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PROGRESS ON DEVELOPMENT OF FUELS AND
TECHNOLOGY FOR ADVANCED REACTORS
DURING JULY THROUGH SEPTEMBER, 1970
PROGRESS REPORT

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REPORTS RELATING TO REACTOR MATERIALS,
TECHNOLOGY, AND SAFETY ISSUED DURING THE PERIOD
JULY THROUGH SEPTEMBER, 1970

- BMI-1884 "Progress on High-Temperature Fuels Technology During February Through April, 1970", by Donald L. Keller and Walston Chubb.
- BMI-1885 "An Evaluation of the Applicability of Existing Data to the Analytical Description of a Nuclear-Reactor Accident. Quarterly Progress Report for April through June, 1970", by Wayne A. Carbiener and Robert L. Ritzman.
- BMI-1887 "Investigation of the Initiation and Extent of Ductile Pipe Rupture. Phase II. Progress Report for April-June, 1970", by Robert J. Eiber, Willard A. Maxey, and Thomas J. Atterbury.
- BMI-1888 "ADEP-1 and ADEP-2, One- and Two-Group Kinetics Codes", by Richard S. Denning.

HIGHLIGHTS OF RESULTS DURING REPORTING PERIOD

Results of studies at Battelle-Columbus during the quarterly period July through September, 1970, on development of fuels and technology for advanced reactors are summarized in this report. The work is being supported by AEC-DRDT and by ANL. The programs under way are concerned with development of uranium-plutonium nitride fuels for LMFBR applications, studies of high-temperature fuels for LMFBR's and advanced fast reactors, and studies of creep of reactor fuel materials. Highlights of the results reported are cited below.

Uranium-Plutonium Nitride Fuels for LMFBR Applications (Task 7)

The high fissile density, high thermal conductivity as compared with oxide fuels, and high breeding ratio of mixed-nitride fuels make them attractive for LMFBR applications. To fully define the potential of UN-PuN fuels, a program involving materials preparation studies, property measurements, long-term thermal and fast irradiations, and supporting studies is under way.

Postirradiation examination of fuel specimens from Capsules BMI-50-6 and BMI-50-11 continued, with emphasis on determining the cause of the two observed cladding failures of sodium-bonded specimens. Irradiation conditions in these capsules had been severe in terms of either burnup (35×10^{20} to 48.6×10^{20} fissions per cm^3 in Capsule BMI-50-6) or average heat generation rates (41.6 to 49.0 kw per ft in Capsule BMI-50-11). Fuel failure in Capsule BMI-50-6 was identified as being associated with the presence of an oxygen-stabilized sesquinitride phase that was present prior to irradiation. Fuel failure in Capsule BMI-50-11 was associated with gas-bubble-induced fuel swelling resulting from high fuel temperatures. Fuel swelling associated with gas-bubble formation was identified as beginning at a temperature slightly below 1400 C.

Capsules BMI-50-8 and BMI-50-10 were discharged from the ETR; postirradiated examination will be initiated. Capsule BMI-50-12 was inserted in the ETR for irradiation at specimen heat generation rates of about 40 kw per ft. This capsule incorporates a new design, permitting irradiation at slightly lower temperatures for a given heat generation rate.

An extensive quality assurance program was undertaken this quarter for the fabrication of fuel pins for a 19-pin subassembly to be inserted in the EBR-11. Loading procedures for mixed-nitride fuel were reviewed; those found inadequate for quality assurance purposes were identified and are being modified. An additional procedure, not utilized previously, involving xenon tagging of each fuel pin will be incorporated.

In the EBR-11 irradiation of mixed-nitride fuels, assembly and testing of the last four capsules of the seven-capsule BMI-3 series experiment continued. A subassembly containing 10 mixed-nitride capsules, three capsules in the BMI-1 series and the seven capsules in the BMI-2 series, was inserted into the EBR-11. Capsule BMI-1-3, containing a helium-bonded fuel pin, was discharged from the EBR-11 after achieving a fuel burnup of 3.8 a/o. This capsule will be shipped to the Battelle-Columbus Hot-Cell Facility for postirradiation examination.

In the preparations for transient irradiation evaluations of (U,Pu)N fuels in the TREAT facility, work progressed toward a definition of the reactor transient conditions for the first test and completion of the irradiation capsule design. Two individually clad specimens operating at different fission rates will be used.

In thermodynamic studies related to development of LMFBR fuels, effort concerned the self-diffusion of nitrogen in UN. A technique was developed where the diffusion coefficient can be determined at pressures and temperatures considerably lower than those previously employed in similar studies, thus negating the need for large extrapolations in pressure and temperature with their concomitant uncertainty. At 1800 C in the pressure range from 0.5 to 200 torr, the diffusion coefficient varies between 10^{-11} and 10^{-10} cm² per sec. The results show a marked dependence of the diffusion coefficient on pressure, and therefore on composition. Roughly speaking, an increase in the diffusion coefficient by a factor of 10 corresponds to a pressure increase by a factor of 100 or to a temperature increase of 150 C.

Technology of Nuclear Fuels for Advanced Fast Reactors (Task 6)

The external swelling behavior of UO₂ has been shown to be temperature dependent at cladding surface temperatures from 1200 to 1900 C. Preliminary data from twelve additional specimens in two capsules confirm the results reported earlier. In addition, the data show that there are no gross differences in the swelling behavior of UO₂ and UO₂-PuO₂.

Observations of gas release from UN at 1700 C suggest that bubbles have to grow to a certain size before they become available for involvement in the gas-release processes. The time delay involved in this growth is decreased by increasing porosity in the UN, suggesting that vacancy flow and other mechanical interactions between bubbles and pores are involved.

A preliminary assessment of the temperature dependency of the swelling of UN suggests that the onset of rapid swelling in UN occurs at a temperature near 1200 C and that the swelling rate of UN at 1350 C is about half that of UC and UO₂ at the same surface temperature.

Studies of the behavior of UN in irradiations at about 1600 C have suggested that much of the external swelling is caused by a few large bubbles in grain boundaries, while most of the fission gas remains in small bubbles within the grains. Similar conclusions were drawn as the result of studies of bubble distributions in lightly irradiated and annealed UC. Large bubbles appear in UC at grain boundaries, at subgrain boundaries, and at UC₂ phase boundaries, but the majority of the gas in lightly irradiated and annealed UC is present in bubbles so small that they can be redissolved by a short re-irradiation at ambient temperatures. A bubble growth model incorporating this concept of homogeneous precipitation followed by heterogeneous growth of bubbles has been developed. A preliminary trial of this model resulted in fortuitously good agreement with bubble size data, but predicted that over 60 percent of the gas was associated with grain-boundary bubbles, in poor agreement with the prior findings.

Creep of Reactor Fuel Materials
(Task 6 and ANL)

In-pile Creep Capsule BRR-4A was fabricated and operated with a UO_2 specimen. Creep tests were made at 4000 psi in the temperature range from 1000 to 1180 C at fission rates of 4×10^{12} and 1.2×10^{13} fissions/(cm^3)(sec). The creep data were in excellent agreement with creep data generated in earlier in-pile capsules. The in-pile creep rate in the temperature range from 1000 to 1180 C at a fission rate of 1.2×10^{13} fissions/(cm^3)(sec) is substantially enhanced as compared to the out-of-pile creep rate. The amount of enhancement increases as the fission rate increases. In addition, the enhanced creep rate in-pile in this temperature range is strongly dependent on temperature, with an activation energy close to the out-of-pile value.

Additional out-of-pile creep tests were performed on UO_2 specimens identical to the ones tested in-pile. The creep rate was found to be strongly dependent on stoichiometry, with the creep rate increasing as the stoichiometry was shifted from 2.001 to 2.10.

Fabrication has started on an in-pile creep capsule which will contain a UO_2 -22 w/o PuO_2 specimen. Specimens have been successfully fabricated from mechanically blended powder in a manner consistent with FFTF fuel preparation procedures.

4

4

4

4

TABLE OF CONTENTS

	<u>Page</u>
A. URANIUM-PLUTONIUM NITRIDE FUELS FOR LMFBR APPLICATIONS (AEC-DRDT)	A-1
Program Management	A-1
Thermodynamics and Transport Characteristics of PuN and Alloys of UN-PuN	A-2
(U,Pu)N Compatibility With Reactor Claddings and Coolants	Inactive
Diffusion in Nitrides	A-4
Fabrication and Cladding of (U _{0.8} Pu _{0.2})N Thermal Irradiation Specimens	Inactive
Thermal Irradiation Studies of Nitride Fuels	A-5
Postirradiation Evaluation of (U _{0.8} Pu _{0.2})N Thermal Irradiation Specimens	A-8
Quality Assurance for EBR-II Pin Fabrication	A-33
Preparation of EBR-II Fuel Pins	A-34
EBR-II Irradiation-Capsule Assembly and Safety Analysis Preparation	A-36
UN-PuN TREAT Experiments	A-39
B. TECHNOLOGY OF NUCLEAR FUELS FOR ADVANCED FAST REACTORS (AEC-DRDT)	B-1
Program Management	B-1
Irradiation Behavior of Oxide Fuels at High Temperatures	B-1
Irradiation Behavior of Nitride Fuels at High Temperatures	B-13
Irradiation Behavior of Carbide Fuels at High Temperatures	B-19
Measurement of Fission-Gas Diffusion and Bubble Growth in UO ₂	B-21
Gas-Bubble Nucleation and Growth	B-25
The Morphology and Migration of Lenticular Voids	B-31
Mechanical Model of Swelling of Nuclear Fuels	B-32
C. CREEP OF REACTOR FUEL MATERIALS (AEC-DRDT AND ANL)	C-1
Program Management	C-1
Design, Fabrication, and Operation of In-Pile Creep Capsules	C-2
In-Pile Creep Experiments	C-7
Mechanical and Thermal Analyses	C-14
Fabrication of UO ₂ -PuO ₂ Preirradiation and IN-Pile Compressive Creep Specimens	C-16
Preirradiation Compressive Creep Testing of UO ₂ -PuO ₂	C-19
Mechanisms of Creep in UO ₂ and UO ₂ -PuO ₂	C-22

URANIUM-PLUTONIUM NITRIDE FUELS FOR
LMFBR APPLICATIONS (AEC-DRDT)Program Management

W. M. Pardue, A. A. Bauer, P. Cybulskis,
and C. A. Alexander

This task has as its objective the evaluation and development of mixed-nitride fuels for advanced LMFBR applications. It is considered, based upon initial results on this program, that sodium-bonded, stainless steel-clad (U,Pu)N fuels offer considerable performance potential beyond that attainable by mixed oxides, the current reference fuel. These advantages can be summarized by the anticipated operating characteristics of (U,Pu)N:

- (1) Linear heat ratings (average) of 30 kw per ft
- (2) Burnup accumulation of 150,000 MWD/T
- (3) No chemical reaction with sodium or cladding
- (4) Extremely low swelling and fission-gas release
- (5) Resultant economy and reliability.

The task was initiated on the basis of the demonstrated excellent irradiation performance of UN reactor fuels and in recognition of the attainable performance from mixed oxides. The major phases of the program have been directed toward synthesis and fabrication, property measurements, and irradiation evaluation.

Synthesis and fabrication have had as their objective the establishment of methods for preparation of reproducible, high-quality mixed-nitride pellets for the other studies and the development of increased production rates. The materials thus prepared have then been used for studies of chemical compatibility, thermal stability, physical properties, and irradiation performance.

The irradiation studies address thermal, fast, and transient environments, and excellent performance has been demonstrated. The property studies are directed toward those subjects considered to be critical for nitride fuel performance. These include investigations of vaporization and decomposition tendencies, thermal conductivity, constituent migration (diffusion), phase relationships, etc., all required for design of irradiation specimens or interpretation of their performance. As information on various properties and irradiation behavior has become available, alterations in fabrication techniques have been undertaken so that fuels with increasingly better behavior might be prepared.

Currently, the available data on (U,Pu)N performance have demonstrated that the original hypothesis concerning its behavior was correct. Because the emphasis is on LMFBR application, the major thrust is directed toward the initiation of a statistically significant program of fast irradiations; thus, a full 19-pin subassembly is being prepared this fiscal year for insertion into the EBR-II. In this conjunction, an extensive quality assurance program has been undertaken to insure optimum fuel performance. Cladding, components, etc., are being obtained, where possible, from sources such as FFTF-approved vendors, and analytical calculational services are being subcontracted to laboratories with well-established

computer codes for this purpose. In the interim, a series of EBR-II pins previously inserted in the reactor is being returned to the Battelle-Columbus Hot-Cell Facility for evaluation.

In order to evaluate the transient response of (U,Pu)N, a series of TREAT experiments has been designed and is being prepared for completion this year. This task has been organized so that a generalized understanding of the transient behavior will be obtained.

In addition to general technical coordination, the program management involves external communication with vendors, establishment of the quality assurance program, and interpretation of programmatic significance in terms of the overall USAEC LMFBR program.

Thermodynamics and Transport Characteristics of PuN and Alloys of UN-PuN

C. A. Alexander, J. S. Ogden, and W. M. Pardue

Objective of Program

To determine those properties pertaining to plutonium nitride-bearing fuel compositions which allow predictions of fuel stability, transport, and other factors needed to evaluate the behavior of the fuel under possible reactor environmental conditions of both transient and steady-state nature.

Approach Being Used

Kinetics and thermodynamics of vaporization processes are being investigated by effusion-thermobalance and mass-spectrometric techniques. The existence of a homogeneity range is being examined under controlled atmosphere conditions. The Oelsen calorimeter is being utilized to experimentally determine heat capacities as a function of temperature and alloy composition.

The experimental apparatus used in the studies consists of a cold-wall furnace constructed of stainless steel which is evacuated through a pipe 10 cm in diameter, a diffusion pump with a capacity of 300 liters per sec, and a mechanical pump with a capacity of 7 liters per sec. The system is equipped with valves so that the furnace chamber can be isolated from the pumps and backfilled with inert gas or any nonoxidizing gas to the desired pressure. The furnace assembly has a maximum leak rate of 10^{-9} std cm³. A Cahn recording microbalance is also incorporated into the vacuum chamber which houses the basic vaporization furnace. The electrobalance is used to continuously monitor the vaporization rates as a function of time and/or temperature.

The double-focusing mass spectrometer used for the surface-ionization studies on neodymium has a focal point about 8 cm on either side of the 25-cm radius of curvature in the electrostatic sector. The magnetic sector is asymmetric, and the distance from the image to the 60-deg, 20-cm sector is shorter than the object distance. The electromagnet incorporates a Rawson field meter in a feedback arrangement to supply short-term stability of about 1 ppm in the magnet power supply. A field sweep is also incorporated in such a manner that the sweep can be made in gaussess rather than in amperes. The design criteria of the machine are such that with 2500 v of accelerating potential, masses up to 1200 are resolvable in the magnetic sector. A resolution of about 2500 at mass 25 was selected as a

reasonable compromise between resolution and transmission. The sensitivity is such that the individual events can be monitored. Data output is on a strip-chart recorder using a Cary Model 31 vibrating-reed electrometer and Faraday cage or an electron multiplier tube and appropriate amplifier.

Progress Reported Previously

The behavior of neodymium in NdN as a measure of burnup revealed that there was neither a lack of thermal stability, nor the existence of a gaseous species, nor an anomaly in chemistry that can account for any lack of agreement when isotopic analyses of mixed-nitride fuels are compared with those obtained from mixed oxides.

Equations for mass transport and general thermochemical properties were obtained. It was also established that the presence of PuN in solution in UN decreases the uranium activity and thereby prevents the formation of a molten phase at temperatures to at least 2000 K. At both 1 and 0.1 atm of nitrogen, no homogeneity range for PuN was detected over the temperature range from 1500 to 2140 C. Pressure-temperature-composition relationships for PuN and (U, Pu)N were also prepared.

The homogeneity ranges for PuN and $(U_{0.8}Pu_{0.2})N$ are very narrow. The nitrogen/metal ratio varies from 1 at 1 atm of nitrogen to 0.968 at 10^{-4} atm of nitrogen.

Work This Quarter

Work this quarter was concentrated on an effort to determine whether the homogeneity range in $(U_{0.8}Pu_{0.2})N$ can give rise to disproportionation reactions with concomitant buildup of high nitrogen pressures. Design and evaluation was undertaken of a cell in which the total nitrogen pressure can be determined under conditions leading to little or no compositional change in the fuel. This latter restriction dictates that the technique be a static one where the gas volume is limited and that the pressure be measured directly. The final design utilizes a tungsten cell about 5 cm high and 2 cm in diameter containing blackbody holes for temperature determinations. A tungsten tube approximately 15 cm long is welded to the cell. This tube has its terminal end threaded and makes an O-ring seal with a water-cooled copper tube leading out of the furnace to a Pirani gage and to a precision diaphragm gage. The Pirani gage has been calibrated for nitrogen over the range from 0 to 100 torr, and the diaphragm gage, up to 2 atm. Provisions are available to protect the Pirani gage at the higher pressures. Total system volume is about 35 cm³; hence, with a charge of 0.03 mole of $(U_{0.8}Pu_{0.2})N$ (about 8 g), the solid would lose 3 percent of its nitrogen to pressurize the system at 1 atm. This appears to be about the limit that can be obtained as far as constancy of metal/nitrogen ratio is concerned.

Static runs have been performed with the cell when the cell is evacuated and the furnace contains nitrogen at 1 atm at 2000 C. The leak rate into the cell has been found to be very low; hence, it is concluded that leakage out of the cell should not be a serious factor in the determinations.

Plans for Future Work

During the next quarter, it is planned to determine total nitrogen pressures over $(U_{0.8}Pu_{0.2})N$.

Diffusion in Nitrides

J. W. Droege and C. A. Alexander

Objective of Program

To investigate the self-diffusion behavior in nitride fuels of interest to LMFBR applications.

Approach Being Used

The self-diffusion behavior of nitrogen in UN and (U, Pu)N is being determined in the temperature range from 1360 to 1800 C and the nitrogen pressure range of 0.5 to 100 torr. An isotopic tracer technique using N_2^{15} is being employed. The sample is sectioned electrolytically after diffusion annealing and the nitrogen evolved is analyzed with a mass spectrometer.

Progress Reported Previously

Self-diffusion of nitrogen in UN was studied over the temperature range from 1360 to 1800 C and the nitrogen pressure range of 0.5 to 10 torr. At the highest temperature and lowest pressure, an argon-nitrogen mixture was used to retard vaporization of the sample. Results of analyses of nitrogen showed straight-line behavior on probability paper up to a maximum penetration of from 3×10^{-4} to 10×10^{-4} cm. Subsequent deviations were presumed to be the result of electrolytic etching at cracks and grain boundaries. The results were lower than those reported by Holt and Almasy⁽¹⁾, making allowance for temperature differences.

Work This Quarter

Vacuum leaks and some difficulty in reconstructing the heating element in the small annealing furnace resulted in delay. The rebuilt furnace element allowed for a maximum temperature slightly lower than the previous furnace, but with better temperature control. Diffusion anneals were carried out on a UN specimen at 1730 C under a nitrogen pressure of 60 torr. On the basis of an estimated temperature-pressure-composition relationship, the composition of this experiment matched the composition of two previous experiments at lower pressures and temperatures. The results showed a diffusion coefficient of 2.5×10^{-11} cm² per sec. A plot of log D versus $1/T$ showed good agreement among the three points and led to an activation energy of 96 kcal per mole. This value is based on only three points and depends on the assumption that the composition of these specimens was the same, i.e., that the estimated pressure-temperature-composition relationship is valid. Short extrapolations make possible a comparison of this result with two previously obtained in this work at 1800 C and with two reported by Holt and Almasy⁽¹⁾ at 1780 C. The comparison (Figure A-1) shows a pressure dependence similar to that reported by the latter authors at 1940 C. This appears to remove the discrepancy previously supposed between the two efforts. The pressure covers the range from 0.5 to 200 torr, and the diffusion coefficient, the range from 10^{-11} to 10^{-10} cm² per sec. These results show a marked dependence of the diffusion coefficient on pressure, and therefore on composition. Roughly speaking, an increase in the diffusion coefficient by a factor of 10 corresponds to a pressure increase by a factor of 100 or to a temperature increase of 150 C. This relationship between pressure and diffusion is not unreasonable in that an increase in pressure by a factor of n^2 should affect the activity by n . Quite possibly, the

activity gradient would also be increased by n , and concomitantly, the diffusion by the same magnitude.

FIGURE A-1. PRESSURE DEPENDENCE OF DIFFUSION COEFFICIENT OF NITROGEN IN UN AT 1800 C

Plans for Future Work

Measurements of the diffusion of nitrogen in (U, Pu)N will be undertaken after installation of equipment in the Battelle-Columbus Plutonium Facility.

Thermal Irradiation Studies of Nitride Fuels

E. O. Fromm, N. E. Miller, and J. W. Ray

Objective of Program

To irradiate UN-PuN fuels to burnups of 150,000 MWD/T at fission-heat-generation rates and temperatures typical of sodium-cooled fast-reactor operations.

Approach Being Used

A series of irradiation-capsule experiments involving $(U_{0.8}Pu_{0.2})N$ specimens is planned for exposure in the high-flux facilities of the ETR or MTR. The guiding criteria include:

- (1) Specimen temperature levels up to 700 C at the cladding surface
- (2) Fuel diameters of about 0.225 in.
- (3) Fission heat generation up to 40 kw per ft of fuel pin
- (4) Fuel burnup levels of 40,000 to 150,000 MWD/T, with accrual of the latter during an in-pile exposure of about 1-1/2 calendar years.

The program includes design, construction, and loading of capsules, monitoring of the irradiation, and interpretation of the conditions of irradiation.

Progress Reported Previously

The initial irradiation-capsule design phase was begun in September, 1965. The first report on this phase of the program appeared in BMI-1745.

The irradiation-capsule design, beginning with Capsule BMI-50-4, is based on a double-walled, finned-annulus concept utilizing electric heaters to keep the sodium molten in the various annuli, as described in BMI-1791. In addition, temperature control capability up to 110 F (60 C) is provided by the electric heaters. For additional temperature control, the capsule is moved to different flux-level positions.

The irradiation history of the first capsule of the new design, designated Capsule BMI-50-4, was reported in BMI-1845. The irradiation summaries for follow-on Capsules BMI-50-5 through BMI-50-10 were reported in BMI-1868 and BMI-1886. Details of the modified capsule design for Capsule BMI-50-11 to permit irradiation at heating rates in excess of 40 kw per ft were reported in BMI-1868; the irradiation history was reported in BMI-1886. The capsule components for the next capsule, designated Capsule BMI-50-12, were fabricated as described in BMI-1886.

Work This Quarter

After a prolonged shutdown of the ETR to change the beryllium in the reflector region, operation was resumed for Cycle 107 (beginning May 27, 1970) with Capsules BMI-50-8 and BMI-50-10 moved to positions of higher neutron flux (from E-5-NE to F-4 and E-5-NW to E-5-NE, respectively).

