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IRRADIATION BEHAVIOR OF BeO-UO_2 FUEL AS A FUNCTION OF FUEL-PARTICLE SIZE

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METALS, CERAMICS, AND MATERIALS

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ABSTRACT

One capsule containing BeO-30 vol-% UO_2 fuel pellets was irradiated in the Materials Testing Reactor to a uranium burnup of approximately 30,000 Mwd/ton. The results of a microscopic examination and of physical measurements that were made during the hot-cell examination of the capsule are described. Some of the implications of these observations are discussed.

I. INTRODUCTION

Many of the factors associated with the use of dispersion-type reactor fuel materials have been discussed in the literature.¹⁻⁴ Some of these factors are (1) increases in the specific power capabilities of the fuel owing to the increased thermal conductivity of the fuel-matrix composite, (2) improved mechanical performance of the composite owing to isolation of the regions in the matrix that are affected by fission-fragment recoils, and (3) reduced fission-product release due to diffusion owing to the decreased average fuel temperature relative to that for solid fuel compounds such as UO_2 . The release of fission products is also minimized by isolation of the fuel particles within an envelope of "impermeable" matrix material.

Some of the early results reported by Yeniscavich and Bleiberg⁵ on the use of UO_2 dispersed in BeO as a reactor fuel indicated that dimensional changes and fission-gas release were substantially greater in fuel bodies that contained the UO_2 in the form of a dispersion of fine (0 to 50 μ) particles than in fuel bodies that contained the UO_2 in the form of a dispersion of coarse ($\sim 150 \mu$) particles. However, these observations were made on fuel bodies that had been irradiated at relatively low average temperatures (440° to 500°C).

In order to determine if the results obtained by Yeniscavich and Bleiberg are representative for high-temperature applications of similar fuels, a program was undertaken at General Atomic as a part of the Experimental Beryllium Oxide Reactor Program to study the relative high-temperature performance of BeO- UO_2 fuel materials containing 30 vol-% UO_2 with the UO_2 present in the form of fine ($< 25 \mu$) and coarse ($\sim 150 \mu$) particles. The irradiation of two capsules containing representative fuel specimens made with the two types of particles was included in this program. The purpose of this paper is to describe the observations that were made

during the post-irradiation examination of the BeO-UO₂ specimens from the first of the two irradiation capsules.

II. MATERIALS AND EQUIPMENT

MATERIALS USED IN FABRICATION OF THE FUEL PELLETS

The fuel pellets used in this irradiation experiment were composed of 70 vol-% BeO-30 vol-% UO₂ and were fabricated at General Atomic. The chemical compositions of the oxide powders used in the fabrication of the fuel pellets are presented in Table I. The pellets were fabricated by dry-pressing in a steel die and subsequently sintering in a molybdenum-wound tube furnace, using an atmosphere of 25% hydrogen and 75% nitrogen. Following the sintering process, the pellets were ground to the required dimensions: approximately 0.4 in. in diameter and 0.5 in. long. The required uranium enrichment of 12 atom-% U²³⁵ was obtained by blending natural-enrichment UO₂ powder and fully enriched UO₂ powder.

Two types of specimens were fabricated. One type contained the UO₂ in the form of a homogeneous dispersion of fine particles. Metallographic examination of a polished cross section of a typical fuel specimen indicated that the UO₂ was present in the form of somewhat irregularly shaped particles with maximum dimensions of approximately 25 μ . The second type of fuel pellet contained the UO₂ in the form of larger particles with maximum dimensions of approximately 150 μ . A comparison of the two types of fuel pellets (see Fig. 1) indicates that in the pellets containing coarse fuel particles, the individual particles are readily discernible by visual inspection, whereas the pellets containing the fuel in the form of fine particles have a uniform, dark-brown appearance. Figure 2a is a photomicrograph of a fine-particle fuel pellet, and Fig. 3 is a photomicrograph of a coarse-particle fuel pellet. From these figures, it may be observed that the distribution of the fuel particles in the BeO matrix was uniform. The densities of the pellets before irradiation are presented in Table II.

