PROGRESS RELATING TO CIVILIAN APPLICATIONS
DURING SEPTEMBER, 1961

by

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BMI-1537 "Electroplates on Thorium and Uranium for Corrosion Protection and to
Aid Joining", by John G. Beach and Charles L. Faust.

BMI-1538 "Determination of Oxygen in Sodium at Concentrations Below 10 PPM", by
Daniel R. Grieser, George G. Cocks, Elton H. Hall, William M. Henry,
and John McCallum.

BMI-1539 "Development of Container Materials for the Fused Chloride-Electrolytic
Fuel-Recovery Process", by Paul D. Miller, Earl L. White, David C.
Drennen, Carl H. Lund, Herbert J. Wagner, A. M. Hall, and
Walter K. Boyd.

BMI-1541 "Progress Relating to Civilian Applications During August, 1961", by
Russell W. Dayton and Clyde R. Tipton, Jr.
Research having the general objective of providing basic structural material and component support to the overall Atomic Energy Commission reactor-development program is reported in the following section.

**High-Pressure High-Temperature Solid-State Studies**

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

**Objective**

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

**Program Approach**

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

**Progress Reported Previously**

Face-centered-cubic type phases with lattice parameters ranging from 5.23 to 5.38 Å were found in U₃O₈-MgO, U₃O₈-CaO, U₃O₈-ZnO, and U₃O₈-CdO mixtures run at 60,000 atm and 1200 °C. The cubic phases were not found in samples of these mixtures run at 1200 °C and normal pressure.

**Work This Period**

The above-mentioned samples, which exhibited face-centered-cubic phases had a U/M ratio of 1:1, where M is the added metallic ion. Mixtures were examined with a U/M ratio of 3:1. Samples from these mixtures run at 60,000 atm and 1200 °C were also face-centered cubic. The X-ray diffraction pattern of the U₃O₈-CaO with a uranium/calcium ratio of 3:1 also showed all the lines of a simple cubic phase with a lattice constant of 5.38 Å. In some instances, lines corresponding to both the face-centered-cubic and simple cubic phases were doublets, indicating that both the face-centered-cubic phase and simple cubic phase were present. Generally, the lattice constants of the face-centered-cubic and simple cubic phases were so close it was difficult to say whether the material was single-phase. The simple cubic pattern has been found in several samples with a uranium/calcium ratio of 3:1. It has not been seen in any samples with a uranium/calcium ratio of 1:1. It is still quite pronounced in a 2:1 mixture.

The simple cubic pattern has not yet been seen in any sample with a MgO, ZnO, or CdO additive. In the case of CdO, this is interesting because calcium and cadmium have nearly the same ion radius and form many analogous compounds. The simplest explanation for the simple cubic phase is that calcium ions are going into the corners and uranium ions into the face centers. The ideal uranium/calcium ratio for this structure is 3 to 1. Since the presence of superlattice lines depends on the difference in scattering between the corner and face-centered ions, these lines might well be detectable in the U₃O₈-CaO sample but invisible in the U₃O₈-CdO sample, where the difference in scattering is much less.
High-pressure samples with a U/M ratio of 1:1 were heated in air at 1000 C. The U3O8-CdC, U3O8-ZnO, and U3O8-MgO samples reverted to U3O8 and added oxide. The U3O8-CaO end product converted to an unidentified phase at present undetected in any other samples.

Plans for Future Work

U3O8-Al2O3 samples will be run with boron nitride gaskets and cylinders, which are now available. The end products will be compared with those obtained with pyrophyllite. U3O8-CaO will be run at lower pressures to determine the effect of pressure on the formation of the phase with the simple cubic pattern.

ZrO2 exists in a face-centered-cubic modification. The effect of pressure and of combined pressure and added oxides on the existence of the face-centered-cubic modification will be examined.

Irradiation-Surveillance Program on Type 347 Stainless Steel

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

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

Program Approach
Subsize tensile, cyclic-strain fatigue, and impact specimens of AISI Type 347 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 x 10^21 nvt (fast). The information thus obtained will be used to predict the safe operating life for in-pile loops constructed of this material.

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

Work This Period
Ten capsules are being irradiated in the I-13 position of the ETR. The ETR has just completed Cycle 39, which was initiated on July 16. To date, the capsules exposed in the maximum fast-flux positions of the ETR have accumulated exposures of about 9.0 x 10^{21} nvt.

Plans for Future Work
The ten capsules will continue to be irradiated in the ETR. Periodically, capsules will be discharged from the reactor for testing at exposures ranging from about 1.0 to greater than 3.0 x 10^{22} nvt.
**Investigation of the Creep Properties of Zircaloy-2 During Irradiation at Elevated Temperatures**

F. R. Shober and P. B. Shumaker

**Objective**

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

**Program Approach**

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

**Progress Reported Previously**

An instrumented capsule, described in an earlier progress report (BMI-1430), was designed, constructed, inserted in a core position of the WTR, and irradiated for one reactor cycle. The test temperature, 650 F, was maintained by a combination of electrical and gamma heating. As the result of heater failures, excessively high gamma heating in the specimen-grip section, and premature failure of the test specimen in the grip section, the influence of irradiation on the creep behavior of Zircaloy-2 could not be evaluated. The results were described in an earlier report (BMI-1541). A second in-reactor capsule was designed and included modifications to improve the heat-transfer characteristics of the grip sections of the test specimen. The fabrication, construction, and bench tests of this capsule were undertaken.

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 a number of factors which include: (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. The influences of grain size, the hydrogen content, the amount of solute retained in solution by heat treatment, and the second-phase distribution were studied.

**Work This Period**

The assembly of the in-reactor capsule was completed, and bench tests were made. This capsule does not contain a LVDT for deformation measurement during test. The completed capsule was shipped to the WTR.

Additional short-time tensile tests were made at 650 F to study the effect of solute concentration on the yield point. Preliminary results indicated that furnace cooling from 1500 F prior to tensile testing produces a yield point of larger magnitude than when the specimens are water quenched from 1500 and 1600 F.

**Plans for Future Work**

The in-reactor capsule will be inserted into the WTR and the irradiation experiment started in October. Some additional tensile tests will be made on Zircaloy-2 at 650 F to further study the influence of grain size, of solute-element concentration, and of second-phase particle size and distribution upon the yield point.
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 to assess the durability of preformed films of known composition and thickness.

Progress Reported Previously
The friction and wear between slowly rubbing molybdenum surfaces in sodium environments were 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) were described in Paper No. 61-LUBS-16 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 behaviors of the surface films are altered by changes in environmental conditions.

Recent results for 2-1/4 Croloy also follow this pattern of decreased friction after sodium is added. Stellite No. 6 did not indicate the same clear-cut improvement. For this material the friction values with sodium lubrication were very erratic and the material exhibited coefficients generally in the same range as obtained with dry sliding.
Work This Period

Friction experiments were completed for specimens of nitrided steel. Average coefficients of friction ranged between 0.20 and 0.45 at temperatures from 80 to 750°F. At higher temperatures — to 1200°F — friction values in the sodium-exposed specimens remained in the same range, whereas friction coefficients for the dry specimens increased to maximum values of \( \mu = 1.00 \). These increasing trends were reversed with a decreasing temperature regime.

Plans for Future Work

Friction experiments will be continued with specimens of Chromallized 52-100 steel. This is the last metal in the group of specially selected structural and bearing materials. Experimentation on improved data-taking techniques, as applied to the slow-speed friction apparatus, will be initiated.

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

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

Objective

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

Program Approach

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

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

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

Work This Period

A strontium-90 source has been prepared and mounted on a beta detecting unit. The strontium is contained in a cup to shield the detector from direct betas. This combination source-detector will be mounted on a specimen test jig now being constructed. The test jig is designed to move the fuel slug past the source-detector at a constant speed with a screw-type motion in order that the entire surface of the slug can be scanned.
Plans for Future Work

Surfaces of fuel slugs known to be of sound quality will be surveyed to provide baseline back-reflection patterns. Slugs suspected of containing areas of excessive uranium and nickel penetration will then be surveyed. Areas where flaws are indicated will be sectioned and examined metallographically to determine the type of flaw capable of detection. Variables to be investigated include test speed and distance of the source-detector from the slug surface.
STUDIES OF FUELS (AEC-DRD)

R. F. Dickerson

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

Irradiation Studies of Niobium-Uranium Alloys

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

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

Program Approach
A program has been in progress to investigate and determine those physical, mechanical, and corrosion properties of niobium-uranium alloys which are of interest in evaluating their usefulness as reactor fuels. These studies have been completed and the irradiation behavior of these alloys is now being investigated. For this purpose, 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 were presented in two topical reports, BMI-1400 and BMI-1536. The planning of and construction of capsules for the irradiation tests were discussed in an earlier progress report, BMI-1464.
Annealing studies of irradiation control samples at 1600 and 1800 F were completed. Examination of the enriched control samples heat treated in sodium showed no appreciable attack after 2400 hr. The first capsule, WR-20, was inserted in the WTR during the shutdown for Cycle 14. During the first cycle the capsule operated with a specimen center-line temperature of 1100 F. A 45 volume per cent neon-helium mixture was introduced into the capsule annulus to raise the specimen center-line temperature to 1600 F during the next cycle.

Work This Period
Microstructural examination of the enriched control specimens annealed at 1600 and 1800 F showed slight grain growth due to the prolonged time at temperature (2400 hr), but no other apparent changes.
A malfunction in the inlet valves of capsule WR-20 occurred prior to Cycle 16 and has prevented the mixing of neon and helium with a resultant lowering of the specimen center-line temperature to around 1300 F. During the shutdown for Cycle 17 attempts will be made to free the valves by exerting pressure on the valves and, if this is unsuccessful, by employing a slight vacuum on the exhaust side of the valves. If the valves cannot be opened an attempt to move the capsule to a higher flux position will be made.

Plans for Future Work
A second capsule containing 15 fuel pins for irradiation at 1880 F has been shipped to the WTR for insertion during the shutdown for Cycle 17 (October 5 to 7, 1961). This
capsule has been modified slightly in the following ways: the gas annulus has been increased from 12 to 15 mils to increase the resistance of the heat path, and the valves have been removed to eliminate the problem encountered in Capsule WR-20.

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

Development of Uranium-Containing Thorium Compounds

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

Objective
To investigate thorium-uranium compounds with the aim of developing high-temperature fuels for thermal breeder reactors, and to determine those properties of the compounds which may indicate their suitability for various reactor applications.

Program Approach
Thorium-uranium aluminides, beryllides, borides, carbides, nitrides, silicides, and sulfides were selected as promising materials for study using melting points, thermal-neutron cross sections, thorium (and uranium) densities, and crystal structures as criteria for selection. These compounds have been evaluated as to ease of preparation, corrosion resistance in various media, and mechanical and physical properties. The four most promising compounds have been selected for more detailed study, primarily on the basis of ease of preparation by casting as determined during the first year of study. These compounds and compositions are (Th,U)B₄, (Th,U)Be₁₃, (Th,U)C, and (Th,U)C₂.

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

Three cylindrical casting (3/8 in. in diameter by 2-1/2 in. long) of each of the four compounds selected for detailed study were cast for use in property determinations. Radiographs of these castings indicated that the (Th,U)C₂ castings were sound; however, two each of the other three compositions showed cracks.

Work This Period
Additional castings of the (Th,U)C compound were radiographed and were found to be crack-free. The preparation of (Th,U)B₄ material has not been completed. Casting of the (Th,U)Be₁₃ compound has been delayed until suitable beryllium of low gas content can be obtained.

The sound castings of the (Th,U)C compound are being prepared for use in property studies.

Plans for Future Work
As castings are produced specimens will be prepared for various physical- and mechanical-property studies. These include thermal-conductivity, thermal-expansion, hot-hardness, and compressive-strength measurements and oxidation studies and compatibility tests.
A Study of Plutonium-Containing Fuel Alloys

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

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

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

Progress Reported Previously
Cast uranium-niobium-plutonium and uranium-niobium-plutonium-zirconium alloys were found to exhibit cored microstructures, as reported last month in BMI-1541. Hardness of the cast ternary alloys increased with increasing plutonium content (BMI-1534), while the quaternary alloys exhibited varying hardnesses (BMI-1541).
A button of PuSi₂, prepared by arc melting stoichiometric proportions of plutonium and silicon, was found to be essentially single-phase (BMI-1541). The PuSi₂ was prepared as a preliminary step in an attempt to prepare a niobium-30 volume percent PuSi₂ alloy.

Work This Period
Uranium-niobium-plutonium and uranium-niobium-plutonium-zirconium alloys heat treated 5 hr at 800 C and furnace cooled exhibit transformed microstructures containing two and possibly three phases. The heat treatment apparently was not sufficient to completely homogenize the alloys, as evidence of the cast dendritic structure was still present. Hardnesses of the heat-treated alloys are listed below.

<table>
<thead>
<tr>
<th>Alloy Composition (Balance Uranium), w/o</th>
<th>DPHN</th>
</tr>
</thead>
<tbody>
<tr>
<td>10 niobium-5 plutonium</td>
<td>438</td>
</tr>
<tr>
<td>10 niobium-10 plutonium</td>
<td>525</td>
</tr>
<tr>
<td>10 niobium-15 plutonium</td>
<td>551</td>
</tr>
<tr>
<td>10 niobium-20 plutonium</td>
<td>551</td>
</tr>
<tr>
<td>10 niobium-10 plutonium-5 zirconium</td>
<td>599</td>
</tr>
<tr>
<td>10 niobium-10 plutonium-10 zirconium</td>
<td>420</td>
</tr>
<tr>
<td>15 niobium-15 plutonium-15 zirconium</td>
<td>370</td>
</tr>
<tr>
<td>20 niobium-10 plutonium-10 zirconium</td>
<td>402</td>
</tr>
</tbody>
</table>

The uranium-10 w/o niobium-5, -10, -15, and -20 w/o plutonium alloys were given an additional heat treatment of 30 min at 800 C and oil quenched. After this heat treatment the hardnesses of these alloys were 356, 371, 371, 402 DPHN, respectively. As metallographic examination of these alloys is not complete, it is not clear if the increased hardness which accompanies increasing plutonium content is a reflection of decreasing gamma stability or of solid-solution hardening by the plutonium.
On the basis of hardness measurements, 90 per cent cold-reduced thorium-5 and -10 w/o plutonium alloys appear to recrystallize within 3 to 5 min at 600 C. Pronounced
decreases in hardness accompanied increasing annealing times up to 5 min at 600 C. A slight decrease observed in hardness when the annealing time was extended to 10 min is probably associated with grain growth.

Master-alloyed PuSi$_2$ has been arc cast with sufficient niobium to provide a nominal niobium-30 volume per cent PuSi$_2$ alloy. The prealloying step was introduced in an attempt to control composition more closely. The oxidation resistance of PuSi$_2$ appears to be poor, as an oxide scale formed on a section of a button exposed to air for several days.

Plans for Future Work
Investigations of uranium-niobium-plutonium and uranium-niobium-plutonium-zirconium alloys will continue with emphasis on phase identification and determination of gamma stability.

The niobium-30 volume per cent PuSi$_2$ will be evaluated by metallography and by X-ray diffraction analysis. Further studies of niobium-plutonium alloys will be dependent on the findings of these studies.

**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 are to be irradiated in the BRR at temperatures to 3000 F. The atmosphere surrounding the irradiation samples will be a streaming mixture of purified helium and hydrogen. Released fission gases will be continuously transported in the gas stream to a single-channel gamma-ray spectrometer for determination of fission-gas concentrations in the streaming atmosphere. Following this continuous activity monitoring, the fission gases will be extracted from the atmosphere stream and concentrated by adsorption on refrigerated charcoal. Gamma-ray spectrometric analysis of the concentrated fission gases will establish the temperature and time dependence of fission-gas release rates.

