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BAW-3809-4
July 1968

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POSTIRRADIATION EXAMINATION OF THORIA-URANIA FUEL RODS

— Quarterly Progress Report No. 4 —

March - May 1968

RESEARCH & DEVELOPMENT
DIVISION

NUCLEAR DEVELOPMENT
CENTER

Lynchburg, Virginia

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Price: Printed Copy \$3.00; Microfiche \$0.65

BAW-3809-4
July 1968
TID-4500
UC-25

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March - May 1968

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AEC Contract No. AT(30-1)-3809
B&W Contract No. 859-3103

Prepared for
THE UNITED STATES ATOMIC ENERGY COMMISSION

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1. INTRODUCTION

The objective of this program is to perform isotopic and metallurgical analyses on thoria-urania fuel, stainless steel cladding, and a Zircaloy fuel can taken from Core A of the Consolidated Edison Indian Point Reactor Number 1. The isotopic composition of the several actinide and fission product elements in the fuel will be determined. The fuel and the cladding will be examined metallographically. Specimens of the stainless steel cladding and the Zircaloy can (channel) will be tensile-tested under several conditions.

This report, the fourth in a series of quarterly progress reports on this program, covers the period from March 1 through May 30, 1968. The previous reports issued are as follows:

- BAW-3809-1 Progress Report No. 1
March - August 1967
- BAW-3809-2 Progress Report No. 2
September - November 1967
- BAW-3809-3 Progress Report No. 3
December 1967 - February 1968

2. SUMMARY

The sectioning of fuel rods to obtain samples of the fuel and cladding for chemical and metallurgical investigations has been completed. All samples for isotopic analysis have been dissolved, and uranium, plutonium, americium-curium, neodymium, promethium, and samarium have been separated. Some results for the isotopic composition of uranium have been compared with preliminary calculated values.

Profilometer measurements on the cladding were completed. Preliminary results of metallographic examinations indicate some porosity on the inner surface of the cladding.

3. ISOTOPIC ANALYSIS OF THORIA-URANIA FUEL

3. 1. Experimental Measurements

3. 1. 1. Sample Selection

The fuel rod selection is described in the first progress report.¹ One specimen was taken from near the center of each rod. This corresponded to the peak burnup area for each rod except 105 F, which had no initial uranium in the central zone. Another specimen was taken from a lower burnup zone for all rods except the second "D" rod from element 105.

3. 1. 2. Dissolution

Dissolution studies on thoria-urania fuel are described in the third progress report.² All of the samples have been dissolved in a mixture of 13M nitric acid and 0.025M hydrofluoric acid under reflux. The samples were fragmented during mechanical decladding operations. This fragmentation contributed to rapid dissolution. None of the samples required more than 1 hour for complete dissolution.

3. 1. 3. Chemical Separation

A sequential separation scheme for uranium, plutonium, americium-curium, neodymium, promethium, and samarium is described in previous reports.^{1,3} All of the fuel samples have been processed in accordance with that scheme. Individual separation procedures will be used for other components of the fuel.

3. 1. 4. Measurements

Mass spectrometric analysis of the isotopic composition of uranium samples was begun. Ten samples were analyzed, and the preliminary results are compared with calculated values in section 3.2. Three plutonium samples were analyzed mass spectrometrically, and a preliminary review of the data shows a high abundance of ^{238}Pu . This

is not surprising since ^{238}Pu is produced from ^{235}U and the fuel was initially enriched to 93% ^{235}U . Alpha spectrometry confirms the presence of ^{238}Pu and also indicates that small amounts of ^{236}Pu were produced.

3.2. Calculated Isotopic Abundances

The isotopic abundances of uranium and plutonium are being calculated based on the reactor operating history and will be compared with experimental values. Some preliminary experimental and calculated values are compared in Table 1. There is generally good agreement for the four rods on which analyses are complete. All of these samples came from the peak burnup area of the rod. The preirradiation uranium composition was 1.0% ^{234}U , 93.3% ^{235}U , 0.3% ^{236}U , and 5.4% ^{238}U .

Table 1. Comparison of Experimental and Calculated Uranium Isotopic Composition of Thoria-Urania Fuel Rods

	<u>Rod 62A</u>		<u>Rod 105B</u>		<u>Rod 105C</u>		<u>Rod 105D</u>	
	<u>Exp</u>	<u>Calc</u>	<u>Exp</u>	<u>Calc</u>	<u>Exp</u>	<u>Calc</u>	<u>Exp</u>	<u>Calc</u>
^{233}U	45.3	43.1	34.2	30.0	11.6	13.7	8.2	8.8
^{234}U	5.1	4.1	3.8	2.9	1.3	1.4	1.3	1.3
^{235}U	30.1	35.4	41.2	48.2	74.7	73.7	79.0	79.2
^{236}U	13.5	11.9	14.3	11.9	6.2	5.7	5.6	5.0
^{238}U	6.0	5.5	6.5	6.1	6.2	5.5	5.9	5.7

4. METALLURGICAL ANALYSES

The Metallurgical Analyses Task of this project involves the detailed postirradiation examination of the $\text{ThO}_2\text{-UO}_2$ fuels, the borated type-304 stainless steel, and the Zircaloy-2 which were in the Consolidated Edison Reactor.

