^3	Fission-Gas[(a)] Release, ppm Xe^{133}
G-18	4E	UO_2	Al_2O_3	9.6	2.04	0.07-0.51
G-19	919A	UC_2	PyC	9.6	1.92	0.20-0.30
G-20	920A	UO_2	PyC	9.9	1.91	0.07-0.14

(a) Values are minimum and maximum release obtained from pellets which are grouped to constitute a "specimen". Release values were obtained by post-neutron-activation heating for 1 hr at 1090 C for G-18 and 1150 C for other types.

In the further preparation of graphite-matrix specimens, procedures similar to those outlined in GAMD-2221 have been used to prepare batch-fueled and unfueled specimen sets. Their evaluation is not complete.

Plans for Future Work

Fuel particles clad at higher temperatures will be dispersed in graphite-matrix specimens. Specimen sets prepared with GAMD-2221 methods and materials will be evaluated.

Beryllia-Matrix Studies

A. K. Smalley, D. J. Bowers, and W. H. Duckworth

Objective

To develop fuel-element bodies consisting of oxide-coated UO_2 particles dispersed in a sintered BeO matrix, the bodies to have high degrees of fission-product containment and oxidation resistance.

Program Approach

The effects of type and amount of fuel particles and the effects of ceramic processing variables on the quality and microstructure of composite bodies are being investigated.

Progress Reported Previously

An experiment comparing the resistance of knobby and smooth coated particles to cracking during compaction in a matrix suggested that the smooth-coated particles were less likely to be damaged. This experiment did not afford a direct comparison of knobby versus smooth coatings, since the coating thicknesses were not equal.

The sintering behavior of a sample of Brush UOX-grade BeO was evaluated. Bodies having densities in a marginally interesting range were achieved by somewhat severe pressing and sintering treatments.

Work This Period

Experiments on the durability of smooth and knobby coatings, initiated last month, were continued during August. Two modifications were made this month: the coating thicknesses of the two groups of particles were the same, 40 μ, instead of the 30 and 40 μ employed last month, and the volume loading of particles in the specimens was extended to 30 volume per cent. Pellets containing 10, 20, or 30 volume per cent of coated particles were compacted at 10,000, 20,000, or 40,000 psi. These pellets were then heat treated in the as-compacted condition for 1 hr at 650 C in air. Evidence of disruption, indicating coating failure and UO_2 oxidation and expansion, was observed.

The salient result of this experiment was that with 40-μ coatings the smoothness or knobbiness of the coatings had little effect on their durability during compaction. There were no visually apparent differences after heat treatment in pellets containing the two types of particles. It thus appears that durability during compaction is related more nearly to coating thickness than to coating smoothness. Pellets containing 30 volume per cent of coated particles showed a higher degree of particle damage than did pellets containing 10 or 20 volume per cent of particles at 40,000 psi. However, pellets containing the higher particle content and compacted at 10,000 or 20,000 psi showed little or no disruption on heat treatment.

Plans for Future Work

The experiment described above will be repeated using Al_2O_3-coated particles of demonstrated coating integrity.

BeO-coated UO_2 particles which were subjected to thermal cycling (BMI-1534) are being examined metallographically in an attempt to establish why they failed in subsequent oxidization tests. Evidence of excessive grain growth, oriented grain boundaries, and fissuring in the coatings will be sought.

Work on BeO-matrix bodies will be begun when reliable BeO-coated UO_2 particles are available.

Explosive Compaction Studies

R. J. Carlson and S. J. Paprocki

This program has been recessed.

Pressure-Bonding Studies

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

This program has been recessed.

FUEL-IRRADIATION PROGRAM

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

Sweep Capsule SP-5

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

Objective

To obtain preliminary fission-gas release information on alumina-coated uranium dioxide particles in a graphite matrix

Progress Approach

Fission-gas-release data on a preliminary sample of alumina-coated UO_2 particles were obtained by periodic sampling during irradiation and by a variety of postirradiation fission-gas-release experiments. These data are correlated considering various release mechanisms and phenomena. The postirradiation examination includes metallographic inspection, X-ray studies, and strength tests.

Progress Reported Previously

Two 1-1/2-in.-diameter graphite spheres fueled with about 10 w/o of fully enriched UO_2 alumina-coated particles were irradiated in Capsule SP-5 to a depletion burnup of about 6.6 per cent of the uranium-235 at the surface of the sphere and 5.5 per cent at the center at a temperature level between 510 and 730 C. A summary of the experimental results from this capsule for the in-pile fission-gas-release behavior of the specimen containing alumina-coated particles (No. 471) and the graphite specimen fueled with uncoated UC particles (E8-7) was presented in a previous progress report, BMI-1480. The R/B values pertinent to the coated-particle specimen for the various fission-product gases were initially in the 10^{-7} to 10^{-8} range, but rose to the 10^{-4} to 10^{-1} level near the end of the irradiation. A tentative correlation of the fission-product-release data from the coated-particle specimen with respect to the disintegration constants of the various species and a discussion of plausible release mechanisms were reported in another previous progress report, BMI-1496. A nondestructive examination after irradiation indicated that the spheres were undamaged but had experienced greater contraction and loss of weight than expected. One sphere was compression tested and the other examined metallographically. The metallographic examination of the particles after separation from the graphite matrix revealed that about 3.6 per cent of the particles in the sphere possessed cracked coatings and that about 86 per cent of the particles with cracked coatings were within 10 particle diameters of the surface of the sphere. Microhardness measurements indicated that a decided decrease in the hardness of the alumina coatings occurred during irradiation.

Postirradiation fission-gas-release measurements and acid-leaching studies with irradiated fuel particles separated from the graphite matrix of Sphere No. 471 were performed. Uranium analyses on nitric acid solution used to leach coated fuel particles removed from various positions along the radius of the sphere indicated that about 14 per cent of the coatings on particles located near the sphere surface were cracked, with the proportion of cracked coatings continuously diminishing to less than 1 per cent at a distance of about 0.5 in. in from the surface. Acid-leached and unleached coated fuel particles heated at temperatures up to 1500 C released up to almost 7 per cent of the contained krypton-85. The temperature dependency of krypton-85 release during postirradiation heating was found to have an activation energy of approximately 70 kcal per g mole. Samples of coated particles released up to 30 per cent of the krypton-85 when crushed. Prior leaching reduced the gas release by a factor of about 1.6.