Table I

COMPOSITION OF OXIDE POWDERS USED IN THE FABRICATION
OF FUEL PELLETS FOR THE MGCR-9 CAPSULE

	Typical Composition (ppm)		
	BeO ^a	UO ₂ (Normal) ^b	UO ₂ (93.24% U ²³⁵) ^c
U		87.32%	87.9%
O/U ^d		2.16	
BeO	99+%		
MgO	50	17	60
Al ₂ O ₃	100		70
SiO ₂	100	100	300
CaO	50	70	10
Fe ₂ O ₃	20	100	140
Na ₂ O	50		
Cr	10	<10	50
Ni	20	<10	50
Co	5		<1
Cu	5	<3	20
Mn	10	<10	<10
Li	1		
B	3	<0.2	1
Cd	2		0.1
Rare earths			<10
Pb		<2	
Sn		<2	<5
V		<1	<10
P			<10
Zn			0
Ba		2	<10

^aUOX grade, Brush Beryllium Company.

^bCeramic grade, Nuclear Materials and Equipment Corp.

^cCeramic grade, Davison Chemical Company.

^dAtomic ratio of oxygen to uranium.

Fig. 1--Photomacrograph of typical BeO-30 vol-% UO_2 fuel pellets used in the MGCR-9 capsule, containing coarse UO_2 particles (left) and fine UO_2 particles (right)

M-754-1-2

100 μ

(a) As-polished cross section of a typical BeO-30 vol-% UO_2 fuel pellet containing fine ($<25 \mu$) UO_2 fuel particles (250 $\times$)

M-754-2-2

100 μ

(b) As-polished cross section of a typical BeO-30 vol-% UO_2 fuel pellet containing coarse ($\sim 150 \mu$) UO_2 fuel particles (250 $\times$)

Fig. 2--Pre-irradiation photomicrographs of typical fuel pellets used in the MGCR-9 capsule

M-754-2-1

250 μ

(a) As-polished cross section
(100 $\times$)

M-754-2-2

100 μ

(b) As-polished cross section
(250 $\times$)

M-754-2-5

100 μ

(c) Polished and etched cross
section illustrating the BeO
grain structure (250 $\times$)

Fig. 3--Pre-irradiation photomicrographs of typical coarse-particle BeO-30 vol-% UO₂ fuel pellets used in the MGCR-9 capsule

Table II
PRE- AND POST-IRRADIATION DENSITY OF FUEL SPECIMENS
FROM THE MGCR-9 CAPSULE

Specimen No.	Pellet No.	Pre-irradiation Bulk Density		Post-irradiation Bulk Density		Change in Density (%)
		g/cm ³	% Theo.	g/cm ³	% Theo.	
1	F-106	5.16	95.6	4.93	91.3	-4.5
	F-107	5.12	94.8	5.00	92.6	-2.3
4	F-108	5.21	96.5	5.03	93.1	-3.5
	F-109	5.02	93.0	----	----	----
2	F-110	5.18	95.9	----	----	----
	F-111	5.13	95.0	----	----	----
3	C-124	5.05	93.5	4.80	88.9	-4.5
	C-125	5.02	93.0	----	----	----
5	C-126	5.04	93.3	4.91	90.8	-2.4
	C-127	5.03	93.1	----	----	----
6	C-128	5.06	93.7	----	----	----
	C-129	5.02	93.0	----	----	----

DESCRIPTION OF THE IRRADIATION CAPSULE (MGCR-9)

The capsule used in the irradiation of the fuel pellets was fabricated at Battelle Memorial Institute. The general features of this type of capsule have previously been described,⁶ although some of the details of the capsule construction were changed slightly for this irradiation. Figure 4 is a photograph of some of the components used in the construction of the capsule, and Fig. 5 is a cross-sectional view of the capsule through the region where the fuel pellets were located.

In this capsule the fuel specimens were clad with Hastelloy-X. Each of the Hastelloy-X fuel claddings was back-filled with helium prior to making the final weld closure. Two fuel pellets were contained in each compartment and comprised a single specimen. A total of six specimens were included in the capsule. The thermal bond between the Hastelloy-X fuel cladding and the inner capsule can was NaK. Thermocouples were inserted in the NaK thermal bond with the hot junctions located adjacent to each fuel specimen in order to determine the temperatures that existed during the irradiation. An electrical resistance heater was brazed to the outside of the inner capsule can to provide a measure of control of the temperature in the capsule during the irradiation.

