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HIGH BURNUP PLUTONIUM OXIDE IRRADIATIONS^(a)

MASTER

J. A. Christensen

Battelle Memorial Institute, Pacific Northwest Laboratory
Richland, Washington^(b)

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Bulk plutonium oxides were irradiated to greater than 50% burnup in MTR under conditions shown in Table I.

Table I - Materials and Irradiation Parameters

Specimen GEH-14-

Material	PuO ₂ 93% T.D.	PuO ₂ 93% T.D.	PuO ₂ 93% T.D.
Sample Size	7 mm dia. x 1.6 mm thick	7 mm dia. x 1.6 mm thick	5 mm square x 1.6 mm thick
Burnup, %Pu Fissioned	55	9	38
Fuel Surface Temp., °C	<350	<350	<350

The irradiation capsule and typical pre-irradiation PuO₂ metallography are shown in Figures 1 and 2.

Postirradiation examination consisted of metallography, autoradiography, and x-ray diffraction analyses for new phases and lattice parameters. Results, as follows, are encouraging in terms of the slight effects on crystal structure. The ability of the oxide to creep extensively at these low temperatures is surprising and may dictate more concern over dimensional stability in the cooler regions of fuel pins.

GEH-14-411 (PuO₂, 55% Burnup)

This was the highest exposure material examined. Calculations indicated that approximately 55% of the Pu should have fissioned. Some fuel cracking and porosity but no significant non-uniformities in Pu or fission fragment distribution were observed. Substantial bulk fuel movement occurred

FIGURE 5.3. High Burnup Plutonium Oxides Irradiation Capsule

100 μ

²
FIGURE 5.4. PuO₂ Preirradiation Microstructure

even though the capsule offered relatively high restraint. This occurred by plastic flow of the oxide as a thin sheet between mating aluminum pieces at the specimen ends. Flow occurred to a diameter approximately 50% greater than that of the original specimen (Figure 3). The prominent microstructure features observed were uniformly distributed large bubbles and a white second phase (Figure 4). No residual fabrication porosity was observed. Bubbles ranged in size to 10 μ and were irregular and had rougher surfaces than expected for fission-gas bubbles. Surprisingly, also, bubble regularity seemed directly proportional to size, with large bubbles being nearly spherical and <1 μ bubbles having random, equiaxed shapes. A white second phase was distributed uniformly in the matrix and in many areas formed a coating on the void surfaces. Although several percent of the microstructure consists of white inclusions, which are presumably fission-fragment metals, none were large enough to show on the β -autoradiographs. An X-ray diffraction pattern showed a well defined PuO_2 fluorite structure with no extraneous lines. The lattice parameter had increased 0.56% over that of the nonirradiated material.

GEH-14-412, PuO_2 , 2.2×10^{12} f/cc

This material was similar to the higher burnup oxide previously discussed except that void and inclusion fractions were lower and void sizes ranged to approximately 1 μ , i.e., one-tenth the maximum void size in Specimen 411 (Figure 5). The X-ray diffraction pattern showed only PuO_2 with a lattice expansion of 0.22% relative to the unirradiated oxide.

GEH-14-413, Pu_2O_3 , 9.0×10^{21} f/cm³

This material was irradiated with essentially zero restraint. The swelling that occurred deformed the originally square specimen to the distorted globular mass shown in Figure 6. As with the PuO_2 , no nonuniformities in plutonium

FIGURE 5.5. Plastic Behavior of PuO_2 During Irradiation to $12.9 \times 10^{21} \text{ f/cm}^3$; Material Extruded Between Mating Aluminum Pieces as a Thin Sheet 50% greater in Diameter than the Fuel Pellet. Temperature 350°C

*FIGURE 5.6. Typical Irradiated PuO₂ Microstructure
2.2 × 10²¹ f/cm³*

or fission-fragment concentration were observed. Fission-gas bubbles were present in colonies of small and large bubbles, with the latter extending to 30 μ diam. The bubbles were more nearly spherical than those in the restrained PuO_2 specimens but, again, became less regular as their size decreased (Figure 7). White precipitates are present in approximately the same quantities and distributions as in the PuO_2 irradiated to 12.9×10^{21} f/cm³. The structure had changed to fluorite; if indexed as PuO_2 , the diffraction pattern indicates a lattice dilation of 0.82% as compared with nonirradiated PuO_2 . This reversion in structure may have occurred during irradiation or in postirradiation handling because of oxidation caused by self-heating in the specimen.

Both Specimens 411 and 413 (PuO_2 and Pu_2O_3) had reacted slightly with the aluminum clad (Figure 8).

100μ

50μ

FIGURE 5.7. Typical Irradiated PuO₂ Microstructure
12.9 x 10²¹ f/cm³

FIGURE 5.9.7 Typical Irradiated Pu_2O_3 Microstructure
 $9.0 \times 10^{21} \text{ f/cm}^3$

FIGURE 5-10. Reaction Between Plutonium Oxides and Aluminum Clad; Temperature at Interface 350 °C