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IRRADIATION TESTING IN SUPPORT OF THE Pu-RECYCLE DEMONSTRATION EXPERIMENT

IN EBR*

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By

W. J. Bailey**

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IRRADIATION TESTING IN SUPPORT OF THE Pu-RECYCLE DEMONSTRATION EXPERIMENTIN EBWR

More than 1500 vibrationally compacted UO_2 (depleted)-1.5 wt% PuO_2 rods were prepared⁽¹⁾ for a cooperative Argonne-Hanford Plutonium Recycle Demonstration Experiment in the Experimental Boiling Water Reactor (EBWR). The purpose of the experiment is to obtain useful information for the utilization of plutonium as a fuel in light water, thermal systems - either an all-thermal plutonium recycle system or as the thermal part of a mixed fast-thermal reactor complex.⁽²⁾ Cutaway views of a typical rod and fuel element box are shown in Figure 1. Each rod is 148 cm long and contains 830 grams of impacted fuel⁽³⁾ (active length - 122 cm). The bulk fuel density is 86-89 percent of the theoretical value. The cladding is low-nickel Zircaloy-2, 0.945 cm ID by 0.064 cm wall. The fission gas plenum, 14.6 cm long, incorporates a stainless steel (302 alloy) spring separated from the fuel by a ZrO_2 pellet, 0.64 cm thick. The fuel element boxes are designed to allow individual rods to be withdrawn and replaced. This essential feature permits isotopic analysis studies to be made as burnup proceeds. These studies constitute the main objective of the experiment.⁽²⁾

Thirty-six short rod specimens, some with prototypic fission gas plenums, are being irradiated (Tables I and II, Figure 2) to produce Zircaloy surface heat fluxes to 315 w/cm^2 or greater and exposures to 6.8×10^{20} fissions/ cm^3 to evaluate fission gas release, sintering, plutonium and/or fission product migration, and spring-pellet-fuel compatibility. Vibrational compaction of the specimens was accomplished by the resonant beam technique. Prototypical fuel mesh size fractions (-6 +10, 55%; -20 +35, 12.8%; -35 +65, 12.2%;

-200 +325, 5.5%; and -325, 14.5%) were employed. High bulk fuel densities were difficult to achieve in the 7.6-10 cm long fuel sections. The Zircaloy-clad specimens had bulk fuel densities ranging from 77 to 85% TD. All specimens were sheathed in secondary aluminum jackets, 2.9 cm OD, to provide additional plutonium containment and to avoid coolant boiling. The aluminum jackets contain four, full-length, replaceable Al-Co alloy flux-monitoring wires. Positions of discharged specimens in Figure 2 are based on wire data. Of the twelve capsules (one with prototypic plenum) under irradiation as of November 1, 1964, the maximum exposure achieved was 3.4×10^{20} fissions/cm³. Twenty capsules (six with plenums), discharged at 1.9×10^{20} fissions/cm³ (maximum), are being examined. Excellent performance is indicated and no failures have occurred.

Early plans had indicated that the EBWR would employ UO₂-2.5 wt% PuO₂ rods operating at a maximum heat flux of 158 w/cm² but control rod problems necessitated design modifications. Under presently proposed 42-Mw EBWR operation, conditions for the UO₂-1.5 wt% PuO₂ fuel rods are: rod power - 206 w/cm (maximum), heat flux - 60 w/cm² (maximum), and exposure $\sim 2.5 \times 10^{20}$ fissions/cm³. This is the exposure of most interest, however, the rods are designed for and may receive an exposure of 6.8×10^{20} fissions/cm³ (25,000 Mwd/ton of UO₂).

Photomacrographs and autoradiographs of longitudinal sections of four capsules near the top and bottom of the fuel region show the effects of rod power and exposure. All autoradiographs in this paper were made using high-resolution film plates (exposures ranged from 10 to 40 seconds) and dark areas in the figures correspond to regions of high radioactivity level. In Figure 3, one notes no apparent change in the fuel structure after operation at a rod