III. EXPERIMENTAL PROCEDURE

The capsule was designed for irradiation in the Materials Testing Reactor in position A-43-NW at a fuel-pellet surface temperature of approximately 1700°F and an estimated average fuel temperature of 2400°F. This position received an estimated unperturbed thermal-neutron flux of approximately 5×10^{13} nv. Physics calculations indicated that the effective flux in the fuel pellets was approximately 2×10^{13} nv. The irradiation of the capsule began with MTR cycle No. 165 and continued for the equivalent of 7020 hr at full reactor power. The capsule underwent a total of 89 thermal cycles during the irradiation. Of the original six thermocouples in the capsule,

Fig. 4--Photograph of components used in the MGCR-9 capsule

Fig. 5--Section of MGCR-9 capsule assembly (full scale)

four continued to operate in a normal manner throughout the irradiation period, one became unreliable after six cycles of irradiation, and one became unreliable after eight cycles of irradiation. Electrical power to the resistance heater in the capsule was used throughout the irradiation as a shim control of the capsule temperature.

At the end of the irradiation period the capsule was shipped to the General Atomic hot cell where it was disassembled using conventional hot-cell procedures. The densities of selected specimens were measured by a liquid-displacement technique. The fission gases that were generated inside the individual fuel claddings during the irradiation were collected and quantitatively analyzed using gamma-spectrometry techniques. Selected specimens were prepared for microscopic examination by normal polishing methods. Photomicrographs were taken using a Bausch and Lomb research metallograph that had been specially adapted for hot-cell use. The nickel-cobalt dosimeters from the capsule were analyzed by radiochemical methods to determine the integrated neutron fluxes and the U^{235} burnup in the capsule.

IV. RESULTS

The results of the analysis of the fission gas released during the irradiation are presented in Table III. It was found that only one of the specimens containing fuel pellets with fine-dispersion UO_2 released measurable quantities of Kr^{85} . The fuel specimens containing coarse-particle UO_2 released between 17% and 24% of the Kr^{85} during the irradiation. The amount of Kr^{85} released during irradiation is plotted in Fig. 6 as a function of specimen position in the capsule, fuel-particle size, and irradiation temperature.

Density changes observed for selected samples are presented in Table II and are plotted in Fig. 7 as a function of UO_2 particle size, specimen position in the capsule, and total fission density in the individual specimens. Density changes in the range of 2.3% to 4.5% were observed for pellets

Table III
FRACTIONAL RELEASE OF Kr⁸⁵

Specimen No.	Fuel-particle Size	Fission Density (f/cm ³ ×10 ⁻²⁰)	Total Fissions (×10 ⁻²⁰)	Kr ⁸⁵ Atoms Formed (×10 ⁻¹⁸)	Fraction Released
4	Fine	2.01	3.95	1.16	N. D. ^a
1	Fine	2.19	4.35	1.27	N. D. ^a
3	Coarse	2.28	4.42	1.30	0.242
6	Coarse	2.28	4.40	1.29	0.176
2	Fine	2.07	4.08	1.20	0.003
5	Coarse	2.01	3.90	1.14	0.232

^aNone detected.

Fig. 6--Fission-gas release and fuel-surface temperature as a function of specimen position and fuel-particle size

Fig. 7--Density change and irradiation flux as a function of specimen position and fuel-particle size

containing both fine and coarse-particle fuel. The maximum fission density obtained in the capsule was 2.28×10^{20} fissions/cm³ (see Fig. 7 and Table IV).

Following the irradiation, photomacrographs of selected pellets were taken. Some of the results are presented in Fig. 8, where part (a) of the figure shows a fuel pellet in its Hastelloy-X cladding and parts (b) and (c) show two views of one of the fragments from the same fuel pellet. Photomicrographs illustrating the appearance of one of the UO₂ particles and of the BeO matrix from a pellet containing the UO₂ in the form of coarse particles are presented in Fig. 9. Similar photomicrographs for selected fine-particle fuel specimens are presented in Fig. 10.