Temperatures of both capsules in the new facilities during most of this cycle were at satisfactory levels. However, on June 27, 1970, on return to full power after a reactor scram, capsule temperatures higher than normal were noted on both Capsules BMI-50-8 and BMI-50-10. Temperatures of both capsules continued to rise, necessitating reduction of reactor power to keep the temperatures below maximum permissible levels. At this time the high temperatures were believed to be caused by something external to the capsules (possibly blockage in the water flow channels of the irradiation facilities). Subsequently, the temperature of Capsule BMI-50-10 dropped several times during periods of relatively stable reactor power, whereas the temperatures of Capsule BMI-50-8 remained constant during this same period. Since it was not possible to satisfactorily explain the behavior of Capsule BMI-50-10, the decision was made by Idaho Nuclear Corporation to discharge the capsule. When the capsule was removed from the reactor, a light brown deposit was found on the outer nickel shell, particularly in the zone adjacent to the maximum heat rejection area of the capsule. The thickness of the deposit was sufficient to cause a noticeable increase in the diameter of the capsule; this would have resulted in reduced coolant flow with attendant high capsule temperatures. The cause for the deposit has not been determined, but may be associated with similar appearing deposits that have been found subsequently on other experiments as well as on the reactor fuel elements themselves. The deposit on Capsule BMI-50-10 was easily scraped off and a sample was removed and analyzed. The preliminary analysis showed the deposit to be primarily nickel with a small amount of nickel hydroxide. The investigation into the cause of the deposit is still being pursued. The capsule is presently in transit to the Battelle-Columbus Hot-Cell Facility for postirradiation examination. The fuel-surface temperatures of specimens in Capsule BMI-50-10 for Cycle 107 are reported in Table A-1; accumulated burnup for each specimen in this capsule is reported in Table A-2.

TABLE A-1. TEMPERATURE HISTORY FOR CAPSULE BMI-50-10 IN POSITION E-5-NE DURING ETR CYCLE 107

Type of Temperature Data	Fuel-Surface Temperatures for Indicated Specimens as Extrapolated from Thermocouple Data ^(a) , C					
	A-41 ^(b)	A-51 ^(b)	B-41	B-42	C-41	C-42
Average	625	620	620	655	630	720
Maximum	770	720	715	760	760	830

(a) Specimen lineup from top to bottom in the capsule is as follows:

Specimens A-41 and A-51: helium bond

Specimens B-41 and B-42: 10-mil sodium bond

Specimens C-41 and C-42: 20-mil sodium bond.

(b) Temperatures reported for the helium-bonded specimens are for the ID of the cladding.

TABLE A-2. ACCUMULATED SPECIMEN BURNUP DATA AT THE TIME OF DISCHARGE FOR CAPSULES BMI-50-8 AND BMI-50-10

Specimen	Total Accumulated Burnup	
	10^{20} Fissions per Cm ³	10^3 MWD/T ^(a)
<u>Capsule BMI-50-8</u>		
A-25	34.6	115
A-26	38.1	109
B-25	46.6	153
B-28	41.9	121
C-25	46.0	132
C-26	53.1	152
<u>Capsule BMI-50-10</u>		
A-41	18.2	67
A-51	18.3	60
B-41	18.5	69
B-42	19.6	60
C-41	20.4	66
C-42	23.2	78

(a) Based on 200 Mev per fission.

The irradiation of Capsule BMI-50-8 in Position F-4 was continued into the first week of ETR Cycle 108 (beginning July 23, 1970), at which time temperature fluctuations were noted for the first time in four thermocouples. Since a somewhat similar situation had been encountered in Capsule BMI-50-10 during Cycle 107 and since the target burnup was nearly reached, the decision was made to discharge this capsule and return it to the Battelle-Columbus Hot-Cell Facility along with Capsule BMI-50-10. Capsule BMI-50-8 was discharged on July 29, 1970. The fuel-surface temperatures of specimens in this capsule for Cycle 107 are reported in Table A-3; accumulated burnup for each specimen is given in Table A-2.

TABLE A-3. TEMPERATURE HISTORY FOR CAPSULE BMI-50-8 IN POSITION F-4 DURING ETR CYCLE 107

Type of Temperature Data	Fuel-Surface Temperatures for Indicated Specimens as Extrapolated from Thermocouple Data ^(a) , C					
	A-25 ^(b)	A-26 ^(b,c)	B-25	B-28	C-25	C-26
Average	560	605	675	605	665	821
Maximum	725	770	835	770	790	965

- (a) The specimen lineup from top to bottom in the capsule is as follows:
 Specimens A-25 and A-26: helium bond
 Specimens B-25 and B-28: 10-mil sodium bond
 Specimens C-25 and C-26: 20-mil sodium bond.
- (b) Temperatures reported for helium-bonded specimens are for the ID of the cladding.
- (c) The thermocouple for Specimen A-26 failed during the early part of Cycle 106A. Therefore, the surface temperature for this specimen was estimated from adjacent Specimen A-25.

Capsule BMI-50-12 was assembled and sent to the reactor for irradiation beginning with ETR Cycle 109 (September 24, 1970). This capsule differs from the previous capsules in the BMI series in that a single outer shell of Type 316 stainless steel was used in place of the combination of a finned intermediate shell of nickel and a water-contacting outer shell of nickel. As in Capsule BMI-50-11, niobium-1 w/o zirconium was substituted for "A" nickel as the material for the heavy-wall compartments. The new design permits irradiation at slightly lower temperatures for a given heat generation rate. In addition, the thermal stresses as well as the stress due to the fins have been reduced greatly. The capsule was inserted into ETR Position E-4 for Cycle 109. Initial temperatures for the cycle indicated a heat generation rate of 36 to 41 kw per ft along the specimen array, with corresponding fuel-surface temperatures of 705 to 770 C.

Plans for Future Work

The irradiation of Capsule BMI-50-12 will continue to a fuel burnup of about 100,000 MWD/T.

The postirradiation examination for Capsules BMI-50-8 and BMI-50-10 will be initiated following neutron radiography at the Battelle Research Reactor. The investigation into the cause for the deposit on the surface of Capsule BMI-50-10 will continue.

(1) Holt, J. B., and Almassy, Y., J. Am. Ceram. Soc., 52, 631-635 (1969).

Postirradiation Evaluation of (U_{0.8}Pu_{0.2})N Thermal Irradiation Specimens

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Objective of Program

To evaluate the stability of mixed-nitride fuels irradiated in a thermal-neutron flux at cladding temperatures up to 700 C, heat generation rates up to 40 kw per ft, and burnups up to 150,000 MWD/T.

Approach Being Used

Helium-bonded and sodium-bonded ($U_{0.8}Pu_{0.2}$)N pellets clad in 15-mil-thick Type 304L stainless steel will be examined following irradiation in the ETR or MTR. The postirradiation examination planned involves (1) measurement of fission-gas release from clad specimens to the capsule and from fuel to the cladding, (2) visual inspection of the cladding for defects or evidence of chemical reaction, (3) measurement of the dimensions and density of the specimens, and (4) metallographic examination of the cladding and fuel to determine cladding-fuel compatibility and changes in the microstructures of cladding and fuel. When appropriate, additional investigation techniques such as gamma scanning, autoradiography, X-ray diffraction analyses, electron-microprobe analysis, and fuel-burnup analyses will be used.

Progress Reported Previously

In Capsules BMI-50-4, BMI-50-5, BMI-50-7, and BMI-50-9, burnups ranged from 11.0×10^{20} to 34.5×10^{20} fissions per cm^3 , and average heat generation rates ranged from 13.5 to 32.2 kw per ft. Fuel swelling rates in these capsules were uniformly low at 0.48 percent per 10^{20} fissions per cm^3 or less. None of the sodium-bonded specimens exhibited any cladding strain and the remaining sodium-filled annuli were large enough to accommodate considerably more fuel swelling. All of the helium-bonded specimens experienced cladding strain and several had failed claddings. Fission-gas release from the high-density fuel was very low, being on the order expected from recoil from the free surfaces. The somewhat higher gas release from low-density specimens was attributed to the greater exposed surface area associated with the high degree of surface-connected porosity.

Consistent with the low swelling and low gas release was the absence of optically resolvable fission-gas bubbles in the fuel.

No evidence of interaction between the cladding and fuel or fission products was found by metallographic examination or electron-microprobe analysis. Electron-microprobe analysis also showed that no segregation of uranium and plutonium had occurred. Kjeldahl analyses indicated that the cladding did not pick up any significant quantities of nitrogen during irradiation.

In Capsules BMI-50-6 and BMI-50-11, the irradiation conditions were more severe in terms of either burnup (35×10^{20} to 48.6×10^{20} fissions per cm^3 in Capsule BMI-50-6) or average heat generation rates (41.6 to 49.0 kw per ft in Capsule BMI-50-11).

Preliminary results of the examination of fuel specimens from Capsule BMI-50-6 indicated a large amount of cladding strain in the helium-bonded specimens, a slight amount of cladding strain in the 10-mil sodium-bonded specimens, and no cladding strain in the 20-mil sodium-bonded specimens. Claddings on both helium-bonded specimens and one 10-mil sodium-bonded specimen were ruptured.

In Capsule BMI-50-11, all specimens experienced some cladding strain. However, only one 10-mil sodium-bonded specimen had a visibly failed cladding. This failure was oriented in a circumferential direction and did not appear to be caused by the hoop stresses generally associated with swelling fuel.

Postirradiation macrographs of specimens from Capsules BMI-50-6 and BMI-50-11 were previously recorded in BMI-1886.

Work This Quarter

The postirradiation examination of fuel specimens from Capsules BMI-50-6 and BMI-50-11 continued this quarter with emphasis on determining the cause(s) of the two observed cladding failures of sodium-bonded specimens. Also, metallographic examinations of sound specimens were performed to more clearly establish the effects of high burnup and high heat generation on fuel swelling, fission-gas release, and fuel-cladding compatibility.

The irradiation conditions and swelling and fission-gas-release data for specimens from Capsules BMI-50-6 and BMI-50-11 are summarized in Tables A-4 and A-5. As can be seen from the neutron radiographs of Figure A-2, a helium-bonded specimen from Capsule BMI-50-11 and sodium-bonded specimens from both Capsules BMI-50-6 and BMI-50-11 contained cracked fuel pellets. Data on the specimen cladding diameters were obtained by performing vernier micrometer measurements along each specimen at 0- and 90-deg notations. Fuel-pellet diameter data for helium-bonded specimens were based upon cladding diameter data, assuming no clearance between fuel and cladding. Fuel-pellet diameter data for sodium-bonded specimens were based on dimensions taken from neutron radiographs. Length changes, in all cases, were measured from neutron radiographs.

Volume changes of mixed-nitride fuel as a function of burnup are plotted in Figure A-3. The fuel behavior, in general, was independent of bonding medium and of fuel density. All of the specimens from Capsule BMI-50-11 and four of the specimens from Capsule BMI-50-6 exhibit swelling rates that exceed the maximum recorded for specimens from previous capsules. However, two features distinguish these specimens from the majority of specimens in previous irradiations. The specimens in Capsule BMI-50-11 were irradiated at abnormally high heat ratings, leading to maximum central fuel temperatures ranging from 1390 to 1665 C; therefore, the maximum temperatures associated with individual specimens in this capsule are given beside each volume-change data point. By comparison, fuel temperatures for all remaining specimens were a maximum of 1260 C. The specimens in Capsule BMI-50-6, while irradiated under conditions within this lower temperature range, are identified as having high oxygen contents (3000 to 4000 ppm) when compared with fuel from most of the remaining specimens, where oxygen contents ranged from 300 to 500 ppm. Exceptions are three specimens from Capsule BMI-50-5 which contained fuel similar to that in Capsule BMI-50-6. The high burnups experienced by the fuel in the latter capsule are not regarded as being responsible for the accelerated swelling because of the similarity in appearance between this fuel and the lower-burnup, high-oxygen fuel of the earlier capsule.

Additional results for the examination of specimens from Capsules BMI-50-6 and BMI-50-11 are discussed below.

Capsule BMI-50-6. Two samples extracted from 10-mil sodium-bonded Specimens B-1 and B-2 were examined metallographically. The characteristic cracking pattern, Figure A-4, with porosity in a different appearing structure concentrated near the fuel OD has been observed before in specimens from Capsule BMI-50-5, as reported in BMI-1868. At that time, the nature and source of this structure were uncertain. X-ray diffraction examination revealed only the mononitride structure and it was concluded that the structure represented a solid solution of oxygen, and possibly carbon, contamination in a (U, Pu)(N, O, C) phase. However, because the structure concentrated near the fuel OD and along cracks it was uncertain whether the contamination stemmed from an external source, such as the

TABLE A-4. IRRADIATION CONDITIONS AND SWELLING DATA FOR FUEL SPECIMENS IN CAPSULE BMI-50-6
(IRRADIATION CONDITIONS BASED ON THERMOCOUPLE DATA)

	Specimen Designation					
	A-4	A-3	B-2	B-1	C-2	C-1
Bond						
Type	Helium	Helium	Sodium	Sodium	Sodium	Sodium
Cold Gap, mils	<1	<1	10	10	20	20
Density, percent of theoretical						
Pellet	83.8	95.5	83.9	95.5	96.5	94.4
Smear	83.8 ^(a)	95.5 ^(a)	73.3	83.0	70.0	68.0
Burnup						
10^{20} Fissions per Cm^3	39.7	47.7	48.6	43.1	46.3	35.0
10^3 MWD/T ^(b)	130	132	157	123	131	102
Irradiation Temperature ^(c) , C						
Fuel Surface						
Average	625	695	690	615	640	545
Maximum	725	800	800	720	750	645
Fuel Center						
Average	945	1070	1070	900	965	915
Maximum	1130	1240	1240	1090	1145	960
Linear Heat Generation ^(c) , kw per ft						
Average	27.8	32.5	32.3	26.1	28.6	22.7
Maximum	34.5	39.7	39.7	34.1	36.2	29.5
Cladding Diameter Increase, percent						
Average	4.1	4.4	0.15	0.6	0	0
Maximum	5.8	5.2	0.3	1.9	0	0
Fuel Diameter Increase ^(d) , percent	6.6	5.9	5.6	12.6	18.5	16.7
Fuel Length Increase ^(e) , percent	8.6	5.9	9.2	9.6	4.9	5.7
Fuel Volume Increase, percent	21.8	17.7	21.4	34.8	41.9	39.1
Fission-Gas Release, percent	--	--	27	--	--	--
Cladding Condition ^(f)	Failed	Failed	Sound	Failed	Sound	Sound

(a) Slip fit of fuel into cladding - essentially no clearance.

(b) Based on 200 Mev per fission.

(c) Calculated for last third of irradiation.

(d) Calculated from cladding diameter increase for helium-bonded specimens; for sodium-bonded specimens, dimensions obtained from neutron radiographs.

(e) Determined from neutron radiographs.

(f) Based on macroappearance.

TABLE A-5. IRRADIATION CONDITIONS AND SWELLING DATA FOR FUEL SPECIMENS IN CAPSULE BMI-50-11
(IRRADIATION CONDITIONS BASED ON THERMOCOUPLE DATA)

	Specimen Designation					
	A-43	A-44	B-43	B-54	C-43	C-44
Bond						
Type	Helium	Helium	Sodium	Sodium	Sodium	Sodium
Cold Gap, mils	<1	<1	10	10	20	20
Density, percent of theoretical						
Pellet	82.7	94.7	82.7	94.4	94.3	94.3
Smear	82.7 ^(a)	94.7 ^(a)	70.2	80.6	69.0	68.8
Burnup						
10^{20} Fissions per Cm^3	17.0	18.2	15.9	19.2	20.1	18.9
10^3 MWD/T ^(b)	55.7	52.5	58.5	55.0	57.5	54.5
Irradiation Temperature, C						
Fuel Surface						
Average	830	880	870	920	960	910
Maximum	930	970	980	1015	1105	1035
Fuel Center						
Average	1230	1310	1295	1375	1440	1360
Maximum	1390	1455	1470	1520	1665	1550
Linear Heat Generation, kw per ft						
Average	41.6	44.5	44.0	46.5	49.0	46.4
Maximum	47.5	50.1	50.3	52.2	57.6	53.1
Cladding Diameter Increase, percent						
Average	0.8	1.7	0	1.2	1.0	0.2
Maximum	1.4	2.2	0	2.8	2.0	0.8
Fuel Diameter Increase ^(c) , percent	1.6	2.4	6.5	8.6	14.6	5.6
Fuel Length Increase ^(d) , percent	5.3	9.9	3.7	5.9	6.0	6.9
Fuel Volume Increase, percent	8.5	14.7	16.7	23.1	35.2	18.1
Fission-Gas Release, percent	6.0	5.9	7.4	8.3	2.8	4
Cladding Condition ^(e)	Sound	Sound	Sound	Failed	Sound	Sound

(a) Slip fit of fuel into cladding - essentially no clearance.

(b) Based on 200 Mev per fission.

(c) Calculated from cladding diameter increase for helium-bonded specimens; for sodium-bonded specimens, dimensions obtained from neutron radiographs.

(d) Determined from neutron radiographs.

(e) Based on macroappearance. The presence of fission gas in all the capsule compartments suggests that pinhole leaks existed in the cladding of at least one specimen in each compartment.

Specimen A-4
(helium bond)

Specimen A-43
(helium bond)

Specimen A-3
(helium bond)

Specimen A-44
(helium bond)

Specimen B-2
(10-mil sodium)

Specimen B-43
(10-mil sodium bond)

Specimen B-1
(10-mil sodium)

Specimen B-54
(10-mil sodium bond)

Specimen C-2
(20-mil sodium)

Specimen C-43
(20-mil sodium bond)

Specimen C-1
(20-mil sodium)

Specimen C-44
(20-mil sodium bond)

BMI-50-6

BMI-50-11

FIGURE A-2. NEUTRON RADIOGRAPHS OF SPECIMENS FROM
CAPSULES BMI-50-6 AND BMI-50-11

FIGURE A-3. VOLUME CHANGE OF (U,Pu)N SPECIMENS AS A FUNCTION OF BURNUP

TEO1928

FIGURE A-4. LOW-MAGNIFICATION COMPOSITE OF METALLOGRAPHIC SAMPLE FROM 10-MIL SODIUM-BONDED SPECIMEN B-1 (CAPSULE BMI-50-6)

A burnup of 123,000 MWD/T was achieved. Note the porosity concentrated near the fuel OD.

sodium, or from the fuel itself. The results from this capsule show that only fuel with an initially high oxygen content exhibits the cracking pattern shown in Figure A-4. Specimens in Capsules BMI-50-5 and BMI-50-6 are the only ones in which the fuel contained 3000 to 4000 ppm oxygen. By comparison, the remainder of the fuel has an oxygen content of between 300 and 500 ppm.

The high oxygen content is not related to a particular fabrication process. High-density pellets for Capsules BMI-50-4, -5, -6, and -7 and low-density pellets for Capsule BMI-50-4 were all fabricated by hot pressing, while all remaining pellets have been pressed and sintered. Rather, the contamination is related to the glove-box atmosphere which, at the time that fuel for these first capsules was fabricated, was less well controlled than at present.

The microstructure of control pellets for this irradiation is illustrated in Figure A-5 along with the microstructure of typical low-oxygen fuel. The contaminant phases appearing in the mixed-nitride microstructure are metallographically identified as a mixture of U(Pu)O_2 and U_2N_3 . Oxygen has been shown to stabilize the sesquinitride, and it is believed that under the specific fabrication conditions employed U_2N_3 formed in the structure. Thus, the normal fabrication procedure involves either hot pressing or sintering at elevated temperature under a nitrogen atmosphere and then cooling to 1400 C before the nitrogen is evacuated to prevent U_2N_3 formation on subsequent cooling. This procedure has been shown to be satisfactory for low-oxygen fuel. However, the temperature of 1400 C may not be sufficiently high with high-oxygen fuel to prevent sesquinitride formation in the presence of nitrogen. Thus, evacuation of nitrogen at some higher temperature may be required to prevent sesquinitride formation in high-oxygen material.

The microstructure of the fuel across a cross section of Specimen B-6 is shown in Figure A-6. Maximum porosity exists near the fuel OD, although porosity exists throughout this originally high-density fuel. The major portion of the structure appears as in Figure A-6c with no evidence of a variation in porosity associated with temperature gradient across the fuel. It appears rather that the fuel porosity is related to decomposition of the sesquinitride during irradiation although once the porosity has formed fission gases may contribute, particularly in the high-burnup region near the fuel OD.

To complement two samples extracted from Specimen B-2 that are being analyzed for retained fission gas and burnup (mass spectrometric analysis), a third sample was extracted for metallographic examination. High-burnup Specimen B-2 (48.6×10^{20} fissions per cm^3) did not experience a cladding failure. The 27 percent fission-gas release (Table A-4) was higher than expected for the estimated average fuel surface temperature of 690 C. However, examination of the fuel microstructure of Specimen B-2, Figures A-7 and A-8, shows that a large proportion of the outer periphery of the fuel contains large bubbles (Figure A-8b). The microstructure of central regions of the fuel, as depicted in Figure A-8a, contains a more normal amount of porosity as associated with low-density (83.9 percent of theoretical) fuel. Increased porosity as a result of sesquinitride decomposition and subsequent release of fission gas to this porosity particularly in the peripheral regions of the fuel, are suggested as the causes for the higher fission-gas release.

Microstructures of the cladding on Specimens B-1 and B-2 appeared as expected. Typical views are given in Figure A-9, where the cladding on Specimen B-1 is depicted. Etched regions containing what appear to be carbides precipitated at grain and twin boundaries are normal and have been observed in prepared claddings from previous capsules.

500X

High Density

AB74

500X

High Density

AB75

a. Fuel Containing 3000 to 4000 ppm Oxygen

500X

High Density

AB76

500X

Low Density

AB77

b. Fuel Containing 300 to 500 ppm Oxygen

FIGURE A-5. MICROSTRUCTURES OF MIXED-NITRIDE FUELS WITH HIGH AND LOW OXYGEN CONTENTS

a. View at the Fuel OD

b. Approaching the Fuel OD

c. Fuel Center

FIGURE A-6. PHOTOMICROGRAPHS OF SELECTED AREAS FROM
10-MIL SODIUM-BONDED SPECIMEN B-1
(CAPSULE BMI-50-6)

TEO1931

FIGURE A-7 LOW-MAGNIFICATION COMPOSITE OF METALLOGRAPHIC SAMPLE FROM 10-MIL SODIUM-BONDED SPECIMEN B-2 FROM CAPSULE BMI-50-6

A burnup of 157,000 MWD/T was achieved Note the porous phase on the OD of the fuel

500X

HC24288

a. Fuel Center

500X

HC24289

b. Outer Periphery of Fuel

FIGURE A-8. SELECTED PHOTOMICROGRAPHS FROM 10-MIL SODIUM-BONDED SPECIMEN B-2 (CAPSULE BMI-50-6)

Note the well-developed fission-gas bubbles depicted in Figure A-8b.

150X

HC24334

a. Low-Magnification View

750X

HC24337

b. High-Magnification View

FIGURE A-9. SELECTED PHOTOMICROGRAPHS DEPICTING MICROSTRUCTURE OF TYPE 304 STAINLESS STEEL CLADDING ON 10-MIL SODIUM-BONDED SPECIMEN B-1 FROM CAPSULE BMI-50-6

Capsule BMI-50-11. All of the specimens in this capsule exhibited some measure of fission-gas-bubble formation and consequent swelling of the fuel above that associated with solid-fission-product-induced growth. Specific swelling rates for the fuel from this capsule are shown plotted in Figure A-10 along with a curve showing the maximum swelling rate for previously irradiated low-oxygen mixed-nitride fuel. The data indicate that swelling associated with fission-gas-bubble formation and growth begins at a temperature slightly below 1400 C.

FIGURE A-10. TEMPERATURE DEPENDENCE OF (U, Pu)N SWELLING

Metallographic examination of Capsule BMI-50-11 consisted of the examination of four samples; one sample from Specimen A-44, two samples (one longitudinal and one transverse) from Specimen B-54, and one sample from Specimen C-34. Although Specimen A-44 experienced a very high heat generation rate (44.5 kw per ft), the cladding remained sound except for a small pinhole-type leak capable of passing released fission gas. The sample cross section is shown in Figure A-11. The fuel microstructure located toward the cladding in Specimen A-44 did not change to any great extent (refer to Figures A-12a and A-12b, depicting unirradiated and irradiated UN-PuN fuel, respectively). This result is not surprising, as maximum fuel surface temperatures for Specimen A-44 were estimated at 970 C, too low for fission-gas bubble growth to occur. However, toward the center of the fuel, estimated temperatures reached 1455 C, a sufficiently high temperature for bubble growth to occur (Figure A-12c).