Progress Reported Previously
Equipment-design experiments were continued in order to improve the high-temperature reliability of the beam-tube furnace. An electrical-resistance furnace with tantalum-wire resistor was selected for use in future experiments in the range up to 3000 F. The program of UO$_2$ irradiations and released fission-gas analysis was resumed with an irradiation of a 93 per cent dense UO$_2$ compact at 600 F. Evaluation of experimental results was incomplete.

Work This Period
The evaluation of fission-gas release measurements obtained during the 600 F irradiation was continued. The same UO$_2$ sample was irradiated in subsequent experiments at 1500 and 1900 F. A similar UO$_2$ sample was irradiated at 2300 F. Estimated
neutron exposure for each of these experiments was $10^{18}$ nvt. Presently, an irradiation at 2700 F is under way. Two experimental methods have been applied to the analysis of released fission-product gases: direct gamma-ray spectrometric determination of krypton and xenon mixtures is being supplemented by a technique in which krypton isotopes are separated from the xenon isotopes by gas chromatography before gamma-ray analysis. Calculation of fission-gas release rates for these experiments and evaluation of the resultant data are continuing.

Plans for Future Work

The experiment now in progress at 2700 F will be completed and will be followed by an experiment at 3000 F. At the completion of this series of experiments, post-irradiation studies will include $UO_2$ sample burnup measurement and metallographic examination.

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 recent 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 capsule was 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 $2 \times 10^{14}$ nvt, and that the desired burnup can be reached in 50 days (three MTR cycles). The capsule, designated as Capsule BMI-41-1, was inserted into an MTR core position during the week of September 4, 1962 (shutdown for MTR Cycle 162).

Work This Period

The irradiation of Capsule BMI-41-1 is continuing. The capsule will be irradiated for three MTR cycles with an expected discharge date of November 6, 1961.
Plans for Future Work

Preparations have been initiated for the postirradiation examination of the nine specimens being irradiated in Capsule BMI-41-1. These examinations will include visual inspection, dimensional-distortion examination, and study of the changes in selected structural properties.

Irradiation Study of Metastable Beta-Phase Uranium Alloys

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

Objective

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

Program Approach

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

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

Progress Reported Previously

The irradiation of the second capsule was begun during Cycle 156 in Position A-26-NE of the MTR, and the capsule remained in this position through Cycle 158. During Cycle 159 the capsule was irradiated in Position A-20-SE. During these cycles, the specimen array was not located across the peak flux and nonuniform specimen temperatures resulted. Monitored temperatures for the top-zone specimens ranged between 425 and 525 F and for the bottom specimens between 625 and 700 F. Heat-transfer considerations for the capsule design indicate that a monitored temperature of 625 F corresponds to a specimen center-line temperature of approximately 850 F.

During Cycles 160 and 161, the capsule was irradiated in Position A-20-NE, and more uniform capsule temperatures were achieved (in the neighborhood of 560 F at the top zone and about 625 F at the bottom zone). Thus, it appears that during the last two irradiation cycles, the center-line temperatures of all specimens were close to or within the 800 to 850 F range.

Work This Period

The capsule was discharged from the MTR on September 4, 1961, after six cycles of irradiation. Estimates based on the available temperature data indicate that the fission burnup accrued by the enriched specimens was about 0.3 per cent of the total uranium atoms originally present in the case of specimens located in the top zone of the capsule and about 0.4 per cent in the case of the bottom-zone specimens. The capsule was received at the Battelle Hot-Cell Facility.
Metallographic examination of sectioned enriched control specimens has shown that all of the gamma-quenched alloys contained a significant number of cracks. In addition, the beta-quenched uranium-0.3 w/o chromium alloy exhibited cracks. Of the specimens irradiated in the BRR and examined metallographically, the enriched uranium-0.3 w/o chromium alloy and the normal uranium-0.3 w/o chromium-0.3 w/o molybdenum alloy, both irradiated in the gamma-quenched state to retain the beta phase, were cracked. The cracks are believed to result from stresses accompanying transformation of the beta phase.

Further X-ray diffraction studies were performed on several of the specimens irradiated in the BRR. However, the diffractometer traces did not exhibit discernible peaks.

Plans for Future Work

Capsule disassembly and postirradiation examination of the specimens irradiated in the MTR will begin shortly.

Effects of Fabrication Variables on the Radiation Stability of Uranium Carbide


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 by 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 surface-to-center line temperature increase of 500 C. The target irradiation exposure is 15,000 MWD/T burnup of the uranium.

Progress Reported Previously

In order to permit studies of the fission-gas-retention properties of these materials, the fuel specimens are to be separately sealed in thin-walled cans. A liquid-metal bond will be incorporated between the fuel specimen and can to insure good heat-transfer conditions. Five such encapsulated specimens are to be aligned axially in each capsule and immersed in a liquid metal. The irradiation capsules are to be of the double-wall type with metallic fins to provide proper heat-transfer conditions. Each capsule will be equipped with thermocouples for monitoring the can-surface temperatures and with auxiliary electrical heaters with a total capacity of 6 kw. The irradiation of these capsules will be performed in the ETR.
Work This Period

The design of the irradiation capsules and the fuel containers has been completed, and fabrication of component parts is progressing. The compatibility behavior of various materials used in these irradiation capsules is being reviewed. The fabrication of a mock-up capsule has been completed and studies of the irradiation behavior of these capsules are about to begin.

Plans for Future Work

The design and fabrication of capsules for this program will be completed, and studies of the physical properties of the fuel materials will be performed. Upon completion of the encapsulation operations, the capsules will be shipped to the ETR for irradiation.

Measurements of the Volatility of PuO₂

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

Objective

To determine the volatility of PuO₂ in the 1400 to 1750 C temperature range.

Program Approach

The volatility of PuO₂ 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 PuO₂ will be the congruently vaporizing plutonium-oxygen compound in nonreducing atmospheres. The vapor pressures are expected to be several orders of magnitude lower than those for uranium oxides under oxidizing conditions. X-ray diffraction studies will be performed subsequent to some of the transpiration runs in an attempt to identify the elevated-temperature structure of the oxides. Such interpretation of room-temperature studies may be justified on the basis of the relatively sluggish phase transformations generally associated with oxide systems.

Progress Reported Previously

The program was initiated during August.

Work This Period

Final modifications were made pertaining to temperature control using a newly installed platinum-6 w/o rhodium versus platinum-30 w/o rhodium thermocouple. In addition, a platinum-30 w/o rhodium alloy shield was fabricated and inserted to prevent heat loss from the region of the apparatus which contains the saturated vapor and thus reduce temperature gradients. Duplicate runs to determine alpha propulsion rates in a static air atmosphere at room temperature showed approximately \(2 \times 10^{-8}\) g of plutonium per hr was deposited in the condenser. This transport rate compares with a value of \(6 \times 10^{-8}\) g per hr under normal gas flow conditions at room temperature. It is concluded that alpha particles are extremely mobile, that gas flow is conducive to this mobility, and that the transport rate must be compensated for in final vapor-pressure calculations.

Plans for Future Work

Vapor-pressure determinations will be initiated at elevated temperatures. Initially, the reproducibility of results obtained in earlier work will be investigated, followed by more diversified and precise investigations.
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 two capsules, BMI-39-1 and BMI-39-2, were completed. Capsule BMI-39-1 was scheduled into the reactor for Cycle 162 and BMI-39-2 for Cycle 163.

**Work This Period**

During September, the two irradiation capsules in this program were inserted at the MTR; Capsule BMI-39-1 was inserted for Cycle 162 while Capsule BMI-39-2 was inserted for Cycle 163. The irradiation facility for the former is A-38-SE while the facility for the latter is A-38-NE.

Temperature data now available for the systems are presented in Table D-1. As Cycle 162 progressed, the thermocouples in BMI-39-1 began to behave erratically and at the end of the cycle it appeared that all were malfunctioning. Consequently, it is doubtful that direct temperature data will be available during the remainder of the experiment.

According to present plans, the irradiation of Capsule BMI-39-1 will continue for a total of three reactor cycles; specimen burnups in the neighborhood of 2 per cent of the original uranium atoms present are expected. Capsule BMI-39-2 will continue for eight reactor cycles to accrue a specimen burnup of approximately 5 per cent.

<table>
<thead>
<tr>
<th>Specimen Pair</th>
<th>Design Temperatures, F</th>
<th>Thermocouple</th>
<th>Cladding Surface</th>
<th>Specimen Thermocouple</th>
<th>Cladding Surface</th>
<th>Specimen Thermocouple</th>
<th>Cladding Surface</th>
<th>Specimen Thermocouple</th>
<th>Cladding Surface</th>
<th>Specimen Thermocouple</th>
<th>Cladding Surface</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nb-UO₂</td>
<td>1570</td>
<td>1620</td>
<td>2750</td>
<td>1600</td>
<td>1650</td>
<td>2800</td>
<td>1420</td>
<td>1470</td>
<td>2600</td>
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<td>Cr-UO₂</td>
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<td>1700</td>
<td>1750</td>
<td>2800</td>
<td>1460</td>
<td>1510</td>
<td>2450</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mo-UO₂</td>
<td>1380</td>
<td>1430</td>
<td>2750</td>
<td>1650*</td>
<td>1700</td>
<td>3100</td>
<td>1320</td>
<td>1370</td>
<td>2700</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

(a) Temperatures listed for Capsule BMI-39-1 are representative of those during the early part of Cycle 162. The starred temperature is estimated from erratic thermocouple readings.

(b) Temperatures listed for Capsule BMI-39-2 are representative of those during the first 3 days of Cycle 163.

Plans for Future Work


Fabrication of Cermet Fuel Elements

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

Objective

To develop fabrication techniques and obtain property data for cermet fuels.

Program Approach

The compatibility of UC with promising matrix materials is being evaluated and 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 are under development. UC-metal couples will be fabricated by hot isostatic pressing to insure intimate contact for heat treatments for determining metal-ceramic compatibility. Mechanical property data are being obtained on UN cermets for direct comparison with UO₂ cermets.

Progress Reported Previously

Fabrication and compatibility data for UC and UN cermets have been presented in previous progress reports. Specimens are being prepared for property studies. Tentative thermal-conductivity values on an 80 volume per cent UN-chromium cermet of 89.5 per cent of theoretical density as well as electrical-resistivity data measured concurrently with the thermal conductivity were presented last month in BMI-1541.

Work This Period

Two samples of 80 volume per cent UN-chromium and three samples of 80 volume per cent UN-molybdenum have been gas-pressure bonded for 3 hr at 2600 F under a pressure of 10,000 psi. After machining the containers from the samples, the chromium and molybdenum cermet densities were measured, respectively, at 92.0 and 93.9 per cent of theoretical.

In addition, UC cores of 4.8 and 5.2 w/o carbon have been assembled in tantalum tubes. Metal cores of chromium, molybdenum, and vanadium were placed in the tubes adjacent to the UC. Tantalum end plugs were welded in place, resulting in a tantalum-jacketed UC-metal couple. The assemblies were gas-pressure bonded for 2 hr at
2200 F under 10,000 psi to insure intimate contact between the UC cores and the metal core being studied for compatibility. Several of the UC-chromium cores were lost during welding, apparently as a result of heating of the chromium-tantalum interfaces above the eutectic temperature. If compatibility results are favorable between UC and chromium, additional samples will be prepared.

Plans for Future Work
Thermal-conductivity measurements will be made on one of the 80 volume per cent UN-chromium cerments. The remaining UN cerments will be used to obtain mechanical-property measurements. Heat treatments will be conducted on the UC-metal coupled at temperatures ranging from 2200 to 3000 F to determine compatibility.

Development of Low-Cost Tubular Fuel Elements
S. J. Paprocki, R. J. Carlson, and E. G. Smith, Jr.

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

Program Approach
Several approaches are under current investigation. One of these involves the assembly of concentric core and cladding components followed by densification, bonding, and sizing by techniques such as explosive forming or pressure bonding. Various loading techniques are being investigated for incorporation of the core powder material into the annulus formed by two concentric stainless steel cladding tubes.

In another approach, multiple-cored fuel plates are fabricated in a conventional manner with each core running the length of the plate. Subsequent to rolling, two such plates are lined up core-for-core and joined together by seam welding in such a manner that the areas between the cores can be expanded to form tubular shapes. The cores from each plate make up half of the circumference of the tubular cross section. Mechanical methods are used to open up the tubular sections in a drawbench operation.

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

Compaction and sizing using explosives resulted in tubes which had the most uniform over-all dimensions as well as smooth inside and outside surfaces. A special two-part split die is used to shape outside of the tubes. The explosive is placed inside a mild steel sleeve that is inserted in the fuel tubes prior to detonation. Use of this sleeve helps to densify the powder core more uniformly and to produce a smooth inside surface. It is subsequently removed by acid pickling. Compacted and sized fuel tubes are then loaded into an autoclave and gas-pressure bonded at 2100 F and 10,000 psi for 3 hr. The wrought end plugs are then removed and the bonded tubular fuel element is complete. Tubes approximately 6 in. in length have been produced to date, but a larger explosive die is being constructed to allow tubes 2 ft in length to be made.
In the multiple-tube assembly concept, seam-welding studies using stainless steel flat plates indicated the need for more rigid fixtures to clamp the plates during welding. Such clamping is necessary to minimize warpage and to more accurately position the seam welds along the edge of the fuel cores. A consequence of nonparallel seam welds is nonuniform expansion of the tube sections when mandrels of increasing diameter are drawn through the welded composites.

Work This Period
Fuel plates employing two cores each of a stainless steel-30 w/o UO₂ dispersion fuel were fabricated to 0.027 in. using the conventional picture-frame technique. The four plates were aligned core for core in two-plate composites with the use of radiographs. Seam welds were made along the side of the cores as described in the previous monthly report (BMI-1541). Expansion of the fuel tubes in the composites resulted in rupturing of the fuel after partial expansion. This was at first attributed to the cold rolling (20 per cent) of the fuel plates during their fabrication. The remaining composite was given an anneal to remove the effects of cold work. Further expansion was made possible, but rupturing occurred when the final and largest mandrel was being drawn through. Previous welded assemblies had been expanded without rupturing, but a stainless steel-20 w/o UO₂ dispersion had been used.

Plans for Future Work
Further tests are planned which will employ stainless steel-20 w/o and -30 w/o UO₂ dispersions to determine the influence of UO₂ loading on rupturing during expansion of the fuel tubes.
A die is being constructed that will make possible the explosive compaction of elements up to 2 ft in length. A series 2-ft-long elements will be fabricated and these elements will be evaluated for straightness, roundness, concentricity, and core-thickness uniformity. Other elements in the series will be defected and checked for waterlogging in a steam autoclave.

**Gas-Pressure Bonding of Ceramic Fuel Elements**

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

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

Program Approach
This program concerns two efforts which represent cooperative programs with Hanford, and Gladding, McBean & Co. In these studies, which will be conducted simultaneously, the gas-pressure-bonding process as combined with the vibratory packing technique and the UO₂ sheet-rolling technique will be evaluated.

Progress Reported Previously
In previous studies, the evaluation of various types of uranium dioxide powders was established on the basis of their initial compacting and pressure-bonding characteristics. These results and the development of pressure-bonding techniques for various fuel geometries were discussed in a phase report, BMI-1475. More recently preliminary process specifications for pressure bonding several basic Type 304 stainless
steel-clad UO$_2$ fuel-element shapes have been partially developed. Four pressure-bonded fuel rods have been supplied for irradiation in the VBWR.

Work This Period

A flat-plate fuel assembly containing rolled UO$_2$ platelet cores fabricated by Gladding, McBean & Co. was pressure bonded for 3 hr at 2100 F and 10,000 psi. Metallographic examination revealed significantly less cracking in the UO$_2$ than noted in previous composites. This effect was attributed to the slightly higher density of the more recent material (94 to 96 per cent of theoretical). Final density and stoichiometry determinations are presently being made.