The detailed results of the postirradiation examinations of these two spheres were included in the following progress reports: BMI-1496, BMI-1504, BMI-1509, BMI-1514, BMI-1518, and BMI-1534.

Work This Period

An electron microscopic examination of irradiated and unirradiated particles is continuing in an effort to determine if changes occurred in the structure of the alumina coating as a result of irradiation. The results obtained to date are difficult to interpret. It is difficult to etch the alumina and reveal any structure. Etchants reported to have been used successfully on sintered alumina have been ineffectual on the vapor deposited alumina used in this program. The most successful etchant found for use on these coatings is a 5 per cent solution of hot sodium hydroxide. Etching times used have been in the order of a few seconds. Although this etchant has not revealed a positively identifiable structure, some interesting observations have been made. The alumina of both the irradiated and the unirradiated particles appeared to have been etched and to have been possibly an amorphous material. Paradoxically, however, there were some indications of grain outlines in certain regions of this "amorphous" material. The size of these outlines was about the size one would expect for grains of alpha alumina. Also, distributed over the surface of the alumina was a crystallinelike material which was relatively unetched. These "crystals" were much smaller than the grain outlines observed in the matrix and may have been residue from the chemical action of the etchant or possibly very small grains of alpha alumina. Both the irradiated and the unirradiated specimens exhibited this same general appearance in all areas of the coating. However, some regions of the alumina coatings on the irradiated particles were more densely covered with the crystalline material.

Examination of irradiated particles, unsuccessfully etched with hydrofluoric acid, did not reveal any grain boundaries in the alumina matrix and very little of the crystalline material. However, about a 1-μ-wide zone existed at the interface of the UO_2 and alumina which may have been a reaction or diffusion zone.

The electron-microscopy examinations are continuing on the material in the as-polished condition in an effort to better identify the phases which have been observed. The results of the X-ray diffraction studies when completed may also aid in the evaluation of the results of the electron-microscopy examination.

Coated fuel particles which had been used for postirradiation fission-gas-release experiments were inspected visually under a microscope and then leached in acid to determine if the high-temperature heating had caused additional coating failures. Both methods failed to detect any evidence of cracked coatings that could be attributed to exposures to temperatures up to about 1550 C. Samples of these particles were also crushed and the fission gas collected to determine if prior heating affects gas evolution from the particles when crushed. The data from the crushing experiments are now being analyzed.

Plans for Future Work

A final appraisal of both the in-pile and postirradiation fission-gas-release data will be performed.

Electron microscopic examination of the irradiated coated particles will be continued, and the Al_2O_3 coating will be studied by X-ray diffraction techniques to determine if any changes occurred in the structure as a result of neutron and fission-fragment attenuation. Planned cracking tests of the separated particles will not be performed. These tests require hand picking of the particles to obtain nearly uniform shapes and sizes. The high radioactivity of the particles makes manual handling impractical, and it is believed that the data would not be meaningful enough to warrant the time and expense of remote particle selection and testing.

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 temperature of irradiation, 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 two types of fuel particles, as described below, supported in two types of graphite matrices, and also unsupported by any matrix material. 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)

(2) UO_2 particles coated first with low-density Al_2O_3 and then with dense Al_2O_3 (Type 721C).

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 in an unsupported condition and also in two types of graphite matrices. 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 measurement 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 two types of Al_2O_3-coated UO_2 particles were incorporated into both an almost fully graphitized matrix and a relatively ungraphitized (carbon) matrix. Since the carbon matrix will tend to graphitize and contract during irradiation, this matrix may give a greater degree of support to the coated particles than the more graphitized matrix. Neutron-activation studies indicated that the specimens made of Type 715A particles in the carbon matrix were unsuitable for irradiation. This specimen has been replaced by unsupported particles which have a pyrolytic carbon coating over an Al_2O_3 coating on UO_2 particles. These particles are designated as Type 921 coated particles and were not included in the original program.

Each of the four capsules, CFP-1A, CFP-1B, CFP-1C, and CFP-1D, were loaded and sealed. They each contain the following specimen types:

(1) Type 715A particles, unsupported (Xe^{133} release: 3.2 ppm at 1300 C)
(2) Type 715A particles, graphite matrix (Xe^{133} release: 0.1 to 2.0 ppm at 1150 C)
(3) Type 721C particles, unsupported (Xe^{133} release: 1.3 ppm at 1300 C)
(4) Type 721C particles, graphite matrix (Xe^{133} release: 0.1 to 5.3 ppm at 1150 C)
(5) Type 721C particles, carbon matrix (Xe^{133} release: 0.1 to 1.3 ppm at 1150 C)
(6) Type 921 particles, unsupported (Xe^{133} release: 0.5 ppm at 1300 C).

Work This Period

The irradiation of all four capsules was initiated on August 9, 1961. They are being irradiated in pairs to two target burnup levels of about 2 and 6 a/o of the uranium.

At each scheduled burnup level, one capsule is operating at a specimen temperature of about 980 C, and the other is operating at a specimen temperature of about 150 C.

Plans for Future Work

The low-burnup capsules, CFP-1C and CFP-1D, will be irradiated for three BRR cycles; Capsules CFP-1A and CFP-1B will be irradiated for six BRR cycles. The post-irradiation examination will include analyses of the gases in the individual specimen cans, visual inspection of the specimens at magnifications to 30 diameters, dry-weight and dimensional measurements (in the case of the matrix specimens), metallographic examination, and fission-gas-release studies.

Sweep Capsule CFP-S-2

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

As discussed in BMI-1524, the irradiation of this capsule was begun in June, 1961, and terminated after 36 hr of operation when a leak occurred between the argon-filled gas annulus and the sweep-gas system. Immediately thereafter, the fabrication of a replacement capsule, designated CFP-S-2R, was begun.

Work This Period

The irradiation of the CFP-S-2R capsule was begun on August 9, 1961. The capsule was operated with a specimen-surface temperature of 1070 ± 15 C. Table L-3 contains the fission-gas-release data available at this time. The "A" samples are adsorption samples which are taken by diverting the sweep gas approximately 1 hr after leaving the capsule through small refrigerated charcoal adsorption traps. The adsorbed gases are subsequently driven from the trap, separated, and analyzed by gamma spectrometry. This type of sample is used for the detection of Kr^{85m}, Kr^{87}, KR^{88}, Xe^{133}, and Xe^{135}. The sample designated "DT" is a daughter-trap or short-half-life gas sample which is taken by diverting the sweep gas, a few seconds after it leaves the capsule, into the daughter trap where the short-half-life Kr^{89}, Kr^{41}, Xe^{137}, Xe^{140}, and Xe^{141} are given a sufficient residence time to decay to their longer half-life solid daughter products. The daughter products are deposited on a charged wire within the trap near their point of origin. The wire is removed from the trap for analysis after the sampling period.