Two metallographic samples, one transverse and one longitudinal, were prepared from Specimen B-54 to aid in discovering mechanisms active in causing the circumferential crack found in this specimen (Figures A-13 through A-15). The transverse metallographic sample (Figure A-13), cut from the lower fuel pellet in Specimen B-54, depicts the partial formation of a center void and a major

FIGURE A-11. LOW-MAGNIFICATION COMPOSITE OF METALLOGRAPHIC SAMPLE FROM HELIUM-BONDED SPECIMEN A-44 FROM CAPSULE BMI-50-11

An average heat generation rate of 44.5 kw per ft was achieved. Note porosity in the fuel center. The maximum fuel temperature was 1455 C.

500X

PL5535

500X

HC24297

a. Unirradiated Fuel

b. Irradiated Fuel Near Specimen OD

500X

HC24285

c. Irradiated Fuel Near Specimen Center

FIGURE A-12. SELECTED PHOTOMICROGRAPHS OF SAMPLES FROM HELIUM-BONDED SPECIMEN A-44 FROM CAPSULE BMI-50-11

Note the fission-gas bubbles that formed near the fuel centerline (Figure A-12c), and the similarity between the unirradiated fuel and the irradiated fuel near the specimen OD.

TEO1927

FIGURE A-13. LOW-MAGNIFICATION COMPOSITE OF TRANSVERSE METALLOGRAPHIC SAMPLE FROM 10-MIL SODIUM-BONDED SPECIMEN B-54 FROM CAPSULE BMI-50-11

An average heat generation rate of 46.5 kw per ft was achieved. Note the porosity in the fuel center. The maximum full temperature was 1520 C.

500X

HC24300

a. Fuel-Cladding Interaction

500X

HC24318

b. Metallic-Appearing Phase in Fuel Grain Boundaries and Voids

500X

HC24303

c. Another View of the Second-Phase Material Found in the Fuel Grain Boundaries and Voids

FIGURE A-14. PHOTOMICROGRAPHS OF SELECTED AREAS OF 10-MIL SODIUM-BONDED SPECIMEN B-54 FROM CAPSULE BMI-50-11

FIGURE A-15. LOW-MAGNIFICATION COMPOSITE OF LONGITUDINAL METALLOGRAPHIC SAMPLE FROM 10-MIL SODIUM-BONDED SPECIMEN B-54 FROM CAPSULE BMI-50-11

Note the location of the circumferential crack in the cladding relative to the fuel pellet-pellet interface.

longitudinal crack that passes through the centerline of the pellet. The estimated maximum fuel center temperature of 1520 C for Specimen B-54 was sufficient for fission-gas bubble formation, which is depicted in higher-magnification photomicrographs (Figure A-14). Second-phase material, detected in grain boundaries and voids (Figures A-14b and A-14c) has the characteristic appearance of a metallic phase. Identification of this metallic second-phase material is uncertain, as it may consist of metallic fission products or it may have formed as a result of fuel-cladding interaction; the cladding reached temperatures of about 900 C. Positive identification will be sought.

One specimen in Capsule BMI-50-11 had a visibly failed cladding (10-mil sodium-bonded Specimen B-54). This failure was oriented in a circumferential direction and did not appear to be caused by the hoop stresses generally associated with swelling fuel. The crack, as depicted in the metallographic sample shown in Figure A-15, is evidence for excessive cladding straining in the longitudinal direction. Insufficient clearance between the fuel column and upper hold-down device to accommodate the longitudinal fuel growth or simple frictional bonding to prevent accommodation of longitudinal growth by axial pellet motion could be responsible for this strain. The closeness of the cladding crack to the gap between fuel pellets suggests that a discontinuity in the pressure exerted on the cladding by the fuel at this point played a role in the nucleation of the circumferential crack.

The specimen in Capsule BMI-50-11 undergoing the greatest volumetric swelling, Specimen C-43, was prepared metallographically for evaluation (Figure A-16). The increased swelling coefficient clearly resulted from the temperature exceeding the temperature at which fission-gas-bubble formation and growth could occur. Still higher temperatures than those estimated may have resulted in this particular specimen because of a loss of sodium bonding between the fuel and cladding. This is evidenced in the formation of columnar grains on one side of the fuel diameter, from which it is estimated that temperatures at least as high as 1600 to 1700 C were reached. Several stages of fission-gas-bubble development in Specimen C-43 are depicted in Figures A-17a and A-17b. An example of fuel-cladding reaction is shown in Figure A-17c. The fuel-cladding reaction is associated with excessive cladding temperatures (which reached 1000 C in this specimen). An electron-beam microprobe analysis is scheduled to establish reaction products in the fuel-cladding reaction zone.

Microstructures of the cladding of several of the specimens with higher heat generation rates, Specimens B-54 and C-43, exhibited unusual structures, i.e., fields of subgrain boundaries (Figure A-18a) and a Widmannstaetten-type structure (Figure A-19a). Such structures are typically obtained by annealing at about 1000 C. Negative results with Murkams' etch suggested that the carbide content of the Widmannstaetten structure was very low and, therefore, that it had a high nitrogen content. Evidence of fuel-cladding reaction was exhibited, as seen in Figures A-18b and A-19b. Electron-beam microprobe analysis of cladding and fuel interfaces should aid in determining if either cladding constituents or fuel and fission products have diffused out of or into the cladding.

Plans for Future Work

Remaining tasks to be completed in the postirradiation examination of Capsules BMI-50-6 and BMI-50-11 include:

FIGURE A-16. LOW-MAGNIFICATION COMPOSITE OF METALLOGRAPHIC SAMPLE FROM 20-MIL SODIUM-BONDED SPECIMEN C-43 FROM CAPSULE BMI-50-11

Fuel temperatures of 1600 to 1700 C are suggested by apparent vapor-transport migration of gas bubbles. Loss of sodium bonding could account for the apparently high temperature on one side of the fuel. The maximum fuel temperature was 1665 C.

500X

HC24316

a. Fission-Gas Bubbles at Fuel OD

500X

HC24295

b. Fission-Gas Bubbles at Fuel Center

500X

HC24296

c. Fuel-Cladding Reaction

FIGURE A-17. PHOTOMICROGRAPHS OF SELECTED AREAS OF SPECIMEN C-43 FROM CAPSULE BMI-50-11

500X

Etched

HC24344

a. Subgrain Boundaries

500X

Etched

HC24346

b. Fuel-Cladding Reaction

FIGURE A-18. MICROSTRUCTURE OF TYPE 304 STAINLESS STEEL CLADDING OF
20-MIL SODIUM-BONDED SPECIMEN C-43 FROM CAPSULE
BMI-50-11

500X

Etched

HC24367

a. Widmannstaetten-Type Structure

150X

Etched

HC24364

b. Fuel-Cladding Reaction

FIGURE A-19. MICROSTRUCTURE OF TYPE 304 STAINLESS STEEL CLADDING OF 10-MIL SODIUM-BONDED SPECIMEN B-54 FROM CAPSULE BMI-50-11

- (1) Burnup analysis
- (2) Retained gas analysis
- (3) Electron-beam microprobe analysis of fuel and cladding.

When Capsules BMI-50-8 and BMI-50-10 arrive at the Battelle-Columbus Hot-Cell Facility, a postirradiation examination will be made similar to that conducted on Capsules BMI-50-6 and BMI-50-11.

Quality Assurance for EBR-II Pin Fabrication

D. E. Lozier

Objective of Program

To establish a coordinated quality assurance program for the fabrication of fuel pins for the 19-pin subassembly to be inserted in the EBR-II.

Approach Being Used

The responsibility for procurement, fabrication, assembly, and evaluation of component parts and the completed fuel pins for the 19-pin subassembly to be inserted in the EBR-II is assigned to specific task engineers. Each engineer has the responsibility of initiating a document of procedures, material, and fabrication specifications for his task commensurate with the established EBR-II fuel-pin specifications. These component documents will then be assembled into a program procedural and specification manual by the quality assurance task leader and reviewed by the program managers prior to initiation of fuel-pin fabrication.

The established quality assurance document will be distributed to the task engineers for their compliance. Appropriate check sheets verifying compliance of each specification or procedure will be generated in addition to the recording of all data in a laboratory record book. The check sheets will be assembled into a quality assurance verification record for each fuel pin.

In addition to compiling and reviewing procedural check lists and recorded data to assure specification compliance, the quality assurance task leader will act as an observer on critical operations in the fabrication of the fuel pins.

Progress Reported Previously

This is the initial report on this program.

Work This Quarter

Specifications on Type 316 stainless steel tubing for cladding material for procurement purposes; fabrication procedures and specifications for the (U, Pu)N pellets and UN end pellets; and procedures for sodium bonding, sodium bond evaluation, and encapsulation of clad fuel pins have been received from the task engineers. Preliminary editing has been done on the task documents received.

Plans for Future Work

The procedural and specification quality assurance document will be compiled, edited, and reviewed by the program managers subsequent to receipt of the remaining task procedural and specification sections from the responsible engineers.

Preparation of EBR-II Fuel Pins

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and O. L. Kruger

Objective of Program

To fabricate solid-solution ($U_{0.8}Pu_{0.2}$)N pellets by powder metallurgical techniques and clad them in well characterized Type 316 stainless steel for irradiation in the EBR-II.

Approach Being Used

Sintering techniques are being employed to fabricate ($U_{0.8}Pu_{0.2}$)N pellets of controlled density, size, and microstructure for EBR-II fuel-pin irradiation experiments. Nominal ($U_{0.8}Pu_{0.2}$)N powder containing fully enriched uranium will be synthesized by reaction of prealloyed metal with nitrogen gas. This powder will be ball milled to reduce the particle size, cold pressed in hardened steel dies, sintered, and centerless ground to produce fuel pellets that are 95.0 ± 3.0 percent of theoretical density and within $+0.0000$ and -0.0015 in. of the required diameters of 0.260 and 0.250 in. The fuel pellets will be right circular cylinders having a length-to-diameter ratio of approximately one. Control pellets from each powder batch will be characterized with respect to microstructure, nitrogen content, and uranium/plutonium ratio. Two additional pellets from each powder batch will be retained as historical samples.

Depleted UN pellets will be prepared for end spacers in the EBR-II pins by techniques similar to those employed for the mixed nitride.

Fuel pins consisting of (U, Pu)N pellets either helium or sodium bonded to tubular cladding will be assembled in an atmosphere of high-purity helium and sealed by TIG welding. Type 316 stainless steel components which have been well characterized and thoroughly inspected for defects and dimensional tolerances will be utilized. After plutonium decontamination, each sealed fuel pin will be helium leak checked, visually examined, and radiographed to verify the position of the fuel pellets and the overall quality of the welds. Sodium-bonded fuel pins are heated to a temperature in excess of the melting point of sodium and centrifuged; an eddy-current inspection is performed to assure a continuous, void-free sodium bond.

The procurement of components (tubing, end-plug bar stock, springs, etc.) for EBR-II fuel-pin assembly and inspection will be performed in a manner consistent with FFTF and RDT established procedures. Loading and inspection of the EBR-II fuel pins will be carried out in a prescribed manner. All procedures and inspections will be well documented for quality assurance purposes.

Specifically, solid-solution ($U_{0.8}Pu_{0.2}$)N pellets for the sodium-bonded fuel pins of the BMI-5 irradiation series will be prepared and characterized. The BMI-5 series constitutes a 19-pin subassembly; however, because of the high quality assurance requirements and to provide for replacement of fuel pins removed for interim examination during the course of the irradiation, a 25 percent overrun is scheduled so that 25 fuel pins will be fabricated for the 19-pin subassembly. Each pin will contain 13.5 in. of fuel. Two configurations will be employed, one involving a 10-mil sodium gap and the other, a 15-mil sodium gap; hence, pellets of two different diameters will be fabricated, 0.260 and 0.250 in.

Progress Reported Previously

Fuel pellets for the seven pins comprising the BMI-3 series were prepared during Fiscal Year 1970. The pellets were fabricated by cold pressing and sintering nominal $(U_{0.8}Pu_{0.2})N$ powders prepared by a hydride-nitride cycle on arc-cast ^{235}U -20 w/o plutonium buttons. The seven pins, each composed of a 10-in. stack of $(U, Pu)N$ pellets within a tube of Type 316 stainless steel cladding, were loaded in an atmosphere of high-purity helium. The bonding medium utilized between fuel and cladding was either helium or sodium. The end plugs were welded in. Each of the pins was tested in accordance with the program requirements and found acceptable for the eddy-current testing preliminary to secondary encapsulation.

Work This Quarter

Pellet Fabrication for 19-Pin Subassembly. Calculations, using shrinkage data obtained last fiscal year, to determine a range of die sizes in an attempt to fabricate fuel pellets to size (0.260 in. in diameter, $+0.0000$ and -0.0015 in.) were completed. Hardened steel dies with diameters of 0.324, 0.326, 0.328, and 0.330 in. have been fabricated. Additional plutonium has been ordered and has arrived.

Two batches of depleted UN powder were synthesized by the direct reaction of prepurified nitrogen with depleted uranium metal. U_2N_3 powder was formed by the reaction at 850 C and decomposed to UN at 1400 C in a vacuum of 1×10^{-5} torr. As-synthesized UN powder of less than $177\text{-}\mu$ diameter was reduced in size to $<2\text{-}\mu$ particles by ball milling in a uranium-lined mill with tungsten carbide-cobalt balls and nanograde hexane for 48 hr. The milled UN powder was dried in vacuum and hydrostatically pressed at 45 tsi into rods. The rods were sintered for 4 hr at 2300 C in a nitrogen atmosphere to approximately 95 percent of theoretical density. Pellets were cut and ground from the sintered rod to the specified dimensions of 0.250 ± 0.0015 in. in diameter by 0.250 ± 0.002 in. long. Cleaning of the pellets was performed in successive ultrasonic washings with acetone and 200-proof ethyl alcohol followed by outgassing in a vacuum of 10^{-5} torr at 1000 C.

All depleted UN end pellets have been characterized as to dimensions, weight, and density. Metallographic examinations were performed on control specimens from each of two sintering batches of pellets. Control specimens from both sintering batches have also been submitted for analyses of nitrogen, oxygen, and carbon.

Quality Assurance of Component Procurement, Loading, and Inspection. The quality assurance package for procurement of Type 316 stainless steel cladding was prepared. Seamless tubing was ordered from Carpenter Technology according to the following specifications:

- (1) Material: Type 316 stainless steel, 20 percent cold worked
- (2) Outside Diameter: 0.310 ± 0.001 in.
- (3) Inside Diameter: 0.280 ± 0.001 in.
- (4) Length: 300 ft in 10-ft pieces
- (5) Manufacturing and Characterization: In full accordance with FFTF specifications (A-0086, Rev. 1, October 1968) as updated, and to be issued as AEC Quality Assurance Report RDT 13-8.

Delivery of this tubing to Battelle-Columbus after interim inspection at WADCO is expected late in December, 1970.

The loading procedures for mixed-nitride fuel have been reviewed. Certain procedures that are inadequate for quality assurance purposes with bare fuel pins

have been identified and are being modified. These include welding, radiography, and helium leak detection. An additional procedure, not utilized previously, involving xenon tagging of each fuel pin has been reviewed and a selection of one of the two or three gas systems designed in other laboratories will be made soon.

BMI-3 Series Pins. A low sodium level was detected in three of the BMI-3 series pins when they were eddy-current inspected; it is assumed that the sodium was absorbed into the open porosity of the low-density pellets. These fuel pins had been loaded in an atmosphere of high-purity helium and sealed by TIG welding. All three pins are sodium bonded, and contain (U, Pu)N fuel of density in the range from 87.1 to 94.5 percent of theoretical. After removal from the welding chamber, each pin had been decontaminated and then leak checked and radiographed to establish the integrity of the weld. Since eddy-current inspection has shown a low sodium level in these three pins, they will be opened, loaded with additional sodium, and welded.

Plans for Future Work

Initial shrinkage studies will be performed during the first part of November to determine the die size to use in fabricating the 0.260-in. -diameter fuel pellets for the 19-pin subassembly. Materials synthesis and pellet production will be initiated following generation of these data.

The analytical result will be evaluated for the depleted UN end pellets. If the nitrogen, oxygen, and carbon contents are found to be satisfactory, the pellets will be stored in an inert atmosphere until needed for the individual fuel pins.

The quality assurance packages for procurement of end-plug bar stock and for loading and inspection procedures for the EBR-II fuel pins will be prepared for review. Upgrading of equipment and procedures will continue.

Work will be completed on the BMI-3 series pins so that they will be ready for eddy-current testing and subsequent secondary encapsulation.

EBR-II Irradiation-Capsule Assembly and Safety Analysis Preparation

E. O. Fromm, N. E. Miller, and J. W. Ray

Objective of Program

To design and fabricate irradiation capsules, encapsulate clad (U, Pu)N fuel pins, perform required nondestructive examinations, prepare safety analyses for the irradiations, and ship the completed capsules to the EBR-II.

Approach Being Used

Standard EBR-II irradiation capsules are currently being used for this program. The capsule components and some cladding material which are of Type 304L stainless steel will be procured from ANL. Additional cladding material consisting of Type 316L stainless steel will be procured commercially. The fuel-pin cladding will be sodium bonded to the outer capsule by centrifuging at 1000 F. Sodium levels and bond soundness will be verified by X-radiography and eddy-current testing.

Progress Reported Previously

The first set of four irradiation capsules, designated as the BMI-1 series, was prepared and delivered to the EBR-II for irradiation, and the documents necessary for approval of the experiments for insertion in the reactor were prepared and forwarded to ANL for review, as described in BMI-1868. The first capsule in this series, designated Capsule BMI-1-3 (a helium-bonded fuel pin), was inserted in the EBR-II in Subassembly X-070 for Run 39B. At the end of Run 42 this capsule had accrued a total burnup of 2.4 a/o, as reported in BMI-1886.

A second series of experiments consisting of seven encapsulated fuel pins was completed and shipped to the EBR-II for irradiation.

Work was initiated on the third set of encapsulated EBR-II irradiation experiments designated as the C-BMI-3 series. Three capsules, all containing helium-bonded fuel pins, were completed and the "Quality Assurance Document" associated with them was prepared. In addition, the "Hazards Analysis" for all of the capsules in Series 3 was prepared, as described in BMI-1886.

Work This Quarter

Capsule BMI-1-3 containing the helium-bonded fuel pin from BMI Series 1 was discharged from the EBR-II at the end of Run 44 after achieving a fuel burnup of about 3.8 a/o. The maximum specific fuel-pin power was estimated to be 25.8 kw per ft. The capsule was neutron radiographed at TREAT prior to transfer to the Test Area North Hot Laboratory for loading into the BCL-4 cask for shipment to the Battelle-Columbus Hot-Cell Facility. The postirradiation neutron radiograph of Capsule BMI-1-3 is shown in Figure A-20. For comparison, the preirradiation X-ray is also shown in the figure.

On August 7, 1970, Subassembly X-086 was loaded into Position 4D3 of the EBR-II grid for Run 45 (50 Mw). The subassembly contained ten mixed-nitride capsules, consisting of three capsules in the BMI-1 series and the seven capsules in the BMI-2 series. Upon completion of Run 45 (September 10, 1970), the subassembly was moved to Position 5D4 for Run 46, the first reactor run scheduled for operation at 62.5 Mw. A summary of the estimated specific fuel-pin power levels for the various capsules for the two subassembly locations in the reactor for Runs 45 and 46 is given in Table A-6.

Eight EBR-II capsules containing UN-PuN fuel pins were originally included in the Series 3 irradiation experiment. However, because of fuel fabrication difficulties one capsule was eliminated, so that there are now seven capsules in this series.

Assembly of the last four capsules in Series 3 was initiated. Capsule C-BMI-3-4 was sodium bonded in the centrifuge bonding apparatus; the eddy-current test showed the sodium bond to be sound.

The "Approval in Principle" for the Series 5 irradiation experiments was prepared and forwarded to the AEC for review. Essentially, the irradiation experiments comprising Series 5 consist of 19 individual fuel pins assembled into an N-type experimental subassembly. Each fuel pin consists of a 13.5-in. length of UN-PuN fuel sodium bonded to 0.310-in. -OD cladding of Type 316 stainless steel with a 0.015-in. -thick wall. Thirteen of the fuel pins will have a 0.010-in. radial sodium bond and six will have a 0.015-in. radial sodium bond. The UN-PuN fuel in pellet form will be immersed in sodium and loaded under a helium atmosphere. A small measured amount of xenon tag gas will be included in each fuel pin, as

FIGURE A-20. CAPSULE BMI-1-3 BEFORE AND AFTER IRRADIATION

TABLE A-6. SPECIFIC POWER LEVELS OF BATTELLE-COLUMBUS MIXED-NITRIDE CAPSULES
IN EBR-II SUBASSEMBLY X-086 FOR RUNS 45 AND 46

Capsule Identification	Location of Capsule in Subassembly	Specific Fuel-Pin Power Rating for Capsule in Indicated Grid	
		Position, kw per ft	
		Position 4D3, Run 45 (50Mw)	Position 5D4, Run 46 (62.5 Mw)
BMI-1-1	13	25.3	27.9
BMI-1-2	2	24.7	27.1
BMI-1-4	17	26.3	28.6
BMI-2-1	14	30.6	32.7
BMI-2-2	10	29.7	32.5
BMI-2-3	6	29.4	32.4
BMI-2-5	3	29.0	32.4
BMI-2-6	18	33.9	36.6
BMI-2-7	15	33.9	36.7
BMI-2-8	11	33.5	36.5

required by the EBR-II Project. Sodium bonding will be accomplished by centrifuging at 1000 F, and verification of the sodium bond will be made by eddy-current testing.

Plans for Future Work

The assembly and testing of the last four Series 3 capsules will continue. The quality assurance documents associated with these experiments will be prepared. The documents for all capsules including the "Hazards Analysis" will be forwarded to ANL for review.

The irradiation surveillance and performance analysis of the ten capsules being irradiated in Subassembly X-086 will continue.

Preparation of the "Hazards Analysis" for the 19-pin subassembly designated Series 5 will be initiated. The fabrication of two new dummy capsules for determination of the eddy-current equipment sensitivity will be initiated. The dummies will simulate the actual fuel pellets, sodium, and cladding of two types of clad assemblies comprising the Series 5 irradiation experiments.

UN-PuN TREAT Experiments

R. Artigas, N. E. Miller, and P. Cybulskis

Objective of Program

To evaluate the performance of (U, Pu)N fuels in transient irradiation exposure in the Transient Reactor Test Facility (TREAT).

Approach Being Used

Analytical studies are being conducted to define the experimental parameters and to establish the basic capsule design requirements for the exposure of (U, Pu)N fuels to transient irradiations in the TREAT facility. These tests will help to

establish the acceptability of mixed-nitride fuels for LMFBR applications under operational transient conditions.

Progress Reported Previously

Transient testing programs for oxide and carbide fuels performed in the TREAT facility were reviewed. Considering differences in physical properties, the results of these tests were used to predict the relative behavior of the nitride to the oxide and carbide fuels under transient conditions. Analytical comparisons were made between the thermal response of nitride fuels during TREAT transients and the response of a nitride-fueled LMFBR to an accident reactivity excursion. It was concluded that TREAT can provide an adequate simulation of the thermal transients that may be expected under LMFBR overpower or accident conditions.