During this report period, the sectioning of all 13 fuel rods was completed; metallographic examinations, profilometer measurements, and preparation of the tensile specimens were initiated. The current status of the overall examination of the fuel rods is summarized in Table 2.

4. 1. Fuel Rod Sectioning

In the previous quarterly report,² it was reported that fuel pellets were free to move into the plenum even in the high burnup rods. After sectioning the rods, fuel pellets could be poured out of the tubing. Two views of a fuel pellet removed from rod 105C are shown in Figure 1. This pellet achieved an estimated burnup of 15 GWd/T. Irradiation did not alter the shape of the pellet or the enrichment identification mark ("C").

A side view of a series of three pellets removed from another portion of rod 105C is shown in Figure 2. The ability to remove apparently undamaged pellets was not limited to the "C" fuel, since it was possible to remove entire pellets from all fuel rods examined regardless of their enrichment or burnup.

Figure 1. End and Side Views of Irradiated Thoria-Urania Pellet

End View

(Tubular piece is a support, not the cladding.)

Side View

(Pellet Diameter: ≈ 0.260 inch;
Pellet Length: ≈ 0.750 inch)

Figure 2. Side View of Three Irradiated Thoria-Urania Pellets

(Pellet Diameter: ≈ 0.260 inch;
Pellet Length: ≈ 0.750 inch)

Table 2. Status of Hot Cell Examination of ThO₂-UO₂ Fuel Rods

Pin No.	Th/U ratio	Estimated peak rod burnup, GWd/T	Visual	Gamma scan	Diameter	Profilometer	Gas puncture	Gas analyses	Sectioning	Metallography	Micro-hardness	Tensile test	Burnup	Isotopic analyses	Specimens for OSU - BMI
20A	33	33	x	x					x			*	*		
61A	33	30	x	x	x	x			x	*	@	*	*	@	
62A	33	28	x	x					x			*	*		
66A	33	28	x	x					x			*	*		
105A	33	22	x	x	x	x			x	*	@	*	*	@	
105B	23	30	x	x					x	*		*	*		x
105C	17	15	x	x	x	x			x	*	@	*	*		x
105D ₁	12	20	x	x	x	x	x	x	x	*		*	*	@	
105D ₂	12	18	x	x			x	x	x	*		*	*		x
105E	10	22	x	x			x	x	x	*		*	*		
105F ^(a)	33	2 ^(b)	x	x			x	x	x	*	@	*	*		x
106A	33	8	x	x	x	x			x	*	@		*	@	x
106B	23	12	x	x					x	*			*		x

(a) One-third of rod contains pure ThO₂ and two-thirds contains ThO₂-UO₂ with a Th/U of 33.

(b) Pure ThO₂ section.

Code

x Completed.
 * In progress.
 @ Planned.
 Blank Not planned.

4.2. Profilometer Measurements

The profilometer measurements for bow, ovality, and diameter of the 7-inch-long segments from rods 61A, 105A, 105C, 105D, and 106A have been completed. The locations from which the specimens were obtained are shown in Table 3. Some bowing was observed in essentially all of the sections from these five fuel rods. However, it is impossible to determine whether the bowing occurred during irradiation or in handling of the rods during the postirradiation examination. Neither ridging at the pellet interfaces nor increase in the rod diameters was observed in any of the fuel rod sections regardless of fuel enrichment or burnup. A profilometer scan of the diameter from a section of fuel rod 105A is shown in Figure 3. The maximum variation in diameter is less than that normally specified for the as-received fuel cladding tube.

4.3. Metallographic Examination

Twenty-one specimens are in various stages of metallographic examination. These specimens are described in Table 4. Thus far, four significant items have been observed during the metallographic examination:

1. There were no major changes in the microstructure of the fuel regardless of the burnup or the heat rate. All fuel samples showed some overall increase in porosity and some migration of the voids to the grain boundaries. The extent of the porosity and the migration of voids to the grain boundaries increased with increasing burnup. Typical examples of the fuel at high and low exposures are shown in Figure 4.