A discussion of the results will be reserved until all of the data are available for the various isotopes. However, two points should be noted: (1) the data from Samples 3A and 5A are effected by the preceding shutdowns, and (2) it is very apparent that a great change in the fission-gas release from the specimen occurred prior to Sample 9A after 7 days of irradiation. This change took place 2-1/2 hr before Sample 9A was taken when the reactor came to full power after being shutdown 1 day by mechanical difficulties. At this time no reason can be given for the sudden change. It may be noted that during reactor shutdowns the fuel specimen temperature is maintained at 750 to 800 C with the auxiliary electrical heater.

At the same time that the increase in fission-gas release occurred the pressure drop through the system began to increase. This pressure increase was believed to have been caused by a plugging of the flow path either in the capsule or in the small-bore outlet tubing downstream from the capsule. The resulting reduction in flow rate has an adverse effect on the gas sampling by increasing the transit time from the capsule to the daughter trap and creating an overlong residence time before sample collection. The longer transit time to the daughter trap resulted in the loss of the 1.7-sec Xe^{141}. Also, the longer holdup time before Samples 9A and 14A resulted in uncertainties of approximately ±100 per cent in the trap analysis.

Shortly after beginning the second reactor cycle of operation, the pressure drop through the system increased to the point that it was necessary to stop the flow of sweep gas.

Plans for Future Work

Plans are being made to attempt to free the system of restrictions during the next scheduled reactor shutdown early in September so that the experiment may be continued. Until that time the capsule will be irradiated under static conditions and maintained at its normal operating temperature.

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

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 to be irradiated were selected and include the following:

(1) UO_2 coated with a thin dense interlayer of Al_2O_3 which in turn is coated with a porous and then a second dense coating of Al_2O_3 (S & P 4E, Xe^{133} release: 1.2 ppm at 1100 C)

(2) UO_2 coated with dense Al_2O_3 (715A, Xe^{133} release: 3.2 ppm at 1300 C)

(3) UO_2 coated with two dense and one porous layer of Al_2O_3 similar to Type (1) above, but differing in coating-layer thicknesses and coating application techniques (721C, Xe^{133} release: 1.3 ppm at 1300 C).

(4) UC_2 coated with columnar pyrolytic carbon (918A, Xe^{133} release: 2.7 ppm at 1300 C
(5) UC_2 coated with laminar pyrolytic carbon (919A, Xe^{133} release: <0.5 ppm at 1300 C)
(6) UO_2 coated with laminar pyrolytic carbon (920A, Xe^{133} release: 0.9 ppm at 1300 C).

Each of the six types were sealed in a helium-filled annulus in individual aluminum cans. The six cans were fitted end to end in a NaK-filled capsule. The specimen cans were centered radially in the capsule by three longitudinal dosimeter wires spaced 120 deg apart. The capsule, designated as Capsule CFP-4, was charged into the BRR on July 10, 1961.

Work This Period

The irradiation of Capsule CFP-4 is continuing. The specimen irradiation temperature is estimated to be about 65 to 120 C. The capsule is scheduled to be discharged the first week in September, 1961, with an estimated fuel burnup of about 25,000 MWD/T of uranium.

Plans for Future Work

After discharge from the reactor, the capsule will be opened, and the specimen cans recovered. The gases inside these cans will be collected and analyzed. Postirradiation examination of the coated particles will include visual inspections at magnifications to 30 diameters, metallographic examinations, and fission-gas-release studies.

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

Four pellets, 0.26 in. in diameter and 0.26 in. long, were machined from Sphere No. 472, which was a prototype of the two graphite spheres containing alumina-coated uranium dioxide particles which were irradiated in Capsule SP-5. The damaged coated particles were picked from the surface of the pellets and attempts were being made to remove the uranium contamination from the pellet surfaces in preparation for encapsulating two of the pellets for this sweep-capsule experiment.

Work This Period

The surfaces of the four pellets were cleaned using an electroetching technique with dilute chromic acid as the electrolyte. An alpha count of the surfaces showed that

TABLE L-3. CFP-S-2R FISSION-GAS-RELEASE DATA

Sample[a]	Time After Start of Irradiation, hr	R/B Value						Remarks
		Kr^{85m}	Kr^{89}	Kr^{91}	Xe^{133}	Xe^{137}	Xe^{140}	
1A	2	4.6×10^{-10}	--	--	--	--	--	--
2A	4	4.3×10^{-9}	--	--	--	--	--	--
3A	24	2.2×10^{-8}	--	--	--	--	--	Sampled 1.5 hr after a 10-hr shutdown
4A	42	3.0×10^{-8}	--	--	--	--	--	Sampled 7.25 hr after a 2.75-hr shutdown
5A	68	8.2×10^{-9}	--	--	--	--	--	Sampled 1.5 hr after a 17.5-hr shutdown
6A	90	3.1×10^{-8}	--	--	--	--	--	Sampled 8 hr after a 1-hr shutdown
7A	114	3.1×10^{-8}	--	--	--	--	--	--
8A	136	2.5×10^{-8}	--	--	--	--	--	Sampled 21.25 hr after a 0.5-hr shutdown
9A	171	2.2×10^{-4}	--	--	3.2×10^{-3}	--	--	Sampled 2.5 hr after a 28-hr shutdown
10A	190	3.6×10^{-6}	--	--	1.1×10^{-3}	--	--	--
11DT	190	--	6.9×10^{-4}	2.7×10^{-5}	--	3.4×10^{-4}	2.1×10^{-5}	--
12A	195	4.7×10^{-5}	--	--	2.5×10^{-4}	--	--	--
13A	210	3.5×10^{-6}	--	--	3.2×10^{-5}	--	--	--
14A	379	4.3×10^{-3}	--	--	3.8×10^{-3}	--	--	Sampled 8 hr after a 100-hr shutdown

(a) "A" samples designate adsorption samples; "DT" samples designate daughter-trap samples.

a satisfactory degree of decontamination was achieved by this method. Three of the pellets were neutron activated and the xenon-133 release was measured in subsequent heat treatment. From these results, two pellets having Xe^{133} release ratios of 2.1 and 5.0 ppm at 1150 C were selected for this sweep capsule. These release ratios compare favorably with the value of 7.7 ppm at 1050 C for the similar Sphere No. 471 prior to irradiation in the SP-5 capsule.