The midplane temperature behavior of the fuel, sodium bond, and cladding resulting from flat-top transients corresponding to 40, 50, 60, and 75 kw per ft was investigated using the ARGUS code and utilizing a conceptual capsule configuration and assuming a radial power profile in the fuel similar to that in previous TREAT experiments. These studies have shown that only moderately high temperatures occur in the fuel until the occurrence of vaporization of the sodium bond; continued energy release after bond vaporization results in essentially adiabatic heating with corresponding rapid rises in fuel temperatures.

A one-dimensional, multigroup, integral transport code (THERMOS) and a two-dimensional diffusion code (2DB) were used to calculate a figure of merit for the conceptual capsule in the TREAT facility. These calculations showed that 1.8×10^{-4} w per cm^3 would be generated in the fuel specimen per watt of reactor power.

Work This Quarter

Work has progressed toward a definition of the reactor transient conditions for the first test and completion of the irradiation capsule design. The fuel for the test is 93 to 95 percent dense ($\text{U}_{0.8}\text{Pu}_{0.2}\text{N}$) with natural uranium. It is planned to use two individually clad specimens, each of which comprises a 2.5-in. stack of 0.225-in. -OD unirradiated fuel pellets sodium bonded to a 0.290 in. -OD Type 304L stainless steel cladding with a 0.020-in. wall. The volume of the gas plenum in each cladding is equal to 25 percent of the fuel volume. Each cladding will be prepressurized to produce a pressure of nominally 8 atm at the pretransient temperature (200 C) which will simulate the pressure of cover gas, fission gas, and sodium vapor in a typical mixed-nitride fuel pin operating in a LMFBR.

The two fuel pins are positioned one above the other along the axis of a 1.0-in. -OD shell of niobium-1 w/o zirconium which serves as the primary containment and pressure vessel. Within the pressure vessel, each specimen is surrounded by a metal sleeve which acts as a heat sink. The lower sleeve is made of beryllium and the upper sleeve is niobium-1 w/o zirconium. The material with the higher cross section is used for the upper sleeve to attenuate the neutron flux in the top specimen, leading to different transient exposures for the two specimens. The annular spaces between the claddings, heat-sink sleeves, and pressure vessel are filled with sodium. Metal-sheathed electric heaters are wrapped on the outer surface of the sleeves for melting the sodium and preheating the specimens to an initial temperature of about 200 C. Small-diameter (0.5 mm) metal-sheathed thermocouples at the specimen claddings and the heat-sink sleeves will be used to

monitor the thermal behavior of the specimens during the transient exposures. The niobium-1 w/o zirconium pressure vessel is surrounded by a massive aluminum outer shell which serves as the secondary containment for the fuel and as the final heat sink for the experiment.

The initial transient tests imposed on the mixed-nitride fuel are selected to simulate a 25 to 50 percent overpower reactor condition which could exist for a period of seconds as opposed to extremely high power bursts of short duration representative of hypothetical accident conditions. For the overpower simulation, a clipped-burst or flat-topped transient operation of TREAT will be utilized. During this exposure general melting of the fuel is not anticipated, nor is it desired on the basis of gaining the most information from the tests. The performance of the sodium bond and its effect on the performance of the fuel is of great interest in these tests. It is desirable to prolong the duration of the transient to the point where sodium boiling and vapor blanketing occurs in some regions of the bond annulus. The large ratio of the sodium-bond-annulus volume to gas-plenum volume (about 1 to 1) will cause a significant change to occur in the internal cladding pressure as liquid sodium is expelled from the annulus. This increase in pressure will tend to suppress further boiling so only localized boiling and localized high fuel temperatures are anticipated. The level and duration of the final TREAT transient commensurate with a simulated overpower condition are being selected on the basis of neutronic and heat-transfer calculations. This selection will be modified as deemed necessary from the results of preliminary lower-level transients. The use of two specimens operating at different fission rates increases the probability of achieving the desired exposure.

Plans for Future Work

The conduct of the TREAT irradiation and the posttest analysis are planned to be completed prior to June 30, 1971. During the next report period, the final neutronic and heat-transfer calculations will be completed pending definition of all capsule dimensions. Preparation of the TREAT hazards analysis and operation documents will be initiated, and assembly of the irradiation capsule will begin.

TECHNOLOGY OF NUCLEAR FUELS FOR ADVANCED FAST REACTORS (AEC-DRDT)

Program Management

W. Chubb

The objective of this program is to obtain information on the basic causes of changes in the properties and dimensions of the most promising fuel candidates for improved or advanced types of fast reactors as well as for current concepts. Data obtained in this program will provide a unique description of the in-pile behavior of those portions of LMFBR fuels which operate at temperatures from 1300 to 2000 C. Such data will allow determination of the relative contributions of those parts of the fuel operating above 1300 C, of those parts operating below 1300 C, and of the cladding to the overall fuel-element swelling behavior. The data will enhance assessment of the performance of present LMFBR fuel concepts under all credible operating conditions and will tend to validate subsequent use of current fuels at more efficient temperature levels. A model describing the mechanical interactions of the various parts of the fuel element will be developed to allow extrapolation of the experimental data to all temperatures of interest to LMFBR's.

Included in the program is (1) an irradiation study in which the dimensions and other physical properties of oxide, and, to a lesser extent, nitride and carbide, fuels are being determined as a function of burnup and irradiation temperature, (2) a study focused on the mechanisms of swelling in nuclear fuels in which theoretical models describing the generation and movement of gaseous fission products, including solubility and chemical diffusivity factors, bubble formation, and overall swelling processes, are being formulated and evaluated by comparison with the behavior and properties of actual fuels, (3) a fuel materials development effort in which fuels suited to the assessment of various technological concepts under study in the program are prepared and exhaustively characterized, (4) a materials properties study in which chemical and mechanical properties that might influence the behavior under irradiation of refractory fuels are investigated, and (5) a capsule development effort in which capsules tailored to the special requirements of the irradiation study are designed and prepared for use in the BRR and the MTR or ETR.

Irradiation Behavior of Oxide Fuels at High Temperatures

R. F. Hilbert, W. J. Zielenbach, D. E. Lozier,
R. B. Clark, and V. W. Storhok

Objective of Program

To investigate the irradiation behavior of oxide fuels at surface temperatures from 1200 to 1900 C.

Approach Being Used

Several UO_2 specimens are being irradiated in order to study swelling, fission-gas release, and restructuring of UO_2 at surface temperatures from 1200 to 1900 C. The effects of density, axial centerline holes, venting of fission gases,

stoichiometry, and the addition of PuO_2 are also being studied. One specimen in each capsule starting with Capsule HT-BRR-9 will be monitored for the pressure of gas collecting inside its cladding during the irradiation to determine the extent and timing of gas release. Fission gas released from the fuel to the interior of the cladding will be conducted to one side of a metal diaphragm located in the cool zone of the capsule. An externally controlled and measured pressure source will act on the opposite side of the diaphragm to balance the fission-gas pressure. The balance of pressures will be determined by sensing the deflection of the diaphragm with an electrical contact.

Four specimens of UO_2 and two specimens of $\text{UO}_2\text{-PuO}_2$ were irradiated in Capsule HT-BRR-11 to determine the effects of stoichiometry, as-fabricated porosity, and the addition of PuO_2 on the swelling and gas-release behavior of UO_2 at 1700 C. In addition, a portion of the experiment was designed to determine the restraint to swelling of these fuels at 1700 C by a cladding with a strength approximating that of stainless steel at 600 C. The fuels irradiated in Capsule HT-BRR-11 are given in Table B-1.

TABLE B-1. DESCRIPTION OF THE SPECIMENS IRRADIATED
IN CAPSULE HT-BRR-11

Fuel	Geometry	Nominal Density, percent of theoretical	Cladding Thickness, in.
$\text{UO}_2\text{-x}$	110-mil axial hole	96	0.020 ^(a)
UO_2	55-mil axial hole	80	0.020
$\text{UO}_2\text{-8 w/o PuO}_2$	110-mil axial hole	96	0.040
UO_2	110-mil axial hole	96	0.040
$\text{UO}_2\text{+x}$	110-mil axial hole	96	0.020
$\text{UO}_2\text{-8 w/o PuO}_2$	110-mil axial hole	96	0.020

(a) Gas pressure monitored.

Three specimens of UO_2 and three specimens of $\text{UO}_2\text{-PuO}_2$ were irradiated in Capsule HT-BRR-12 to study the temperature dependency of restructuring, columnar grain growth, and gas-bubble formation. The capsule was neutron radiographed at selected times early in the irradiation and at the end of the cycle (about 500 hr) in an attempt to determine whether restructuring occurred (as evidenced by center void formation). The duration of the irradiation was based primarily on the timing and the nature of changes seen on the neutron radiographs. The irradiation was discontinued after two cycles of irradiation to minimize the chances of the structure being altered by inadvertent overtemperature conditions. The fuels irradiated in Capsule HT-BRR-12 are described in Table B-2.

TABLE B-2. DESCRIPTION OF THE SPECIMENS IRRADIATED
IN CAPSULE HT-BRR-12

Fuel	Design Cladding Temperature, C	Description
UO ₂	1500	90 percent dense, gas-pressure monitor
UO ₂ -8 w/o PuO ₂	1700	90 percent dense
UO ₂	1700	90 percent dense, 110-mil axial hole
UO ₂ -8 w/o PuO ₂	1500	90 percent dense
UO ₂	1300	90 percent dense
UO ₂ -8 w/o PuO ₂	1300	90 percent dense

Capsule HT-BRR-14 will contain oxide specimens and will be primarily aimed at determining the mechanism of gas release from and swelling of UO₂. The capsule will contain the specimens shown in Table B-3.

TABLE B-3. DESCRIPTION OF THE SPECIMENS TO BE IRRADIATED
IN CAPSULE HT-BRR-14

Fuel	Nominal Density, percent of theoretical	Centerline Hole Diameter, in.	Design Cladding Temperature, C	Description
UO ₂	96	0.110	1300	Gas monitor
UO ₂	95	0.055	1450 ± 50	Annular tungsten foil at fuel midradius
UO ₂	95	0.055	1450 ± 50	Annular region of de- pleted UO ₂ on fuel OD at fuel axial midplane
UO ₂	100	0.110	1700	--
UO ₂	100	0.110	1700	Vented cladding
UO ₂ -8 w/o PuO ₂	90	None	1500	--

The fully dense UO₂ specimens are designed to study lenticular pore movement and columnar grain growth as a function of temperature and in the absence of as-fabricated porosity. The specimen with the annular tungsten foil at the mid-radius and the specimen with the depleted UO₂ annulus are designed to interrupt the sweeping action of lenticular voids. Hopefully, interrupting the flow of lenticular voids will permit the formation of observable fission-gas bubbles in the columnar grains. The UO₂ specimen with the gas monitor is designed to determine the timing and extent of fission-gas release at temperatures where lenticular void migration by vapor transport of UO₂ is slow or nonexistent.

Four specimens of UO_2 and two specimens of UO_2 -20 w/o PuO_2 will be irradiated in Capsule HT-BRR-16 with the following experimental objectives:

- (1) To determine the stress-strain relationships of fuel and cladding at high temperatures by varying the strength through control of cladding thickness and use of different cladding materials (niobium-1 w/o zirconium, tungsten-26 w/o rhenium)
- (2) To compare swelling and gas release of UO_2 with that of UO_2 - PuO_2 at a cladding temperature of about 1700 C
- (3) To test the theory of continuing void generation and movement by vaporization-condensation throughout an irradiation
- (4) To study redistribution of UO_2 - PuO_2 at high temperatures as a function of oxygen/metal ratio
- (5) To reproduce behavior previously observed.

Specifically, the specimens listed in Table B-4 will be irradiated.

TABLE B-4. DESCRIPTION OF THE SPECIMENS TO BE IRRADIATED
IN CAPSULE HT-BRR-16

Fuel	Cladding	Cladding Thickness, in.	O/M Ratio	Irradiation Temperature, C	Experimental Objectives
$\text{UO}_2^{(a)}$	Tungsten-26 w/o rhenium	0.020	2.00	1700-1200-1600 ^(b)	Test theory of continuing void generation and movement by vaporization-condensation
UO_2	Niobium-1 w/o zirconium	0.020	2.00	1450	Determine fuel-cladding stress-strain relationships
UO_2	Tungsten-26 w/o rhenium	0.040	2.00	1750	Determine fuel-cladding stress-strain relationships
UO_2	Tungsten-26 w/o rhenium	0.020	2.00	1750	Determine fuel-cladding stress-strain relationships; reproduce behavior previously observed
UO_2 -20 w/o PuO_2	Tungsten-26 w/o rhenium	0.020	1.98	1750	Compare swelling and gas release of UO_2 with that of UO_2 - PuO_2 ; study redistribution of UO_2 - PuO_2 as a function of O/M ratio
UO_2 -20 w/o PuO_2	Tungsten-26 w/o rhenium	0.020	1.92-1.94	1750	Ditto

(a) Gas pressure to be monitored.

(b) Two cycles at 1700 C, then three to four cycles at 1200 C, then one cycle at 1600 C.

Most of the specimens in HT-BRR series capsules consist of a column of pellets or slugs of fuel 1 in. long contained in tungsten-26 w/o rhenium claddings approximately 1.44 in. long with an OD of 0.25 in. and a wall thickness of 20 mils, as shown in Figure B-1. Two specimens in Capsule HT-BRR-11 and one

FIGURE B-1. STANDARD CONFIGURATION OF SPECIMENS IRRADIATED IN THE BRR

specimen in Capsule HT-BRR-16 have a cladding thickness of 40 mils and an OD of 0.29 in.

Dimensional changes in the specimens are determined after each cycle of irradiation by measurements from neutron radiographs. The postirradiation examination includes (when appropriate) dimensional measurements, fission-gas-release measurements, and metallography.

Progress Reported Previously

Four specimens of bulk UO_2 were irradiated in Capsule HT-BRR-5 and the results were reported in BMI-1848 (August, 1968) and in the open literature⁽¹⁾. This irradiation provided the first indication of the rapid swelling behavior of clad bulk UO_2 at surface temperatures of 1450 to 1750 C. Unfortunately, decreasing temperatures throughout the irradiation prevented a quantitative analysis of the results.

Six specimens of UO_2 were irradiated in Capsule HT-BRR-9 to study swelling, fission-gas release, and restructuring of UO_2 as a function of surface temperature. Examination of Capsule HT-BRR-9 was completed in Fiscal Year 1970 and reported in BMI-1886 (July, 1970) and in several publications⁽²⁻⁴⁾.

On the basis of the swelling rates observed on a cycle-by-cycle basis (a cycle consists of 3 weeks of irradiation) for a number of specimens in this program, the external swelling of UO_2 was observed to be temperature-dependent. At cladding surface temperatures below about 1600 C, external swelling rates generally were less than 8 v/o per 10^{20} fissions per cm^3 . At about 1750 C external swelling rates were two to four times as great. This behavior shows that UO_2 can severely deform its cladding at these temperatures even though gas release is high.

A gas-pressure monitor indicated that a UO_2 specimen which operated at a cladding surface temperature of 1840 C released fission gas at nearly the same rate as it was generated. This high gas release was continuous after the first few hours of irradiation. Retained-gas measurements performed during the post-irradiation examination indicated that all the specimens that operated at cladding surface temperatures of 1475 C and above released about 90 percent of their fission gas. The UO_2 in these specimens formed columnar grains extending from the cladding to the center of the sample. At a cladding surface temperature of 1325 C, the UO_2 released about 75 percent of the gas produced during the irradiation and consisted of short columnar and enlarged equiaxed grains.

The above-mentioned specimen of UO_2 which operated at a cladding surface temperature of 1840 C with a gas-pressure monitor attached continued to swell even though the cladding failed after a burnup of 0.5×10^{20} fissions per cm^3 . This behavior demonstrates that the diametral increase of this specimen was not caused by an increase of the gas pressure contained within the cladding. The gas-pressure monitor registered 29 psig before failure and zero after the cladding rupture. A review of the irradiation history of all the specimens revealed no relation between the amount of swelling and the number of thermal cycles (to ambient temperature). Thus, thermal ratcheting can probably be discounted as a major contributor to the swelling. On the basis of these observations, it was concluded that the swelling of UO_2 at high temperatures is caused by the precipitation and growth of fission-gas bubbles within the UO_2 prior to the release of the gas at external surfaces of the UO_2 .

A theory to explain these observations postulated that the uranium dioxide swells by the formation of small, relatively immobile fission-gas bubbles. This swelling results in small, permanent external dimensional changes. However, periodically, portions of the uranium dioxide are swept by lenticular voids. This vapor-phase zone-refining process reconstitutes the uranium dioxide down the temperature gradient, maintaining the density of the oxide essentially constant. The vapor-phase transport and redensification of the oxide occur in such a way that external dimensional changes are not recovered. The large external dimensional changes occurring when this sequence is repeated many times represent the summation of a large number of repetitive steps involving small density decreases in the uranium dioxide followed by redensification by the zone-refining process. The zone-refining process is instrumental in collecting fission gas and in forming channels for gas release. Although probably not the sole mechanism of gas release, the process is a major mechanism of gas release from uranium dioxide at temperatures over 1600 C.

Work This Quarter

During the quarter, the postirradiation examination of Capsule HT-BRR-11 was continued, and four cycles of the irradiation of Capsule HT-BRR-14 were completed.

The irradiation conditions and dimensional changes of the specimens irradiated in Capsule HT-BRR-11 and Capsule HT-BRR-14 are given in Tables B-5 and B-6, respectively. Note that some of the actual preirradiation specimen densities given in Table B-6 are slightly different from the design densities given in Table B-3. External swelling rates as a function of burnup for specimens from Capsules HT-BRR-9, -11, -12, and -14, Figure B-2, show that the external dimensions of UO_2 and $\text{UO}_2\text{-PuO}_2$ clad with 20 or 40 mils of tungsten-rhenium alloy increase with increasing temperature from 1250 to 1900 C. (It should be noted that the data shown in Figure B-2 represent the external volumetric swelling rate of each specimen for the entire irradiation and that the temperatures represent an average of the temperatures of each reactor cycle.) Variables in the experiments included cladding thickness, cladding vents, centerline-hole diameter, fuel composition, fuel stoichiometry, and fuel density. The data given in Figure B-2, however, suggest that temperature is the most significant variable with respect to swelling of uranium-plutonium oxides at high temperatures. The swelling data given in Figure B-2 for Capsules HT-BRR-9, -11, and -12 were presented earlier, and can be separated into three loosely defined temperature regimes based on microstructural observations. Gas-release data obtained this quarter on specimens from Capsule HT-BRR-11 showed a gas release of 87 percent from UO_2 Specimen 359 at a cladding surface temperature of 1540 C and a gas release of 56 percent from $\text{UO}_2\text{-PuO}_2$ Specimen 5P at a cladding surface temperature of 1300 C. The swelling data from the specimens in Capsule HT-BRR-14 after only four cycles of irradiation are in good agreement with the swelling data obtained on the earlier capsules (Figure B-2). An evaluation of the effect of density, stoichiometry, and cladding thickness on the behavior of the specimens irradiated in Capsule HT-BRR-11 will be deferred until the microstructures are examined.

TABLE B-5. IRRADIATION CONDITIONS AND DIMENSIONAL CHANGES FOR SPECIMENS IN CAPSULE HT-BRR-11

Specimen	Description	BRR Cycle	Cladding Surface Temperature, C	Estimated Accumulated Burnup, 10^{20} fissions per cm^3	External Dimensional Changes ^(a) , percent			
					Maximum Cladding OD	Average Fuel OD	Fuel Length	Average Fuel Volume
358	UO _{2-x} : 96 percent dense, 0.110-in. axial hole, gas-pressure monitor	1	1680	0.21	0.7	0.8	2.7	4.3
		2	1410	0.45	-0.1	-0.1	2.0	1.8
		3	1430	0.73	0.9	1.1	1.7	3.9
		4	1420	0.97	0.3	0.4	3.3	4.1
		5	1380	1.19	0.5	0.6	2.7	3.9
		6	1380	1.41	0.4	0.5	1.5	2.5
		7	1380	1.64	1.8	1.2	1.8	4.2
		8	1370	1.86	2.8	1.6	2.0	5.2
			1400 avg		[0.8]	[0.5]		[3.0]
360	UO ₂ : 80 percent dense, 0.055-in. axial hole	1	1530	0.23	0.4	0.5	-1.7	-0.7
		2	1370	0.50	0.3	0.3	0.5	1.1
		3	1390	0.81	1.3	1.5	-0.1	2.9
		4	1380	1.08	0.8	1.0	0.0	1.9
		5	1400	1.33	0.6	0.7	0.3	1.1
		6	1400	1.58	0.4	0.5	-0.3	0.7
		7	1400	1.84	1.8	1.3	-0.3	2.9
		8	1390	2.09	1.9	1.8	0.4	4.0
			1390 avg		[1.2]	[0.8]		[2.0]
4P	UO ₂ -8 w/o PuO ₂ : 96 percent dense, 0.110-in. axial hole, 0.040-in. -thick cladding	1	1460	0.18	-0.1	-0.2	1.2	0.8
		2	1280	0.39	0.3	0.4	0.9	1.7
		3	1300	0.63	-0.3	-0.5	1.8	0.8
		4	1290	0.84	0.2	0.3	1.5	2.1
		5	1300	1.03	0.7	1.0	1.3	3.3
		6	1300	1.22	0.2	0.2	1.5	1.9
		7	1290	1.42	0.6	0.5	1.5	2.5
		8	1270	1.61	0.9	0.7	1.7	3.1
			1290 avg		[0.5]	[0.5]		[2.7]
359	UO ₂ : 96 percent dense, 0.110-in. axial hole, 0.040-in. -thick cladding	1	1695	0.23	0.3	0.4	0.9	1.8
		2	1500	0.51	0.3	0.4	2.3	3.2
		3	1530	0.84	0.2	0.2	1.4	2.2
		4	1530	1.12	0.6	0.8	1.9	3.5
		5	1550	1.38	0.7	0.9	1.2	3.0
		6	1550	1.64	0.6	0.8	0.6	2.2
		7	1550	1.91	2.3	2.2	0.7	5.1
		8	1550	2.17	3.1	2.6	1.0	6.2
			1540 avg		[3.4]	[3.0]		[6.0]
303	UO _{2+x} : 96 percent dense, 0.110-in. axial hole	1	1630	0.21	0.2	0.2	2.6	3.1
		2	1500	0.48	0.3	0.4	2.3	3.1
		3	1510	0.79	0.2	0.2	1.8	2.2
		4	1510	1.06	0.6	0.7	2.5	3.9
		5	1520	1.31	0.4	0.4	2.5	4.3
		6	1520	1.56	0.4	0.5	2.4	3.4
		7	1510	1.82	1.4	1.6	4.0	7.2
		8	1500	2.07	2.7	1.8	4.1	7.7
			1510 avg		[2.8]	[2.3]		[8.7]

TABLE B-5. (Continued)

Specimen	Description	BRR Cycle	Cladding Surface Temperature, C	Estimated Accumulated Burnup, 10^{20} fissions per cm^3	External Dimensional Changes ^(a) , percent			
					Maximum Cladding OD	Average Fuel OD	Fuel Length	Average Fuel Volume
5P	UO ₂ -8 w/o PuO ₂ : 96 percent dense, 0.110-in. axial hole	1	1425	0.14	0.6	0.7	0.9	2.1
		2	1320	0.33	0.1	0.1	1.2	1.4
		3	1320	0.55	0.7	0.9	2.4	4.2
		4	1310	0.74	0.6	0.8	2.0	3.6
		5	1320	0.92	0.8	0.9	1.6	3.4
		6	1300	1.10	0.3	0.4	1.4	2.2
		7	1280	1.28	0.6	0.3	1.7	2.3
		8	1260	1.45	1.5	0.8	1.5	3.1
			1300 avg		[0.7]	[0.6]		[2.7]

(a) Values in brackets are based on micrometer measurements of diameter; all other measurements were taken from neutron radiographs.