Three additional assemblies were prepared for further pressure-bonding tests. With this group of specimens the effects of slight pressure-cycle variations on the extent of cracking in the oxide cores will be examined. These assemblies are currently being bonded.

Plans for Future Work

Further evaluation of pressure-bonded rolled UO$_2$ sheet materials will be conducted. Pressure bonding of vibration-packed UO$_2$ fuel rods will be initiated.

Factors Affecting Pressure Bonding

D. C. Carmichael and J. W. Spretnak

Objective

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

Program Approach

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

Progress Reported Previously

The mechanism for the self-bonding of copper has been established and the effects of pressure, temperature, time, 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

Study is being continued on the relationships between the hot hardness of a metal at a given temperature, the applied load in hot pressing a metal couple, and the resulting area of contact between the mating surfaces. Platelets of pure nickel which have surfaces shaped to a roughness of 500 µm. rms are being hot pressed in vacuum into annealed copper platelets with smooth surfaces. The results are being compared with those previously obtained using copper-copper couples. An additional copper-nickel specimen has been hot pressed at 1000 F for 5 min using a pressure of 10,000 psi. The resulting area of contact in the couple has been measured using a Hurlbut counter and is given in Table D-2 along with the previously obtained results for comparison. Additional specimens are being run to check the results obtained at the two highest pressures, since small amounts of bond-line contamination or nonuniform pressure applications could significantly affect the observed results.

### Table D-2. Effect of Hot-Pressing Pressure on the Fractional Area of Contact Produced in Copper-Nickel Couples

<table>
<thead>
<tr>
<th>Applied Pressure, psi</th>
<th>Resulting Fractional Area of Contact</th>
</tr>
</thead>
<tbody>
<tr>
<td>2,000</td>
<td>0.18</td>
</tr>
<tr>
<td>4,000</td>
<td>0.30</td>
</tr>
<tr>
<td>8,000</td>
<td>0.55</td>
</tr>
<tr>
<td>10,000</td>
<td>0.79</td>
</tr>
<tr>
<td>12,000</td>
<td>0.82</td>
</tr>
</tbody>
</table>

Note: The specimens were vacuum hot-pressed at 1000 F for 5 min using the pressures indicated.

The experimental results obtained are being examined in terms of theoretical relationships between the applied pressure and resulting area of contact, particularly the Hill-Lee-Tupper* relationship. Their treatment is based on the penetration of a hard wedge into a ductile flat surface. This model is much more closely approximated by the nickel-copper specimens than the copper-copper couples previously bonded. Comparison of calculated data and the experimental data for both types of couples indicates that there is a systematic error in the application of the theory at least up to a point where 0.6 of the possible area of contact has been produced. Examination of the nickel-copper couples shows that the included angle of the wedge-shaped asperities does not change appreciably during the deformation, which was not the case with the copper-copper couples and complicated the analysis. It now appears that the systematic variation of the experimental data from the theoretical results is due to the fact that the asperities on the machined surfaces have somewhat flat rather than sharp tips. This produces an appreciable area of contact between the surfaces at essentially zero load. Account can be made of this in applying the theory and the experimental and theoretical results are then in agreement. Additional results will have to be obtained before the analysis can be extended to the higher experimental pressures.

Plans for Future Work

The investigation of the bonding of copper-nickel specimens will be continued to study the deformation relationships involved. Additional results will be obtained at high pressures in order to extend the analysis of experimental and theoretical results to greater degrees of deformation. Later, the formation and subsequent elimination of the bond interface will be investigated for other metals, including aluminum.
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 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 due to tip erosion.

The effects of various atmospheres on the carbon content of melts produced in this manner were investigated. The atmospheres of interest were a dynamic vacuum, an atmosphere of helium at 1/3 atm abs, and a mixture of 3 parts helium and 1 part argon at 1/3 atm abs. The visual and metallographic evaluations of these melts were reported previously.

Work This Period

Chemical analyses of samples from the 1-in.-diameter by 6-in.-long castings, weighing approximately 1 kg each, produced under various atmospheres were completed. These analyses are given in the Table F-1.
TABLE F-1. ANALYSES OF SERIES OF SKULL-CAST INGOTS

<table>
<thead>
<tr>
<th>Furnace Atmosphere</th>
<th>Ingot</th>
<th>Specimen Location</th>
<th>Carbon Content, w/o</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dynamic vacuum</td>
<td>7</td>
<td>Top</td>
<td>5.16</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Bottom</td>
<td>5.19</td>
</tr>
<tr>
<td></td>
<td>8</td>
<td>Top</td>
<td>5.22</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Bottom</td>
<td>5.21</td>
</tr>
<tr>
<td></td>
<td>9</td>
<td>Top</td>
<td>5.13</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Bottom</td>
<td>5.16</td>
</tr>
<tr>
<td>1/3 atm of helium</td>
<td>10</td>
<td>Top</td>
<td>5.19</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Bottom</td>
<td>5.18</td>
</tr>
<tr>
<td></td>
<td>11</td>
<td>Top</td>
<td>5.09</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Bottom</td>
<td>5.14</td>
</tr>
<tr>
<td></td>
<td>12</td>
<td>Top</td>
<td>5.40</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Bottom</td>
<td>5.12</td>
</tr>
<tr>
<td>1/3 atm of 3 parts helium and 1 part argon</td>
<td>13</td>
<td>Top</td>
<td>4.95</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Bottom</td>
<td>4.95</td>
</tr>
<tr>
<td></td>
<td>14</td>
<td>Top</td>
<td>4.96</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Bottom</td>
<td>4.91</td>
</tr>
<tr>
<td></td>
<td>15</td>
<td>Top</td>
<td>4.97</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Bottom</td>
<td>4.91</td>
</tr>
</tbody>
</table>

These analyses substantiate the visual observations concerning tip erosion which indicated that the addition of argon to the helium atmosphere substantially reduced tip erosion during melting. The very narrow spread in carbon content from melt to melt within a given series of melts suggests that, with proper adjustment of the starting composition, any of the atmospheres investigated will yield castings of predictable carbon concentration.

A series of 12 melts was prepared in the modified skull-type arc-melting furnace utilizing the recently installed charging mechanism described in the previous monthly report (BMI-1541). The first melt in this series was designed to evaluate the ability of the furnace to prepare skulls from virgin material. A charge of 4 kg of unalloyed material was placed in the skull and melted. The molten pool obtained during melting was extremely small because of the low power level of 3700 amp and to the unalloyed uranium adjacent to the crucible wall which was not incorporated into the skull. The inability of the arc to penetrate the charge and completely alloy the virgin material indicated a need for an initial charge of prealloyed material to form a bed upon which a skull could be deposited.

The second melt in the series was designed to yield a homogeneous skull with a predictable carbon level. An initial charge of 4 kg of prealloyed material containing 5.0 w/o carbon was melted in the crucible, and, when the molten pool reached a size approximately one-half the diameter of the crucible, elemental additions of uranium and 4.5 w/o carbon were added slowly to the melt. A power level of 3700 amp proved insufficient to supply the required superheat to completely melt the charge at the rate
the material was added. The casting from this skull on analysis showed 4.5 w/o carbon, and the residual skull proved to be inhomogeneous. The next six melts were made in a similar manner using a charge composition of uranium-4.8 w/o carbon and adjusting the rate of addition and power level with each melt. In every case some unmelted material was evident in the skull after pouring the castings, and the composition of the castings was approximately the composition of the starting charge of uranium-4.8 w/o carbon. It is believed that the material that was consolidated consisted predominantly of uranium, and therefore, the tip erosion only compensated for the unmelted carbon.

The four final melts were made using a power level of 4300 amp and 28 v. The initial skull was built up from 4 kg of prealloyed uranium-5.0 w/o carbon as in the preceding melts. However, the additions to the skull of 4 to 6 kg of elemental uranium and carbon were not begun until the molten pool covered approximately 7/8 of the crucible area, and the rate of additions was controlled so that it varied throughout the melt cycle, the rate increasing as the size of the molten pool increased. The results of increasing the power level were to increase the depth of the molten pool to approximately two times that obtained at the lower power level and to permit the complete solution of all material added to the melt. Castings 2 in. in diameter by 6 in. long with good surface definition and excellent compositional control, as determined by metallographic techniques, were produced. The castings weighed from 4.5 to 5 kg each. Radiographic examinations revealed secondary pipes in each of the castings extending approximately one-half the length. It is believed that refinements in molding techniques could improve the yield obtained from these castings.

Plans for Future Work
The series of melts described above complete this phase of the Fuel-Cycle Development Program. No further work in this area is contemplated at this time.

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 (U₂C₃), and uranium dicarbide (UC₂) 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 to 0.7 a/o of the uranium. The specimens were contained in six NaK-filled capsules. Evaluations of these irradiation specimens were based on their dimensional stability and microstructural stability and on measurements of their electrical resistivities and lattice parameters.

Progress Reported Previously
The irradiation of the six capsules of this program has been completed. The post-irradiation examinations of the specimens recovered from four of these capsules have been completed and the results reported in a previous progress report, BMI-1524. In general, it was found that a saturation of lattice strain had 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 of these four failed during irradiation.

The postirradiation examination of the specimens contained in Capsules 5 and 6 has been completed. The isotopic analyses of representative specimens from both capsules have indicated that the specimens sustained a uranium burnup of about 0.5 a/o. These specimens had experienced average surface temperatures ranging from 1100 to 1400 F. The two centrally positioned specimens of uranium-5.0 w/o carbon, essentially UC\textsubscript{2}, in Capsule 5 were completely disintegrated, and one of two specimens of uranium-6.7 w/o carbon, essentially U\textsubscript{2}C\textsubscript{3}, was partially broken. The remaining specimens from both Capsules 5 and 6 were in excellent condition with no apparent evidence of cracking or deterioration. Analysis of the gases contained in the capsules by gamma-ray spectrographic techniques indicated that about 0.32 per cent of the total krypton-85 formed in the six specimens in Capsule 6 was released. This corresponds to about 2.5 times that release expected from recoil. Analysis of the gas recovered from Capsule 5, using gamma-ray spectrographic techniques, indicated a release of only 2 per cent in spite of the fact that two of the specimens had disintegrated. Mass spectrometric analyses of the gases from Capsule 5 indicated that the gas release data for this capsule were too low.

The physical measurements of the uranium-5.0 w/o carbon specimens of Capsules 5 and 6 were presented in an earlier progress report, BMI-1541.

The microstructural studies of the uranium-5.0 w/o carbon specimens from these capsules did not reveal any detrimental effects resulting from irradiation. In most instances, a depletion of the UC\textsubscript{2} second phase was observed in a band around the outer circumference of each specimen. No microstructural changes were observed in the specimens of uranium-6.7 w/o carbon.

Work This Period

Attempts to reprepare the metallographic samples for electron microscopic studies were unsuccessful; therefore, these studies were abandoned. Since no reasonable explanation for the disintegration of the uranium-8.5 w/o carbon specimens during irradiation could be determined from the postirradiation examination, compatibility studies of UC\textsubscript{2} in NaK at temperatures near 1600 F were performed. Control specimens of identically heat treated uranium-8.5 w/o carbon alloy were found to contain precipitated graphite in the microstructure. This resulted in complete disintegration of the specimens in NaK after only 10 hr at 1600 F. Therefore, it is believed that the disintegration of the uranium-8.5 w/o carbon specimens contained in Capsule 5 was a compatibility effect rather than an effect of irradiation. The X-ray diffraction studies have been completed and will be reported in the final evaluation of the results of this program.

Results of mass spectrometric analyses of the gases collected from Capsule 6 have confirmed the release data previously reported, 0.32 per cent of the krypton-85, as determined by gamma-ray spectrometry. In the case of Capsule 5, however, the mass-spectrometer data indicate the presence of air, which suggests that some of the gas recovered from the capsule was lost through leakage from the sample vials. The gas release calculated from gamma-ray spectrometry data for the specimens in this capsule are, therefore, too low, and it will not be possible to determine the fraction of gas released from the specimens.

Plans for Future Work

A final evaluation of the effects of irradiation on the carbide specimens of this program will 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. An apparatus to allow crystal growth from the melt by the Bridgman technique is being developed.

**Progress Reported Previously**
Several high-temperature sources have been investigated, and a tungsten-resistance furnace has been selected for use in the program. Effort has been expended in improving shielding, electrode, and resistance-element design so that the 2800°C temperature necessary to melt UO$_2$ may be achieved. Temperatures in excess of 2800°C have been reached for periods of 1 hr and UO$_2$ has been successfully melted.

**Work This Period**
During September, effort was spent on determining the melting characteristics of UO$_2$ in the capped tungsten crucible. With a temperature of about 2830°C, it was shown that fusion of UO$_2$ can be obtained with the experimental conditions. A starting charge of chips and powdered UO$_2$ was removed as a large fused mass. However, the center portion still had the appearance of a sintered mass. It is now interpreted that the center may have been fused, but contained vapor (UO$_2$ or possibly dissolved oxygen) in the form of small bubbles which were frozen in during cooling. It is thought that the viscosity of UO$_2$ may be a factor in the movement of these gases.

A short-term experiment with rapid cooling from a temperature of about 2930°C showed that very large bubbles were formed in the center of the charge. It is believed that greater mobility of the gas pockets is obtained at the higher temperature.

**Plans for Future Work**
Bridgman drop experiments will be made at temperatures above 2830°C in an attempt to reduce the porosity of the crystals. Heavier heating elements will be employed in these experiments to gain greater high-temperature strength.
RADIOISOTOPE AND RADIATION APPLICATIONS (AEC-DID)

D. N. Sunderman

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

Use of Intrinsic Radioactive Tracers for Process Control

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

Objective

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

Program Approach

It is planned to determine 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. Experiments will then be performed under the optimum conditions to determine general stream activation efficiencies for different process streams as a function of neutron-source strength. 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. After an initial survey of neutron sources and source technology, a 10-curie polonium-beryllium source was ordered from Mound Laboratory. Concurrently, a combination neutron source storage shield-activation cell holder was designed and construction was started.

Work This Period

The source storage shield was completed this month. Shielding around the neutron source consists of 16-1/2 in. of borated paraffin. It is expected that this will be adequate for the 70-curie source to be obtained later in the program.

The holder for the neutron-activation cell is ready for the preliminary optimization experiments. The design provides a ready means of varying geometry during these experiments.

The 10-curie polonium-beryllium source has been received from Mound Laboratory and is ready for insertion into the storage shield.

Construction of the multiprobed beta counter has been delayed because some of the components have not been received. This detector will be assembled as soon as all parts are available.
Plans for Future Work

Next month the preliminary optimization experiments will begin. Standard solutions of a suitable manganese salt will be used in these experiments to determine the optimum geometry for the activation cell.

Graft-Polymerization Studies


Objective

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

Program Approach

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

Progress Reported Previously

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

Work This Period

During September, experimental work included: (1) continuation of the investigation of procedural techniques employed in the analytical methods used for the determination of volatile-product concentrations from irradiated polymethacrylates, (2) additional grafting studies, (3) comparison of free-radical concentrations in PMMA determined as a function of dose by EPR and by changes in number-average molecular weight, and (4) preparation of several additional vinyl polymers.

The investigation of procedural techniques for determining volatile-product concentrations from irradiated polymethacrylates was continued during the month. Earlier, evidence had been obtained which indicated that absorption and/or solubilization of the low-molecular-weight fragments had made their removal from the polymer difficult. This is particularly true in the case of the ter-butylmethacrylate polymer. Techniques have now been devised for the quantitative removal of CO, hydrogen, CO₂, and the low-molecular-weight hydrocarbons. Some difficulties are still being experienced in the case of some of the higher molecular-weight constituents, particularly the alcohols and esters. It is anticipated that the required changes can be completed during October.