In addition to the sweep-gas specimen, this capsule will contain four static specimens as follows:

1. Specimen G-18, a 0.25-in.-diameter by 0.20-in.-long graphite-matrix pellet containing alumina-coated uranium dioxide particles from the same lot (4E) used to fabricate the SP-5 spheres and is one of several pellets fabricated as a backup effort for the sweep-capsule specimen (Xe^{133} release: 0.07 ppm at 1150 C)
2. Unsupported columnar-carbon-coated uranium dicarbide particles of Lot 918A (Xe^{133} release: 2.7 ppm at 1300 C)
3. Unsupported laminar-carbon-coated uranium dicarbide particles of Lot 919A (Xe^{133} release: <0.5 ppm at 1300 C)
4. Unsupported pyrolytic-carbon-coated uranium oxide particles of Lot 920A (Xe^{133} release: 0.9 ppm at 1300 C).

The fabrication of the capsule for this experiment, which incorporates the one sweep-gas specimen and the four static specimens, is now in progress.

Plans for Future Work

This irradiation experiment is expected to be initiated in the BRR during September and operated at a nominal temperature of 1050 C.

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 pyrolytic-carbon-coated uranium dioxide particles supported 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

This is a new program.

Work This Period

The planning of this experiment was initiated late in this report period.

Plans for Future Work

The fuel specimen is expected to be available in September for encapsulation. Fuel specimens for static irradiation in this capsule will be designated at a later date. The irradiation is expected to be initiated in October, 1961.

Intermediate-Temperature Design Studies

N. E. Miller and W. J. Zielenbach

This work has been temporarily recessed and is expected to resume in September.

Fission-Gas Analytical Studies

R. L. Ritzman and T. S. Elleman

Objective

To develop improved techniques for the analysis of fission-gas mixtures.

Program Approach

The measurement of fission-gas-release rates from in-pile fuel specimens requires accurate sampling of the offgas and a suitable method for analyzing the fission gases. Using the gamma-ray-spectrum analysis method, it is necessary to know the size of the gas sample collected, the detection efficiency of the gamma-ray spectrometer for the various fission-gas gamma rays, and the accuracy with which the gamma-ray spectrum can be resolved into contributions from individual fission gases.

The present program is directed toward the improvement and expansion of current techniques for fission-gas analysis. The retention time of fission gases on charcoal traps will be measured to determine the maximum sampling times in collecting gas samples. Fission gases with gamma-ray photopeaks in the energy range from 80 to 230 kev will be standardized in the gamma-ray spectrometer to check the presently used efficiency values for this energy range. New techniques for resolving complex gamma-ray spectra will be studied as possible replacements for present methods.

Progress Reported Previously

Helium-elution data were gathered for krypton and xenon from charcoal traps at liquid-nitrogen and dry-ice temperatures. Also, the gamma-ray spectrometer detection efficiencies of xenon-133 and xenon-133m on charcoal traps were determined. A krypton and xenon fission-gas mixture, produced by irradiating uranium foil, was separated by gas chromatographs and adsorbed on charcoal traps. Spectrometer scans of the separated krypton and xenon fractions were divided into a number of energy intervals, such that each interval included at least one gamma-ray photopeak from one of the gases of interest. The contribution of each xenon and krypton radioisotope to each selected energy interval was determined by resolving the time-decay curve for each energy interval. These isotopic contributions can now in principle be used to resolve any fission-gas spectra into the counting-rate contributions of each component.

Work This Period

Resolution of the recorded gamma-ray spectra from a second mixture of krypton isotopes has been completed which considerably improves the individual isotopic gamma-ray contribution values for a mixture consisting of krypton-85m, krypton-87, and krypton-88.

Plans for Future Work

The gamma-ray-spectrometer detection efficiencies for gamma rays having energies of 0.14 to 0.84 Mev will be determined. It is expected that all of the techniques necessary for the analysis of complex fission-gas mixtures will have been completed by the end of the coming month.

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

A literature survey was completed to assist in evaluating the amount of fission-product release which can be tolerated in a system without interfering with the normal reactor maintenance. The effects of oxide fuel-particle coatings on the reactivity of a graphite-moderated pebble-bed reactor (PBR) were presented in a recent progress report, BMI-1504. 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 oxide or carbon films were reported in BMI-1524. A preliminary investigation was started to determine the range of fuel loadings of interest in the development of reactor fuel elements containing coated particles.

Work This Period

Compilation of reactor data leading to the establishing of the range of fuel loadings of interest in the development of fuel elements containing coated particles was continued. The gas-cooled reactors being investigated include the Experimental Gas-Cooled Reactor (EGCR), the High-Temperature Gas-Cooled Reactor (HTGR), and the Gas-Cooled Reactor Experiment (GCRE).

A preliminary investigation of the feasibility of employing coated particles in current superheater reactor designs, e.g., the Pathfinder and BONUS reactor concepts, was performed, indicating that coated particles might have several marked advantages in this application.

The investigation to examine the nuclear-criticality problems involved in designing a chemical reactor to be employed to coat fuel particles was expanded to include the effects of gradual buildup of moderating material during the coating process, with the objective of determining the maximum safe reactor diameter.

Plans for Future Work

Compilation of data to establish range of fuel loadings of interest in the development of fuel elements will be continued.

The application of coated fuel particles to nuclear superheater design will be investigated more thoroughly. The initial investigation will consider fuel elements fabricated of fuel particles coated with high-temperature-steam corrosion-resistant material (ZrO_2, MgO, or Al_2O_3) and canned moderator.

The investigation to determine the maximum safe diameter for the coating reactor will be continued.

Fission-Gas-Diffusion Studies

R. H. Barnes, 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 dispersed 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 reported in BMI-1504. Xenon-133 diffusion coefficients for pyrolytic carbon between 700 and 1300 C were presented in two recent progress reports, BMI-1514 and BMI-1518.