TABLE B-6. IRRADIATION CONDITIONS AND DIMENSIONAL CHANGES FOR SPECIMENS IN CAPSULE HT-BRR-14

Specimen	Description	BRR Cycle	Cladding Surface Temperature, C	Estimated Accumulated Burnup, 10^{20} fissions per cm^3	External Dimensional Changes ^(a) , percent			
					Maximum Cladding OD	Average Fuel OD	Fuel Length	Average Fuel Volume
383	UO ₂ : 96 percent dense, 0.110-in. axial hole	1	1330	0.22	0.5	0.6	0.2	1.4
		2	1250	0.46	0.3	0.3	-0.1	0.5
		3	1300	0.71	0.8	1.0	0.2	2.2
		4	1320	0.96	0.6	0.8	0.5	2.1
385	UO ₂ : 95 percent dense, 0.055-in. axial hole, tungsten foil at midradius	1	1500	0.24	0.5	0.6	0.5	1.7
		2	1440	0.51	1.0	1.1	0.7	2.9
		3	1460	0.79	1.8	1.9	1.5	5.3
		4	1470	1.07	2.7	2.7	1.3	6.7
387	UO ₂ : 95 percent dense, 0.055-in. axial hole, depleted fuel at axial midplane	1	1370	0.25	-0.1	-0.1	0.5	0.3
		2	1320 ^(b)	0.53	0.4	0.5	1.1	2.1
		3	1350	0.82	1.2	0.8	1.6	3.2
		4	1370	1.11	1.3	1.3	1.8	4.4
389	UO ₂ : >99 percent dense, 0.110-in. axial hole	1	1650 ^(b)	0.31	0.5	0.6	1.8	3.0
		2	1600	0.65	0.5	0.6	0.9	2.1
		3	1640	0.99	2.0	1.8	1.3	4.9
		4	1660	1.33	3.3	2.8	1.5	7.1
390	UO ₂ : >99 percent dense, 0.110-in. axial hole, vented cladding	1	1630 ^(b)	0.30	0.4	0.5	5.6	6.6
		2	1580	0.63	0.5	0.6	5.5	6.7
		3	1620	0.94	3.6	3.8	5.6	13.2
		4	1640	1.25	5.3	5.5	6.4	17.4
6P	UO ₂ -8 w/o PuO ₂ : 89 percent dense	1	1550	0.18	-0.1	-0.1	0.9	0.7
		2	1460	0.38	0.0	0.0	1.5	1.5
		3	1500 ^(b)	0.57	0.5	0.6	1.2	2.4
		4	1520	0.76	0.6	0.7	1.7	3.1

(a) Measurements were taken from neutron radiographs.

(b) This temperature, and all succeeding temperatures, estimated from inner-shell thermocouple after specimen thermocouple failed.

As shown in Figure B-2, the swelling data obtained from the oxide specimens in Capsule HT-BRR-14 are consistent with oxide swelling observed in previous capsules. Neutron radiographs of Capsule HT-BRR-14 (Figure B-3) show the specimens to be in good condition. A "bridge" of fuel across the top of the fuel in Specimen 390, apparently caused by vapor transport of fuel (mainly during the first cycle of irradiation) from the central regions of the specimen and subsequent condensation at the upper end of the fuel, Figure B-3, accounts for the large initial length increase reported in Table B-6. This phenomenon has been observed in other UO₂ specimens and usually is the result of a large axial temperature gradient.

FIGURE B-2. EXTERNAL VOLUMETRIC SWELLING RATE FOR UO_2 AND $\text{UO}_2\text{-PuO}_2$ SPECIMENS FROM CAPSULES HT-BRR-9, HT-BRR-11, HT-BRR-12, AND HT-BRR-14

FIGURE B-3. NEUTRON RADIOGRAPHS OF CAPSULE HT-BRR-14

Plans for Future Work

The postirradiation examination of the specimens from Capsule HT-BRR-11 will continue with metallography and retained fission-gas analyses on selected specimens.

The irradiation of Capsule HT-BRR-14 will continue toward a target burnup of 2×10^{20} fissions per cm^3 .

Fabrication of the specimens for irradiation in Capsule HT-BRR-16 will be completed and the irradiation will be initiated.

- (1) Storhok, V. W., and Chubb, W., "Swelling of UO_2 at 1700 C", Trans. Am. Nucl. Soc., 12, 76 (June, 1969).
- (2) Hilbert, R. F., Storhok, V. W., and Chubb, W., "High Temperature Irradiation Behavior of UN, UO_2 , and UC", Record of the 1969 Thermionic Specialist Conference, October, 1969, Carmel, California, IEEE, New York, pp 331-340.
- (3) Hilbert, R. F., Storhok, V. W., and Chubb, W., "Irradiation Behavior of UO_2 at Surface Temperatures from 1300 to 1900 C", Trans. Am. Nucl. Soc., 13, 132 (June, 1970).
- (4) Hilbert, R. F., Storhok, V. W., Chubb, W., and Keller, D. L., "Mechanisms of Swelling and Gas Release in Uranium Dioxide", J. Nucl. Mater., in publication.

Irradiation Behavior of Nitride Fuels at High Temperatures

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Objective of Program

To investigate the irradiation behavior of nitride fuels at surface temperatures from 1200 to 1900 C.

Approach Being Used

Several specimens of UN and tungsten-UN were or are being irradiated to study the swelling and fission-gas-release behavior of UN at high temperatures. Variations of the amount and configuration of porosity, the effect of cladding restraint, and the effect of grain size on the swelling behavior of UN are being investigated. The timing of gas release, the effects of a nitrogen overpressure (to control the stoichiometry), and the effects of additions of PuN are also being studied.

Capsule HT-BRR-13 contains three specimens of UN and one specimen of UN-15 w/o PuN designed to be irradiated at 1700 C. Two of the UN specimens are 93 percent dense, one with a sound cladding and one with a cladding slit every 90 deg (to reduce cladding restraint), to test the effect of cladding restraint (or lack of restraint) on the swelling and gas-release behavior of UN. A 73 percent dense UN specimen has a gas-pressure monitor to investigate the timing and magnitude of gas release from porous UN. The effect of PuN additions on the swelling and gas-release behavior of UN is being studied by comparison of the above specimen with a 78 percent dense specimen of UN-15 w/o PuN.

Except for the variations mentioned, each specimen consists of a column of slugs of fuel 1 in. long contained in a tungsten-26 w/o rhenium cladding with an OD of 0.25 in. and a wall thickness of 20 mils, as shown in Figure B-1.

Dimensional changes in the specimens are determined after each cycle of irradiation by measurements from neutron radiographs. The postirradiation examination includes (when appropriate) dimensional measurements, fission-gas-release (retention) measurements, and metallography.

Progress Reported Previously

Capsules HT-BRR-3, -4, and -6 contained seven specimens of bulk UN and two tungsten-UN cermet. Examination of these specimens was completed in Fiscal Year 1968 and the results were reported in BMI-1848 (August, 1968). Capsules HT-BRR-7 and HT-BRR-8 contained six bulk UN specimens and three tungsten-UN cermets. Examination of these specimens was completed in Fiscal Year 1969 and the results were reported in BMI-1870 (August, 1969). Capsules HT-BRR-10 and LRL-2 contained 12 specimens of bulk UN. Examination of these specimens was completed in Fiscal Year 1970 and the results were reported in BMI-1886 (July, 1970) and in several publications⁽¹⁻³⁾.

Two different types of swelling behavior apparently corresponding to two different types or compositions of UN have been observed in UN-base fuels at about 1700 C. One type is characterized by initial swelling rates at 1700 C of the order of 10 to 20 v/o per 10^{20} fissions per cm^3 caused by the accumulation of a large fraction of the fission gases at the grain boundaries. After development of a high degree of interconnected porosity, there is little or no additional swelling. The second type of UN is characterized by initial swelling rates of the order of 2 to 4 v/o per 10^{20} fissions per cm^3 , a low mobility of fission gases, and gradually increasing swelling rates throughout the irradiation. The swelling of this second type of UN also was observed to take place primarily by growth of fission-gas bubbles at the grain boundaries. However, the growth of these bubbles involved only a small fraction of the fission gas available in the fuel. Decreased swelling rates were realized by minimizing grain-boundary area. In the second type of UN this was accomplished through the use of very large-grained cast material. In both types of UN-base fuels, the location of interconnected porosity at grain boundaries seemed to eliminate the formation of large gas bubbles by venting the gas that was mobile enough to appear at grain boundaries. However, the porosity also reduced the ability of the swelling fuel to stress the cladding, so that the two effects cannot be completely separated.

The gas-pressure monitor attached to a 73 percent dense UN specimen operating at temperatures near 1700 C in Capsule HT-BRR-13 indicated that the gas-release rate was negligible to a burnup of about 0.25×10^{20} fissions per cm^3 . At this stage of the irradiation, the gas-release rate increased to nearly 100 percent until the transducer ceased operation at a burnup of 1.04×10^{20} fissions per cm^3 . The cumulative gas release at this burnup was about 65 percent. The gas-pressure monitor on an 85 percent dense UN specimen, which was irradiated at 1700 C in Capsule HT-BRR-10, indicated a low gas-release rate until a burnup of about 0.9×10^{20} fissions per cm^3 was achieved. Then, the gas-release rate increased to about 25 percent until the cladding failed at a burnup of 1.14×10^{20} fissions per cm^3 . The cumulative gas release at this burnup was about 15 percent. A comparison of these data indicated that increasing the amount of porosity

in UN at 1700 C has: (1) decreased the time delay before increased gas release occurs, (2) increased the gas-release rate after the delay, and (3) increased the cumulative gas release. The increased time before significant gas release from the 85 percent dense UN specimen, as compared to that for the 73 percent dense UN specimen, suggests that the processes of fission-gas agglomeration and channel opening take more time to occur as the specimen density increases.

The UN specimens in Capsule LRL-2 showed cladding diameter increases of less than 1 percent and external volume increases of less than 4 percent upon irradiation to about 2.1×10^{20} fissions per cm^3 at 1400 to 1600 C. Fission-gas release was found to be 5 percent or less in all cases, even though two specimens contained 25 v/o porosity, indicating that fission-gas mobility in the UN was low.

Work This Quarter

The irradiation of Capsule HT-BRR-13 was completed, and the capsule is cooling in the BRR pool in anticipation of the postirradiation examination. The irradiation conditions and dimensional changes of the UN and UN-PuN specimens in Capsule HT-BRR-13, as obtained from neutron radiographs, are given in Table B-7. Preirradiation and postirradiation neutron radiographs of the specimens are shown in Figure B-4. Because of the anomalous appearance of some of the fuel in the neutron radiographs, dimensional measurement data are not reported for the fuel in Specimens 371 and 380. Apparently, most, if not all, of the fine, cold-pressed UN in Specimen 371 sintered and shrank away from the cladding early in the first cycle of irradiation, and has since been redepositing by vapor transport on the cladding (Figure B-4). This type of vapor transport had been observed in several low-density UN specimens irradiated previously. Further details of the nature of this behavior will be given after the specimens have undergone a complete postirradiation examination.

Early in the present irradiation a high neutron-absorbing material, presumably plutonium-rich, transferred to the inside of the top end cap of UN-PuN Specimen 374 (Figure B-4). This effect is in agreement with data which show that PuN has a higher vapor pressure than UN⁽⁴⁾.

A graphical comparison of external volume increase versus burnup for UN Specimen 382 at 1710 C and UN-15 w/o PuN Specimen 374 at 1670 C from Capsule HT-BRR-13 and UN Specimens 351 and 352 at 1600 C from Capsule HT-BRR-10 is shown in Figure B-5. The data presented in Figure B-5 show a fortuitously small amount of scatter and exhibit the type of swelling behavior associated with nitrogen-rich UN. It should be noted that although UN-PuN Specimen 374 was only 78 percent dense at the start of the irradiation, the fuel densified and formed a large central void in the first cycle of irradiation. Thus, the fuel in Specimen 374 was probably about 90 percent dense after the first cycle of irradiation, and its behavior can be logically compared with that of the higher density specimens, as shown in Figure B-5.

A summary plot of external swelling rate versus cladding surface temperature data obtained from several capsule irradiations is shown in Figure B-6. This preliminary indication of the swelling of UN versus temperature may be compared with the estimates of the swelling rates of UO_2 and UC. Such a comparison is particularly useful at a temperature just above the temperature of "breakaway swelling". Figure B-2 shows that UO_2 exhibits external volumetric swelling rates in the range of 1 to 5 percent per 10^{20} fissions per cm^3 at cladding surface

temperatures between 1250 and 1350 C. Previous work at Battelle⁽⁵⁾ has shown swelling rates for UC between 0.8 and 4.5 percent per 10^{20} fissions per cm^3 at a surface temperature of 1350 C. Figure B-6 suggests that the swelling rates of UN at a surface temperature of 1350 C range between 0.4 and 2.5 percent per 10^{20} fissions per cm^3 . This preliminary result suggests that the behavior of UN is superior to that of UO_2 and UC.

TABLE B-7. IRRADIATION CONDITIONS AND DIMENSIONAL CHANGES FOR THE UN AND UN-PuN SPECIMENS IN CAPSULE HT-BRR-13

Specimen	Description of Fuel	BRR Cycle	Average Temperature, C	Estimated Accumulated Burnup, 10^{20} fissions per cm^3	External Dimensional Changes, percent			
					Maximum Cladding OD	Average Fuel OD	Fuel Length	Average Fuel Volume
371	UN: 73 percent dense Gas-pressure monitor	1	1660	0.30	1.6	No fuel measurements made ^(b)		
		2	1675	0.59	2.4			
		3	1690	0.89	3.2			
		4	1650	1.18	3.6			
		5	1715	1.50	4.8			
		6	1725	1.82	5.6			
382	UN: 93 percent dense	1	1690 ^(a)	0.33	-0.4	-0.5	-0.3	-1.3
		2	1700	0.64	-0.2	-0.2	0.2	-0.2
		3	1700	0.97	0.4	0.5	0.8	1.8
		4	1680	1.29	2.9	2.3	1.0	5.6
		5	1740	1.64	3.7	3.8	1.8	9.4
		6	1730	1.99	4.7	5.0	2.6	12.6
374	UN-15 w/o PuN: 78 percent dense	1	1710	0.3	0.0	0.0	1.0 ^(c)	1.0
		2	1690	0.59	0.2	0.2	-1.9	-1.5
		3	1650	0.89	0.7	0.8	-2.7	-1.1
		4	1620	1.18	0.8	0.9	2.3	5.5
		5	1675	1.50	3.8	3.7	2.1	9.5
		6	1675 ^(a)	1.82	5.3	5.5	3.0	14.0
380	UN: 93 percent dense Slit cladding	1	1575 ^(a)	0.27	0.1	No fuel measurements made ^(b)		
		2	1575	0.53	3.6			
		3	1550	0.80	10.3			
		4	1530	1.06	13.6			
		5	1580	1.35	20.3			
		6	1580	1.64	36.1			

(a) Temperature estimated from inner-shell thermocouple after specimen thermocouple failed.

(b) Due to the anomalous behavior of the fuel in this specimen, no dimensional measurements on the fuel are reported. See text for an explanation of specimen behavior.

(c) These values do not account for the fuel transferred to the inside of the top end cap.

UN Specimen 371
73 percent dense
Gas-pressure monitor

UN Specimen 382
93 percent dense

UN-15 w/o PuN Specimen 374
78 percent dense

UN Specimen 380
93 percent dense
Slit cladding

Preirradiation

Postirradiation

FIGURE B-4. NEUTRON RADIOGRAPHS OF THE UN AND UN-PuN SPECIMENS
IN CAPSULE HT-BRR-13

FIGURE B-5. EXTERNAL FUEL VOLUME INCREASE AS A FUNCTION OF BURNUP FOR UN AND UN-PuN SPECIMENS FROM CAPSULES HT-BRR-10 AND HT-BRR-13

FIGURE B-6. EXTERNAL VOLUMETRIC SWELLING RATE AS A FUNCTION OF CLADDING SURFACE TEMPERATURE FOR UN AND UN-PuN

Plans for Future Work

The UN and UN-PuN specimens in Capsule HT-BRR-13 will be examined at the Battelle-Columbus Hot-Cell Facility.

- (1) Hilbert, R. F., Storhok, V. W., and Chubb, W., "Irradiation Behavior of UN at High Temperatures", Trans. Am. Nucl. Soc., 12, 547-548 (December, 1969).
- (2) Hilbert, R. F., Storhok, V. W., and Chubb, W., "High Temperature Irradiation Behavior of UN, UO₂, and UC", Record of the 1969 Thermionic Specialist Conference, October, 1969, Carmel, California, IEEE, New York, pp 331-340.
- (3) Hilbert, R. F., Storhok, V. W., and Chubb, W., "The Role of Grain Boundaries in the Swelling of UN at 1600 to 1700 C", Trans. Am. Nucl. Soc., 13 (November, 1970).
- (4) Alexander, C. A., Ogden, J. S., and Pardue, W. M., "Vaporization of Nitride and Carbide Fuels", Trans. Am. Nucl. Soc., 12, 581-582 (November, 1969).
- (5) Melehan, J. B., Markworth, A. J., Perrin, J. S., Wullaert, R. A., Chubb, W., and Gates, J. E., "Irradiation Behavior of Uranium Monocarbide", BMI-1806 (June, 1967).

Irradiation Behavior of Carbide Fuels at High Temperatures

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Objective of Program

To investigate the irradiation behavior of carbide fuels at surface temperatures from 1200 to 1900 C.

Approach Being Used

Early experiments in the HT-BRR irradiation series showed that dense UC and tungsten-UC cermet swelled steadily at high rates when irradiated at temperatures above 1600 C to burnups up to 2.5×10^{20} fissions per cm³ (BMI-1848, August, 1968). Only slight tendencies toward decreased swelling rates with increasing burnup were observed and no improvements in swelling behavior were realized by the introduction of centerline holes. The high swelling rates were primarily attributed to a relatively high fission-gas retention (58 percent at 1675 C) that was associated with a tendency for the fission gases to precipitate and form relatively large, discrete bubbles within UC grains. Also, a portion of the high external swelling was attributed to cladding deterioration caused by a chemical reaction between the UC and the tungsten-rhenium cladding.

A variety of modifications of UC were or are being irradiated in attempts to either accommodate swelling internally (within the cladding) or to enhance fission-gas release or to enhance the strength of the cladding relative to that of the fuel. Because of the extremely high fuel temperatures that resulted (2000 to 2200 C), the use of a 10-mil annular gap around a bulk UC specimen proved to be totally ineffective as a method of preventing cladding strain at a cladding surface temperature of about 1600 C (BMI-1870, August, 1969). Swelling of a copper-UC cermet was much greater than expected because the cermet separated into an annular ring of solid UC on the inside surface of the cladding plus a central core of copper-rich liquid (BMI-1870).

A 60 percent dense specimen of crushed, minus 230-mesh particles of UC was irradiated in Capsule HT-BRR-10 to determine whether the gas-release behavior of UC can be modified by providing built-in gas-release channels and by lessening the distance fission gas must migrate to reach these channels, or whether the porosity is used to accommodate swelling and reduce the stress on the cladding. A 1-mil-thick tungsten foil was placed at the fuel-cladding interface to reduce the tendency for chemical reaction between the fuel and the cladding.

Capsule HT-BRR-13 contains two specimens of 60 percent dense UC to test the effect of buffering with tungsten on fuel-cladding reactions and on the swelling and gas-release behavior of porous UC. One of the specimens contains a 1-mil-thick tungsten foil external buffer at the fuel-cladding interface, while the other is internally buffered with 5 w/o tungsten in the UC and no tungsten foil.

All specimens consist of a column of fuel 1 in. long contained in either tungsten or tungsten-26 w/o rhenium claddings with an OD of 0.25 in. and a wall thickness of 20 mils, as shown earlier in Figure B-1.

Dimensional changes in the specimens are determined after each cycle of irradiation by measurements from neutron radiographs. The postirradiation examination includes dimensional and density measurements, fission-gas-release measurements, and metallography.

Progress Reported Previously

The 60 percent dense specimen of crushed UC particles irradiated at 1530 C in Capsule HT-BRR-10 showed a low initial swelling rate and a final external volume increase of 7 percent after a burnup of 2×10^{20} fissions per cm^3 . This result compared favorably with external volume increases of about 35 percent at the same burnup at 1680 C in dense UC. However, the relatively high initial swelling rates exhibited by the two 60 percent dense UC specimens at 1690 C in Capsule HT-BRR-13 suggested that a significant part of the improved swelling behavior of the specimen that operated at 1530 C was attributed to its lower irradiation temperature. The postirradiation microstructure of the specimen that operated at 1530 C suggested that the lack of extensive bonded interparticle contacts and the attendant inability of the swelling fuel to transmit stresses to the cladding was a major factor in eliminating swelling during the first half of the irradiation. The formation of solid bonds at the interparticle contacts by particle swelling and sintering led to a gradual increase in swelling rates during the last half of the irradiation. It was suggested that the specimens of loose-packed UC particles operating at 1690 C experienced increased rates of swelling of individual particles and bonding at interparticle contacts and, thus, higher swelling rates.

Work This Quarter

The irradiation of Capsule HT-BRR-13 was completed after six cycles. The irradiation conditions and dimensional changes of the 60 percent dense UC specimens in Capsule HT-BRR-13 are given in Table B-8. Neutron radiographs (Figure B-7) show that Specimen 377 is in good condition, but that internally buffered Specimen 379 has a region of asymmetrically swollen fuel. Apparently, the use of a 1-mil-thick tungsten foil at the fuel-cladding interface is the more successful of the two proposed methods of enhancing fuel-cladding compatibility.

TABLE B-8. IRRADIATION CONDITIONS AND DIMENSIONAL CHANGES FOR 60 PERCENT DENSE UC SPECIMENS IN CAPSULE HT-BRR-13

Specimen	Description of Fuel	BRR Cycle	Average Temperature, C	Estimated Accumulated Burnup, 10^{20} fissions per cm^3	External Dimensional Changes, percent			
					Maximum Cladding OD	Average Fuel OD	Fuel Length	Average Fuel Volume
379	UC: 60 percent dense, loose-packed particles, internal buffer of 5 w/o tungsten	1	1680(a)	0.25	0.4	0.4	-1.4	0.6
		2	1690	0.49	2.8	1.7	-1.0	2.4
		3	1660	0.74	4.7	3.5	-0.8	6.2
		4	1630	0.98	5.5	3.8	-0.3	7.3
		5	1690(b)	1.25	5.6	4.3	0.5	9.1
		6	1690	1.52	9.0	8.5	2.0	19.0
377	UC: 60 percent dense, loose-packed particles, 1-mil-thick tungsten foil	1	1675(a)	0.26	2.7	1.7	-2.6	0.8
		2	1700	0.51	2.1	1.6	-0.7	2.5
		3	1680	0.77	3.7	2.7	-0.1	5.3
		4	1650(b)	1.02	4.1	2.6	0.1	5.3
		5	1700	1.30	4.4	2.9	1.8	7.6
		6	1700	1.58	7.0	4.8	2.8	12.4

(a) Temperatures were approximately 225 C hotter for the first 40 min of the first irradiation cycle.

(b) Temperatures were estimated from inner-shell thermocouple after specimen thermocouple failed.

External volume changes as a function of burnup for the 60 percent dense UC specimens from Capsules HT-BRR-10 and HT-BRR-13 are compared in Figure B-8 with those for specimens of high-density, cast UC from earlier capsules. An explanation of the behavior of these specimens will be deferred until the postirradiation examination is completed; however, the increasing swelling rates probably result from increased rates of swelling of individual particles and bonding at interparticle contacts.