*I. A. E. A. Fellow.
As described earlier, the grafting procedure has been revised to include a 24-hr extraction with water to insure complete removal of vinyl pyrrolidone monomer and homopolymer. Approximately 25 graft copolymers have been prepared at 25 C using the revised method. Nitrogen concentrations and number-average molecular weights are being determined for each copolymer. Table H-1 lists $M_N$ values obtained for a number of the copolymers. These values are generally much lower than might be expected. However, no attempt will be made to analyze the results until all of the data are available. The additional data should be obtained in time for the inclusion of a complete analysis of the results in the next report.

Free-radical concentrations as a function of dose by EPR and by changes in number-average molecular weight are compared in Table H-2. Agreement between the two methods is poor at all doses (sites calculated from molecular-weight changes being greater), and a graph of the data indicates that the two curves are diverging. This is to be expected, since the site concentration calculated from molecular-weight changes should increase regularly with dose whereas that determined by EPR, although increasing with dose, should not increase at the same rate. The retardation would be due to the increasing effect of molecular mobility on free-radical formation and/or destruction.

During the month, the preparation of several additional polymers has been undertaken. Polymethyl-$\alpha$-cyanoacrylate has been prepared by irradiation polymerization. Work has also been initiated on the $\alpha$-bromo and $\alpha$-iodo derivatives.

Plans for Future Work

During October, work will be continued in the several areas described above. In addition, the study of the chemical measurement of free-radical sites, initiated in August, will be continued. The use of DPPH alone, rather than in combination with hydroquinone, will be examined.
TABLE H-1. GRAFT-COPOLYMERIZATION STUDIES ON PMMA(a) AND VINYL PYRROLIDONE(b)

<table>
<thead>
<tr>
<th>Run</th>
<th>Weight of Base Polymer, g</th>
<th>Total Dose, rads x 10^7</th>
<th>Sites per Monomer Unit x 10^3(c)</th>
<th>Copolymerization Time(d), hr</th>
<th>MN of H2O- Insoluble Product</th>
<th>Nitrogen(e), per cent</th>
</tr>
</thead>
<tbody>
<tr>
<td>G-39</td>
<td>1.15</td>
<td>--</td>
<td>--</td>
<td>96</td>
<td>2490</td>
<td>---(f)</td>
</tr>
<tr>
<td>G-23</td>
<td>1.35</td>
<td>0.03</td>
<td>0.05</td>
<td>24</td>
<td>1315</td>
<td>---(f)</td>
</tr>
<tr>
<td>G-4</td>
<td>2.88</td>
<td>0.11</td>
<td>1.2</td>
<td>24</td>
<td>1692</td>
<td>---(f)</td>
</tr>
<tr>
<td>G-26</td>
<td>1.46</td>
<td>0.60</td>
<td>3.5</td>
<td>24</td>
<td>2560</td>
<td>---(f)</td>
</tr>
<tr>
<td>G-32</td>
<td>1.31</td>
<td>0.60</td>
<td>3.5</td>
<td>3</td>
<td>2716</td>
<td>0.42</td>
</tr>
<tr>
<td>G-30</td>
<td>1.80</td>
<td>3.60</td>
<td>8.0</td>
<td>96</td>
<td>1370</td>
<td>---(f)</td>
</tr>
</tbody>
</table>

(a) The initial molecular weight of the PMMA was 4125.
(b) In all cases, grafting was carried out at 25 C using 100 cm^3 of vinyl pyrrolidone.
(c) These values were determined by EPR measurements.
(d) These values represent the contact time between monomer and "active" polymer.
(e) This represents the per cent nitrogen in the water-insoluble copolymer. Multiplication of this value by 7.93 gives per cent vinyl pyrrolidone in copolymer.
(f) These analyses have not been completed.

TABLE H-2. SITE FORMATION IN PMMA

<table>
<thead>
<tr>
<th>Dose, rads</th>
<th>Sites per Monomer Unit x 10^3</th>
<th>By EPR</th>
<th>By MN Decrease(a)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.7 x 10^4</td>
<td>--</td>
<td>2.7</td>
<td></td>
</tr>
<tr>
<td>5 x 10^4</td>
<td>--</td>
<td>5.2</td>
<td></td>
</tr>
<tr>
<td>1 x 10^5</td>
<td>--</td>
<td>20.9</td>
<td></td>
</tr>
<tr>
<td>1.7 x 10^5</td>
<td>0.31</td>
<td>--</td>
<td></td>
</tr>
<tr>
<td>2 x 10^5</td>
<td>--</td>
<td>17.5</td>
<td></td>
</tr>
<tr>
<td>3.4 x 10^5</td>
<td>0.49</td>
<td>--</td>
<td></td>
</tr>
<tr>
<td>5 x 10^5</td>
<td>--</td>
<td>24.9</td>
<td></td>
</tr>
<tr>
<td>1 x 10^6</td>
<td>--</td>
<td>21.5</td>
<td></td>
</tr>
<tr>
<td>1.4 x 10^6</td>
<td>1.3</td>
<td>--</td>
<td></td>
</tr>
<tr>
<td>5 x 10^6</td>
<td>--</td>
<td>23.8</td>
<td></td>
</tr>
<tr>
<td>6.1 x 10^6</td>
<td>3.9</td>
<td>--</td>
<td></td>
</tr>
<tr>
<td>1 x 10^7</td>
<td>--</td>
<td>24.2</td>
<td></td>
</tr>
<tr>
<td>1.3 x 10^7</td>
<td>6.0</td>
<td>--</td>
<td></td>
</tr>
<tr>
<td>3 x 10^7</td>
<td>--</td>
<td>43.2</td>
<td></td>
</tr>
</tbody>
</table>

(a) Assuming one site per chain scission.
Research and development are being conducted on materials to be used in conjunction with construction of the NPR and operation of the PRTR 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 cycling on the creep properties of this material.

**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 \( \mu \) of mercury. Test temperatures are 550, 650, and 750 F. Creep measurements are taken approximately daily by means of an optical system. In all respects, the tests meet or exceed the requirements for creep testing specified in ASTM Standard E-139-58T.

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

**Progress Reported Previously**

All 20 tests now in progress have been in progress for times ranging from about 3000 to 33,500 hr. Therefore, data have been accumulating for some time and they have been reported in monthly progress reports from time to time. The latest summary was presented in the April progress report (BMI-1514).

**Work This Period**

Current test results on Zircaloy-2 sheet are shown in Table J-1. Since all tests are long term, there is little change in test results, other than accumulated time, from one month to the next. Comparison of the data in Table J-1 with similar data in BMI-1514 will indicate the change taking place over a period of 5 months.

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

All temperature-cycle tests have been concluded for the present but will be resumed later. These tests were made on annealed and 15 per cent cold-rolled Zircaloy-2 sheet at 550 and 650 F. Indications are that stress-temperature cycling accelerates creep, and temperature cycling alone accelerates it still more when compared with static (no cycle) tests. The major effect of temperature and stress-temperature cycling is noted at the higher stress levels with very limited effects being noted at the lower stresses.
### TABLE J-1. CURRENT STATUS OF CREEP TESTS ON ZIRCALOY SHEET

<table>
<thead>
<tr>
<th>Specimen</th>
<th>Temperature, F</th>
<th>Stress, psi</th>
<th>Time in Progress(a), hr</th>
<th>Deformation, per cent On Loading</th>
<th>Deformation, per cent At Present</th>
<th>Tentative Minimum Creep Rate, per cent per hr</th>
</tr>
</thead>
<tbody>
<tr>
<td>7-2-1</td>
<td>650</td>
<td>25,000</td>
<td>33,500</td>
<td>0.230</td>
<td>1.795</td>
<td>0.00003</td>
</tr>
<tr>
<td>7-4-3</td>
<td>750</td>
<td>12,500</td>
<td>31,000</td>
<td>0.134</td>
<td>2.465</td>
<td>0.00006</td>
</tr>
<tr>
<td>Zr-A-10</td>
<td>550</td>
<td>22,500</td>
<td>21,500</td>
<td>3.180</td>
<td>4.348</td>
<td>0.00004</td>
</tr>
<tr>
<td>Zr-A-13</td>
<td>550</td>
<td>20,000</td>
<td>19,000</td>
<td>1.511</td>
<td>2.862</td>
<td>0.00002</td>
</tr>
<tr>
<td>Zr-A-14</td>
<td>550</td>
<td>17,500</td>
<td>19,000</td>
<td>1.250</td>
<td>1.961</td>
<td>&lt;0.00001</td>
</tr>
<tr>
<td>Zr-A-15</td>
<td>650</td>
<td>20,000</td>
<td>19,000</td>
<td>2.240</td>
<td>22.78</td>
<td>0.00010</td>
</tr>
<tr>
<td>Zr-A-17</td>
<td>650</td>
<td>17,500</td>
<td>19,500</td>
<td>1.778</td>
<td>3.018</td>
<td>0.000065</td>
</tr>
<tr>
<td>Zr-A-25</td>
<td>650</td>
<td>15,000</td>
<td>17,500</td>
<td>0.971</td>
<td>1.455</td>
<td>0.000030</td>
</tr>
<tr>
<td>Zr-A-28</td>
<td>650</td>
<td>12,500</td>
<td>14,500</td>
<td>0.321</td>
<td>0.473</td>
<td>0.000005</td>
</tr>
<tr>
<td>Zr-A-16</td>
<td>750</td>
<td>12,500</td>
<td>19,000</td>
<td>0.447</td>
<td>5.955</td>
<td>0.00025</td>
</tr>
<tr>
<td>Zr-A-21</td>
<td>750</td>
<td>10,000</td>
<td>18,000</td>
<td>0.113</td>
<td>2.190</td>
<td>0.00008</td>
</tr>
<tr>
<td>Zr-A-26</td>
<td>750</td>
<td>9,000</td>
<td>16,000</td>
<td>0.082</td>
<td>0.687</td>
<td>0.00002</td>
</tr>
<tr>
<td>Zr-A-36</td>
<td>550</td>
<td>15,000</td>
<td>8,500</td>
<td>0.785</td>
<td>1.154</td>
<td>0.000005</td>
</tr>
<tr>
<td>Zr-A-31</td>
<td>550</td>
<td>12,500</td>
<td>11,500</td>
<td>0.170</td>
<td>0.535</td>
<td>0.000005</td>
</tr>
<tr>
<td>Zr-A-34</td>
<td>650</td>
<td>15,000</td>
<td>9,000</td>
<td>0.949</td>
<td>1.279</td>
<td>0.000020</td>
</tr>
<tr>
<td>Zr-A-32</td>
<td>650</td>
<td>12,500</td>
<td>11,500</td>
<td>0.265</td>
<td>0.650</td>
<td>0.000015</td>
</tr>
<tr>
<td>Zr-A-35</td>
<td>650</td>
<td>9,000</td>
<td>8,500</td>
<td>0.063</td>
<td>0.113</td>
<td>0.000005</td>
</tr>
<tr>
<td>11-A-5-1</td>
<td>550</td>
<td>20,000</td>
<td>5,000</td>
<td>0.444</td>
<td>0.631</td>
<td>0.000007</td>
</tr>
<tr>
<td>11-A-5-3</td>
<td>550</td>
<td>15,000</td>
<td>3,000</td>
<td>0.187</td>
<td>0.304</td>
<td>0.000005</td>
</tr>
<tr>
<td>11-A-5-4</td>
<td>550</td>
<td>12,500</td>
<td>4,000</td>
<td>0.129</td>
<td>0.195</td>
<td>NH</td>
</tr>
</tbody>
</table>

(a) Less than actual test time but within 500 hr.
Plans for Future Work

The 20 tests in progress are scheduled to continue. Creep data will be accumulated until it is believed 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. In addition, more tests are scheduled to be made in which the stress and/or temperature will be cycled.

Materials of Construction for the
Fused Chloride-Electrolytic UO₂ 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 800 °C equimolar solutions of sodium and potassium chlorides containing U₃O₈ and sparged with chlorine.

Progress Reported Previously

Progress in the program has been summarized in the several recent 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 interface was somewhat greater.

Work This Period

A topical report covering the preparation, corrosion properties, and metallographic studies of the alloys has been issued as BMI-1539.

Plans for Future Work

No additional work is contemplated.

Thermal Conductivity and Electrical Conductivity of UO₂

H. W. Deem and C. F. Lucks

Work on this program has been concluded. A draft of Battelle's contribution to a joint BMI-HAPO report on fabrication and irradiation of UO₂ has been forwarded to HAPO.
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 UO₂, UC, and UC₂ 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 dicarbide powder (UC₂-E₂) was found to contain 9.45 w/o total carbon and 1.62 w/o free carbon. The major phase was UC₂ with a small amount of UC present throughout and graphite as a 4-μ surface layer. In appearance, 90 per cent of the particles were found to be in the classification most closely corresponding to perfect spheres.

Work This Period

Further micro- and macrographic evaluations of 3M dicarbide (UC₂-E₁) are in progress. These evaluations will include estimates of grain size, phase constitution, and porosity, as well as a quantitative characterization as to shape.

A method of measuring pore diameter of loose powder by means of mercury intrusion under pressure is being studied. Preliminary indications show that pore diameters from 0.015 to 50 μ can be determined. Tests are now in progress on two batches of ANL natural UO₂ that have previously been evaluated as to surface area.

Neutron-activation tests have been completed on UO₂-E₃ and UC₂-E₂. Postirradiation heat treatments were conducted at temperatures of 815, 1090, and 1320 °C. Whereas the uncoated UC₂ (which had an approximately 4-μ-thick graphite coating as produced) released nearly equal amounts of Xe₁³³ during 60 min at all the temperatures, about 60 per cent of the Xe₁³³ release from UO₂ occurred at 1320 °C. The total amount of Xe₁³³ released from 1.06 g of UO₂ was 996 ppm and 5900 ppm of Xe₁³³ was released from 1.09 g of UC₂.

Plans for Future Work

Evaluation of pycnometer, sedimentation, petrographic, and Coulter counting devices will continue.
Objective
To establish conditions for coating fuel particles with pyrolytic carbon of optimum quality.

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

Progress Reported Previously
In a previous program, pyrolytic-carbon coatings were applied to UC, UC₂, and UO₂ 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.

Pyrolytic-carbon coatings prepared at temperatures above 1500°C have thus far had crushing strengths and densities similar to coatings deposited at temperatures of approximately 1400°C. These coatings show columnar grains with apparent soot inclusions. Uranium contamination of the carbon coatings prepared above 1700°C has been more of a problem than with those coatings prepared at 1400°C.

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

Lot 918A, columnar coating on enriched UC₂-E₁ (Xe¹³³ release: 2.7 ppm at 1300°C)
Lot 919A, laminar coating on enriched UC₂-E₁ (Xe¹³³ release: <0.5 ppm at 1300°C)
Lot 920A, laminar coating on enriched UO₂-E₃ (Xe¹³³ release: 0.9 ppm at 1300°C)
Lot 921, laminar-columnar mixture PyC coating on dense Al₂O₃-coated UO₂-E₂ (Xe¹³³ release: 0.5 ppm at 1300°C)
Lot 922, laminar-columnar mixture PyC coating on dense:porous Al₂O₃-coated UO₂-E₃ (Xe¹³³ release: 3.1 ppm at 1300°C).

Work This Period
Most of the preparations made to relate coating properties to processing conditions have been completed. The remaining preparations and evaluation of the coatings should be concluded by the end of October.