Work This Period

Measurements of diffusion coefficients for xenon-133 in Linde Company alpha-alumina spheres were performed at temperatures of 700, 900, and 1100 C. Some data have already been obtained in this temperature range; however, the present results are required to fully establish the temperature dependency of diffusion below 1200 C, which appears to have a lower activation energy than above 1200 C. A second series of xenon-133 diffusion measurements was conducted using spheres from which about 20 per cent of the xenon-133 had been removed by grinding a uniform layer from the surface of the sphere. The objective of these experiments was to study surface effects and their relation to the concentration profile of the diffusing species in the sphere.

The data from the above experiments are currently being analyzed and the results will be reported next month.

Plans for Future Work

Diffusion experiments are to be performed using alpha-alumina spheres, vapor-deposited alumina particles, and vapor-deposited pyrolytic-carbon particles at temperatures in the range from 700 to 1600 C. The purpose of these experiments will be to indicate if differences in fission-gas diffusion rates exist between vapor-deposited alumina and alpha-alumina spheres and if diffusion coefficients calculated from the back-diffusion of recoiled fission gases in pyrolytic carbon correspond to the diffusion coefficients obtained for diffusion of fission gas through pyrolytic carbon. Additional experiments will be conducted to measure xenon-133 release during postirradiation heating from alumina and pyrolytic-carbon spheres irradiated for periods of 4 to 8 weeks in a thermal-neutron flux of 1×10^{13} nv. These experiments will demonstrate the effect of prolonged irradiation on observed fission-gas release.

Diffusion studies involving other potential coating materials for fuel particles are planned.

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 used to allow observation of 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 a defected cross-sectional 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 2μ. Longer, thinner regions 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 smaller. Decoration by precipitated silver in AgCl has revealed dislocations and grain boundaries.

Work This Period

Studies of NaCl handled under controlled-humidity conditions are still in progress.

Direct decoration of radiation damage by precipitated silver has been observed in the AgCl samples prepared by melting and resolidifying crystals between glass plates. Fueling the under side of the upper plate with UO_2 served as a source of fission recoils. Insufficient irradiations have been analyzed to date to definitely separate the fission events from other effects. Rolled sheets of AgCl have also been irradiated and studied with replica techniques very similar to those used with the NaCl. Fission damage in the AgCl appears to be less extensive than that in NaCl and to possess a structure that is less fine.

Plans for Future Work

Continuation of the studies of NaCl under controlled-humidity conditions as well as the studies of decoration by silver precipitation in AgCl is 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 operation 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 compared to weight changes 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 UC_2 in a temperature gradient for specified periods of time.

Progress Reported Previously

Carbon-coated UC was heat treated at 1400, 1600, and 1800 C for varying time periods for use in determination of 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 UC_2 in a few hours. At lower temperatures, UC and U_2C_3 also exist. Oxidation studies on Al_2O_3-coated UO_2 particles coated at 1000 C indicated that no oxidation occurred after 100 hr at 1180 C, but rapid oxidation was observed after heating the particles for short periods of time at 1340 C or higher.

Work This Period

Data from measurements of uranium diffusion through pyrolytic-carbon coatings on UC particles at 1800 C using autoradiographic and alpha-counting techniques are currently being analyzed. Preliminary analysis of alpha-counting measurements on particles heated for 1 and 2 hr indicated a diffusion coefficient of about 2.3×10^{-10} cm^2 per sec at 1800 C for uranium in the pyrolytic-carbon coatings. This value assumes a carbon-coating density of 1.7 g per cm^3 with a corresponding alpha-particle range of 19 μ. Analysis of results of autoradiographic measurements on the same particles is in progress and will be reported next month.

Studies on the oxidation of Al_2O_3-coated particles continue to suggest that failure of the coatings due to the different thermal expansion of the Al_2O_3 and UO_2 is the major cause of the UO_2 oxidation. The rate of oxidation appears to be too rapid to be controlled by a volume diffusion mechanism and too dependent upon a critical temperature range to be controlled by diffusion of gas through pores or by other direct paths. For example, a specimen of UO_2 powder coated with 53 μ of Al_2O_3 heated at 1300 C for 1 hr in 1 atm of oxygen showed no detectable weight change, but showed a weight gain after 5 min at 1350 C. This same sample after being held for only 15 min at 1350 C was then cooled to 1300 C and a continuous weight change of about 0.1 per cent per hr was observed.

Other specimens show that failure is dependent upon coating thickness and density of the UO_2. At a heating rate of approximately 12 C per min in 1 atm of oxygen, the UO_2 coated with 4 to 5 μ of Al_2O_3 begins to show a weight gain at about 1250 C while the UO_2 coated with 53 μ of Al_2O_3 does not show a weight gain before 1400 C. Porous UO_2 (~85 per cent of theoretical density) coated with 35 to 50 μ of Al_2O_3 and heated at the same rate showed no change in weight before reaching a temperature of 1700 C. All of the above specimens were coated at a temperature of 1000 C.

Plans for Future Work

Analysis of data from autoradiographic and alpha-counting measurements of uranium diffusion in carbon coatings is being continued. Further diffusion measurements will be planned after both methods have been fully evaluated. On the basis of the favorable preliminary results, it is anticipated that both techniques will be employed in future measurements to serve as a means of obtaining more reliable diffusion data.

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 will be conducted in which steam will be passed through an outer jacket under controlled pressure and then through an electrically heated inner jacket, the reactor. Volatile reaction products will be condensed in the cool portion of the reactor and analyzed. These experiments will yield vapor-pressure and free-energy data which can then be used to calculate erosion losses.

Progress Reported Previously

Initial plans for the experimental program were being made.

Work This Period

Computations were made concerning the length of time transpiration runs would have to be made to obtain enough condensation for analysis. These calculations indicated that run times in the order of days would be necessary to obtain about 20 μg of condensate at temperatures of 1200 to 1700 C. These calculations are based upn existing free-energy data and postulated vaporization mechanisms, neither of which can be assumed to be accurately known.

It has been concluded that a basis is necessary in order to interpret the results obtained in the high-temperature steam investigation. The basis selected is to determine the vapor transport over a range of steam pressures. Consider, for example, this reaction:

$$MgO(c) + H_2O(g) \longrightarrow Mg(OH)_2(g).$$

The equilibrium constant is given by:

$$K_p = \frac{p[Mg(OH)_2]}{p(H_2O)}.$$

Doubling the water pressure would then double the pressure of $Mg(OH)_2(g)$, but, in order to obtain significant changes in the vapor pressure of the oxide, a change in steam pressures of the order of factors of ten would be desirable. The maximum dry steam pressure that can be maintained for extended periods in the autoclave is about 60 atm. This then forms the upper limit of steam pressure to be explored in this investigation. Selecting 0.03 atm steam as a lower limit would produce a variation of 2000 in the vapor pressure of the volatile oxide between the two steam pressures. Using steam at a 1-atm pressure would then give an intermediate value between the two extremes, being 33 times higher than the low steam pressure vapor pressure and 60 times lower than the high steam pressure.