V. DISCUSSION

The objective of this experiment was to provide some initial data on the relative performance of BeO-30 vol-% UO₂ fuel bodies with the UO₂ present in the form of fine (<25 μ) particles and coarse (~150 μ) particles. Results obtained by Yeniscavich and Bleiberg⁵ in a similar experiment where the fuels were irradiated at average temperatures below 1000°F indicated that the fine-particle fuels exhibited substantially larger dimensional change and fission-gas release than the coarse-particle fuels. In their experiment, the fuel loading was less than 30 wt-% UO₂ as compared with approximately 61 wt-% UO₂ in the present experiment. Yeniscavich and Bleiberg observed fission-gas releases as high as approximately 20% for the fine-particle fuels as compared with 7% fission-gas release for the coarse-particle fuel. Dimensional-change data indicated even more marked differences in the performance of the two types of fuel material.

In some of the early fuel-irradiation work conducted at General Atomic as a part of the Experimental Beryllium Oxide Reactor Program, fuel pellets containing 20 vol-% UO₂ and having a density of approximately 89% of theoretical density released less than 0.5% of the Kr⁸⁵ formed during the irradiation.² In that case the fuel-cladding surface temperature was

Table IV
DOSIMETRY DATA FOR THE MGCR-9 CAPSULE

Specimen No.	Fuel- particle Size	Fission Density (f/cm ³ ×10 ⁻²⁰)	Total Absorptions in U ²³⁵ (×10 ⁻²⁰)	Integrated Fast Flux (nvt×10 ⁻²⁰ >1 Mev)	Integrated Thermal Flux (nvt×10 ⁻²⁰)
4	Fine	2.01	2.37	2.00	4.79
1	Fine	2.19	2.58	2.24	5.22
3	Coarse	2.28	2.69	2.36	5.46
6	Coarse	2.28	2.68	2.36	5.42
2	Fine	2.07	2.44	2.08	4.94
5	Coarse	2.01	2.37	2.00	4.79

M-1151

(a) Irradiated BeO-30 vol-% UO_2 fuel pellet as mounted and polished in its Hastelloy-X cladding (6 $\times$)

M-1155

(b) Fragment of an irradiated BeO-30 vol-% UO_2 fuel pellet illustrating the nature of fracturing that occurred during irradiation (6 $\times$)

M-1153

(c) End view of the fuel-pellet fragment shown in (b) at left (6 $\times$)

Fig. 8--Photomacrographs of irradiated BeO-30 vol-% UO_2 fuel pellets from the MGCR-9 capsule; fuel-particle size of $\sim 150 \mu$

M-0869

50 μ

(a) As-polished pellet (500 $\times$)

M-0867

50 μ

(b) Another area of the pellet shown at the left, illustrating the appearance of the BeO matrix (as-polished, 500 $\times$)

Fig. 9--Photomicrographs of a BeO-30 vol-% UO_2 fuel pellet after irradiation to $\sim 2 \times 10^{20}$ fissions/cm³; the UO_2 was present as coarse ($\sim 150 \mu$) particles

M-0864

100 μ

(a) Unirradiated pellet (250 $\times$)

M-0890

100 μ

(b) As-polished irradiated
pellet (250 $\times$)

M-0897

50 μ

(c) As-polished irradiated
pellet (500 $\times$)

Fig. 10--Photomicrographs of BeO-30 vol-% UO_2 fuel pellets before and after irradiation to $\sim 2 \times 10^{20}$ fissions/cm³; fine (<25 μ) fuel particles

approximately 1550°F, and the average fuel-pellet surface temperature was estimated to be in the range of 2000° to 2300°F. The UO_2 particles were present in the fuel bodies in the form of approximately 125- μ particles. More recent results obtained at General Atomic on the irradiation performance of BeO-UO_2 fuel pellets indicate excellent dimensional stability to high burnups of the uranium when the UO_2 is present in the form of approximately 125- μ particles. As before, the fuel temperatures were estimated to be in the range of 2000° to 2300°F.⁴

The irradiation performance of BeO-UO_2 fuel pellets containing 70 and 80 wt-% UO_2 in the form of fine-particle dispersions has been reported by Titus and Saling.⁷ They found that bodies containing both 70 and 80 wt-% UO_2 exhibited minor dimensional changes and, in general, a relatively small release of Kr^{85} (<3%). Of this gas release, approximately 0.25% could be accounted for on the basis of recoil.