Three additional lots of pyrolytic-carbon-coated particles prepared at temperatures above 1500°C were coated in the inductively heated carbon-tube reactor described last month (BMI-1541).
These materials were prepared as candidates for in-pile study. They are as follows:

<table>
<thead>
<tr>
<th>Lot</th>
<th>Fuel</th>
<th>Average Coating Thickness, Cm</th>
<th>Coating Temperature, C</th>
</tr>
</thead>
<tbody>
<tr>
<td>1008A</td>
<td>U02-E3</td>
<td>47</td>
<td>1700</td>
</tr>
<tr>
<td>1009B</td>
<td>UC2-E2</td>
<td>64</td>
<td>1600</td>
</tr>
<tr>
<td>1010A</td>
<td>U02-E3</td>
<td>66</td>
<td>1600</td>
</tr>
</tbody>
</table>

The coatings of Lot 1008A appeared to be free of voids and of uniform thickness with a columnar structure. However, unexplained porosity was revealed by the acid-leach test (1.2 w/o UO₂ removed). The coatings of Lots 1009B and 1010A appeared to be uniform, free of voids and of a just perceptible laminar structure. Coating rates for these lots were higher than that for Lot 1008A (46 μg per hr versus 11 μg per hr). Acid-leach data indicated that the coatings of Lots 1009B and 1010A were less porous than those of Lot 1008A with the UO₂ removal being only 0.017 and 0.002 w/o, respectively. However, the coating contamination as determined by alpha assay was 500 ppm for Lot 1009B and only 5.5 ppm for Lot 1010A. Lot 1010A, a candidate for irradiation evaluation, is being tested for fission-gas release on postirradiation heating.

Plans for Future Work

The planned program relating coating properties and coating conditions should be concluded by the end of October. The cause of uranium contamination of coatings prepared above 1600 C will be determined and a limited amount of experimentation is planned on silicon-doped-carbon coatings during October to obtain more definitive information on the strengthening effect of the silicon addition.

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 UO₂ shot by hydrolysis of BeCl₂ 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 Al₂O₃ 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 BeCl₂ were continued. Coatings deposited at 1400°C in a Mullite reactor by both steam hydrolysis (Lot 826A) and the CO₂-hydrogen reaction (Lots 828A and 829A) appeared to be translucent, dense, and of low porosity. The 0.08 w/o UO₂ removed from a sample of Lot 826A (30 μ coating) by the standard acid-leach test indicated that this coating was essentially as nonporous as any previous BeO coating. However, in contrast, only 0.001 to 0.003 w/o UO₂ would be removed from a good Al₂O₃ coating of similar thickness by the same acid-leach test. Oxidation at 1100°C for 48 hr showed the coating of Lot 826A to be sufficiently porous to oxygen that the UO₂ equivalent to 0.7 w/o of the particles was exposed by this treatment. Oxidation for a second 48-hr period exposed the UO₂ equivalent to an additional 8 per cent of the particles. The density of this coating was approximately 97 per cent of theoretical as calculated from density of the whole particles measured by a xylene pycnometer. This would indicate that the leakage could be due to general porosity rather than to failure of a fraction of the particles.

Since the coating of Lot 826A was relatively thin and the thickness nonuniform, it would be desirable to double the average thickness of this coating and to evaluate the resulting sample before deciding the merits of a coating prepared in this manner.

The BeO coatings deposited from BeCl₂ and the CO₂-hydrogen reaction have not been representative of the quality which can be expected nor have they been of sufficient thickness to merit evaluation. This is thought to have resulted from the use of a new supply of BeCl₂ which is more highly contaminated with aluminum, iron, niobium, silicon, and zinc than the chlorides from the same suppliers used previously. This appeared to affect the quality of the coating and to aggravate plugging of the apparatus.

An effort was made to deposit a BeO coating by the decomposition of beryllium basic acetate at 1400°C in water vapor as a mildly oxidizing atmosphere, since the carbon contamination could thus be minimized. Premature decomposition of the beryllium compound plugged the vapor-inlet line.

**Plans for Future Work**

The coatings prepared at 1400°C by steam hydrolysis will be doubled in thickness and the product re-evaluated. The investigation of the effect on coating properties of the use of the CO₂-hydrogen reaction at 1400°C will be continued.

Since it is possible that an intermediate reaction evolving Be(OH)₂ may contribute to the formation of porous BeO deposited by hydrolysis of BeCl₂, an effort will be made to prepare BeO coatings by the direct oxidation of BeCl₂. Measures will be taken to prevent oxidation of the UO₂.

Decomposition of beryllium basic acetate at 1400°C under mildly oxidizing conditions will be investigated further as time permits.
Experimental work was deferred during September.

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$_2$O$_3$-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$_2$O$_3$-clad UO$_2$ in graphite by conventional processing, except for the use of solvent mixing. Use of this method to disperse dense Al$_2$O$_3$-clad, enriched UO$_2$ (Lot 715A) in a carbon matrix, and porous-dense Al$_2$O$_3$-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). These specimen sets were selected for static capsule irradiation.

Specimen, or small-quantity, dispersal methods have been developed and used in combination with pressure-baking procedures similar to those outlined in GAMD-2221. Four types of clad, enriched fuel (Lots 4E, 918A, 919A, and 920A) have been dispersed by these methods. The resulting fueled specimens had neutron-activated fission-gas released at temperatures of about 1150 C of the same order as, or lower than, those of the respective undispersed fuels at temperatures of 1300 C.
Work This Period

A higher temperature Al₂O₃-clad enriched UO₂ (Lot 725D) was dispersed in graphite-matrix specimens. Specimen-quantity fuel-dispersal and pressure-baking techniques were used. Coating integrity in these specimens is being assessed by the neutron-activation technique.

Strength and electrical-resistivity data were obtained for specimens representative of three sets of fueled-matrix specimens now undergoing static-capsule irradiation. Neutron-activation test results were reported previously for these specimen types in BMI-1534. The set designated Type 1 consists of carbon-bonded coke specimens fueled with dense-porous Al₂O₃-clad UO₂ (Lot 721C). Type 3 specimens are carbon-bonded graphite fueled with dense Al₂O₃-clad UO₂ (Lot 715A). Type 4 specimens are the same as Type 3 except that the fuel is Lot 721C. The strength and resistivity data obtained to date are summarized in Table L-1.

<table>
<thead>
<tr>
<th>TABLE L-1. STRENGTH(a) AND ELECTRICAL RESISTIVITY OF FUELED-MATRIX SPECIMEN TYPES NOW UNDERGOING STATIC-CAPSULE IRRADIATION</th>
</tr>
</thead>
<tbody>
<tr>
<td>Type</td>
</tr>
<tr>
<td>------</td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td>Unfueled</td>
</tr>
<tr>
<td>Fueled</td>
</tr>
<tr>
<td></td>
</tr>
</tbody>
</table>

(a) Data were obtained from diametral-compression tests on small cylindrical specimens 0.25 in. in diameter by 0.21 in. in length or 0.30 in. in diameter by 0.30 in. in length.

Ideally, fuel particles should be spaced equidistantly in the matrix. In this geometry, each fuel-particle cladding would receive maximum support from the matrix, composite strength would be maximized, and operating-temperature gradients within the composite minimized.

Work has been started on an approach to the preparation of equidistant fuel dispersion in graphite. This approach is to coat individual clad fuel particles with the desired quantity of matrix, then mold these pellets into a matrix specimen.

Plans for Future Work

As new clad fuel types become available, they will be dispersed in graphite by present random dispersion methods combined with pressure baking. The preparation of equidistant fuel dispersions by the above-mentioned pelletizing approach will be explored.
**Explosive Compaction Studies**

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

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

**Program Approach**

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

**Progress Reported Previously**

Experimental work has just been initiated.

**Work This Period**

Two 15-g batches of pyrolytic-carbon-coated UC₂ particles have been obtained for evaluation. The structure of the pyrolytic-carbon coatings in one of the batches is columnar and in the other it is laminar. Acid-leaching tests in hot nitric acid have shown that both coatings will protect the UC₂ particles from acid attack. Both types of coatings will be evaluated to determine their relative susceptibility to loss of integrity during explosive compaction in a graphite matrix.

Both batches of material were blended with natural graphite in the ratio of 20 volume per cent coated particles to 80 volume per cent graphite and were loaded by tamp packing into mild steel tubes for compacting. The as-loaded density of these specimens was calculated to be about 55 per cent of theoretical. Samples of the blended powders are undergoing leaching tests to determine if the particle coatings were damaged by the blending operation.

**Plans For Future Work**

These specimens will be leak checked and explosively compacted at ambient temperatures. After the mild steel tubes have been removed, density determinations will be made on all specimens. Specimens containing each of the two types of pyrolytic-carbon-coated particles will be subjected to microstructural studies. The remaining specimens will be subjected to acid-leaching studies in hot nitric acid to determine if the coatings continue to protect the UC₂ particles from acid attack. Additional investigations will be conducted on the effect of different types of graphite matrices.

**Pressure-Bonding Studies**

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

This program has been recessed.
Objective

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

Program 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 being 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. An electron microscopic examination did not reveal any difference in the structure of the alumina coatings after irradiation. Tentatively, the alumina appears to be an amorphous material, but it is possible that small crystals of alpha alumina are randomly dispersed throughout the amorphous material.

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 after a distance of about 0.5 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-1534, and BMI-1541.

Work This Period

Electron microscopic examinations of the alumina coated particles were discontinued. It was decided to await the results of X-ray diffraction studies since it is believed that these studies will be more helpful than electron microscopy in revealing any change in the alumina during irradiation. The X-ray diffraction studies have been initiated.

Plans for Future Work

A final appraisal of both the in-pile and postirradiation fission-gas-release data will be performed. The Al$_2$O$_3$ coatings from irradiated and unirradiated particles will be studied by X-ray diffraction techniques in order to determine any change in the structure of the coating during irradiation.

\[ \text{(Static Capsules CFP-1A, -1B, -1C, and -1D)} \]

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$_2$O$_3$ may be improved by using various combinations of high- and low-density UO$_2$ and Al$_2$O$_3$. It is planned to irradiate two types of fuel particles, as described below, both supported in a graphite matrix and also unsupported by any matrix material. The combinations of coated fuel contemplated for study in this set of experiments are:

1. Dense UO$_2$ particles coated with dense Al$_2$O$_3$ (715A)
2. Dense UO$_2$ particles coated with low-density Al$_2$O$_3$ and then with dense Al$_2$O$_3$ (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, both in graphite matrices and in an unsupported condition. The specimens in the four capsules will be irradiated at temperatures of about 150°C 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 examination, metallographic examinations, and strength tests. In addition, the quantity of fission gases released from each specimen will be determined. Dosimeter wires within the capsules 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 BMI-1489 and BMI-1496. The two types of Al$_2$O$_3$ coated UO$_2$ particles were incorporated into both a near fully graphitized matrix and a relatively ungraphitized (carbon) matrix. Since the
carbon matrix will tend to graphitize and contract during irradiation, it is expected that this matrix will give a greater degree of support to the coated particles than the more graphitic 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 Al₂O₃ coating on UO₂ 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, was loaded and sealed. They each contain the following specimen types:

1. Type 715A particles, unsupported (Xe¹³³ release: 3.2 ppm at 1300 C)
2. Type 715A particles, graphite matrix (Xe¹³³ release: 0.1 to 2.0 ppm at 1150 C)
3. Type 721C particles, unsupported (Xe¹³³ release: 1.3 ppm at 1300 C)
4. Type 721C particles, graphite matrix (Xe¹³³ release: 0.1 to 5.3 ppm at 1150 C)
5. Type 721C particles, carbon matrix (Xe¹³³ release: 0.1 to 1.3 ppm at 1150 C)
6. Type 921 particles, unsupported (Xe¹³³ release: 0.5 ppm at 1300 C)

All four capsules were charged into core position CP-12 of the BRR on August 9, 1961.

Work This Period

The irradiation of all four capsules is continuing. 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, are being irradiated for three BRR cycles and are scheduled for discharge from the reactor on October 2, 1961. Capsules CFP-1A and CFP-1B will be irradiated for six BRR cycles. The postirradiation 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 and -2R

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

Objective

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

Program Approach

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

Progress Reported Previously

The CFP-S-2 capsule, containing pyrolytic-carbon-coated uranium dicarbide particles in graphite-matrix pellets designated G-1 and G-2, was operated during June, 1961, with a specimen temperature of 1070 ± 15 C. After 36 hr of operation, a gas leak
occurred between the sweep-gas system and the argon-filled gas annulus necessitating termination of the experiment. As described in BMI-1541, a replacement capsule, CFP-S-2R, containing a similar specimen, designated G-10, was placed in operation in the BRR early in August, 1961. This specimen had been tested by neutron activation and showed a \( \text{Xe}^{133} \) release ratio of 0.18 ppm at 1150°C. The initial fission-gas-release rate in the sweep capsule was very low as shown by the data in Table L-2. One week after starting the experiment, when the reactor was brought to power after a 28-hr shutdown, the fission-gas release suddenly increased by approximately three orders of magnitude. Concurrently, the pressure drop through the sweep-gas system increased with a resulting decrease in the flow of sweep gas.

Work This Period

During the scheduled reactor shutdown early in September, attempts were made to improve the flow through the sweep-gas system. Although a reasonable flow could be maintained through the system when the capsule was cool, when the temperature within the capsule was increased the pressure drop would increase, resulting in a decrease in the flow. This temperature effect on the pressure drop indicated that the restriction was in the heated portion of the capsule, that is, the fuel-containing inner can. The experiment was resumed upon reactor startup on September 6, 1961, but within a few hours the flow had dropped to below the detectable value at the maximum input pressure of 75 psi. The sweep-gas system was shut down and the irradiation of the capsule continued under static conditions with the fuel surface temperature maintained at 1070 ± 15°C.

Table L-2 contains the fission-gas-release fraction (R/B) data from the analysis of all the samples taken during the dynamic operation of this experiment. The data from samples after 8A suffer from uncertainties in the transit time through the holdup tank and nonequilibrium conditions within the capsule, hence errors of ±100 per cent may be expected. Efforts to correlate the R/B data from Samples 6A, 7A, and 8A with the diffusion of fission products through a thick uranium-free spherical coating were unsuccessful. The R/B values were less dependent on the half-life of the diffusing species than is predicted by this model.

Plans for Future Work

Efforts to correlate the fission-gas-release data will be continued in order to describe the nature of the gas-release mechanism.

**Sweep Capsule CFP-S-3**

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

This work is currently recessed.