2. Metallic-appearing inclusions were found on all types of fuel. Examples of various sizes and shapes of the inclusions are shown in Figure 5. It seems likely that these inclusions were introduced during fabrication since relatively large agglomerates were found in the pure ThO_2 . The low burnup achieved by the ThO_2 (≈ 2 GWd/T) essentially eliminates fission product accumulation as the source of inclusions. Furthermore, most of the available data indicate that fission product agglomeration does not occur unless fuel temperatures exceed 2000 C. The lack of significant grain growth in any of the fuel rods indicates that the center line temperatures were considerably below 1800 C. Additional work is planned in order to identify these inclusions.

Figure 3. Profilometer Scan of 7-Inch Section From Rod 105A

Figure 4. As-Polished Appearance of Irradiated Thoria-Urania Fuel

250X

Rod 105A

(Estimated Burnup: 22 GWd/T)

250X

Rod 106A

(Estimated Burnup: 8 GWd/T)

Figure 5. Metallic Inclusions in Thoria-Urania Fuel
(As-Polished, 250X)

Rod 105A

(ThO_2 - 3% UO_2 ; Peak Rod
Burnup: 22 GWd/T)

Rod 105C

(ThO_2 - 5.5% UO_2 ; Peak Rod
Burnup: 15 GWd/T)

Rod 105F

(Pure ThO_2 ; Peak Rod
Burnup: 2 GWd/T)

Table 3. Location of Profilometer Specimens

<u>Rod No.</u>	<u>Estimated rod burnup, GWd/T</u>	<u>Specimen location</u>		
		<u>High</u>	<u>Average</u>	<u>Low</u>
61A	30	1	1	1
105A	22	1	1	1
105C	15	1	0	0
105D ₁	20	1	0	0
106A	8	1	0	1

Table 4. Location of Metallographic Specimens

<u>Rod No.</u>	<u>Estimated peak rod burnup, GWd/T</u>	<u>Specimen location</u>			
		<u>High</u>	<u>Low</u>	<u>Ferrule</u>	<u>Spacer</u>
61A	30	1	1	0	0
105A	22	1	1	1	1
105B	30	1	1	0	0
105C	15	1	1	0	0
105D ₁	20	1	1	0	0
105E	22	1	1	0	0
105F	2	1	0	0	0
106A	8	1	1	1	1
106B	12	1	1	0	0

3. There was no noticeable deterioration of the Kannigen braze joints during irradiation. A braze joint from fuel rod 106A is shown in Figure 6. The porosity and cracking observed at the extremities of the braze joints are very similar to those observed in the as-fabricated joints. The porosity is caused by shrinkage of the liquid during solidification of the braze alloy.

4. There was considerable porosity on the inside surface of the cladding from most of the fuel rods. This porosity assumed a variety of shapes including small radial cracks, irregular voids, and spheres. The zone of porosity is generally limited to about 0.002 inch in depth. Typical examples of the types of voids observed in several samples are shown in Figures 7 and 8. These voids were not observed in the cladding adjacent to the pure ThO_2 (rod 105F) as shown in Figure 9. The thermal and fast neutron exposures in rod 105F are similar to those observed in rods 105B and 105C, in which significant degrees of porosity were observed. Several samples of the unirradiated cladding were examined, but no porosity was found. Typical photomicrographs of the unirradiated cladding are shown in Figure 10. Currently, the course of this porosity and cracking is unknown. Additional data will be required before any attempt is made to explain this phenomenon.

4.4. Tensile Testing

Essentially all of the necessary preparations for tensile-testing the cladding have been completed. Suitable grips and an extensometer have been developed and evaluated. Additional details on these items will be included in the next progress report after testing has been completed.

The sectioning of the Zircaloy-2 can was completed, and the preparation of the tensile blanks was initiated. Specimens of the can were prepared for metallographic examination and hydrogen analyses.

Figure 6. Cladding-to-Ferrule Kannigen Braze Joint

Etch: Oxalic Acid
Electrolytic

75X

Figure 7. Porosity in Irradiated Stainless Steel Cladding
(As-Polished, 250X)

Rod 105B

(Peak Rod Burnup: 30 GWd/T)

Rod 105C

(Peak Rod Burnup: 15 GWd/T)

Figure 8. As-Polished and Etched Appearance of
Cladding From Rod 105C

As-Polished

250X

Etch: Oxalic Acid
Electrolytic

250X

Figure 9. As-Polished Appearance of Cladding
From Rod 105F

250X

Figure 10. As-Polished and Etched Appearance of
Unirradiated Cladding

As-Polished

150X

Etched

150X

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- ¹ Bishop, W. N., Marsh, S. F., and Baroch, C. J., Postirradiation Examination of Thoria-Urania Fuel Rods — Quarterly Progress Report No. 1 — March - August 1967, The Babcock & Wilcox Company, BAW-3809-1, Lynchburg, Virginia, October 1967.
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