Plans for Future Work

The 60 percent dense UC specimens in Capsule HT-BRR-13 will be examined in the Battelle-Columbus Hot-Cell Facility.

Measurement of Fission-Gas Diffusion and Bubble Growth In UO_2

R. L. Ritzman, R. B. Price, and A. J. Markworth

Objective of Program

To determine fission-gas diffusion rates and to examine the dynamics of fission-gas bubble growth in uranium dioxide at low and intermediate fission exposures.

UC Specimen 379
60 percent dense
Internal buffer of 5 w/o tungsten

UC Specimen 377
60 percent dense
1-mil-thick tungsten foil

Preirradiation

Postirradiation

FIGURE B-7. NEUTRON RADIOGRAPHS OF 60 PERCENT DENSE UC SPECIMENS FROM CAPSULE HT-BRR-13

FIGURE B-8. A COMPARISON OF THE EXTERNAL VOLUME INCREASE OF UC SPECIMENS OF DIFFERENT DENSITIES AND TEMPERATURES FROM THE HT-BRR SERIES

Approach Being Used

Postactivation annealing experiments are being employed to produce measurable amounts of fission gas and to generate fission-gas bubbles in single-crystal UO_2 as a function of fission exposure, annealing temperature, and annealing time. The release data will be analyzed to obtain fission-gas diffusivity values, and electron microscopy will be used to examine fission-gas-bubble distribution. Results of both efforts will be correlated with current theories of fission-gas precipitation and bubble growth in nuclear fuels.

Progress Reported Previously

Previous studies of this type have been conducted primarily with uranium monocarbide. However, all work on UC has been terminated, and an effort to develop similar data on UO_2 has commenced. Preliminary results were reported last period which indicated that fission-gas diffusion proceeds at a significantly lower rate in UO_2 than in UC.

Work This Quarter

Improvements were made in the equipment used for performing high-temperature postactivation anneals, and several trial runs were made to establish the conditions which will be used with the single-crystal samples of UO_2 . On the basis of this work, the UO_2 will be heated under vacuum in a tungsten crucible within an induction furnace. The small UO_2 cylinders will be supported on chips of single-crystal UO_2 and also will be completely surrounded by stoichiometric UO_2 microspheres. The arrangement permits heating at temperatures up to 1600 C and for times of at least 8 hr without changes in specimen weight or appearance.

Small UO_2 cylinders, each weighing approximately 1 g, were machined from pieces of stoichiometric, single-crystal UO_2 that had been obtained from Battelle-Northwest. The samples were cleaned by agitation in a chemical etching solution in preparation for neutron activation. Most of the specimens are currently being encapsulated for irradiation. These will be used for fission-gas-release measurements.

However, an extended activation of two of the specimens has been completed in preparation for SEM examinations for fission-gas bubbles. The irradiation and annealing conditions used for these two specimens were selected on the basis of data reported by Cornell⁽¹⁾ in order to produce a fission-gas-bubble distribution which would be readily observable with SEM techniques.

Quantitative predictions of bubble-growth characteristics in these single-crystal specimens were made under the following assumptions:

- (1) An initially homogeneous gas-solid solution, with a dissolved-gas concentration of $0.25 \times 10^{18} \text{ cm}^{-3}$
- (2) A specimen temperature, during the anneal, equal to 1700 C and a gas-diffusion coefficient of $1.7 \times 10^{-14} \text{ cm}^2 \text{ per sec}$ calculated from Cornell's data⁽¹⁾ extrapolated to this temperature
- (3) A vacancy supply to bubbles rapid enough to maintain the gas pressure approximately equal to the surface tension restraint [assuming 600 erg per cm^2 as the UO_2 surface tension⁽²⁾]

- (4) The duration of the anneal (about 10 days) assumed to be sufficiently short, and the rate of fission-gas diffusion in UO_2 at 1700 C sufficiently low, that only a small fraction of the gas initially in solution diffuses into bubbles.

The fourth assumption implies that effects, upon precipitation kinetics, of competition between bubbles for the dissolved gas, are not likely to be felt during the experiment, since such effects are manifested only when the dissolved-gas concentration has been significantly reduced below its initial value.

Under these assumptions, the time-dependent diffusion equation for volume diffusion of gas to a spherical, stationary bubble was solved assuming bubbles of initial radius 5 A. The results of this calculation are shown in Figure B-9.

FIGURE B-9. PREDICTION OF BUBBLE PRECIPITATION IN UO_2 DURING ANNEAL AT 1700 C AFTER ACTIVATION TO 1×10^{18} FISSIONS PER CM^3

Curve A is based on a modified van der Waals equation of state for the gas in the bubble, and solution of the time-dependent diffusion equation for volume diffusion of gas in the fuel; Curve B is based on ideal gas behavior and quasi-stationary diffusion of gas.

Curve A represents the prediction based on solution of the time-dependent diffusion equation and assuming a modified van der Waals equation of state for the gas within bubbles [with a van der Waals constant of 51 cm^3 per mole assumed for xenon⁽³⁾]. Curve B represents an analogous prediction based on assumed quasi-stationary gas diffusion within the fuel and ideal gas behavior within bubbles.

The difference between the two curves is due, almost entirely, to the higher rate of bubble growth predicted using the van der Waals equation of state. Because of the initially small bubble radii, conditions of quasi-stationary diffusion are achieved soon after the start of the anneal.

Of course, on a physical basis, Curve A represents a more realistic prediction, and it can be seen that after 10 days (8.64×10^5 sec) at 1700 C, the predicted bubble diameter is approximately 340 Å. Bubbles of this size should be visible via SEM.

In addition, the calculated gas content of one of these bubbles was found to be about 5×10^4 gas atoms after 10 days. Thus, for example, if 10^{11} bubbles are nucleated per cm^3 , only about two gas atoms per hundred will be in bubbles after the anneal (making the rough assumption of a uniform bubble size distribution), the rest still being in solution. Consequently, our initial assumption, regarding the negligible depletion of gas atoms from solution, appears to be well founded, at least if the bubble concentration is not in excess of roughly 10^{11} cm^{-3} . This simulation of bubble behavior will be checked experimentally by measurements of bubble size and distribution in the two samples of UO_2 irradiated to 10^{18} fissions per cm^3 after they are annealed at 1700 C.

Plans for Future Work

The UO_2 single-crystal specimens to be used for fission-gas-release measurements will be activated to a nominal exposure of 10^{15} fissions per cm^3 . Then individual specimens will be heated out-of-pile at temperatures ranging from 1200 to 1600 C with measurement of the evolved Xe^{133} . The resulting Xe^{133} release curves will be analyzed in terms of gas immobilization mechanisms to obtain fission-gas-diffusivity values.

The pair of specimens irradiated to 10^{18} fissions per cm^3 for SEM work will be recovered and annealed for an extended period near 1700 C under static helium to produce fission-gas bubbles. Then, the samples will be fractured and examined via SEM to determine the gas-bubble concentration and size distribution.

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Gas-Bubble Nucleation and Growth

A. J. Markworth, W. Oldfield, and G. T. Hahn

Objective of Program

To describe the growth of fission-gas bubbles in nuclear fuels in terms of atomic diffusion processes, and thereby to predict the swelling and gas-release behavior of the fuels.

Approach Being Used

The growth of inert-gas bubbles resulting from the coupled influx of gas atoms and lattice vacancies is being investigated in solids. Analytical and numerical models, which take into account such parameters as nonuniformity of bubble size, gas diffusivity and solubility in the fuel, and the self-diffusivity of the fuel, are being developed for both the postirradiation and in-pile cases. The validity of the models is tested by comparison with bubble-growth and swelling data accumulated as another part of this program.

Progress Reported Previously

This effort was begun during Fiscal Year 1966 and was first reported in BMI-1783. A computer simulation of fission-gas-bubble precipitation in nuclear fuels, under both in-pile and out-of-pile conditions, has been constructed. The simulation analyzes the evolution in time of a size distribution of stationary bubbles, growing through the coupled volume diffusion of gas atoms and lattice vacancies. In certain limiting situations, analytical approximations are possible, and descriptions of these special cases have been published⁽¹⁻⁴⁾. Predictions of the model have generally been in good accord with swelling data and with experimental observations of gas bubbles in fuels. For example, it was found⁽⁵⁾, using the results of the analysis contained in Reference (3), that the model predicts that fission-gas swelling in UC, at a given burnup, is virtually nil at temperatures below roughly one-half the absolute melting point, but rises rapidly as the temperature increases beyond that point. In addition, scanning-electron-microscopic observations of gas bubbles in neutron-activated and annealed UC⁽⁶⁾ were consistent with simulations of these anneals obtained using the computer model. The latter model has also been used to show that in-pile bubble precipitation in UO₂ depends strongly upon the input data chosen for uranium self-diffusion, indicating, for instance, that the restraint offered to bubble growth by the UO₂ matrix may play a significant role in bubble-growth kinetics (and also indicating a need for better understanding of vacancy diffusion mechanisms in UO₂).

Work This Quarter

In polycrystalline nuclear fuel specimens, two distinct populations of fission-gas bubbles are generally observed: one within grains and one situated on the grain boundaries. The bubbles on grain boundaries are often observed to be larger than those within grains, and this is usually attributed to the relatively rapid rates of diffusion of gas atoms and vacancies along the boundaries, which can be several orders of magnitude greater than corresponding volume-diffusion rates. It is therefore to be expected that the presence of grain boundaries can have a significant effect on the overall bubble-growth kinetics, keeping in mind the fact that all the bubbles within the fuel are competing with each other for the available dissolved gas. Consequently, efforts were undertaken this past quarter to include grain-boundary bubbles in the bubble-growth model, and thus to provide a more realistic picture of the simultaneous growth of these dual bubble populations in a polycrystalline fuel specimen.

Initially, it will be assumed that vacancy flow to all bubbles is rapid enough to maintain the gas pressure equal to the surface tension restraint. This assumption will be relaxed in subsequent, more general treatments. The flow of gas atoms to intragranular and grain-boundary bubbles is illustrated schematically in

Figure B-10. For an intragranular bubble, the flow is assumed to be symmetrical about the center of the bubble (assumed spherical). On the other hand, for a bubble on a grain boundary, gas atoms first flow, via volume diffusion, to the grain boundary from the interior of the adjacent grains, and then flow rapidly along the boundary into the bubble. Note that the shape of the bubble on the grain boundary is not spherical, but rather is determined by free-energy considerations associated with "attachment" of the bubble to the grain boundary^(7, 8).

On this general foundation, an analytical expression was derived for the rate of influx of gas atoms into grain-boundary bubbles. This expression, and an analogous equation for intragranular bubbles⁽²⁾, was then applied to the construction of a simple model which describes the simultaneous growth of the dual bubble populations. This initial model is based on the following assumptions: (1) uniform size distributions of bubbles (but generally different sizes for intragranular bubbles relative to grain-boundary bubbles); (2) gas pressure in bubbles restrained only by surface tension; and (3) zero thermodynamic solubility of the gas in the fuel. These restrictions will be relaxed when the grain-boundary-diffusion analysis is sufficiently refined to permit its inclusion into the more general computer model. An approximate expression for the conservation of gas atoms is given as follows:

$$m = C + n_v N_v + n_b N_b ,$$

where

m = total concentration of gas in fuel (m is a constant for out-of-pile bubble growth, and variable in-pile)

C = instantaneous average concentration of gas dissolved in grain volume

n_v, n_b = volume concentration of intragranular and grain-boundary bubbles, respectively

N_v, N_b = instantaneous number of gas atoms in an intragranular and a grain-boundary bubble, respectively.

The expressions for the time rate of change of N_v and N_b were incorporated in the above equation with the equation of state for the respective types of bubbles, in order to deduce a system of simultaneous differential equations describing the overall growth kinetics. Unfortunately, this system cannot be integrated in exact form, so a computerized numerical analysis was carried out using the second-order Runge-Kutta approximation.

In the initial test of this model, conditions were chosen which corresponded roughly to the postirradiation annealing of a lightly irradiated specimen of uranium monocarbide, as reported previously⁽⁶⁾. These assumed conditions based on the above-mentioned specimen included: (1) $m = 1 \times 10^{17}$ gas atoms per cm^3 ; (2) $n_v = n_b = 1 \times 10^9$ bubbles per cm^3 ; (3) temperature = 1700 C; (4) volume diffusion coefficient for xenon in UC as determined by Ritzman, et al.⁽⁹⁾, grain-boundary diffusion coefficient for xenon in UC estimated to be three orders of magnitude greater than the corresponding volume diffusion coefficient; (5) all bubbles growing from initial time zero with an initial radius of 10 Å; (6) the gas in all bubbles satisfying the ideal gas law. Obviously, the latter assumption is not valid for very small bubbles (radius ≤ 100 Å), and the rate of growth of small bubbles is underestimated by assuming perfect gas behavior^(4, 10, 11).

FIGURE B-10. SCHEMATIC REPRESENTATIONS OF THE DIFFUSION OF GAS ATOMS INTO VARIOUS TYPES OF BUBBLES

It must be emphasized, at this point, that the analysis involves several crude assumptions and approximations, and this should be kept in mind especially when comparing predicted bubble growth kinetics with experimental data. On the other hand, qualitative differences in behavior between intragranular and grain-boundary bubbles, and any influence that these two bubble populations have upon each other, should be readily demonstrable.

The results of the analysis are illustrated in Figure B-11. The log-log scale of Figure B-11 is particularly useful for exhibiting the relatively rapid rates of growth found for grain-boundary bubbles at "early" times. The triangular and circular data points represent data from UC Specimens 13 and 10, respectively⁽⁶⁾, the triangular point representing primarily grain-boundary bubbles, the circular point representing intragranular bubbles.

The relatively good agreement between the predictions of the model and the experimental data are probably fortuitous. In this regard, a number of important questions need to be answered, including the following: (1) a numerical value for the grain-boundary diffusion coefficient was chosen rather arbitrarily; what would be the overall effect on the predicted precipitation kinetics of increasing this value by, say, one or two orders of magnitude? (2) likewise, values for n_v and n_b were chosen to correspond very roughly with experimental data; what would be the effect of varying one or another (or both) of these parameters? Answers to these questions are presently being obtained.

It is also interesting to note that the simulation shown in Figure B-11 concludes with about 36 percent of the gas being contained in intragranular bubbles, and about 64 percent in grain-boundary bubbles. In other words, the assumed initially homogeneous distribution of dissolved gas undergoes precipitation such that nearly two-thirds of the gas is associated with grain-boundary bubbles after an anneal of several hours. This is not in good agreement with the available data. The lack of agreement probably derives from the assumption of equal numbers of bubbles on grain boundaries as exist within grains.

Plans for Future Work

Efforts to include the coupled growth kinetics of grain-boundary and intragranular bubble populations in the general computer model will be continued. Special emphasis will be placed upon describing the in-pile growth of bubbles in polycrystalline UO_2 .

In addition, computer simulations of bubble precipitation during postirradiation heat treatment of lightly irradiated UO_2 will be carried out in support of experimental work in this area⁽¹²⁾. These combined studies are aimed toward increased understanding of the kinetics of bubble growth in oxide fuels.

The computer program describing the in-pile growth of gas bubbles will be expanded in a number of additional directions. First, it is well known that vacancy concentrations within a fuel under irradiation can be well in excess of those characterizing thermal equilibrium. This essentially gives rise to a mechanism for irradiation-enhanced creep, and the program will be extended in an attempt to include this phenomenon. Second, the behavior of very small bubbles can be more realistically described by a van der Waals equation of state than by the perfect gas law, and the program will be correspondingly modified.

FIGURE B-11. SIMULATION OF SIMULTANEOUS GROWTH OF INTRAGRANULAR AND GRAIN-BOUNDARY BUBBLES IN UC COMPARED TO DATA FOR GRAIN-BOUNDARY BUBBLES (Δ) AND INTRAGRANULAR BUBBLES (O)

To more realistically account for the in-pile behavior of oxide fuels, the effect upon bubble-precipitation kinetics of periodic sweeping of bubbles by moving lenticular pores will be evaluated. Results obtained from another program⁽¹³⁾ will be of value here.

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- (12) See the preceding section of this report entitled "Measurement of Fission-Gas Diffusion and Bubble Growth in UO_2 ".
- (13) See the following section of this report entitled "The Morphology and Migration of Lenticular Voids".

The Morphology and Migration of Lenticular Voids

A. J. Markworth, W. Oldfield, and G. T. Hahn

Objective of Program

To experimentally study the morphology and migration of lenticular voids in UO_2 , to evaluate the results in terms of current theory, and to investigate possible means for controlling fuel swelling resulting from void migration.

Approach Being Used

A theory⁽¹⁾ has been developed to explain the morphology and migration of lenticular voids in UO_2 in terms of crystal-growth phenomena. Concurrently, a laser image furnace was constructed and is being used to generate large temperature gradients in unirradiated UO_2 specimens in order to experimentally evaluate the theory on the basis of the behavior of fabricated porosity in the specimens.

Progress Reported Previously

The laser image furnace has been used to measure the temperature dependence of the lenticular-void migration velocity in UO_2 and the results have been interpreted in terms of a subdivision of the total available driving force for migration into various components (e.g., relating to faceting, diffusion through the UO_2 vapor, impurities, capillarity, atomic attachment kinetics, etc.). The details of previous theoretical and experimental work have been reported in two publications^(1,2).

Work This Quarter

Experiments have been undertaken in an attempt to observe the redistribution of impurities in the wakes of lenticular voids migrating up a thermal gradient.

The theory postulates that impurities encountered by a migrating lenticular void are preferentially redistributed along the boundaries of the columnar grain which is generated by the void as it migrates.

Specimens of UO_2 are being used having densities of about 92 percent of the theoretical value. Hence, a sizable fraction of the fabricated porosity can be expected to be interconnected; in particular, porosity near the sample surface is likely to be interconnected to the surface, an important observation in view of the fact that the voids observed to migrate in these experiments lie just a few mils from the outer surface. Impurities such as zirconia and platinum will be deposited on the surfaces of this interconnected porosity from a liquid solution.

Pore migration in the specimen is subsequently induced via the laser image furnace. Migrating pores overtake regions containing the impurity, producing partitioning. The partitioning is observed by taking microprobe traces, on a polished section of the specimen, across columnar grains generated by migrating pores. Preferential concentration of the impurities near the grain boundaries is the effect being sought.

Plans for Future Work

Other experiments will be carried out using the laser image furnace. Successive traversals of lenticular voids will be induced by producing void migration at a given laser power, then increasing the laser power to extend the region in which voids are generated. The newly generated voids will move into regions already traversed, and their behavior within these regions will be examined. This experiment is most important, since a current theory⁽³⁾ describing fission-gas swelling in oxide fuels is based upon the hypothesis that fission gas is continually being swept to the interior fuel surface by continuous generation of, and traversals by, rapidly moving lenticular voids.

The effect of surface active agents (impurities which reduce surface energy) will be evaluated. Surface active agents should increase the rate of step nucleation and thus lead to higher rates of void migration. Experimentation of this nature could ultimately enable us to control the migration rates of lenticular voids in fuels under irradiation, and thus lead to optimum swelling resistance.

Some efforts will also be devoted to model construction as applied to lenticular void migration in oxide fuels. In particular, these studies will be used to evaluate the effect of these voids on overall fuel swelling. Comparisons will be made with oxide swelling data from the HT-BRR-capsule series.

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Mechanical Model of Swelling of Nuclear Fuels

S. K. Batra, S. S. Bupara, and R. L. Jentgen

Objective of Program

To develop a mathematical model for the external dimensional change of nuclear fuels whose mechanical behavior is representative of that believed to be occurring in-pile.

Approach Being Used

The model is to be applicable to both high-temperature irradiations and LMFBR conditions so that it can be used both as a vehicle for interpreting data obtained under each of the above conditions and for relating high-temperature irradiation data to LMFBR behavior.

A purely mechanistic approach is adopted in meeting this objective. A quantitative analysis of an idealized model of a clad fuel element is used to provide further insight into the radial swelling problem and to suggest ways in which such dimensional changes can be controlled.

Progress Reported Previously

During Fiscal Year 1970, a nonisothermal model was developed which took into account the influence of temperature on the creep behavior of oxide fuels. A series of simplifying assumptions was made to make the problem tractable through numerical analysis. These assumptions have been listed in previous reports.

At the end of the fiscal year the model was essentially operational and preliminary runs were made. Although it was found that no significant swelling could be predicted for the set of input parameters used, the general trends appeared to be physically reasonable. On this basis it was believed that a parametric study using the model was in order. However, the computation times involved were quite large. It was proposed that an effort should first be made to shorten the computational time before undertaking parametric studies.

Work This Quarter

The nonisothermal program (SWLTHR) was analyzed in great detail and the tolerances in the iterative schemes were tightened. This resulted in significant reduction of the computing time; a run which took about 50 central processor units to compute now takes about 15 such units.

The particular experimental result that the model is being required to analyze shows that the inner boundary of a hollow (annular) fuel moves inward while the cladding shows an overall swelling (i. e. , a radial motion outward). The mass conservation requirement in the present mathematical model makes this kind of behavior impossible; it requires the inner and outer surfaces to move in the same direction. This difficulty arises because, in practice, the fuel material is at liberty to change the shape and volume of the grains, and this is very difficult to duplicate in a mathematical model that treats the fuel as a continuum. On the other hand, it may be possible to incorporate the effect into the mathematical model by an appropriate assumption concerning the compressibility of the fuel. Efforts are currently under way to assess this possibility by means of a subterfuge in the analysis such that motion in opposite directions of the inner and outer surfaces of the fuel is possible.

Plans for Future Work

In the next quarter, attempts will be made to bring the mathematical model more in line with the experimental observations such that swelling of the fuel material both inwardly and outwardly is feasible.

CREEP OF REACTOR FUEL MATERIALS
(AEC-DRDT AND ANL)

Program Management

J. S. Perrin

Substantial fuel-element dimensional changes can occur in-pile during reactor operation. A major portion of the dimensional changes can be caused by fuel swelling from the accumulation of fission-product atoms. The gaseous atoms will agglomerate or flow into bubbles, the fuel matrix is stressed, and deformation or growth occurs in a manner thought to be analogous to creep deformation. As the fuel swells, both from the gaseous and solid fission products, growth is restrained by the fuel-element cladding and the cooler, outer region of fuel. Fuel porosity is introduced to accommodate part of this swelling, thus reducing the total strain on the cladding. This fuel swelling accommodation factor will depend primarily on the relative creep strength of the fuel and cladding. These considerations require that the influence of the fission process on the creep strength of ceramic fuels be determined before the most meaningful fuel design studies can be made.

In recognition of these factors, a program was set up to study the in-pile and out-of-pile creep of ceramic fuels as support for the LMFBR program. The program is being conducted for the AEC Division of Reactor Development and Technology and Argonne National Laboratory. Initial emphasis is on UO_2 and $\text{UO}_2\text{-PuO}_2$, since $\text{UO}_2\text{-PuO}_2$ is the reference fuel for the Fast Flux Test Facility.

The in-pile creep experiments will be conducted in compression, and a constant load will be maintained during testing. In order to reduce the radial temperature gradient, specimens will be hollow cylinders. Capsules will be operated in both the Battelle Research Reactor (BRR) and in a higher flux reactor.

Preirradiation creep tests will be conducted on UO_2 and $\text{UO}_2\text{-PuO}_2$ to determine to what extent out-of-pile behavior can be used to predict in-pile behavior, and to help provide information on basic creep mechanisms which can be used in the design of fuels of increased creep strength. The data obtained will be used in the selection of stresses and temperatures for the in-pile creep tests and will later be compared to the in-pile data to determine the effect of the fission process on the creep rate. In the case of UO_2 , tests will be run on solid cylindrical specimens as well as hollow cylindrical specimens to determine whether specimen geometry affects creep behavior.