**Static Capsule CFP-4**

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

Objective

To evaluate the performance of unsupported fuel particles of uranium compounds coated with various ceramic materials during low-temperature irradiation.
<table>
<thead>
<tr>
<th>Sample(a)</th>
<th>Time After Start of Irradiation, hr</th>
<th>Xe$^{141}$</th>
<th>Xe$^{140}$</th>
<th>Xe$^{137}$</th>
<th>Kr$^{91}$</th>
<th>Kr$^{89}$</th>
<th>Kr$^{87}$</th>
<th>Xe$^{135}$</th>
<th>Kr$^{88}$</th>
<th>Kr$^{85m}$</th>
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<td>--</td>
<td>(c)</td>
<td>(c)</td>
<td>(c)</td>
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<td>(c)</td>
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<td>4.6x10$^{-10}$</td>
</tr>
<tr>
<td>2A</td>
<td>4</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>(c)</td>
<td>(c)</td>
<td>(c)</td>
<td>--</td>
<td>(c)</td>
<td>--</td>
<td>4.3x10$^{-9}$</td>
</tr>
<tr>
<td>3A</td>
<td>24</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>9.2x10$^{-9}$</td>
<td>1.2x10$^{-8}$</td>
<td>(c)</td>
<td>6.9x10$^{-9}$</td>
<td>(c)</td>
<td>--</td>
<td>1.1x10$^{-8}$</td>
</tr>
<tr>
<td>4A</td>
<td>42</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>1.4x10$^{-8}$</td>
<td>3.3x10$^{-9}$</td>
<td>1.1x10$^{-8}$</td>
<td>2.9x10$^{-8}$</td>
<td>(c)</td>
<td>--</td>
<td>1.1x10$^{-8}$</td>
</tr>
<tr>
<td>5A</td>
<td>68</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>3.4x10$^{-9}$</td>
<td>7.9x10$^{-9}$</td>
<td>6.4x10$^{-9}$</td>
<td>8.2x10$^{-9}$</td>
<td>(c)</td>
<td>--</td>
<td>1.1x10$^{-8}$</td>
</tr>
<tr>
<td>6A</td>
<td>90</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>7.4x10$^{-9}$</td>
<td>1.1x10$^{-8}$</td>
<td>1.7x10$^{-8}$</td>
<td>3.1x10$^{-8}$</td>
<td>1.1x10$^{-8}$</td>
<td>--</td>
<td>1.1x10$^{-8}$</td>
</tr>
<tr>
<td>7A</td>
<td>114</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>6.2x10$^{-9}$</td>
<td>1.3x10$^{-8}$</td>
<td>1.7x10$^{-8}$</td>
<td>3.1x10$^{-8}$</td>
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<td>1.1x10$^{-8}$</td>
</tr>
<tr>
<td>8A</td>
<td>136</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>6.0x10$^{-9}$</td>
<td>1.6x10$^{-8}$</td>
<td>1.3x10$^{-8}$</td>
<td>2.5x10$^{-8}$</td>
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<td>1.1x10$^{-8}$</td>
</tr>
<tr>
<td>9A</td>
<td>171</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>1.1x10$^{-4}$</td>
<td>--</td>
<td>1.6x10$^{-4}$</td>
<td>3.2x10$^{-3}$</td>
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<td>1.1x10$^{-3}$</td>
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<tr>
<td>10A</td>
<td>190</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>2.9x10$^{-6}$</td>
<td>1.7x10$^{-5}$</td>
<td>--</td>
<td>3.6x10$^{-6}$</td>
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<td>--</td>
</tr>
<tr>
<td>11DT</td>
<td>190</td>
<td>(c)</td>
<td>2.1x10$^{-5}$</td>
<td>3.4x10$^{-4}$</td>
<td>2.7x10$^{-5}$</td>
<td>6.9x10$^{-4}$</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>12A</td>
<td>195</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>3.3x10$^{-5}$</td>
<td>--</td>
<td>4.7x10$^{-5}$</td>
<td>2.5x10$^{-4}$</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>13A</td>
<td>210</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>6.3x10$^{-8}$</td>
<td>6.2x10$^{-6}$</td>
<td>9.4x10$^{-7}$</td>
<td>3.5x10$^{-6}$</td>
<td>3.2x10$^{-5}$</td>
<td>--</td>
</tr>
<tr>
<td>14A</td>
<td>379</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>5.3x10$^{-4}$</td>
<td>6.6x10$^{-3}$</td>
<td>5.0x10$^{-3}$</td>
<td>5.3x10$^{-3}$</td>
<td>--</td>
<td>--</td>
</tr>
</tbody>
</table>

(a) "A" samples designate adsorption samples; DT samples designate daughter-trap samples.
(b) Species listed in order of increasing half-life; Xe$^{135}$ listed according to its apparent half-life.
(c) Values below limits of detection.
Program Approach

Fuel particles of UO$_2$ and UC$_2$ coated with Al$_2$O$_3$ 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

The six types of coated fuel particles to be irradiated were selected and include the following:

1. UO$_2$ coated with a thin, dense innerlayer of Al$_2$O$_3$ which is in turn coated with a porous and then a second dense coating of Al$_2$O$_3$ (S&P 4E, Xe$^{133}$ release: 1.2 ppm at 1100 C)
2. UO$_2$ coated with dense Al$_2$O$_3$ (715A, Xe$^{133}$ release: 3.2 ppm at 1300 C)
3. UO$_2$ coated with two dense and one porous layer of Al$_2$O$_3$ 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: 2.0 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 was sealed in a helium-filled annulus in an aluminum can. The six cans were placed 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

Capsule CFP-4 was discharged from the BRR on September 18, 1961. It was irradiated for five reactor cycles. This is two more cycles than was originally scheduled. The irradiation was extended because of extended downtime of the reactor during two cycles because of core refueling and instrument calibration.

The capsule was opened, and the six specimen cans were recovered and visually examined. All six specimen cans appeared clean and in good condition. Dosimeter wires were recovered from the capsule, and analyses were initiated. A hole was drilled in the top of each specimen can, and gases within the can were collected and analyzed for fission-gas content. Preliminary results indicated that detectable amounts of xenon-133 were present in all specimen cans 7 days after discharge of the capsule from the reactor. Krypton-85 was not detected. The release fraction of xenon cannot be calculated pending the completion of gas-activity analyses and dosimeter-wire analyses. Recovery of the particles from the specimen cans has been initiated.

Plans for Future Work

Particles irradiated in Capsule CFP-4 will be recovered and visually examined. The fraction of particles with cracked coatings will be determined by metallographic examination and by analysis of leach solutions of the particles. Particles with intact coatings will be heat treated to determine gas-diffusion and -release characteristics of the coatings.
Objective  
To obtain high-temperature high-burnup fission-gas-release information on alumina-coated uranium dioxide particles supported in a graphite matrix.

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

Progress Reported Previously  
The description of the sweep-gas specimen and the four static specimens was presented in BMI-1541. The sweep specimen consists of two pellets machined from S&PCF Sphere 472 and containing alumina-coated uranium dioxide particles with a porous alumina interlayer in the coating (Lot 4E). The pellets were designated M-2 and M-4 and had an average Xe133 release ratio of 4.2 ppm at 1150 C after neutron activation. The following materials were selected for the static compartments:

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 one of several pellets fabricated as a backup effort for the sweep-capsule specimen (Xe133 release: 0.07 ppm at 1150 C)
2. Unsupported columnar-carbon-coated uranium dicarbide particles of Lot 918A (Xe133 release: 2.7 ppm at 1300 C)
3. Unsupported laminar-carbon-coated uranium dicarbide particles of Lot 919A (Xe133 release: <0.5 ppm at 1300 C)
4. Unsupported laminar-carbon-coated uranium oxide particles of Lot 920A (Xe133 release: 0.9 ppm at 1300 C).

Work This Period  
A modification in the sweep-gas compartment was made to try to prevent the pressure-drop difficulties that have been experienced in the CFP-S-2R experiment. The sweep-gas specimen compartment was increased in diameter to allow a 0.020-in. annulus between the specimen and the compartment wall. This annulus was filled with 65 to 100-mesh graphite powder, the graphite being retained by a 200-mesh nickel screen at the gas inlet and outlet. It is believed that this powder annulus will permit some shifting and distortion of the specimen without greatly influencing the gas flow. In addition, a 100-mesh stainless steel cloth filter, located in the cool zone on the gas outlet tube, has been added to prevent plugging of the small-bore outlet tubing by particles carried in the gas stream.

The capsule experiment was begun on September 20, 1961, in the BRR. The sweep-gas-specimen surface temperature has been controlled at 1070 ± 15 C. The static specimens are estimated to be operating approximately 100 C higher than the sweep specimen. The fission-gas-release fractions (R/B) from the first gas samples are given in Table L-3. The presence of I131 and I133 was evident in these four
samples. The amount of I$_{131}$ was nearly equivalent to that of the other gaseous species but no R/B values were calculated for these two isotopes because it is not known what fraction of the iodine that is released actually reaches the sample traps. A daughter-trap sample was taken late in September but the data are not available at this time.

### TABLE L-3. CFP-S-5 FISSION-GAS-RELEASE DATA

<table>
<thead>
<tr>
<th>Sample</th>
<th>Time After Start of Irradiation, hr</th>
<th>Kr$_{87}$</th>
<th>Xe$_{135}$</th>
<th>Kr$_{88}$</th>
<th>Xe$_{133}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1A</td>
<td>1</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>1.4 x 10$^{-6}$</td>
</tr>
<tr>
<td>2A</td>
<td>13</td>
<td>2.5 x 10$^{-6}$</td>
<td>2.4 x 10$^{-6}$</td>
<td>3.6 x 10$^{-6}$</td>
<td>6.4 x 10$^{-6}$</td>
</tr>
<tr>
<td>3A</td>
<td>32</td>
<td>1.0 x 10$^{-6}$</td>
<td>2.9 x 10$^{-6}$</td>
<td>2.7 x 10$^{-6}$</td>
<td>5.5 x 10$^{-6}$</td>
</tr>
<tr>
<td>4A</td>
<td>57</td>
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<td>1.1 x 10$^{-6}$</td>
<td>3.5 x 10$^{-6}$</td>
<td>6.0 x 10$^{-6}$</td>
</tr>
</tbody>
</table>

Plans for Future Work

The operation of this experiment is planned to be continued until mid-December, 1961, unless terminated earlier by specimen or capsule failure.

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

The initial planning of this experiment was presented in BMI-1541.

Work This Period

The capsule components for this experiment have been fabricated and are ready for the final assembly. The specimen of pyrolytic-carbon-coated uranium dioxide particles in a graphite matrix, designated G-20 and having a Xe$_{133}$ release ratio of 0.10 ppm at 1150 C after neutron activation, which will be contained in the sweep-gas compartment, has been made available for encapsulation. However, the selection of specimens for the static compartments of the capsule has been deferred until postirradiation examination data are available from Capsules CFP-4, CFP-1C, and CFP-1D.

Plans for Future Work

The initiation of this experiment has been postponed to late November, 1961.
Intermediate-Temperature Design Studies

N. E. Miller and W. J. Zielenbach

The effort on this phase of the encapsulation studies was resumed late in this report period, and there is no progress to report at this time.

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 efficiency values which are currently being used 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. Krypton and xenon fission-gas mixtures, produced by irradiating uranium foil, were separated by gas chromatography and adsorbed on charcoal traps. Spectrometer scans of the separated xenon and krypton 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 for each fission gas.

Work This Period

Gamma-ray spectrometer detection efficiencies were determined using standardized sources of cerium-141, chromium-51, manganese-54, and strontium-85, which were placed in charcoal beds within metal tubes having the same dimensions and wall thickness as the metal fission-gas traps.

Plans for Future Work

The techniques necessary for the analysis of complex fission-gas mixtures are essentially complete. However, efforts to improve these techniques will be continued.
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 normal reactor maintenance. The effects of $\text{Al}_2\text{O}_3$ and $\text{BeO}$ fuel-particle coatings on the reactivity of a graphite-moderated pebble-bed reactor (PBR) were reported in a previous 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-$\mu$-diameter fuel particles with $\text{Al}_2\text{O}_3$, $\text{BeO}$, or carbon were reported in BMI-1524. A preliminary investigation was begun to determine the range of fuel loadings of interest in the development of reactor fuel elements containing coated particles.

The feasibility of employing coated particles in current superheater reactor designs (Pathfinder and BONUS) was studied, the results indicating that particles coated with a low-cross-section corrosion-resistant material might have certain advantages over materials currently employed. The investigation of criticality problems associated with a chemical process reactor was expanded to determine how the maximum "safe" reactor diameter varies with coating thickness.

Work This Period

Completion of the expanded study of criticality problems of the chemical process reactor resulted in curves showing how the maximum "safe" diameter varies with coating thickness of $\text{BeO}$ and $\text{Al}_2\text{O}_3$ coatings.

Work continued on the feasibility investigation of coated-particle application to the Pathfinder nuclear superheater concept. Simplified analysis implies that low-enrichment fuel particles, coated with a low-absorption-cross-section steam-corrosion-resistant material, may be directly substituted for the stainless steel-clad stainless steel-highly enriched $\text{UO}_2$ cermet currently being employed in the Pathfinder superheater cell configuration without sacrificing nuclear characteristics of the core. Advantages of the substitution are cost and nuclear safety improvements.

Plans for Future Work

A more sophisticated nuclear analytical study of the application of coated particles to the Pathfinder superheater reactor is to be undertaken and compilation of data
to establish the range of fuel loadings of interest in the development of fuel elements will be continued.

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 the fission tracks with optical and electron microscopical techniques. Alpha Al₂O₃ 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 Previously Reported
Direct carbon-replica techniques with the electron microscope have revealed fission damage on the surfaces of NaCl, KBr, and LiF. The damage region is usually the defected cross-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 2 μ. Longer, thinner areas have also been observed which are due to the fission recoils which enter the surface at lower angles. Pit damage is also seen but is much smaller. Decoration by precipitated silver in AgCl has revealed dislocations, grain boundaries, and fission damage. In the NaCl crystals, a considerable change in the registered damage has been observed and explained in terms of a humidity and/or temperature effect.

Work This Period
Investigations of this period were concerned primarily with LiF and AgCl. These materials (which have a very low solubility in water) were studied in an attempt to verify that the change in the register of damage as observed in NaCl was due to a humidity and/or temperature effect. Both LiF and AgCl showed consistent damage structures (consistent with the earliest results) when irradiated under the same conditions that produced the anomalous results in NaCl. The same observation techniques were used in all cases.

The observation of decorated fission tracks in AgCl is continuing. At present, samples are being observed with low-energy red light in an attempt to minimize the background decoration effects produced by optical illumination.

Plans for Future Work
LiF and NaCl specimens are to be fueled in the same fuel elements in various humidity conditions in an attempt to reproduce the original data in NaCl. The LiF will serve as a check on the experimental conditions.

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 discussed in a previous progress report, BMI-1504. Xenon-133 diffusion coefficients for pyrolytic carbon at temperatures ranging from 700 to 1300 C were also presented in previous progress reports, BMI-1514 and BMI-1518.

Work This Period

Preliminary analysis of data from measurements of xenon-133 diffusion from Linde Company alpha-alumina spheres at temperatures below 1200 C has been completed. The results of the diffusion measurements, expressed in terms of diffusion coefficients, are presented in Table L-4. These results appear to verify the change in activation energy that was observed between 1000 to 1100 C in earlier diffusion measurements. Above 1200 C, xenon-133 diffusion in alpha alumina has been observed to follow an activation energy of about 70 kcal per g mole, while below 1200 C the activation energy was found to be about 10 kcal per g mole. Analysis of data from xenon-133 diffusion measurements that were performed using alpha-alumina spheres from which about 20 per cent of the xenon-133 had been removed by grinding uniform layers from the surfaces of spheres is in progress. These experiments were conducted to study surface effects and their relation to the concentration profile of the diffusing species in the spheres. Another series of experiments designed to study the effects of xenon-133 concentration on diffusion in alpha-alumina is in progress and will be completed early next month. For these measurements, spheres were used with xenon-133 concentrations about 2000 times smaller than the concentrations in any of the previous work.

TABLE L-4. XENON-133 DIFFUSION IN ALPHA ALUMINA

<table>
<thead>
<tr>
<th>Temperature, C</th>
<th>Diffusion Coefficient, cm² per sec</th>
</tr>
</thead>
<tbody>
<tr>
<td>703</td>
<td>$6.3 \times 10^{-18}$</td>
</tr>
<tr>
<td>898</td>
<td>$1.4 \times 10^{-17}$</td>
</tr>
<tr>
<td>1101</td>
<td>$1.2 \times 10^{-16}$</td>
</tr>
</tbody>
</table>

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 800 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 in contact with fully enriched uranium dioxide for periods of 4 to 8 weeks in a thermal-neutron flux of $1 \times 10^{13}$ nev. These experiments will demonstrate the effect of prolonged irradiation on observed fission-gas release.

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

**Fuel-Compatibility Studies**

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

**Objective**

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

**Program Approach**

The stability of oxide-coated UO$_2$ particles in an oxidizing atmosphere will be studied by measuring weight changes and by determination of changes in oxygen content of the UO$_2$ in the temperature range of 1000 to 1800 C. Specimens will be examined by metallographic and X-ray diffraction techniques and the results 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**

Particles of enriched uranium carbide coated with pyrolytic carbon were heat treated at 1400, 1600, and 1800 C for varying time periods to study uranium diffusion through the coatings. Results of X-ray diffraction analyses on the particles were presented in a recent progress report, BMI-1524. At temperatures of 1600 C or above, the UC is converted to UC$_2$ in a few hours. At lower temperatures, UC and U$_2$C$_3$ also exist. Alpha-counting and autoradiographic measurements were performed on these specimens to obtain data to be used for calculating the rate of diffusion of uranium through the coatings. Studies of the oxidation of Al$_2$O$_3$-coated UO$_2$ particles indicated that the observed failure of particles at a critical temperature is due to the difference in thermal expansion of the Al$_2$O$_3$ and the UO$_2$. The temperature of failure was found to depend on the coating thickness and UO$_2$ particle density.