power of 501 w/cm, more than double the proposed EBWR rod power. Spring and pellet were intact in this specimen (GEH-14-646). The second capsule (GEH-14-706) in Figure 3 exhibits evidence of pellet-fuel interaction and fuel slumping. There appears to be no fission product migration into the ZrO_2 . Rod power of this specimen was 1013 w/cm. Note the absence of plutonium-rich areas (e.g., black dots of the type which are present at the fuel periphery) at the axial fuel-pellet interface. Solid solution formation is indicated by the areas of uniform coloration on the autoradiographs. Zirconia forms a wide range of solid solutions with both UO_2 and PuO_2 .⁽⁴⁾⁽⁵⁾⁽⁶⁾ In Figure 4 the specimen (GEH-14-650) which operated at a rod power of 1037 w/cm shows definite evidence of a fuel-pellet-spring interaction. The pellet, as such, has vanished. Only about one inch of the spring end nearest the pellet was severely damaged. The second specimen (GEH-14-651) in Figure 4 operated at a rod power of 1276 w/cm and to three times the exposure of the last capsule. The pellet and spring were missing. Note the highly-radioactive mass at the upper end of the fuel. To date, the mass is unidentified; x-ray diffraction work has not disclosed the presence of uranium, zirconium, or stainless steel. Some fuel-cladding reaction has occurred.

Transverse sections at the fuel midpoint of five specimens further illustrate rod power effects. The specimen (GEH-14-714) in Figure 5 operated at a rod power of 327 w/cm. It exhibits little or no sintering and no apparent change on the autoradiograph. At a slightly higher rod power (376 w/cm), grain growth became evident (Figure 6) and some homogenization of the fuel in the specimen (GEH-14-525) was noted at the thermal center on the autoradiograph. The next specimen (GEH-14-526) displays on the autoradiograph (Figure 7) a concentration of highly-radioactive particles in the central

region. Identification of the particles is in process. Based on the observations made by numerous investigators studying irradiated UO_2 , UO_2 - PuO_2 , and other ceramic fuels, it is believed that the particles in this specimen are probably a combination of uranium, plutonium, and fission products. It is anticipated that the particles will show a high concentration of Ru^{106} . The particles are located in the grain growth region of this specimen which operated at a rod power of 425 w/cm. At a higher rod power (1013 w/cm), the particles agglomerate and form an annular ring of high radioactivity level as shown in Figure 8 on the autoradiograph of the next specimen (GEH-14-706). This capsule operated with a molten core (note porous central region) and the fuel reacted with nearly half of the single ZrO_2 pellet. The effect of high rod power (1010 w/cm) and high exposure (1.9×10^{20} fissions/cm³) is reflected in the appearance of the specimen (GEH-14-707) in Figure 9. The long narrow columnar grains in this specimen (in reactor for 115 days) are typical of the type found in highly irradiated UO_2 pellet or packed-particle fuels and UO_2 - PuO_2 pellet fuels.

Fission gas release values (Table I,II; Figure 10) have been obtained for twelve specimens to date. The triangular symbols are plenum-equipped capsules which had bulk fuel densities to 80% TD (range: 77-80% TD, average: 79% TD). Circular and square symbols are non-plenum capsules which had bulk fuel densities to 85% TD (range: 77-85% TD, average: 81% TD). The four points aligned vertically above ~ 500 w/cm illustrate the effect of fuel bulk density on fission gas release; the lower square symbol has the highest density (85% TD). The three specimens which operated above 1000 w/cm all underwent fuel-pellet or fuel-pellet-spring interactions. At proposed

EBWR rod power, 206 w/cm, it appears that one may encounter fission gas release values on the order of five percent or less with full-scale rods (bulk fuel density 86-89% TD).

The capsule tests continue to indicate that the full-size $\text{UO}_2\text{-PuO}_2$ rods will perform satisfactorily under proposed 42-Mw EBWR conditions and to the exposure of most interest, $\sim 2.5 \times 10^{20}$ fissions/cm³. Thermal-hydraulic tests have been conducted, irradiation test proposals evaluated, fuel rod basket and cluster assemblies prepared, and over 60 full-scale prototypic rods selected and examined for forthcoming tests in Hanford reactors at near-EBWR coolant conditions.

Table I. Irradiation Testing of Vibrationally-Compacted
Impacted UO₂-1.5 wt% PuO₂ Capsules*