During the in-pile creep tests, specimens will be under a temperature gradient. In order to determine whether there is an effect of the fission process on the creep behavior, it will be necessary to compare isothermal preirradiation creep behavior with anisothermal in-pile creep behavior. An analysis will be developed for making this comparison and will include a thermal analysis to predict the temperature distribution of in-pile test specimens.

In order to predict the results of the fission process on the creep behavior of UO_2 and $\text{UO}_2\text{-PuO}_2$, the known effects of the fission process on those properties of UO_2 and $\text{UO}_2\text{-PuO}_2$ that may influence the creep behavior will be considered. This will be combined with additional creep experiments on UO_2 to determine the

effect of variables such as stoichiometry, grain structure, and porosity. Etch pitting and transmission-electron microscopy will be used to determine structural changes.

Design, Fabrication, and Operation of In-Pile Creep Capsules

R. A. Robinson, S. J. Basham, and J. S. Perrin

Objective of Program

To design, fabricate, and operate capsules for determining the in-pile compressive creep behavior of UO_2 - PuO_2 fuels as affected by fission rate, stress, temperature, and burnup.

Approach Being Used

A series of irradiation capsules is planned for creep studies of UO_2 - PuO_2 during in-pile exposure. Environmental conditions include specimen surface temperatures up to 1300 C with compressive stresses up to 10,000 psi. The experimental variables include temperature, stress, burnup, and fission rate.

A stepwise procedure has been developed which involves the following series of experiments:

- (1) Capsule assembly operated in the laboratory to check loading and strain measurement systems
- (2) Capsules operated in the BRR to check the creep behavior of UO_2 and UO_2 - PuO_2 at lower fission rates, i. e., $< 2 \times 10^{13}$ fissions/(cm^3)(sec)
- (3) Capsules with high heat-removal capability operated in a high-flux facility to obtain the creep behavior of UO_2 - PuO_2 at high fission rates.

Initial experiments will be conducted with UO_2 specimens.

Progress Reported Previously

The initial effort on this program was reported in BMI-1851. A low-fission-rate creep capsule was designed for in-pile experimentation at the BRR. The design incorporated electrical heating; an unclad, hollow, cylindrical specimen; a gas-flow transducer for specimen-deformation measurement; and a bellows-sealed, gas-actuated compressive loading system.

After the successful operation of various heater mock-up tests and capsule prototype experiments in the laboratory, four creep capsules were assembled with UO_2 specimens and operated in the BRR. Information on the fabrication and operation of these assemblies is contained in BMI-1886.

The UO_2 specimen in Creep Capsule BRR-4 failed during the startup of the initial experiment. The capsule was removed from the BRR pool, decontaminated and returned to the fabrication laboratory for reassembly with a new UO_2 specimen. Since the components in the vicinity of the specimen had been heated to about 1200 C, it was necessary to replace all the refractory-metal components (heaters, thermocouples, and platens). In addition, the alumina components were cleaned and the creep-sensing probes and transducer were replaced.

The design of an apparatus for measuring creep properties of LMFBR reference fuels at fission rates up to 5×10^{13} fissions/(cm^3)(sec) was initiated.

An analysis of existing test reactor facilities led to the selection of the Oak Ridge Reactor (ORR) for the location of the high-flux experiment.

Work This Quarter

Creep Capsule BRR-4A Fabrication. When the specimen prematurely failed before irradiation of Capsule BRR-4 the unit was returned to the laboratory and disassembled. A new specimen was installed, as were all specimen thermocouples, heaters, baffles, platens, displacement probes, and displacement transducer.

The reassembly was completed in the Battelle-Columbus Capsule Laboratory and the capsule was transferred to the BRR where the instrument leads, gas lines, and lead tube were installed. The assembly was set up in the reactor pool and preoperation checks were initiated. The transducer output was found to be erratic, giving indications of intermittent plugging of the lower orifice.

The assembly was taken from the pool and a section of the secondary containment was removed (at the transducer housing). The transducer housing was manually raised, lifting the lower orifice from its seating position around the needle. Large volumes of gas were passed through the transducer. Pressure drops across the lower orifice varied for a short time and then became stable. The high flow was continued for a 24-hr period with no further variations in pressure drop. The housing was resealed and the normal flow rate was maintained for 48 hr. The transducer readings were normal and did not vary during this period. It was believed that the erratic behavior of the transducer system was caused by particulates introduced into the transducer gas lines during reassembly of the lead tube. Lifting the orifice from the needle apparently permitted the particles to pass through the orifice into the chamber below the transducer.

The opening in the secondary containment was then resealed. The capsule was placed in the reactor pool and a neutron radiograph was taken. The specimen appeared to be intact and well centered between the heater shells and platens. The transducer system was rechecked and performed satisfactorily.

Creep Capsule BRR-4A Operation. A series of experiments was conducted in Capsule BRR-4A, as shown in Table C-1. Test 4A was conducted out-of-capsule in order to characterize the specimen before loading into the creep capsule.

Test 4B was planned as a short-time test (≤ 24 hr) with the objective of seating the UO_2 specimen for the subsequent series of experiments. Upon completion of this test it was found that the specimen had deformed only slightly (~ 0.12 percent).

The out-of-pile characterization curve was established for the UO_2 specimen at 1200 C, 4000 psi, and zero fission rate (Test 4C). The specimen stress was then reduced to about 600 psi and the linear-motion displacement transducer was monitored for about 60 hr to determine its stability. The UO_2 specimen should not deform a measurable amount at 1200 C and 600 psi in 60 hr. Since the transducer indicated no deformation over this time period, it was concluded that the deformation system was operating satisfactorily, and the first in-pile experiment, Test 4D, was begun.

TABLE C-1. SUMMARY OF CREEP EXPERIMENTS PERFORMED IN CREEP CAPSULE BRR-4A

Test	Fission Rate, fissions/(cm ³)(sec)	Measured Temperature ^(a) , C	Effective Temperature ^(b) , C	Stress, psi	Creep Rate, hr ⁻¹	Objective of Test and Comments
4A	Out of capsule	1180	1180	3500	3.6×10^{-5}	To establish out-of-capsule creep behavior
4B	0	1200	1200	1500	--	To seat specimen against platens
4C	0	1200	1200	4000	8.22×10^{-5}	To establish out-of-pile creep rate in capsule; test did not get out of primary creep
4D	$\sim 4 \times 10^{12}$	1100	1085	4000	5.10×10^{-6}	To determine whether creep-rate enhancement occurs at low fission rate
4E	$\sim 1.2 \times 10^{13}$	1135	1110	4000	3.54×10^{-5}	To determine creep rate at high fission rate
4F	$\sim 1.2 \times 10^{13}$	1025	1000	4000	2.66×10^{-6}	To determine activation energy with data from Test 4E
4G	$\sim 1.2 \times 10^{13}$	1175	1150	4000	--	To determine activation energy with data from Tests 4E and 4F; specimen failed
4H	$\sim 1.2 \times 10^{13}$	1155	1130	900	--	To check output of strain-measuring system; neutron radiographed; capsule transferred to hot cell

(a) Measured temperature is temperature of middle thermocouple on specimen.

(b) Effective temperature of in-pile specimen is the temperature at which an isothermal specimen would have the same overall measured creep rate, assuming no in-pile enhancement of creep rate.

The capsule was located in the BRR core so that the UO₂ specimen would be irradiated at about 4×10^{12} fissions/(cm³)(sec). The specimen temperature was brought up to 1100 C with the electrical heaters and the load-gas pressure was set to apply a compressive stress of 4000 psi.

After the completion of Test 4D, the capsule was lowered in its test cell location to achieve a fission rate of about 1.2×10^{13} fissions/(cm³)(sec) in the UO₂ specimen. Tests 4E and 4F were then conducted at measured specimen temperatures of 1135 and 1025 C, respectively. The compressive stress was held at 4000 psi for both experiments.

After the completion of Test 4F, Test 4G was begun at 1175 C, 4000 psi, and a fission rate of about 1.2×10^{13} fissions/(cm³)(sec). The large amount of deformation observed in the first 1-1/2 hr of the test indicated that the specimen probably failed. In order to check the functionality of the deformation measurement system, Test 4H was conducted during the remainder of the reactor cycle. This experiment was run at 1155 C, 900 psi, and a fission rate of about 1.2×10^{13} fissions/(cm³)(sec). The low stress condition was selected to monitor the output

of the transducer system. As in the low stress condition in Test 4C, the transducer indicated very little specimen deformation, and it was concluded that the deformation system was performing satisfactorily. Experiment 4H was terminated by the scheduled shutdown period for the BRR cycle. The data obtained will be discussed in the following section of this report.

A neutron radiograph obtained of the specimen region of the capsule showed the specimen to be barrel-shaped and cracked. Since the UO_2 specimen was no longer in a suitable test condition, the capsule was shipped to the Battelle-Columbus Hot-Cell Facility for posttest examination.

The operation of the auxiliary systems in Capsule BRR-4A was excellent. All of the thermocouples except the one placed on the axial centerline of the specimen performed satisfactorily. The centerline specimen thermocouple provided a good temperature measurement about half of the time. It appeared to be intermittently shorting in a lower temperature region.

Creep Capsule BRR-5 Fabrication. Creep Capsule BRR-5 is currently being fabricated and assembled in the Battelle-Columbus Capsule Laboratory. The partially assembled unit will be shipped to the Battelle-Columbus Plutonium Facility for installation of the mixed-oxide specimen and final fabrication.

Facility for Installation of UO_2 - PuO_2 into Creep Capsules. The design and construction of a handling facility for use in the assembly of creep capsules with unclad UO_2 - PuO_2 specimens has been completed. The facility is located in the Battelle-Columbus Plutonium Laboratory.

The handling facility consists of a Plexiglas box positioned on a platform located about 7 ft above floor level. A working platform surrounds the box to provide access to operating personnel. The containment box is elevated to allow the partially assembled capsules (about 6 ft in length) to be held in a vertical position while the mixed-oxide specimens are being installed and the primary fission-gas containment are being sealed. The containment box is provided with access doors and glove ports and is connected to an absolute filter that maintains the pressure inside the box below atmospheric pressure throughout the entire assembly procedure. The floor of the box is equipped with a port opening onto which a sealed plastic bag is attached. The capsule leads are inserted into the bag during the time the mixed-oxide specimen, thermocouples, and loading system are being installed in the primary containment region of the apparatus.

A vacuum diffusion pump capable of maintaining a vacuum pressure of about 10^{-6} torr is located within the handling box. The pumping system is used to evacuate the primary containment region of the capsules prior to backfilling with high-purity helium gas and sealing the chamber.

High-Flux Creep-Capsule Design. The conceptual design phase of the high-flux apparatus has been completed. The design incorporates a hollow UO_2 - PuO_2 specimen in a liquid-metal bath, gas-gap temperature control, pneumatic transducer for measurement of specimen deformation, and a bellows-sealed pneumatic loading system that applies a uniaxial compressive stress to the specimen.

Additional heat-transfer analyses have been completed for the high-flux capsule, with temperature distributions being calculated in reference mixed-oxide and mixed-nitride fuels. Three specimen geometries were evaluated: a stack of hollow disks or washers; a hollow cylinder 0.25 in. in OD, 0.15 in. in ID, and 0.75 in. long; and a hollow cylinder 0.25 in. in OD, 0.19 in. in ID, and 0.75 in. long.

Table C-2 provides a summary of the calculations. The three cases comprising the first series of calculations in Table C-2 are hand calculations for materials with a purely radial mode of heat transfer and a temperature gradient equal to zero at the inner radius of the specimen. For the first two cases, representing mixed-oxide materials, the decrease in the calculated radial ΔT from 450 to 160 C shows the amount of change that is available by decreasing the wall thickness of the hollow cylinder from 50 to 30 mils.

TABLE C-2. SUMMARY OF TEMPERATURE DISTRIBUTIONS CALCULATED FOR
SELECTED SPECIMEN MATERIALS AND GEOMETRIES

Material ^(a)	L/D Ratio ^(b)	Temperature Differential (ΔT), C		Surface Temperature, C
		Radial	Axial	
<u>Series 1: General Cases</u>				
UO ₂ -PuO ₂	∞	450	--	900
UO ₂ -PuO ₂ ^(c)	∞	160	--	900
UN-PuN	∞	65	--	900
<u>Series 2: Stacked Washers in Helium</u>				
UO ₂ -PuO ₂	0.28	30	245	1050
UO ₂ -PuO ₂	0.20	15	115	950
<u>Series 3: Stacked Washers in Liquid-Metal Bath</u>				
UO ₂ -PuO ₂	0.28	60	--	200
UO ₂ -PuO ₂	0.28	80	--	400
UO ₂ -PuO ₂	0.28	120	--	800
UO ₂ -PuO ₂	0.28	130	--	1000
<u>Series 4: Standard Specimen in Liquid-Metal Bath</u>				
UO ₂ -PuO ₂	3	235	270	800
UN-PuN	3	60	120	800
<u>Series 5: Standard Specimen in Helium</u>				
UO ₂ /UO ₂ -PuO ₂ ^(d)	3	40	400	1000

(a) Except where noted, all specimens are hollow cylinders with an OD of 0.25 in. and an ID of 0.15 in. subjected to a fission heating rate of 5×10^{13} fissions/(cm³)(sec) and a gamma heating rate of 8 w per g.

(b) D is taken as the outer diameter.

(c) ID = 0.19 in.

(d) Under BRR conditions of 1×10^{13} fissions/(cm³)(sec) and gamma heating of 0.3 w per g.

In the third case, it is shown that for a specimen material of mixed nitride (with a 50-mil wall thickness) the radial ΔT can be further reduced to 65 C.

The second series of calculations shows the axial and radial ΔT 's that will exist in the mixed-oxide material for a relatively short specimen that is surrounded by helium gas. The internal heat generated in these two cases is forced to flow in the axial direction to finned tungsten platens. A two-dimensional heat-transfer computer code was used to obtain these results.

The third series of calculations shows the ΔT 's associated with the mixed-oxide material for a stacked-washer specimen surrounded by a liquid metal. Four cases were hand calculated to show the dependency between surface temperature and ΔT . The increase in ΔT with increasing surface temperature is a result of the decrease in the thermal conductivity of the material with increasing temperature. These ΔT 's are also applicable for specimens in the shape of a spring or a long, slender beam.

The ΔT 's calculated by the computer code for the standard specimen (OD = 0.25 in., ID = 0.15 in., length = 0.75 in.) immersed in a liquid-metal bath are shown as the fourth series in the table. Again, the higher thermal conductivity of the nitride material leads to a more isothermal specimen.

For completeness, the ΔT 's associated with the low-flux experiments at the BRR are listed in the last row of the table. These ΔT 's are calculated values based on a two-dimensional solution of the BRR capsule geometry. The radial ΔT is not measured in the experiment; however, the axial ΔT is measured from the center line to about 1/16 in. in from the ends of the specimen. The axial ΔT has been measured to be from 100 to 150 C. This indicates that the majority of the axial ΔT takes place in the fuel regions close to the platens, as is predicted by the computer solution.

In view of the large number of ΔT 's that have been presented in Table C-2, some subjective comments are in order. It is clear that large thermal gradients will exist in the mixed-oxide material regardless of the test geometry or temperature at fission rates of 5×10^{13} fissions/(cm³)(sec). However, the ΔT can be minimized in the standard hollow cylinder by either reducing the wall thickness and surrounding the cylinder by liquid metal, or using a stacked-washer specimen.

Plans for Future Work

Creep Capsule BRR-5 will be fabricated in the Battelle-Columbus Capsule Laboratory and sent to the Battelle-Columbus Plutonium Facility, where a bare UO₂-PuO₂ fuel specimen will be installed in the capsule. The capsule will be operated in the BRR for its design lifetime during the next quarter.

The conceptual design for the high-flux creep capsule will be detailed, and preliminary laboratory experiments will be initiated.

In-Pile Creep Experiments

J. S. Perrin, R. A. Robinson, and S. J. Basham

Objective of Program

To generate and analyze creep data obtained from in-pile creep capsules.

Approach being Used

In-pile creep data will be generated for UO_2 and $\text{UO}_2\text{-PuO}_2$. The in-pile creep data will be analyzed both with respect to all in-pile tests and out-of-pile reference creep tests on both UO_2 and $\text{UO}_2\text{-PuO}_2$.

Progress Reported Previously

A series of creep experiments has been performed on UO_2 specimens in Creep Capsule BRR-2 and BRR-3. BMI-1872 contains a summary of results for Creep Capsule BRR-2; a summary of results for Creep Capsule BRR-3 is given in BMI-1886. The creep data from these two capsules were analyzed. At a fission rate of approximately 1.2×10^{13} fissions/(cm^3)(sec), a stress range of 1500 to 4000 psi, and a nominal temperature range of 1100 to 1200 C, an in-pile enhancement of creep rate of a factor of eight to ten was observed.

Work This Quarter

A series of out-of-pile and in-pile creep experiments was conducted in Creep Capsule BRR-4A on a UO_2 specimen. These tests are summarized in Table C-1 of the previous section. The creep rates listed in Table C-1 were determined by a statistical analysis of the creep data (within the linear steady-state region) using a regression analysis computer code. With the use of a least-squares analysis, written in ALGOL, a regression equation was deduced based on an assumed linear relation between creep and time. The regression coefficient of interest (i. e., the slope of the curve treating creep as the dependent variable and time as the independent variable) represents the creep rate.

Figure C-1 shows out-of-pile Test 4C, which was run at a temperature of 1200 C and a stress of 4000 psi. This test was stopped at 53 hr when the specimen had deformed 3.4 mils; at this point, it appeared that the specimen had not yet achieved steady-state creep, for the slope of the curve was still decreasing. The test was terminated in order to keep the out-of-pile deformation low, since these ceramic specimens only deform a moderate amount before fracture. The calculated creep rate for Test 4C is $8.22 \times 10^{-5} \text{ hr}^{-1}$; when this creep rate is compared with the creep data obtained from the other tests conducted in Capsule BRR-4A and the test run in Capsules BRR-2 and BRR-3, it is substantially higher than would be predicted from all the other creep test results. This substantiates the conclusion that the specimen was still in primary creep.

Test 4D was the first in-pile test for Capsule BRR-4A, and was run at a fission rate of approximately 4×10^{12} fissions/(cm^3)(sec). The creep curve is shown in Figure C-2. The stress was 4000 psi, the measured temperature (the temperature of the middle thermocouple on the specimen) was 1100 C. The effective temperature of an in-pile creep specimen is the temperature at which an isothermal specimen would have the same overall measured creep rate as the athermal in-pile specimen, assuming no in-pile enhancement of creep rate. The effective temperature for Test 4D is approximately 1085 C, about 15 C lower than the measured temperature. (For comparison, the effective temperature of a creep test done at the higher fission rate of 1.2×10^{13} fissions/(cm^3)(sec) is 25 C lower than the measured temperature as a result of the steeper temperature gradient.) The creep rate for Test 4D is $5.10 \times 10^{-6} \text{ hr}^{-1}$.

Tests 4E and 4F were run at the higher fission rate of 1.2×10^{13} fissions/(cm^3)(sec) at 4000 psi. Test 4E, run at an effective temperature of 1110 C, had a

FIGURE C-1. DEFORMATION AS A FUNCTION OF TIME FOR OUT-OF-PILE TEST 4C

FIGURE C-2. DEFORMATION AS A FUNCTION OF TIME FOR IN-PILE TEST 4D

creep rate of $2.66 \times 10^{-6} \text{ hr}^{-1}$. These two tests are shown in Figures C-3 and C-4.

The current creep data from Capsule BRR-4A and the data from Capsules BRR-2 and BRR-3 are shown in Figure C-5. These tests were all conducted in the effective temperature range from 1000 to 1185 C. All tests have been normalized to a temperature of 1100 C, using activation energies calculated from the out-of-pile tests and in-pile tests.

The line denoting zero fission rate in Figure C-5 has a slope of one, indicating a creep rate dependence on stress to the first power ($\dot{\epsilon} \propto \sigma$). This is consistent with other out-of-pile UO_2 creep results determined in this program, which had the same stress dependence at low stresses such as these (1500 to 4000 psi). This stress dependency is indicative of a creep mechanism such as Nabarro-Herring or some grain-boundary sliding process, rather than a creep mechanism such as dislocation climb where creep rate is proportional to stress to about the fifth power.

The top line in Figure C-5 is drawn for creep tests conducted at a fission rate of 1.2×10^{13} fissions/(cm³)(sec). The slope of the line through these points is one, i. e., $\dot{\epsilon} \propto \sigma$. Also, there is remarkably good agreement among the data from the three capsules for creep tests. This line for high fission rate indicates a consistent, substantial enhancement of the in-pile creep rate over the out-of-pile creep rate by a factor of 8 to 10. Furthermore, the in-pile enhancement due to the fission process in this temperature range is definitely not athermal. The in-pile creep rate at 4000 psi and 1000 C is $2.66 \times 10^{-6} \text{ hr}^{-1}$; at 4000 psi and 1110 C, it is $3.54 \times 10^{-5} \text{ hr}^{-1}$. This is in contradiction to theories that predict the in-pile enhancement is independent of temperature at temperatures where there is significant enhancement.

The middle line in Figure C-5 is drawn through the single test (4D) which was run at an intermediate fission rate of 4×10^{12} fissions/(cm³)(sec). There is an in-pile creep rate enhancement of slightly over two at this fission rate. The indication is that the creep rate enhancement increases linearly or slightly greater than linearly with increasing fission rate.

The previous results show that $\dot{\epsilon} \propto \sigma$. In general, for a constant fission rate,

$$\dot{\epsilon} = K \sigma e^{-Q/RT}, \quad (\text{C-1})$$

where K is a constant depending on such factors as grain size, density, etc. Equation (C-1) can be rewritten as

$$\dot{\epsilon}/\sigma = K e^{-Q/RT}, \quad (\text{C-2})$$

where $\dot{\epsilon}/\sigma$ is the stress-compensated creep rate. Figure C-6 is a plot of the stress-compensated creep rate as a function of reciprocal temperature for fission rates of 0, 4×10^{12} , and 1.2×10^{13} fissions/(cm³)(sec). It can be seen that there is a strong temperature dependence of the stress-compensated creep rate for the in-pile tests as well as for the out-of-pile tests. The activation energy for the out-of-pile tests is approximately 92 kcal per mole. The activation energy for the in-pile tests at 1.2×10^{13} fissions/(cm³)(sec) is about the same, approximately 98 kcal per mole. These results show that the curve for in-pile creep rate in this temperature range of 1000 to 1180 C is displaced above and approximately parallel to the curve for out-of-pile creep rate.

FIGURE C-3. DEFORMATION AS A FUNCTION OF TIME FOR IN-PILE TEST 4E

FIGURE C-4. DEFORMATION AS A FUNCTION OF TIME FOR IN-PILE TEST 4F

FIGURE C-5. CREEP RATE VERSUS STRESS FOR IN-PILE AND OUT-OF-PILE UO_2 TESTS SHOWING EFFECT OF FISSION RATE (ALL TESTS HAVE BEEN NORMALIZED TO 1100 C)

FIGURE C-6. STRESS-COMPENSATED CREEP RATE AS A FUNCTION OF TEMPERATURE FOR UO_2 TESTS CONDUCTED IN-PILE AND OUT-OF-PILE

Plans for Future Work

The analysis of the in-pile creep data and out-of-pile reference creep data will continue as additional data become available.