In view of the results obtained previously at General Atomic and at other laboratories, the magnitude of the observed density changes and the quantities of Kr^{85} released in the present irradiation from the fuel specimens containing coarse-particle UO_2 is surprising. While dimensional changes were not determined in the present irradiation experiment, they can be inferred from density measurements, and indicate linear dimensional increases of approximately 1.5%. This dimensional behavior is in good agreement with results obtained in earlier irradiations at General Atomic and at other laboratories.

One of the objectives of using a material such as BeO as the matrix for reactor fuels is to provide a relatively impermeable path through which the fission products must migrate before they escape from the fuel bodies. Density-permeability relationships for BeO have been reported by British workers.⁸ In their study, it was found that a marked change in the permeability of BeO occurred at densities between 90% and 95% of theoretical density. Above 95% of theoretical density the BeO was very nearly impermeable to gases. In the present experiment, the starting densities of the fuel pellets

were 94% to 95% of theoretical density. These values decreased to approximately 90% of theoretical density in some cases during the irradiation. Fuel pellets containing fine-particle UO_2 and fuel pellets containing coarse-particle UO_2 exhibited similar changes in density. In view of the measured gas-permeability-density relationship for BeO, the observed fission-gas release behavior for the two kinds of fuels irradiated in this experiment does not appear to be consistent.

A comparison of the microstructure of the irradiated pellets with that of unirradiated pellets indicates that for the fine-particle UO_2 material, some agglomeration of the UO_2 particles appears to have occurred during irradiation. In general, the appearance of the fine-particle UO_2 materials from this experiment is very similar to that reported for UO_2 by Titus and Saling.⁷ No marked change in the structure of the BeO was observed. For the fuel pellets containing the UO_2 in the form of both coarse and fine particles, it was observed that many UO_2 particles were removed from the matrix during the polishing operation. Furthermore, pieces of material appeared to have been pulled out of the sample from the region of the boundary between the UO_2 particles and the BeO matrix. Examples of this behavior, which was rather pronounced, are illustrated in Figs. 9 and 10. A comparison of the structure of the BeO from the irradiated pellets with that of the BeO in the pellets before the irradiation may be made by referring to Figs. 2, 9, and 10. The occurrence of extensive grain-boundary separation such as that reported⁹ to occur in BeO during low-temperature irradiation was not observed by optical-microscopy techniques. (Electron-microscopy examination of the pellets has not been performed.) On the basis of results from the optical examination of the microstructure of the BeO, it does not appear that marked changes occurred during the irradiation.

VI. SUMMARY AND CONCLUSIONS

The results of the irradiation experiment may be summarized as follows:

1. Fuel pellets composed of BeO-30 vol-% UO_2 were irradiated to

integrated exposures of approximately 2×10^{20} fissions/cm³. Fuel pellets containing the UO₂ as fine (<25 μ) particles and coarse ($\sim 150 \mu$) particles were included in the experiment. The estimated fuel-surface temperatures were approximately 1700°F.

2. As inferred from the results of density measurements, the dimensional changes in the pellets did not appear to vary as a function of UO₂ particle size, and represented an increase of the order of 1% to 1.5%.

3. Measurements of the quantity of Kr⁸⁵ fission gas that was collected inside the metallic fuel-pellet claddings during the irradiation indicated that approximately 1% of the Kr⁸⁵ was released from fuel pellets containing fine-particle UO₂, while approximately 20% of the Kr⁸⁵ was released from fuel pellets containing coarse-particle UO₂.

4. Major changes were not observed in the microstructure of either type of fuel pellet included in the test. Pull-out in the neighborhood of the fuel particles during the preparation of specimens for microscopic examination appeared to be more pronounced as a result of the irradiation.

The conclusions reached from this irradiation experiment are:

1. The results obtained on fission-gas release from the two different types of fuel tested do not appear to agree with results obtained in previous irradiation experiments at several laboratories or with the results that would be predicted on the basis of theory.

2. Changes that occurred in the density and microstructure of the BeO-UO₂ fuel pellets during irradiation indicate that the performance of such pellets should be satisfactory for EBOR-type applications.

3. Additional experiments are required to obtain information that will assist in developing an explanation for the fission-gas release behavior that was observed in the present experiment. In this connection, it should be noted that a second capsule containing similar fuel pellets was irradiated in the Materials Testing Reactor to an exposure approximately 1-1/2 times that obtained in this irradiation experiment, and that the examination of specimens from the second capsule is presently under way in the General Atomic hot cell.

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