GEH-14 No.	Bulk Fuel Density (%TD)	Heat Flux** (w/cm ²)	Rod Power** (w/cm)	Exposure**		Fission Gas Release (%)	Remarks
				(fissions/ cm ³)x10 ⁻²⁰	(Mwd/ ton UO ₂)		
517	82	53	180	0.44	(1,690	5	No sintering.
518	82	113	380	0.40	(1,530	14	No sintering.
519	83	340	1160	0.61	(2,290	Lost	Molten core.
520	82	(158)	(535)	(2.0)	(7,500)		Being examined.
521	83	(158)	(535)	(4.1)	(15,000)		In reactor
522	82	(158)	(535)	(4.0)	(15,000)		In reactor
523	84	(158)	(535)	(6.8)	(25,000)		In reactor
524	83	(158)	(535)	(6.8)	(25,000)		In reactor
525	80	112	376	1.0	4,010		Columnar grain growth
526	82	125	425	1.2	4,450		Columnar grain growth
527	79	(158)	(535)	(2.6)	(10,000)		In reactor
528	84	(158)	(535)	(2.7)	(10,000)		In reactor
529	84	(158)	(535)	(5.5)	(20,000)		In reactor
646***	80	147	501	0.24	920	30	Gr. growth, center void
647***	77	147	500	0.72	2,900	22	Col. grains, center void
648***	81	(158)	(535)	(6.6)	(25,000)		In reactor
649***	79	188	641	1.5	5,880		Being examined
650***	80	304	1037	0.46	1,800	81	Molten core
651***	79	374	1276	1.6	6,330	84	Full-rad. col.grs., ctr voi
706	81	298	1013	0.55	2,110	51	Molten core
707	83	298	1010	1.9	7,050		Narrow col.grains, ctr voi
708	81	125	424	0.81	3,110		Slight sintering
709	81	(126)	(427)	(1.3)	(5,000)		Narrow col.grains, ctr voi
710	81	269	915	1.1	4,390		Narrow col.grains, ctr voi
711	81	(236)	(803)	(2.7)	(10,000)		In reactor
712	84	(236)	(803)	(4.1)	(15,000)		In reactor
713	85	(315)	(1071)	(4.2)	(15,000)		In reactor
714	77	96	327	0.51	2,050	10	No sintering
715	78	100	341	0.84	3,360		No sintering
716	78	(95)	(323)	(1.3)	(5,000)		Discharged, at MTR
717	80	132	448	0.48	1,860	14	Slight sintering
718***	79	218	745	0.40	1,590	52	Being examined

*All capsules contain impacted fuel. Tabular values in parenthesis are proposed or design values.

**Values based on flux-monitoring wire data. Zircaloy cladding, 1.08 cm OD by 0.064 cm wall.

***Plenum-equipped capsules.

Table II. Irradiation Testing of Vibrationally-Compacted
Impacted UO₂-2.5 wt% PuO₂ Capsules*

GEH-14 No.	Bulk Fuel Density (%TD)	Heat Flux** (w/cm ²)	Rod Power** (w/cm)	Exposure**		Fission Gas Release (%)	Remarks
				(fissions/ cm ³)x10 ⁻²⁰	(Mwd/ ton UO ₂)		
421	81	151	512	1.2	4830	17	Some sintering.
422	85	151	509	0.38	1390	10	Some sintering.
423	82				1630***		Some sintering.
424	82				4700***		Some sintering.

*Impacted fuel in 421, 422; physically mixed fuel in 423, 424.

**Values based on flux-monitoring wire data. Zircaloy cladding, 1.07 cm OD by 0.048 cm wall

***Estimated from wire data. Over half of sample dissolved for burnup analysis.

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FIGURE 1

BBWR, $\text{UO}_2\text{-PuO}_2$ FUEL ROD AND FUEL ELEMENT BASKET

EBWR PLUTONIUM FUEL ELEMENT

UO₂-15 PuO₂

Negative No. 0642180-5

FIGURE 2

IRRADIATION TESTING OF VIBRATIONALLY-COMPACTED,
IMPACTED, $\text{UO}_2\text{-PuO}_2$ FUEL

IRRADIATION TESTING OF ZIRCALOY-CLAD (1.08cm OD), VIBRATIONALLY-COMPACTED, IMPACTED UO_2 -1.5wt % PuO_2 CAPSULES

Negative c. 0645180-1

FIGURE 2

PHOTOMICROGRAPHS AND AUTORADIOGRAPHS OF LONGITUDINAL SECTIONS,
TAKEN NEAR TOP AND BOTTOM OF THE FUEL CORE, IN IRRADIATED SPECIMENS
(GEH-14-646 and -706)

***FUEL-PELLET (ZrO_2) COMPATIBILITY IN
IRRADIATED, VIBRATIONALLY-COMPACTED,
IMPACTED, UO_2 -1.5wt % PuO_2 CAPSULES***

ROD POWER, (W/cm)

501

1013

HEAT FLUX (W/cm²)