Mechanical and Thermal Analyses

W. R. D. Wilson

Objective of Program

To provide the analytical means for determining compressive creep properties of UO_2 and $\text{UO}_2\text{-PuO}_2$ from the anisothermal in-pile creep experiments so that they can be compared to the properties obtained in isothermal out-of-pile tests of the fuels, and therefore the effect of the fission process on creep can be determined.

Approach Being Used

An analysis for predicting creep behavior under anisothermal conditions based on input creep properties and specimen temperature distributions has been developed. A heat-transfer code is used to predict the temperature distributions in the test specimens under in-pile test conditions using input temperature data from thermocouples located in the capsule. When these analytical tools are used, the effect of anisothermal conditions on the creep rate of the in-pile specimen may be estimated and appropriate corrections made.

Progress Reported Previously

A computer program has been written to determine stress and strain distributions in an in-pile specimen having arbitrary creep properties and temperature distribution. In the program (which is called INPIC and is written in G. E. Time-Sharing FORTRAN) the following assumptions have been made:

- (1) The deformation is axisymmetric
- (2) Plane sections remain plane
- (3) The temperature is steady
- (4) Strains are small
- (5) Strain rate is a function of the current local stress, temperature, radiation level, and the time since loading and irradiation commenced.

The TRAHT-2 heat-transfer code has been modified to allow its use in determining the in-pile specimen temperatures. Provision has been made to convert the output temperature distribution from TRAHT-2 into a form suitable for input into INPIC via paper tape. Thus, the basic means for separating irradiation effects from those due to anisothermal conditions is available.

A parametric study of the influence of the type of creep model and temperature distribution on the magnitude of the effect of anisothermal conditions on the creep of the in-pile specimen has been made. In discussing this work, it is useful to define an "effective" specimen temperature. This is the temperature at which an isothermal specimen would have the same bulk creep rate as the anisothermal in-pile specimen under the same conditions of loading and for a given model of creep behavior. Thus, the effective specimen temperature varies with both the specimen temperature distribution and creep model used. A parametric study of

the effect of different temperature-distribution and creep models on the effective specimen temperatures indicated that for the in-pile tests at 1.2×10^{13} fissions/(cm³)(sec), the effective specimen temperature was about 25 C lower than the nominal specimen temperature measured on the midplane at the outside diameter.

The application of the 25 C correction for anisothermal conditions to the in-pile test results from Capsules BRR-2 and BRR-3 indicated that the creep rate in-pile at 1.2×10^{13} fissions/(cm³)(sec) was greater by an order of magnitude than that measured out-of-pile.

Work This Quarter

The study of the effects of anisothermal conditions has been extended to include lower fission rates. Figure C-7 shows a plot of the variation of the temperature correction with measured temperature for a fission rate of 5×10^{12} fissions/(cm³)(sec). Also included are the previously reported curves for 1×10^{13} and 2×10^{13} fissions/(cm³)(sec). It can be seen that the correction required decreases with decreasing flux, as might be expected. Extrapolation of the data presented in Figure C-7 indicated that a correction for anisothermal conditions of -15 C was appropriate in the case of Test 4D, which was run in-pile at 4×10^{12} fissions/(cm³)(sec).

FIGURE C-7. EFFECT OF FISSION RATE AND MEASURED SPECIMEN TEMPERATURE ON TEMPERATURE CORRECTION

Plans for Future Work

Analysis of the in-pile results and comparison with out-of-pile data will continue.

Fabrication of UO₂-PuO₂ Preirradiation and
In-Pile Compressive Creep Specimens

M. D. Houston, O. L. Kruger, and W. M. Pardue

Objective of Program

To fabricate and characterize UO₂-PuO₂ specimens for preirradiation and in-pile compressive creep experiments.

Approach Being Used

UO₂-22 w/o PuO₂ specimens will be fabricated at Battelle-Columbus in a manner consistent with fast-reactor fuel procedures. The process will utilize mechanically blended and coprecipitated powders with normal ceramic cold-pressing and sintering techniques. Powder compacts will be sintered in the range of 90 to 98 percent of theoretical density in a flowing atmosphere of argon-8 v/o hydrogen. The stoichiometry of the final specimens will be controlled by adjustment of the oxygen partial pressure during the sintering cycle or during an equilibration process performed at 900 to 1000 C after sintering. The stoichiometry of the mixed oxide will correspond to the stoichiometry expected in the fuel of the FTR, which is operating in the same temperature range as the creep tests. Specimens will be characterized by measurements of the following properties in accordance with well-established procedures: stoichiometry, composition (uranium/plutonium ratio), homogeneity, metallic and nonmetallic impurity content, grain size, and density.

Progress Reported Previously

This program was first reported in BMI-1851. Compressive creep specimens of UO₂-22 w/o PuO₂ were fabricated from mechanically blended Eldorado UO₂ and Arhco PuO₂ (oxalate derivative) powders and from G. E. coprecipitated UO₂-PuO₂ powder by cold-pressing and sintering techniques. Mechanical blending of the oxides was accomplished by wet ball milling for 6 to 8 hr in a Viton-lined mill with uranium slugs and oxalene. Solid cylindrical specimens were compacted at 20,000 psi in a tungsten carbide die and sintered in the temperature range from 1500 to 1760 C in argon-8 v/o hydrogen. Final material characteristics were in the ranges that follow: grain size, 3 to 40 μ ; density, 87.3 to 96.3 percent of theoretical; and oxygen/metal ratio, 1.95 to 1.98. Ultrasonic cutting techniques were developed to produce the hollow-cylinder configuration required for in-pile specimens. These specimens were nominally 0.750 ± 0.005 in. long, 0.150 ± 0.0005 in. in ID, and 0.250 ± 0.0005 in. in OD.

Work This Quarter

Fabrication studies with both mechanically blended and coprecipitated mixed-oxide materials continued. The general relationship for density-grain size as a function of sintering temperature is shown in Figure C-8. With mechanically blended UO₂-22 w/o PuO₂, the grain size increased with a slight decrease in density as the sintering temperature was increased. With coprecipitated powder, a density maximum was apparent in the temperature range from 1550 to 1700 C and the average grain size increased with increasing sintering temperature. The density decrease with the coprecipitated powder is attributed to the presence of

certain impurities in the material which volatilize at the higher temperatures. The impurity analysis is presented in Table C-3. Further evidence of this phenomenon is illustrated in Figure C-9, where large elongated voids and other associated grain-boundary porosity are found at the higher sintering temperatures. Attempts to ultrasonically machine specimens prepared from coprecipitated powder that had been sintered at 1700 to 1760 C were unsuccessful as a result of brittle fracture during cutting. Twelve specimens sintered in the range from 1500 to 1625 C were machined to the hollow-cylinder configuration.

Additional mechanically blended specimens were prepared from a new batch of UO_2 -22 w/o PuO_2 . Sintering trials with this material indicated that a slower firing cycle was required for densities in excess of 90 percent of theoretical. The normal heating rate of 500 C per hr was reduced to 175 C per hr. Specimens were sintered at 1550 and 1625 C for 18 hr to final densities of 95.0 ± 0.5 and 93.5 ± 0.5 percent of theoretical, respectively. Ultrasonic and abrasive machining of these specimens is in progress.

TABLE C-3. IMPURITY ANALYSIS OF COPRECIPITATED
 UO_2 - PuO_2 MATERIAL

Impurity	Amount of Impurity Found in Indicated Material, ppm	
	Powder	Sintered (1700 C)
Aluminum	20	30
Boron	5.2	<0.3
Bismuth	1	<1
Chromium	42	8
Copper	5	10
Iron	625	200
Lead	100	5
Magnesium	30	30
Manganese	35	22
Molybdenum	<6	<6
Nickel	20	20
Silicon	50	35
Silver	0.2	0.1
Tin	2	10
Zinc	<50	<50
Total	~890	~425

Plans for Future Work

Fabrication and characterization of UO_2 - PuO_2 specimens prepared from mechanically blended and coprecipitated materials will continue. Creep specimens fulfilling the required specifications will be prepared for preirradiation and in-pile compressive creep experiments.

FIGURE C-8. SINTERING CHARACTERISTICS FOR $\text{UO}_2\text{-22 w/o PuO}_2$ MATERIALS

250X

PL8163

FIGURE C-9. MICROSTRUCTURE OF COPRECIPITATED $\text{UO}_2\text{-22 w/o PuO}_2$ SINTERED AT 1760 C

Preirradiation Compressive Creep Testing of $\text{UO}_2\text{-PuO}_2$

M. D. Houston, O. L. Kruger, and W. M. Pardue

Objective of Program

To determine the creep behavior of unirradiated $\text{UO}_2\text{-22 w/o PuO}_2$ as a reference for the in-pile creep behavior of identical specimens.

Approach Being Used

Compressive creep tests at stresses of 1000 to 5000 psi and at temperatures of 900 to 1400 C will be performed in a vacuum furnace adapted for plutonium containment. The compressive load will be applied by a compression-cage creep-testing apparatus fabricated from TZM. Deformation will be measured continuously by a mechanical device in conjunction with an LVDT recording unit. The test specimens will have the same physical and chemical properties and dimensions as the in-pile creep specimens.

The generated data will be utilized in the selection of stresses and temperatures for the in-pile creep tests and will be compared with both the in-pile creep data on $\text{UO}_2\text{-PuO}_2$ and the out-of-pile and in-pile creep data on UO_2 .

Progress Reported Previously

The initial report for this program was BMI-1851. The compressive creep behavior of various types of $\text{UO}_2\text{-22 w/o PuO}_2$ specimens was studied. For fine-grained (3 to 5 μ , 94 percent of theoretical density) mechanically blended material, the activation energy for creep was determined to be 77 kcal per mole and the stress exponent (n) at 1210 C for stresses in the range from 1500 to 3200 psi appeared to be 1.4. Erratic behavior experienced with the fine-grained material during the long-term creep trials was attributed to thermal- and stress-induced grain growth. This phenomenon was minimized at lower temperatures and stresses, 1100 C and 1500 psi. Coarser-grained (10 to 15 μ , 90 percent of theoretical density) mechanically blended material had a lower creep rate which could be correlated with the properties of the fine-grained material by the reciprocal of the square of the grain size. Cursory results had been obtained with coprecipitated specimens (25 μ , 96 percent of theoretical density).

Work This Quarter

Compressive creep trials with the three types of $\text{UO}_2\text{-22 w/o PuO}_2$ materials continued. In the stress range from 1500 to 2500 psi at 1310 C, the stress exponent (n) for the coarse-grained mechanically blended material and the coprecipitated material was found to be near unity. Results from these and previous trials are shown in Figure C-10. It is apparent from this plot that the creep rate for the coprecipitated material is higher than the rate for the coarse-grained mechanically blended material, even though the density-grain size relationship would predict the opposite. The plot given in Figure C-11 for creep rate as a function of temperature indicates that the creep rate of the coprecipitated material is almost equivalent to that of the fine-grained mechanically blended material. The activation energy for creep for the fine-grained material was given previously as 77 kcal per mole; from the current plot, the value for the coarse-grained material is calculated to be 102 kcal per mole.

FIGURE C-10. CREEP RATE AS A FUNCTION OF STRESS FOR VARIOUS UO₂-22 w/o PuO₂ MATERIALS

FIGURE C-11. CREEP RATE AS A FUNCTION OF TEMPERATURE FOR VARIOUS UO₂-22 w/o PuO₂ MATERIALS (O/M = 1.98) UNDER A COMPRESSIVE STRESS OF 2500 PSI

a. Contact End Showing Pressure Cone Extending in From Outside Corner at 45 Deg

b. Tensile-Boundary Separation

FIGURE C-12. MICROSTRUCTURE OF LARGE-GRAINED COPRECIPITATED UO_2 -21.77 w/o PuO_2 HOLLOW CYLINDER AFTER CREEP TESTING TO A STRAIN OF 2.06 PERCENT

Sintered specimens from coprecipitated material have performed differently than expected in regard to both fabrication (as discussed previously) and creep behavior. To date, a total of three such specimens in two configurations, hollow and solid, have been tested and all have proceeded into tertiary creep with approximately 2 percent total strain. Unlike the UO_2 -22 w/o PuO_2 mechanically blended specimens studied, these coprecipitated specimens have a very short tertiary range before fracture and complete collapse. Fracture occurred with less than 1 percent additional strain. The microstructural details of a hollow cylindrical specimen at 2.06 percent strain are shown in Figure C-12. The creep test had been terminated in the tertiary region prior to fracture. A pressure cone is apparent in the low-magnification view. This appears as a line extending inward at approximately 45 deg and follows closely the density pattern exhibited in powder compacts. Regions outside the conical configuration show a parting of the tensile boundaries as the mechanism leading to fracture. This phenomenon at a fairly low strain may be associated with the initial impurity content and its subsequent behavior during sintering. That is, the impurity depletion as a gaseous phase during the sintering process may leave the grain boundaries in a weakened condition. Further details are necessary to specify the particular cause.

Plans for Future Work

Compressive creep testing of UO_2 -22 w/o PuO_2 specimens in the temperature range from 900 to 1400 C and with stresses of 1000 to 5000 psi will continue.

Mechanisms of Creep in UO_2 and UO_2 - PuO_2

M. S. Seltzer, A. H. Clauer, and B. A. Wilcox

Objective of Program

To provide a method for predicting the results of neutron irradiation on the compressive creep behavior of UO_2 and UO_2 - PuO_2 by combining a program of creep experiments on unirradiated materials with an analysis of the known effects of the fission process on the properties and microstructures of these materials.

Approach Being Used

The experimental phase of the program involves studies on the creep of unirradiated single-crystal and polycrystalline UO_2 as influenced by (1) temperature, (2) compressive stress, (3) stoichiometry, and (4) microstructure, including grain size and voids (porosity). Tests are being performed in compression in a controlled-atmosphere creep apparatus, using cylindrical specimens prepared by ultrasonic machining. Creep properties are being examined in light of structural changes as revealed by transmission electron microscopy and etch pitting.

The theoretical analysis includes two phases: (1) an analysis of creep data gathered in this program to ascertain the modes and mechanisms of creep in UO_2 , and (2) a collaborative effort with other task scientists to examine the known effects of the fission process on those properties of UO_2 and UO_2 - PuO_2 which may influence creep rates in these materials, and from this survey to make predictions about the effects of neutron irradiation on the creep behavior of UO_2 and UO_2 - PuO_2 .

Progress Reported Previously

Studies of the influence of stress, temperature, and stoichiometry on the creep behavior of single-crystal uranium dioxide were completed. Values for creep rates of UO_{2+x} and the activation energies for high-temperature creep of this material were found to be strongly dependent upon the oxygen/uranium ratio of the specimens tested. In the power-law stress regime (10,000 psi) the activation energy, Q , decreases from 135 kcal per mole to 57.5 kcal per mole as the oxygen/uranium ratio increases from 2.0001 to 2.062. Over this same range of compositions creep rates increase by some three to five orders of magnitude in the temperature range of 1100 to 1300 C. This dependence of creep rates on oxygen/uranium ratio can be expressed as $\dot{\epsilon} \propto x^2$ for values of x above 0.003.

At lower stresses, the upper limit of which depends on temperature and composition, creep rates for UO_{2+x} single crystals are found to decrease linearly with decreasing stress. At a typical stress in this linear stress region (3000 psi) creep rates appear to be nearly independent of stoichiometry and the creep activation energy has a value of approximately 130 kcal per mole, nearly identical to that found in the power-law stress region for "stoichiometric" uranium dioxide single crystals.

Some transmission electron microscopy has been performed on nonstoichiometric UO_{2+x} specimens deformed about 10 percent in compression. Many features of these unirradiated specimens resemble features which have previously been attributed to neutron irradiation.

Work was initiated to study the effects of temperature, stress, grain size, density, and stoichiometry on the creep polycrystalline uranium dioxide. At 1100 C, creep rates were found to increase with increasing stress and with increasing oxygen/uranium ratio and to increase with decreasing grain size. For specimens having a grain size of 6 to 7 μ and a density of 95 percent of theoretical, creep rates increase linearly with stress for low stresses ($\dot{\epsilon} \propto \sigma$) and follow a power-law dependence where $\dot{\epsilon} \propto \sigma^4$ at high stresses. Creep tests with specimens having a 4- μ grain size and a density of 92 percent of theoretical show only the linear stress dependence. In the linear stress region $\dot{\epsilon} \propto x$, while in the power-law stress region $\dot{\epsilon} \propto x^{1.75}$, approximately the same dependence on stoichiometry as was found for single-crystal creep. For the specimens with a 6 to 7- μ grain size, the stress at which the transition in stress dependence occurs decreases from 20,000 psi to 5,500 psi as the oxygen/uranium ratio increases from 2.001 to 2.05.

Work This Quarter

Tests were conducted this quarter to establish the influence of stress, temperature, and composition on the creep behavior of enriched polycrystalline UO_2 specimens similar to those employed in the in-pile compressive creep studies. This information will be used to interpret in-pile effects and to serve as a measure of specimen composition for experiments conducted in-pile.

The procedures used to fabricate the high-density UO_2 creep specimens were described in detail in BMI-1868. The final sintering operation consisted of pressing the 24.48 percent enriched powder at 10 tsi and 1700 C for 15 hr in flowing dry hydrogen. Cylindrical specimens 0.315 in. high and 0.152 in. in diameter were ultrasonically trepanned from the sintered rods, which had a density of

97.8 $\pm$ 0.5 percent of theoretical and an average grain size of 27 μ . The UO₂ specimens had an initial oxygen/uranium ratio of 2.002 $\pm$ 0.003, with iron and silicon being the major impurities at 100 and 150 ppm, respectively.

Results obtained from creep tests conducted at 1300 C are included in Figure C-13, in which steady-state creep rates, $\dot{\epsilon}$, are plotted versus stress, σ . Tests at constant oxygen/uranium values of 2.001, 2.01, and 2.10 were performed by equilibrating the creep specimens in flowing CO/CO₂ gas mixtures and maintaining the desired oxygen partial pressure as the test progressed. Also presented in this figure are data obtained by Stellrecht and Moak (see BMI-1868) under vacuum using the same enriched material as utilized in this study.

Somewhat more limited data obtained at 1100 C are included in Figure C-14. The creep rates for oxygen/uranium ratios of 2.001, 2.01, and 2.10 were determined in this study. The value for the test performed in vacuum was determined by Stellrecht and Moak (see BMI-1868), and the creep rates obtained in a helium atmosphere were established in Creep Capsules BRR-2 and BRR-3 at zero flux (see BMI-1886).

The data for an oxygen/uranium ratio of 2.10 at 1300 C (Figure C-13) best illustrate how the stress dependence for steady-state creep varies with stress. At the lowest stresses used, 150 to 500 psi, the creep rates increase linearly with stress, but this dependence increases with increasing stress so that above 3000 psi creep rates are best fit by a power-law stress dependence of the form $\dot{\epsilon} \propto \sigma^n$, where $n = 7$. These results are, of course, similar to those obtained by other investigators⁽¹⁻³⁾, where the high stress dependence suggests a dislocation creep mechanism based on diffusion-controlled dislocation climb or some other diffusion-controlled process while the linear stress dependence indicates the predominance of Nabarro-Herring diffusional creep or some grain-boundary sliding process. For the lower oxygen/uranium ratios at 1300 C and in all cases at 1100 C, the linear stress dependence was not observed because creep rates were too low at the stress levels required to reach this regime.

The absolute values for the creep rates at a given temperature, stress, oxygen/uranium ratio, and grain size are several orders of magnitude larger than those reported by investigators who studied creep of uranium dioxide containing natural or depleted uranium⁽¹⁻³⁾. This appears to be related to the high silicon content of the enriched uranium dioxide. The influence of silicon at grain boundaries on the creep of uranium dioxide has been investigated in some detail by Goodyear, Speidel, and Kizer (see BMI-1868); scanning electron microscopy was utilized to show that very small amounts of a phase containing silicon were present at the grain boundaries of the enriched UO₂ specimens used in this investigation. If this silicon exists as a glassy phase such as SiO₂, it might be expected to appreciably weaken the uranium dioxide creep specimens. In an early study, Armstrong and Irvine⁽⁴⁾ showed that deflection rates from bending experiments were higher for UO₂ containing SiO₂ as compared with undoped UO₂.

Although the absolute values for creep rates obtained from the enriched specimens are not in agreement with data from natural or depleted uranium-bearing specimens, the observed dependence for creep rates of enriched UO₂ on composition is similar to that found for the unenriched specimens. Furthermore, the activation energies for creep of enriched, polycrystalline uranium dioxide are similar to those for creep UO_{2+x} single crystals. Figure C-15 shows the creep activation energies for UO_{2+x} single crystals obtained previously on this program

FIGURE C-13. STEADY-STATE CREEP RATES OF ENRICHED UO_{2+x} SPECIMENS TESTED AT 1300 C UNDER VARIOUS APPLIED STRESSES

FIGURE C-14. STEADY-STATE CREEP RATES OF ENRICHED UO_{2+x} SPECIMENS TESTED AT 1100 C UNDER VARIOUS APPLIED STRESSES

(see BMI-1886) as a function of $\log_{10} x$. Superimposed on these data are the activation energies for creep of enriched polycrystalline UO_{2+x} as determined from the high-stress portions of the data given in Figures C-13 and C-14 for oxygen/uranium ratios of 2.001, 2.01, and 2.10. These values are 117, 75, and 54 kcal per mole. The good agreement between activation energies determined from single crystals and polycrystalline material suggests that the same creep mechanism is operative in both cases. Since the single crystals are free of grain boundaries, the implication is that creep in the high-stress region is controlled by uranium volume diffusion. There are too few data available to establish creep activation energies in the region where creep varies linearly with stress. It is hoped that this information will be obtained from tests with fine-grained uranium dioxide.

FIGURE C-15. ACTIVATION ENERGIES FOR CREEP OF UO_{2+x} AS A FUNCTION OF $\log_{10} x$

The dependence of creep rates on oxygen/uranium ratio is also similar to that found for creep single crystals. At 1100 C, $\dot{\epsilon} \propto x^2$ in the high-stress region, while at 1300 C, the value for the exponent is between 1 and 2. In the low-stress region the stoichiometry dependence is not as well defined; at 1300 C and 1000 psi, $\dot{\epsilon}_{2.10}/\dot{\epsilon}_{2.01} \approx 4.5$ and $\dot{\epsilon}_{2.01}/\dot{\epsilon}_{2.001} \approx 2.5$.

The creep rates obtained by Stellrecht and Moak in a vacuum apparatus are uniformly lower than those obtained in this study under conditions of controlled stoichiometry. On the basis of the controlled-composition data, an oxygen/uranium ratio of 2.0001 can be assigned to the specimens tested by Stellrecht and Moak. Such a low oxygen/uranium ratio is not unreasonable in view of the fact that the hot portion of the creep furnace used for the vacuum tests contains tantalum, molybdenum, and tungsten, all oxygen getters; this would tend to keep the oxygen partial pressure surrounding the creep specimen very low. Some of the scatter in the vacuum creep data may be attributed to variations in stoichiometry among the specimens tested in this system.

By using the controlled-stoichiometry data of Figure C-14 as a measure of specimen composition, an oxygen/uranium ratio of approximately 2.0003 can be assigned to the UO_2 specimens tested in the in-pile creep capsules prior to irradiation. This is a slightly higher oxygen content than is found for specimens tested in the vacuum furnace, but it is nearly equivalent to the oxygen concentration for "stoichiometric" uranium dioxide.

Plans for Future Work

During the next quarter, experiments will be conducted to determine the activation energies for creep as a function of composition for polycrystalline hyperstoichiometric uranium dioxide in the linear-stress-dependence region. Later experiments will be conducted with specimens of varying density and grain size to establish the effects of stress, temperature, and composition on specimens in the density range from 84 to 97 percent of theoretical and having grain sizes from 1 to 30 μ .

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