It is expected that data obtained over an extended range of steam pressures will aid materially in determining the mechanism of vapor transport in cases where this is not known. This information will then make it possible to predict vapor pressures and erosion rates at steam pressures other than those used in the current investigation.

Plans for Future Work

Experimental determinations of UO_2 and possibly MgO vaporization in 0.03-atm steam are planned for September.

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 to select the most promising technique. On the basis of this survey, suitable equipment is to be constructed for use in a hot cell.

Progress Reported Previously

The work has been directed toward the selection of a method suitable for measuring the thermal diffusivity of specimens of a size dictated by coated-particle size, encapsulation considerations, and irradiation conditions. A breadboard setup was made in order to check out certain ideas for making the measurements.

A study was made of various radiation detectors, including the Golay infrared detector, thermocouple beads, thermister bolometer, pyroelectric detector, and lead sulfide cell.

A decision was made to use lead sulfide cells as receivers, and the specific cells and instrumentation were selected. A review was made of arrangements at the Battelle Hot-Cell Facility to see how best to design the apparatus for use there. Work was progressing on the assembly of the apparatus and the checking out of various instruments.

Work This Period

Studies were continued to check out equipment for use with the IR detectors and set up the two-detector bridge system. Preliminary checks indicate a scope sensitivity of about 1 cm per deg change in temperature without the use of preamplifiers. While the preamplifiers have a maximum gain of 1000, problems arising from increasing signal to noise ratio may be encountered at high gains. The mountings for the detectors are being made and various means of reducing the signal to noise ratio are being tried.

The lead-shielded table and vacuum chamber (except cooling coils) have been completed. Work on the remote-controlled specimen-loading and unloading apparatus is about 60 per cent completed, and all optical parts are ready for assembly on the apparatus.

Construction of the table and vacuum chamber were completed. The vacuum system and gages were mounted and the can checked for leaks. One leak in a weld was located and repaired. The system can be operated at about 2 x 10^{-5} mm of mercury, or lower if the cold trap is filled.

Operation of the chopper-timing motor was checked, and it was found possible to set its speed from the oscillator dial control to ±0.8 per cent. This speed control is considered sufficiently precise for the motor's intended use. A more precise control can be had with some additional instrumentation. The range of chopper-motor speeds is such that chopper frequencies ranging from 10 to 330 cps can be had with but two different chopper disks.

Plans for Future Work

Work will be directed toward a trial of the specimen measuring method without completing all equipment needed to handle irradiated specimens. The molybdenum thermal-shielding material and furnace tube have been received. The furnace and thermal shielding will be assembled in the vacuum chamber with a temporary specimen holder in place. Optical benches will be mounted for the optics, chopper disk, and thermal-radiation receivers. It is hoped to make a trial run during September.

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 this 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 August.

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. L. White, 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

A combined hydrofluorination-corrosion study involving ZrO_2 dissolutions and INOR-8 corrosion specimens was described in an earlier progress report, BMI-1518. A similar investigation using Hastelloy X and tungsten was described in BMI-1534. Fluorination-corrosion studies have also been in progress utilizing INOR-8 and HyMu-80 corrosion specimens as reported in BMI-1524 and BMI-1534.

Work This Period

A fluorination study with "L"-Nickel corrosion specimens in LiF-NaF-ZrF_4 melts, sparged with fluorine at 500 C, has been in progress for 150 hr. The corrosion specimens will be examined and the results reported next month.

Plans for Future Work

A corrosion evaluation of HyMu-80 specimens under hydrofluorinator conditions in the presence of dissolving Zircaloy-2 will be conducted.

Metallographic examinations will be made on components from the ORNL pilot plant.

DEVELOPMENT OF COLD-BONDING PROCESSES (SRO)

S. W. Porembka

Work is under way relating to the CANDU program of SRO.

Friction Bonding

S. J. Paprocki, E. S. Hodge, H. D. Hanes, 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. Metallographic techniques are presently employed as the primary method of evaluation. Satisfactorily joined specimens are subsequently tested in corrosion and their mechanical properties are determined. A burst test is being considered as a means for inspecting the integrity of end caps.

Progress Reported Previously

It was reported in BMI-1518 that samples were designed to study the effects of relative size of the mating material in friction bonding Zircaloy-2 and stainless steel tubular components. Results of this study were inconclusive, primarily due to limitation of the modified lathe setup; therefore, tube joining studies were suspended until the completion of the friction bonder, as reported in BMI-1524.

Work This Period

Since interest in the use of friction techniques for tube joining has diminished at AECL, the construction of the friction bonder was suspended by SRO. Therefore, the tube-joining studies were also terminated. Primary interest was shifted to the end capping of SAP fuel sheaths. Additional specimens are presently being prepared for further end-capping tests.

Plans for Future Work

SAP materials have been received for the end-capping studies, and this phase of the program will be initiated.

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-2 tubular components. Variations in surface design and the use of intermediate barrier materials are being considered. Flat-plate samples of stainless steel and Zircaloy-2 are being used in initial studies to determine the nature of the joint interface obtained when employing barrier materials. The most promising material combinations will then be investigated as full-size joint components in a roll-forming operation.

Progress Reported Previously

Surface preparations for Zircaloy-2 and 410 stainless steel flat plates have been established for apparent joining when cold rolling at 50 per cent reduction taken in a single pass. Roll-expanding equipment operation and capabilities have been determined using aluminum and mild steel components. Reduction limitations imposed by the roll-forming equipment have been insufficient to produce the desired bonding in the various designs considered to date, including those providing for a locally high deformation. Metallographic examination of roll-formed joints has demonstrated that the bonds are mechanical in nature.

Work This Period

Metallographic examination of cold-rolled stainless steel-Zircaloy-2 flat-plate couples with an alumina coating indicated that good contact was achieved at reductions of 45 to 65 per cent. The alumina coating was observed to be fragmented and agglomerated at the interface. No definite evidence of metallurgical bonding was seen in as-polished or etched specimens. The reductions necessary for adherence of these plates were the same as those determined previously for plates without the alumina coating. Surface roughness, as before, appears to be an important factor.