147

298

File No. 06-7051-1

FIGURE 1

PHOTO MICROGRAPHS AND AUTORADIOGRAPHS OF LONGITUDINAL SECTIONS,
TAIL FEELER TOP AND BOTTOM OF THE FUEL CORE, IN IRRADIATED SPECIMENS
(GEM-11 -650 and -651)

***FUEL-PELLET (ZrO_2) COMPATIBILITY IN
IRRADIATED, VIBRATIONALLY-COMPACTED,
IMPACTED, UO_2 -1.5wt % PuO_2 CAPSULES***

ROD POWER, (W/cm)

1037

1276

HEAT FLUX, (W/cm²)

304

374

Negative No. 0643027-2

FIGURE 5

TRANSVERSE SECTION OF IRRADIATED UO_2 - PuO_2 SPECIMEN
(GEH-14-714)

***IRRADIATED, ZIRCALOY-CLAD (1.08cm OD),
VIBRATIONALLY-COMPACTED, UO_2 -1.5wt % PuO_2 CAPSULE***

ROD POWER- 327 w/cm

EXPOSURE- 0.51×10^{20} FISSIONS/cm³

PHOTOMACROGRAPH (30 X)

AUTORADIOGRAPH

Negative No.

FIGURE 6

PHOTOMACROGRAPH AND AUTORADIOGRAPH OF TRANSVERSE SECTION OF IRRADIATED

UO₂-1.5 WT% PuO₂ SPECIMEN (GEH-14-525).

Rod Power - 376 w/cm.

Exposure - 1.0×10^{20} fissions/cm³.

Negative No. 0643180-3

FIGURE 7

TRANSVERSE SECTION OF IRRADIATED $\text{UO}_2\text{-PuO}_2$ SPECIMEN
(GEH-14-526)

***IRRADIATED, ZIRCALOY-CLAD (1.08 cm OD),
VIBRATIONALLY-COMPACTED, UO_2 -1.5wt % PuO_2 CAPSULE***

ROD POWER- 425 w/cm

EXPOSURE-1.2 $\times 10^{20}$ FISSIONS/cm³

PHOTOMACROGRAPH (30X)

AUTORADIOGRAPH

Negative No. 0643027-3

FIGURE 8

TRANSVERSE SECTION OF IRRADIATED UO_2 - PuO_2 SPECIMEN
(GEH-14-706)

***IRRADIATED, ZIRCALOY-CLAD (1.08cm OD),
VIBRATIONALLY-COMPACTED, UO₂-1.5wt % PuO₂ CAPSULE***

ROD POWER- 1013 w/cm

EXPOSURE- 0.55×10^{20} FISSIONS/cm³

PHOTOMACROGRAPH (30X)

AUTORADIOGRAPH

Negative No.

FIGURE 9

PHOTOMACROGRAPH AND AUTORADIOGRAPH OF TRANSVERSE SECTION OF IRRADIATED

UO₂-1.5 WT% PuO₂ SPECIMEN (GEH-14-707).

Rod Power - 1010 w/cm².

Exposure - 1.9×10^{20} fissions/cm³.

Negative No. 0643027-1

FIGURE 10

FISSION GAS RELEASE FROM IRRADIATED $\text{UO}_2\text{-PuO}_2$ SPECIMENS

***FISSION GAS RELEASE FROM IRRADIATED VIBRATIONALLY-
COMPACTED, IMPACTED UO_2 -1.5wt % PuO_2***

REFERENCES

1. Sharp, R. E., C. A. Burgess, J. R. Hague, and R. J. Lobsinger, Development and Fabrication of UO_2/PuO_2 Fuel Elements for the Pu Recycle Demonstration Experiment in EBWR, HW-SA-3639A. July 1964. (To be published in Trans. Am. Nuclear Soc., November 1964).
2. Argonne National Laboratory, Private Communication, March 1964.
3. Brite, D. W. and C. A. Burgess, High-Energy-Rate Pneumatically Impacted UO_2/PuO_2 Fuels, HW-SA-3616A. July 1964. (To be published in Trans. Am. Nuclear Soc., November 1964.)
4. Williams, J., "Dispersion of Oxides in Oxide Matrices as High Temperature Reactor Fuels," IAEA Symposium on New Reactor Materials, Prague, July 1963.
5. Carroll, D. F., The System $\text{PuO}_2\text{-ZrO}_2$, HW-69305. April 1961.
6. Freshley, M. D. and D. F. Carroll, "The Irradiation Performance of $\text{ZrO}_2\text{-PuO}_2$ Nuclear Fuel," Trans. Am. Nuclear Soc., 6 (2), 348-349 (1963). (HW-SA-3128).

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