**Work This Period**

Work has continued on the determination of the rate of diffusion of uranium through pyrolytic carbon coatings. For this study, data were obtained from alpha-counting and autoradiographic measurements of heat-treated particles of uranium.
carbide coated with pyrolytic carbon. A general equation for the diffusion coefficient for this type of specimen has been derived. However, suitable approximations for the equation must be determined in order to simplify the expression to permit calculations.

Additional study of the behavior of Al₂O₃-coated UO₂ in an oxygen or air atmosphere has shown that the temperature at which rapid oxidation occurs is also dependent upon the original temperature of application of the coating. Specimens coated at 1400 C have been examined, and failure appears to have occurred by diffusion at 1500 C and by thermal stress at 1700 C. It had previously been found that similar particles coated at 1000 C failed by thermal stress at about 1350 C.

An attempt is also being made to estimate the rate of permeation of oxygen through the Al₂O₃ coatings on UO₂ particles at temperatures below that at which rapid failure occurs due to cracking of the coatings by thermal stress. Particles of dense UO₂ coated with 53-µ-thick Al₂O₃ at 1000 C have been heated at 1200 and 1300 C for periods of 100 hr in oxygen and the weight change followed with a recording microbalance. In both cases the weight gain appeared to be a result of diffusion or permeation of oxygen through the coatings rather than cracking followed by sudden oxidation as reported for this material heated at 1350 C. The weight gain suggests the formation of UO₂,6₂ after 120 hr at 1300 C and UO₂,2₆ after 96 hr at 1200 C. The compositions have not yet been determined from X-ray data.

Plans for Future Work

The investigation concerned with the determination of the rates of diffusion of uranium through carbon coatings will be continued. Further diffusion measurements will be made after a suitable diffusion equation has been determined and the data already obtained have been completely analyzed. The equipment constructed for the investigation of the reactions between oxides and pyrolytic carbon will be calibrated, and the equipment which has been designed and built for the thermal-diffusion studies will be tested. Oxidation studies will be continued on Al₂O₃ coated UO₂.

Coolant-Compatibility Studies

C. A. Alexander and A. Levy

Objective

To establish experimentally the compatibility of high-pressure steam with Al₂O₃, MgO, ZrO₂, and UO₂.

Program Approach

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

Progress Reported Previously

Available free-energy and kinetic data applicable to the coolant compatibility problem were appraised. A topical report covering the study was issued as BMI-1530.

Work This Period

Investigation of the effect of water vapor on the volatilization of UO₂ has been made from 1400 to 1708 C. Initial determinations were made using argon containing 0.03 atm of H₂O, but values higher than predicted were obtained. The observed vapor
pressure at 1660 C was $4 \times 10^{-5}$ atm, which is an order of magnitude higher than predicted on the basis of assumed equilibria in the UO$_2$-H$_2$O system. This high value was considered to be due to the presence of trace amounts of oxygen in the argon flow. Oxygen contaminations of the order of 60 ppm would be sufficient to cause this effect.

As a means of reducing the oxygen pressures (if present) argon-hydrogen-water vapor mixtures were employed. The reaction studied was the same:

$$
\text{UO}_2 + \text{H}_2\text{O(g)} \rightarrow \text{UO}_3(g) + \text{H}_2
$$

and $K_p = \frac{p_{\text{UO}_3} \cdot p_{\text{H}_2}}{p_{\text{H}_2\text{O}}}$ (atm).

The data at low hydrogen concentrations (0.01 atm and less) can be represented by the equation: $\log K_p = -27,600/T + 6.362$, where $K_p$ is in atmospheres and $T$ is in degrees Kelvin. Deviation of any point from this equation is less than 50 per cent. This equation for $K_p$ can be compared with the predicted equation: $\log K_p = 25,200/T + 7.18$. The precision of such measurements does not appear to be as good as in direct vapor-pressure determinations. Consideration of possible side reactions between the gases present and materials of construction would indicate that $\pm 50$ per cent is about the degree of precision to be expected.

It was observed that with hydrogen pressures of 0.1 atm, the volatility of the oxide was greater than could be predicted on the basis of either vaporization as UO$_3$ or congruent vaporization of UO$_2$. At 1580 C the observed volatility was $4 \times 10^{-7}$ atm which is about an order of magnitude higher than predicted on the basis of UO$_3$ volatility.

Although UO$_2$ is thermodynamically stable with respect to uranium liquid and traces of oxygen in a hydrogen-steam atmosphere at the temperature employed in this investigation, it would appear that an additional vaporization process is operative. It is considered that UO may be contributing to the volatility, and may be the principal vapor product with high hydrogen concentrations, i.e., $\text{UO}_2(c) + \text{H}_2 \rightarrow \text{UO(g)} + \text{H}_2\text{O(g)}$. In a run using hydrogen at 1 atm as the carrier gas, for example, the vapor pressure at 1510 C was observed to be $5 \times 10^{-7}$ atm, about 40 times higher than calculated from the predicted UO$_3$ vapor-pressure equation. Water vapor at 0.02 atm was also observed in the exit stream.

The volatility of uranium oxide then appears to be influenced by the atmosphere, either reducing and oxidizing, with the minimum in the volatility occurring for the stoichiometric UO$_2$, maintained in a neutral atmosphere.

Plans for Future Work

It is planned to set aside temporarily further work on the UO-UO$_2$ problem in order to move on to the study of other oxides. During the next month an investigation of volatilization of MgO with water vapor will be begun.

Thermal-Conductivity Studies

H. W. Deem and C. F. Lucks

Objective

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

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

Progress Reported Previously

As discussed in previous progress reports, effort in the program so far has been concerned with selection of a method and construction of apparatus suitable for measuring the thermal conductivity of specimens of a size dictated by coated-particle size, encapsulation conditions, and irradiation conditions. The IR detector bridge system was checked and found to be satisfactory. The table and vacuum system were assembled and checked. Operation of the chopper-timing motor was checked and found to operate within ± 0.8 per cent of the oscillator dial control with frequencies ranging from 10 to 330 cps, using two different chopper disks.

Work This Period

The detector bridge circuit was checked using 18 ft of shielded extension cable to determine possible noise pickup during remote operation. The noise level showed no significant difference from operation with direct connection and appeared to be satisfactory throughout.

Work on the remote controlled specimen loading and unloading apparatus has been suspended until after a trial run of the apparatus. The detector mounting brackets and the optical mounts have been completed, and all optical parts are being mounted in preparation for a trial run of the apparatus.

Shutters, to protect the quartz windows of the vacuum chamber, have been completed and installed. The tubular furnace has been completed and mounted within the chamber. A "single-hole" chopper disk has been constructed and balanced, and good control of frequency has been obtained down to 1.5 cps. For lower frequencies a simple gear-down system can be used. Feed-through seals have been soldered into the vacuum-chamber wall for use with the heater and thermocouple leads, and the vacuum has been rechecked. No additional leaks were found.

Plans for Future Work

As soon as the apparatus is assembled, a trial will be made of the specimen measuring method. It is hoped this trial run will be completed early in October, and any necessary modifications can be determined in preparation for final assembly.

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 September.

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

Various aspects of the work have been described in previous progress reports. A combined hydrofluorination-corrosion study involving ZrO₂ dissolutions and INOR-8 corrosion specimens was described in 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 was carried on in molten LiF-NaF-Zr for 150 hr under a fluorine sparge. Weight-loss measurements showed a maximum corrosion rate of about 4 mils per month for specimens exposed at the vapor-liquid interface. However, a metallographic study showed that all specimens suffered a relatively severe intergranular attack. The corrosion rates based on the amount of sound metal found in the microsections were about 25 mils per month for specimens exposed at the interface and in the liquid and about 7 mils per month for specimens exposed in the vapor. No preferential attack was found at welds.

Plans For Future Work

A hydrofluorination-corrosion study with HyMu-80 and INOR-8 specimens will be conducted at 650 °C in the presence of dissolving Zircaloy-2.

Metallographic examinations will be made on components from ORNL pilot plants.
ASSISTANCE TO THE CANDU REACTOR PROGRAM (SRO)

S. W. Porembka

Work at Battelle relating to the CANDU program of SRO is discussed in this section.

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. A burst test is being considered as a means for inspecting the integrity of end caps.

Progress Reported Previously

Primary interest in this effort has been shifted to end capping of fuel sheaths. Preliminary end-capping studies of Zircaloy-2 fuel sheaths were discussed in a recent progress report, BMI-1524. Termination of the Zircaloy-2 end-capping studies has directed all effort toward the joining of SAP material.

Work This Period

A preliminary study of the joining of SAP material by friction bonding was made on the modified lathe setup using a protective atmosphere of argon. The joining of specific end-cap configurations was not attempted in these tests. Metallographic examination of a joined specimen from this group showed that spotty metallurgical bonding was obtained. The dispersion hardening effect of the oxide in the SAP material narrows the range in which plastic deformation takes place readily. Therefore, close contact of this process variable is necessary.

A new support for the chuck holding the stationary member is being fabricated. The support will allow the use of shorter specimens than was possible previously. The shorter specimens will minimize any radial deflection during the bonding cycle.

Plans for Future Work

Specific end-cap configurations are being scheduled for evaluation. These will not be joined until the new support described above is completed.

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 in this portion of the program. 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 Previously Reported

Surface preparations for Zircaloy-2 and stainless steel flat-plate composites have been established which produce mechanical bonding when cold rolling at 50 per cent reduction taken in a single pass. Roll-expanding equipment and capabilities have been determined for both the aluminum-mild steel and stainless steel-Zircaloy-2 systems. Reduction limitations imposed by the roll-forming equipment have been such that it has not been possible to achieve the desired bonding in the various designs considered to date, including those providing locally high deformations. Metallographic examination of roll-formed joints has indicated that the bonds are mechanical in nature. Flat-plate composites of stainless steel and Zircaloy-2 employing an alumina coating yielded rolling results similar to those obtained in previous tests.

Work This Period

Additional Type 410 stainless steel-Zircaloy-2 joint assemblies were roll formed for further explosive-joining experiments. A 1500-μin. rms surface was utilized on the inside surface of the steel tubes along with three groove designs. Surface finishes of 300 and 100-μin. rms were machined on the outside surface of the Zircaloy-2 tubes. Surface preparation for all components consisted of degreasing in acetone followed by scratch brushing with a rotating wire brush. The joints were expanded just enough to fill the grooves of the stainless steel, which represented approximately 26 per cent reduction in area of the Zircaloy tubes.

Further cold pressure-welding studies were conducted using flat-plate composites of stainless steel and Zircaloy-2. Electroplatings of gold and silver were applied to the stainless steel surfaces finished to 300-μin. rms. Surface preparations consisted of degreasing in acetone followed by scratch brushing of the Zircaloy plates with a wire brush wheel. Reductions of 24 to 57 per cent were employed in a single pass through the 16 by 20-in. rolling mill. Samples with both gold and silver platings were joined using 40, 43, and 57 per cent reductions. Upon sectioning of the rolled composites, however, only the sample reduced 57 per cent remained together. Metallographic results are not yet completed on this sample.

Plans for Future Work

A more extended and comprehensive evaluation of surface roughness is planned on the basis of results to date of cold-rolling and explosive-joining studies. More extensive use of scratch brushing will be made in these tests. The ultimate objective of these studies is to lower the required reductions necessary for joining so that they may fall into the capability of the roll-forming equipment.
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 bond strength as determined by metallographic examination and mechanical testing.

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

Work This Period
Additional Zircaloy-to-stainless steel tubular specimens possessing surface finishes of from 500 to 1500-µin. rms on the stainless steel were prepared. These specimens were roll formed prior to explosive compacting. Stand-off charges and both water and air transfer media were used during this testing. Charge weights were varied from 30 to 42 g. These specimens are presently being sectioned for metallographic examination.

Plans for Future Work
Additional specimens which will contain surface finishes ranging from 500 to 1500-µin. rms will be tested in an attempt to determine the optimum surface finishes required to produce good bonding by a combination of roll-forming and explosive joining.

Long-Term Creep-Rupture Program on SAP Alloys

J. A. VanEcho

Objective
To obtain long-term creep-rupture data on three sintered aluminum powder (SAP) alloys.

Program Approach
The three alloys of immediate interest are M-257, M-583, and SAP895. Test temperatures are scheduled to be 700, 800, and 900 F for all three materials. Long-term in this instance means 100,000 hr; however, tests will be limited to 20,000 hr of running time.

Initially, stresses will be chosen to cause failure in about 1, 10, 100, and 1000 hr. On the basis of these results, tests will be started in which the specimens would be expected to fail in 10,000 and 100,000 hr. The 100,000-hr tests will be discontinued at 20,000 hr. Then, on the basis of all available creep and rupture data, stresses will be estimated which would cause failure in 100,000 hr in each alloy at each test temperature.
Work This Period
To date, two of the materials (M-583 and SAP895) have been received. These two alloys are being machined into 0.505-in.-diameter test bars.

Plans for Future Work
Actual testing should begin during the first week in October. The first group of the tests will be initiated with the two presently available SAP alloys.
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 is being prepared for the irradiation at the MTR of pellets of 30 volume per cent UO₂ (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 x 10¹³ nv.

Work This Period
Capsule BMI-36-1 was received at the MTR in September. Upon installation, a gas leak in the communicating lines to the gas annulus was discovered which permitted leakage of annulus gas to the lead tube. It was necessary, because of this, to discharge the capsule and return it to Battelle for repair.

Plans for Future Work
Repairs will be made to Capsule BMI-36-1 in 3 to 6 weeks. Irradiation will probably start in mid-October or early in November.
RADIATION STUDIES OF SM-2 FUELS (ALCO)


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 noninstrumented 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 three progress reports: 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 197 full-power in-pile days. Peak-flux specimen burnups were estimated at 65 to 75 a/o (total uranium) in the two capsules. Recent operating performance of the four remaining in-pile capsules was reported in BMI-1534.

Work This Period
The irradiations of BMI-32-5, BMI-32-7, BMI-32-9, and BMI-32-10 in the ETR were continued. Reactor Cycle 39 went to power on August 24, 1961, and is scheduled to shut down late in September. Cycle 40 will then run until October 2, and Cycle 41 until November 13. Performance of the four capsules remains essentially as reported previously. Estimated peak-flux specimen burnups as of the end of Cycle 40 are:

<table>
<thead>
<tr>
<th>Capsule</th>
<th>Estimated Burnup on 10-2-61, a/o</th>
</tr>
</thead>
<tbody>
<tr>
<td>BMI-32-5</td>
<td>65</td>
</tr>
<tr>
<td>BMI-32-7</td>
<td>69</td>
</tr>
<tr>
<td>BMI-32-9</td>
<td>89</td>
</tr>
<tr>
<td>BMI-32-10</td>
<td>63</td>
</tr>
</tbody>
</table>

A request was submitted to extend these irradiations until the November 13, 1961, shutdown.

Plans for Future Work
The ETR irradiations of BMI-32-5, BMI-32-7, BMI-32-9, and BMI-32-10 will be continued.
The Army Reactor Branch (ARB) is developing small portable reactor systems for military applications. Aerojet-General Nucleonics (AGN) is the prime contractor under the Army Gas-Cooled Reactor Systems Program (AGCRSP) and is charged with the design, construction, and operation of the GCRE and ML-1. The studies below are in support of 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$_2$O$_3$-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$_2$O$_3$. In this program, the coated powder will be used as a starting material. Various methods of packing the coated powder into steel tubes will be investigated. The tubes will be evacuated and under isostatic pressure will be collapsed around the ceramic core. The maximum Al$_2$O$_3$ diluent is restricted to about 40 volume per cent. The main concern is the retention of coating integrity during densification. This will be evaluated by nitric acid leach tests, oxidation tests, and metallographic studies.