Eight Type 410 stainless steel plates have been electroplated with gold or silver barrier layers. Four plates were plated with the gold and four with the silver to a thickness of about 0.03 mil. Gold plating was accomplished in an acid gold-plating bath while a silver cyanide bath was employed for plating the silver. A nickel strike was first applied to the plates for better adherence of the gold and silver deposits. Attempts will be made to join these plates with Zircaloy-2 plates using single-pass reductions as before.

Initial roll-forming of Type 410 stainless steel-Zircaloy-2 tubular joints was accomplished during this period. The reduction-per-pass limitations for this system are only slightly lower than for the aluminum-mild steel system.

Several joints were rolled for subsequent use in explosive welding studies.

Plans for Future Work

Rolling and evaluation of the flat plates with gold and silver deposits will be accomplished during the next reporting period. Promising materials combinations will be considered for application to full-size tubular components.

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 join Zircaloy-2 to Type 410 stainless steel. Evaluation is based upon bonding 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. The results of this testing are presented in three previous reports, BMI-1518, BMI-1524, and BMI-1534.

Work This Period

Bonding experiments with additional aluminum-mild steel and two Zircaloy-2-stainless steel tubular specimens were run using a combination of the roll-forming and explosive-joining techniques. Both stand-off and contact charges along with water and air transfer media were used. The charge densities for these experiments were varied from 1 to 4 g per in.^{2}. The roll forming of these specimens prior to explosive joining is described in the section on roll forming of this report. Examination after the subsequent explosive-joining operation revealed that no bonding was achieved during this series of tests. The higher charge densities employed with the current aluminum-mild steel assemblies are apparently in excess of that required for successful joining since previous tests with lower charge densities produced apparent bonding. With the Zircaloy-stainless steel components it appears that even lower charge densities must be used because of the sensitivity of Zircaloy-2 to explosive shock.

Plans for Future Work

Attempts will be made to optimize charge densities for explosive joining after roll forming.

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

P. M. 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. This capsule is being prepared for the irradiation at the MTR of pellets of 30 volume per cent UO_2 (12 per cent enriched) in BeO encased in Hastelloy X. Target irradiation conditions include a specimen-surface temperature of 1700 F and an in-pile period of about 18 months at a specimen-effective peak flux of about 2×10^{13} nv. Heater and thermocouple malfunctions caused a delay in delivery to the MTR.

Work This Period

Capsule BMI-36-1. Heater and thermocouple malfunctions were resolved; new protective measures were incorporated into the heater circuits. No new major capsule components were required. The capsule will be shipped to the MTR in September.

Plans for Future Work

Capsule BMI-36-1. It is expected that capsule insertion into the MTR will occur during the September shutdown for Cycle 163. Capsule thermocouple temperatures and auxiliary-heater operation will be monitored continuously.

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.

Progress Reported Previously

Irradiations of BMI-32-1, BMI-32-2, BMI-32-3, and BMI-32-4 were completed early in 1960; irradiation histories and results of specimen analyses were reported in BMI-1442, BMI-1448, and BMI-1464. Capsules BMI-32-6 and BMI-32-8 were discharged from the ETR on July 10, 1961, after 200 in-pile days. Specimen-surface temperatures in the latter capsules were maintained near the 600 F design level for the first 140 days; temperatures during the final 60 days ranged as low as 250 F because of reduced fission-heat generation and auxiliary heater limitations. Peak-flux-specimen burnups were estimated at 65 to 70 a/o (total uranium) in BMI-32-6 and 70 to 75 a/o in BMI-32-8.

Work This Period

The ETR accumulated less than average on-steam time during July and August. Hence, the recent rate of burnup accrual for specimens still in-pile appears to be somewhat lower than predicted a few weeks ago. Current performance of each of the four remaining capsules is summarized below:

BMI-32-5. All thermocouples and auxiliary heaters are operating satisfactorily. Surface temperatures of the instrumented specimens (top four) range from 550 to 620 F; estimates of specimen burnups as of August 15 range up to 60 a/o.

BMI-32-7. Two thermocouples have failed and the electric heaters are not operable. Specimen-surface temperatures are approximately 425 F; burnups, 65 to 70 a/o.

BMI-32-9. One thermocouple and all electric heaters have failed. Specimen-surface temperatures are in the range 350 to 450 F; burnups, 85 to 90 a/o.

BMI-32-10. One thermocouple has failed, the auxiliary heaters are still operable. Specimen-surface temperatures are in the range 420 to 575 F; burnups are estimated to range up to 60 a/o.

Plans for Future Work

The irradiations of BMI-32-5, BMI-32-7, BMI-32-9, and BMI-32-10 will be continued until October 2, 1961. Interest in the effects of very high burnup levels resulted in extension of these irradiations beyond the originally desired 70 a/o of the uranium.

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. These studies below are in support of this 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 the starting material. Various methods of packing the coated powder into stainless steel tubes will be investigated. The steel 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 150 and 50 μ in average diameter have been coated by a vapor-deposition process with from 5 to 10 μ of Al_2O_3. Characteristics of the as-received material were reported in previous progress reports, BMI-1489, BMI-1504 (Del.), and BMI-1518. Defective particles are 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, showed 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.

The factors contributing to the unexpected low oxidation resistance of the fabricated structures are considered to be: (1) defective coatings on the original particles, (2) particle breakup associated with cold-pressing operations, (3) differences in thermal-expansion coefficients of the UO_2 and Al_2O_3, and (4) low densities obtained during fabrication. The first two factors can now be controlled, the differential-expansion problem is being investigated, and it is expected that better particle sizing and the use of "active" Al_2O_3 coatings will permit better densities. The ultimate mechanism of failure consists of exposed UO_2 being converted to U_3O_8 with an accompanying 42 volume per cent expansion which causes brittle fracturing of the Al_2O_3 and continually exposes more UO_2.

Work This Period

A series of isostatically hot-pressed specimens was prepared with a variation of densification parameters to determine whether differential thermal expansion or low density is causing the poor oxidation resistance. The parameters and results are given in Table U-1.