**Progress Reported Previously**

Spherical UO$_2$ particles with average diameters of 150 and 50 $\mu$m have been coated by a vapor-deposition method with from 5 to 10 $\mu$m of Al$_2$O$_3$. Characteristics of the as-received material were described in three previous progress reports: BMI-1489, BMI-1504 (Del.), and BMI-1518. Defective particles were removed by selective leaching in nitric acid followed by heavy-liquid flotation.

Evaluation of fabrication processes utilized prior to isostatic hot pressing, as reported in BMI-1518 and BMI-1524, shows that vibratory compaction eliminates particle breakup associated with other processes such as cold pressing. However, a particle mixture of more ideal packing size distribution is necessary to obtain the desired high densities. The ultimate mechanism of failure consists of exposed UO$_2$ being converted to U$_3$O$_8$ with an accompanying 42 volume per cent expansion which causes brittle fracturing of the Al$_2$O$_3$ and continually exposes more UO$_2$.

The elimination of particles with defective coatings and the reduction of particle breakup associated with cold-pressing operations has improved the oxidation characteristics of the fuel slightly. The large difference in thermal-expansion coefficients between the UO$_2$ and Al$_2$O$_3$ seem not to be detrimental, and the oxidation rate in 1200 F air seems comparatively unaffected by the relative amounts of porosity present. The weight loss during nitric acid leaching is, however, dependent on the percentage of theoretical density attained during fabrication.

The addition of 8 w/o chromium increases the time to complete oxidation by a factor of three. The increase in oxidation resistance may be due to the extrusion of chromium into any Al$_2$O$_3$ defects or to the increased matrix strength.
Specimens containing undiluted UO$_2$, 20 volume per cent chromium-UO$_2$ cermets, cold-pressed Al$_2$O$_3$-coated UO$_2$, vibratory compacted Al$_2$O$_3$-coated UO$_2$, or Al$_2$O$_3$-coated UO$_2$ with chromium diluent were prepared in stainless steel cladding. The specimens contain a small gas plenum which was deliberately defected with a 16-mil hole.

Work This Period

The specimens described above were scheduled to be heat treated in air at 1750 F. The weight gained was to be recorded and calculations made as to the extent of UO$_2$ oxidation which occurred in the various specimens. However, before running these tests, a stainless steel tube was heated in air at 1750 F and the weight gain recorded continuously. The rate of oxidation of the stainless tube (representing the cladding of the fueled specimens) was high enough to completely mask any anticipated weight gains in the fueled specimens. The scheduled tests were, therefore, dropped and alternative tests planned.

Two new lots of coated UO$_2$ have been prepared. One consists of 300-$\mu$-diameter UO$_2$ with a 25 to 30-$\mu$ coating of Al$_2$O$_3$. The second consists of 128-$\mu$-diameter UO$_2$ with 18 microns of porous Al$_2$O$_3$.

The coating integrity of the first batch was evaluated by first heating the powder in air at 1700 F. This was followed by a boiling nitric acid leach for 2 hr. Approximately 15 per cent of the material, assumed to be defective, was removed during these treatments. Both batches of powder were examined metallographically to confirm coating thickness.

Plans for Future Work

The defected specimens described earlier will be evaluated by techniques other than thermogravimetric weight gains. The specimens will be exposed to 1500 F air and periodically removed for examination. Such examinations will include diametrical measurements, radiographic determination of longitudinal fuel swelling, and visual observation.

The newly received lots of coated UO$_2$ will be evaluated during standard fabrication cycles to determine their characteristics.

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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 gas-cooled 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 and on X-ray diffraction studies and determinations of burnup and fission-gas retention.

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 were 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 fuel region of the specimens were slightly larger than those in the outer regions. There also appeared to be a decoration of the UO₂ grains at the boundaries by an unidentified phase and a system of voids.

Work This Period

X-ray diffraction studies of the UO₂ indicated that there was very little change in the UO₂ crystal structure during irradiation. A lattice contraction of 0.36 per cent and a 0.1 per cent internal strain in the crystal were measured.

Plans for Future Work

This completes the evaluation of the bulk UO₂ irradiated in Capsule AGN-BRR-3.

Radiation Stability of UO₂-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 UO₂ for possible use as a fuel for gas-cooled reactors.

Program Approach

Four specimens of BeO fueled with fully enriched 70 w/o UO₂ and two specimens of BeO fueled with fully enriched 80 w/o UO₂ 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 surface 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 and on X-ray diffraction data and determination 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 UO₂, had ruptured during the irradiation. The remaining five specimens were in good condition. Dimensional measurements indicated that the diameter of Specimen 11 had increased approximately 10 per cent while the diameter of the other five specimens had 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 analyses and microstructural examinations. Visual examinations 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 UO₂ was not appreciably affected by irradiation. There was some indication, however, that the BeO matrix had been embrittled in areas surrounding the UO₂ particles.
Work This Period

Samples of gases contained in the main body of the capsule and from five of the six specimens contained in the capsule were analyzed. Specimen 11 had ruptured during the irradiation, and all of the gas which was found in the main body of the capsule was assumed to have been released from this specimen. Analyses of the gas were performed by gamma-ray spectrometry and by mass-spectrographic techniques. These results along with the percentage of the gas that could have escaped from the fuel by the recoil mechanism are presented in Table U-1. Also included in this table are the burnups obtained from isotopic analyses, the burnups obtained by correlating the analyzed burnup data with the gamma-scan data, and the specimen-surface temperatures.

**TABLE U-1. FISSION-GAS RELEASE, IRRADIATION TEMPERATURE, AND FUEL BURNUP OF HASTELLOXY X-CLAD SPECIMENS OF 70 wt% UO₂ AND 80 wt% UO₂ DISPERSED IN BeO AND IRRADIATED IN CAPSULE BMI-38-1**

<table>
<thead>
<tr>
<th>Specimen</th>
<th>Fuel Loading, wt% UO₂</th>
<th>Amount of Krypton-85 Released, per cent</th>
<th>Estimated Amount of Krypton-85 Released by Recoil, per cent</th>
<th>Specimen-Surface Temperature (From Reactor Temperature Charts), °F</th>
<th>Estimated Uranium-235 Fission Burnup a/o</th>
</tr>
</thead>
<tbody>
<tr>
<td>Capsule</td>
<td>--</td>
<td>68.5(a)</td>
<td>17.8(b)</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>3</td>
<td>70</td>
<td>2.8</td>
<td>2.2</td>
<td>0.24</td>
<td>1355</td>
</tr>
<tr>
<td>11</td>
<td>80</td>
<td>--(a)</td>
<td>--(a)</td>
<td>0.24</td>
<td>1520</td>
</tr>
<tr>
<td>4</td>
<td>70</td>
<td>1.3</td>
<td>1.1</td>
<td>0.24</td>
<td>--(d)</td>
</tr>
<tr>
<td>6</td>
<td>70</td>
<td>1.5</td>
<td>1.2</td>
<td>0.24</td>
<td>1425</td>
</tr>
<tr>
<td>13</td>
<td>80</td>
<td>2.8</td>
<td>2.7</td>
<td>0.24</td>
<td>1300</td>
</tr>
<tr>
<td>10</td>
<td>70</td>
<td>0.6</td>
<td>0.6</td>
<td>0.24</td>
<td>1325</td>
</tr>
</tbody>
</table>

(a) Specimen 11 ruptured during irradiation and all gas found in capsule was assumed to have been released from this specimen.
(b) Mass spectrographic results believed to be unreliable in this case.
(c) Obtained by correlating isotopic-analysis burnups with gamma-scan profile.
(d) Thermocouple failed during assembly of capsule. No surface temperatures recorded.

Plans for Future Work

Additional metallographic examinations are being performed along with X-ray diffraction studies. From these data and those already obtained, a final evaluation of the irradiation stability of these fuels will be made.
CORROSION OF THORIUM AND URANIUM UNDER STORAGE CONDITIONS (LIVERMORE)

P. D. Miller

These studies are a part of a program in support of the Lawrence Radiation Laboratory and of the Sandia Corporation.

Electrodeposited Coatings for Thorium (SC)

J. G. Beach and C. L. Faust

Objective
To develop a coating for thorium that prevents corrosion and to evaluate techniques developed for thorium for the protection of uranium.

Program Approach
Various factors for producing sound, protective electroplated coatings on thorium are being studied: (1) prefinishing, to remove surface contamination and defective basis metal; (2) activating, to provide maximum adherence of the plated coatings; (3) coating system, to provide the needed protection and/or surface properties; and (4) postplating treatment, to improve the properties of the composite system. Metallography, bond tests, and corrosion tests are being used to evaluate the coated thorium. A cursory study toward protecting uranium by using the techniques developed for thorium is included so as to define the factors that need further research.

Progress Reported Previously
Corrosion protection of thorium and uranium was obtained using nickel or nickel-plus-chromium electroplates. Joining of thorium and uranium was facilitated by a plated coating.

The procedures for coating uranium and thorium with protective nickel electroplates (discussed in a topical report, BMI-1537) involve electropolishing as a prefinishing step prior to activation, plating, and outgassing. The uranium and thorium generally were in the cold-rolled or vapor-blasted condition for these previous studies.

Work This Period
The effect of mechanical surface finishing of the uranium and thorium on the characteristics of the subsequent nickel coating has become of interest.

Uranium that was grit polished and electropolished prior to activating and plating showed a better-quality nickel coating (based on metallographic examination) than did uranium that was not electropolished. On the other hand, thorium that was grit polished prior to activating and nickel plating showed a better-quality nickel coating than did thorium that was grit polished and electropolished.

The uranium and thorium were grit polished with 150 through 180 or 240-mesh abrasives to simulate a good-quality lathe-tool finish, or with 150 through 600-mesh abrasives to simulate a good-quality mechanically polished lathe-tool finish. Areas of the grit-polished surfaces were electropolished (~3 mils) for comparison.

The roughness of the surfaces, before activating and after plating with 2.5 mils of nickel, is related to the method of prefinishing as shown in Table V-1.
TABLE V-1. SURFACE ROUGHNESS OF URANIUM AND THORIUM AS INFLUENCED BY SURFACE FINISH, PRIOR TO ACTIVATION, AND BY NICKEL PLATING

<table>
<thead>
<tr>
<th>Metal</th>
<th>Surface Finish</th>
<th>Profilometer Readings, μin. rms</th>
<th>Before Activating and Nickel Plating</th>
<th>After Activating and Nickel Plating (2.5 Mils)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uranium</td>
<td>As rolled</td>
<td>6 to 8</td>
<td>Not measured</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Grit polished</td>
<td>5 to 10</td>
<td>65 to 80</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Grit polished and electropolished (-3 mils)</td>
<td>190</td>
<td>125</td>
<td></td>
</tr>
<tr>
<td>Thorium</td>
<td>As rolled and vapor blasted</td>
<td>40</td>
<td>60</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Grit polished</td>
<td>8 to 13</td>
<td>40 to 80</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Grit polished and electropolished (-3 mils)</td>
<td>20 to 35</td>
<td>30 to 50</td>
<td></td>
</tr>
</tbody>
</table>

Electropolishing of the uranium, to remove 3 mils of the surface, roughens the 10-μin. rms grit-blasted surface to a 190-μin. rms surface. Activation for adherent plating also roughens the uranium surface, but to a lesser degree. Nickel plating (leveling-type nickel-plating process) levels out some of the surface roughness.

Electropolishing and activating of thorium also roughen the surface, however, to a smaller extent.

Since electropolishing generally connotes smoothening, the cause of increasing roughness is of interest. But, in this work, electropolishing has been used primarily to remove metal without introducing mechanical working and without introducing hydrogen. The roughness observed after electropolishing of uranium and thorium appears similar to that observed when electropolishing other metals, such as aluminum-silicon alloys, containing nonmetallic or insoluble constituents. Thus, the roughening of thorium and uranium during electropolishing and activation is attributed to the nonmetallic inclusions and/or to intergranular attack of the currently available metal.

The protective quality of 2.5-mil nickel coatings (based on 100-hr exposure to water-saturated air at 200 F) as influenced by surface finish was less clearly defined. The protective quality of the nickel coatings appears to be more related to surface and subsurface defects in the basis metal than to the method of surface finishing prior to activating and plating.

Surface finishing of uranium and thorium by mechanical means, such as a good-quality lathe-tool finish, and/or abrasive polishing, appears to be a satisfactory preparation prior to plating sound and protective nickel coatings. Electropolishing of uranium after mechanical finishing and before activating for plating is desirable, whereas electropolishing of thorium after mechanical finishing is unnecessary.

Plans for Future Work

Electropolishing methods and activation methods for uranium and thorium will be further studied for providing smoother nickel-plated surfaces. Nickel-plated thorium and uranium samples (2 x 3 in.) are being prepared for studies at Sandia-Livermore.
Corrosion Evaluation (LRL)

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

Objective

To determine the durability of thorium, uranium, and related alloys under storage conditions.

Program Approach

Bare and coated coupons of the metals are being exposed under simulated storage conditions and under accelerated laboratory conditions. The work is being carried out in three phases.

Progress Reported Previously

A topical report, BMI-1507, details the results obtained on Phase I and Phase II programs.

In the Phase I program, thorium, uranium, and uranium-10 w/o molybdenum-alloy specimens were exposed to three accelerated laboratory conditions. The attack was localized and resulted in some pitting. The uranium was the most severely attacked. In general, the alloy was the most durable of the materials studied.

The Phase II study involved exposure of materials to 100 per cent relative humidity at 120 F for 2500 hr. A greater variety of specimen types was included in this program. Again, the uranium-10 w/o molybdenum alloy was the most durable material.

In a continuing Phase III program, specimens are being exposed in a warehouse and in a closed container at Columbus, Ohio, and at Daytona Beach, Florida. Results after 6 months were presented in two progress reports, BMI-1514 and BMI-1518.

Work This Period

The Phase III exposure program was continued.

Plans for Future Work

The Phase III program will be continued.
Objective
To develop fabrication techniques for cladding $\text{UO}_2$ fuel cores with beryllium.

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

Progress Reported Previously
Studies of the effect of surface condition and bonding parameters on the self-bonding of beryllium have been made. Good bonds were obtained. Specimens containing $\text{UO}_2$ cores or various barrier materials in contact with beryllium were bonded at various parameters to determine compatibility. No reaction was found between beryllium and $\text{UO}_2$ coated with vapor-deposited chromium. Moderate reactions occurred with all candidate barrier materials, but most could be parted easily.

Work This Period
Beryllia was investigated as a potential barrier material to prevent reaction between cladding and spacer materials. The coatings were applied to stainless steel shims by spraying a slightly acidic slurry of BeO and water. After spraying, the coated spacers were dried at approximately 140 F under an infrared heat lamp. A coating estimated to be 1 mil thick did not prevent reaction, but a 9-mil-coating was effective.

Surface-preparation samples were hand abraded on 40-grit SiC paper with a water coolant to a surface of 30 to 50 $\mu$m, rms and were bonded 4 hr at 1850 F and 10,000 psi. There was no apparent additional grain growth across the bond interface achieved with these conditions. Another sample received from ORNL was shaper surface finished to a surface finish of 210 $\mu$m, rms under a kerosene coolant. After bonding 4 hr at 1650 F and 10,000 psi, no improvement in the amount of growth across the bond interface was observed with this surface roughness.

Plans for Future Work
Work will be concentrated on surface-preparation studies of beryllium surfaces in an attempt to achieve complete grain growth during bonding. Efforts will be concentrated on shapered and grit-blasted surfaces.