TABLE U-1. PARAMETERS FOR ISOSTATIC HOT PRESSING OF Al_2O_3-COATED UO_2PARTICLES

Specimen	Method of Preparation	Core Diluent	Cladding Material	Isostatic Pressing Temperature (3 Hr at 10,000 PSI), F	Density, per cent of theoretical
HP-3	Hot pressing	7 w/o Al_2O_3	Mild steel	2300	78.4
VP-3	Vibration	None	Mild steel	2300	84.1
VP-4	Vibration	None	Mild steel	2300	83.6
VP-5	Vibration	8 w/o Cr	Mild steel	2300	86.6
VP-6	Vibration	None	Niobium	2700	89.7
VP-7	Vibration	None	Niobium	2700	89.5

Metallographic examination of Specimens VP-3 and VP-6 showed that 2700 F treatment was required to accomplish healing of any cracks in the Al_2O_3 and to develop a recrystallized structure.

The cladding was pickled from the remaining specimens, and all were exposed to 1200 F air on a recording thermogravimetric balance. All specimens showed approximately 85 per cent conversion of UO_2 to U_3O_8. The weight gain was linear in all cases; however, VP-5 (containing 8 w/o chromium) required 50 min for complete conversion, while all other specimens had oxidized within 15 min. This threefold increase in oxidation resistance may be attributed to the extrusion of the chromium into the Al_2O_3 defects or may be due to the increased matrix strength. The increased ductility and strength would conceivably reduce the rate of matrix fracture which constantly exposed UO_2. The use of such metallic diluents thus looks promising.

Specimens are being prepared in stainless steel cladding which will be defected with a 10-mil hole and exposed to 1750 F air. Such an evaluation simulates ML-1 reactor conditions. These specimens contain undiluted UO_2, 20 volume per cent chromium-UO_2 cermets, cold-pressed Al_2O_3-coated UO_2, vibratory compacted Al_2O_3-coated UO_2, and Al_2O_3-coated UO_2 with chromium diluent.

Plans for Future Work

An evaluation of the various defected specimens will be undertaken. Procurement of several new types of Al_2O_3-coated particles is expected, and fabrication of these will be undertaken.

Radiation Stability of UO_2 Specimens

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

Objective

To study the radiation stability of bulk UO_2 for possible use as a fuel for ML-1 reactors.

Program Approach

Six specimens of fully enriched bulk sintered UO_2 contained in sealed cans of Hastelloy X were irradiated in a NaK-filled capsule designated AGN-BRR-III. The capsule was irradiated in the Battelle Research Reactor to specimen uranium-235 burnups ranging from 2.3 to 3 per cent at surface temperatures ranging from 1200 to 1725 F.

The evaluation of the irradiated specimens is to be based on changes in dimensions, microstructure, and visual appearance on X-ray diffraction studies and on determinations of burnup and fission-gas-retention properties.

Progress Reported Previously

Samples of gases contained in the main body of the capsule and from each of the specimens contained in the capsule revealed that no fission gas was released to the capsule, but that up to 26 per cent of the fission gases formed was released from the fuel. Visual examination and dimensional measurements indicated that the specimens had not swelled. Macroexamination of the fuel after sectioning revealed that cracking had occurred which was essentially radial in nature although some instances of circumferential cracking were also observed.

Representative samples from specimens which had experienced the highest and lowest irradiation temperatures were examined metallographically. The results of these examinations revealed that the grains in the central region of the specimens were slightly larger than those in the outer regions. There also appeared to be a decoration of the UO_2 grains at the boundaries by an unidentified phase or void system.

Work This Period

X-ray diffraction studies of the irradiated UO_2 are in progress.

Plans for Future Work

X-ray diffraction studies will be completed. From these data and those already obtained, a final evaluation of the irradiation stability of this fuel will be made.

Radiation Stability of UO_2-BeO Specimens

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

Objective

To study the radiation stability of BeO fueled with 70 and 80 w/o UO_2 for possible use as a fuel for ML-1 reactors.

Program Approach

Four specimens of BeO fueled with 70 w/o of fully enriched UO_2 and two specimens of BeO fueled with 80 w/o of fully enriched UO_2 were sealed in separate cans of Hastelloy X and irradiated in Capsule AGN-38-1 at the MTR. These specimens were irradiated to uranium-235 burnups of about 10 per cent at temperatures ranging from 1300 to 1700 F. The evaluation of these irradiated specimens is to be based on changes in dimensions, microstructure, and visual appearance, on X-ray diffraction data and on determinations of burnup and fission-gas-retention properties.

Progress Reported Previously

The postirradiation examination of the specimens irradiated in Capsule AGN-38-1 was initiated in June, 1961. Samples of the gas were collected and analyzed from the main body of the capsule and from five of the six specimens contained in the capsule. Specimen 11, containing 80 w/o UO_2, ruptured during the irradiation. The remaining five specimens were in good condition. Dimensional measurements indicated that the diameter of Specimen 11 increased approximately 10 per cent, while the diameter of the other five specimens increased less than 1 per cent.

Each specimen was gamma scanned to determine the relative gamma activity as a function of specimen length and to assist in selecting specimens for burnup analysis and microstructural examinations. Visual examination of the fuel exposed during sectioning revealed that radial cracking had occurred with some instances of circumferential cracking. Preliminary examination of the microstructure of both fuel compositions indicated that the UO_2 was not appreciably affected by irradiation. There was some indication, however, that the BeO matrix had been embrittled in areas surrounding the UO_2 particles.

Work This Period

Analysis of the dosimeter wires and burnup samples has been completed, and calculations to determine specimen burnups from these data are being conducted.

Plans for Future Work

Burnup calculations will be completed and fission-gas-release rates will be determined. Also, X-ray diffraction studies and additional microstructural examinations will be completed. From these data and those already obtained, a final evaluation of the irradiation stability of these fuel materials will be performed.

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

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

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 on bond and cladding properties after bonding.

Progress Reported Previously

Studies of the effect of surface condition and bonding parameters on the self-bonding of beryllium have been made. Specimens containing UO_2 cores in contact with beryllium have been bonded at various parameters to determine the compatibility of beryllium and UO_2. Additional specimens were bonded to determine the compatibility of beryllium with various barrier materials.

Work This Period

A topical report of the work accomplished to date is being prepared. A specimen containing two plates of machined beryllium with a surface roughness of 210 μ in. rms has been bonded at 1650 F for 4 hr at 10,000 psi. This specimen is being evaluated to determine the effect of roughened surfaces on self-bonding of beryllium.

Plans for Future Work

Beryllium components which have been recently received will be used to continue the self-bonding studies and to fabricate fuel-plate specimens incorporating UO_2 cores.

RWD:CRT/all