

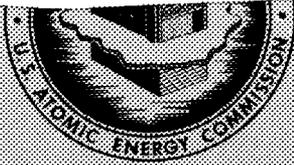
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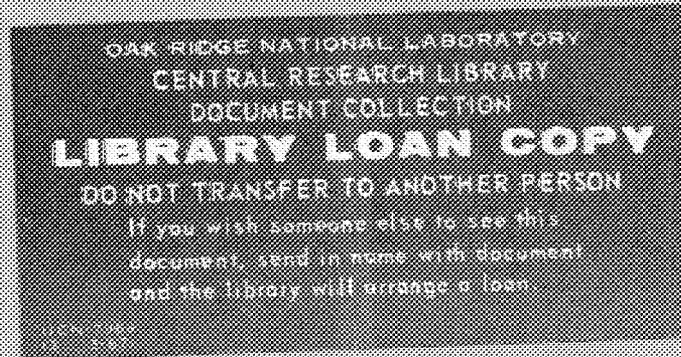
U. S. ATOMIC ENERGY COMMISSION



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THE DISTRIBUTION OF FISSION PRODUCT TRITIUM IN A
ZIRCALOY-CLAD UO_2 BLANKET ROD FROM PWR-1

J. H. Goode and C. M. Cox



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CHEMICAL TECHNOLOGY DIVISION
METALS AND CERAMICS DIVISION

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J. H. Goode
Chemical Technology Division

C. M. Cox
Metals and Ceramics Division

JUNE 1970

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Oak Ridge, Tennessee
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CLAD UO₂ BLANKET ROD FROM PWR-1

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ABSTRACT

The given Zircaloy-2-clad UO₂ blanket rod had been irradiated to 3.41% FIMA at a time-averaged linear heat rate of 4.84 kw/ft, with a clad temperature of 259°C, for the full life of the first core of the Shippingport Pressurized Water Reactor. It was destructively examined to determine the distribution of fission product tritium. There was apparently no loss of tritium from the rod; we found 7% of the yield of tritium in the Zircaloy metal cladding (apparently as the tritide) and 93% in the UO₂ fuel.

INTRODUCTION

Studies are in progress at ORNL to determine the fate of tritium that is produced in fuel elements by ternary fission of uranium and plutonium. Previous studies,¹ with stainless steel and Zircaloy-2-clad UO₂ samples from LWR fuels and with stainless steel-clad PuO₂-UO₂ samples of LMFBR fuels, indicated that tritium diffused through stainless steel cladding when the linear heat rating was higher than 5 kw/ft. The clad temperature was 105 to 350°C for the < 5 kw/ft ratings and 500 to 1000°C for the higher ratings (10 to 25 kw/ft). Thus, it is uncertain whether the primary variable affecting tritium loss is clad temperature or the linear heat rating. An experimental Zircaloy-2-clad fuel rod retained about 50% of the tritium at an estimated heat rating of 5 kw/ft and a surface temperature of 125°C.

This report presents additional information on the fate of tritium; the subject of the study was an irradiated blanket rod from the first core of the Shippingport Pressurized Water Reactor (PWR). This 0.410-in.-diam by 10.26-in.-long Zircaloy-2-clad rod (Rod 10) contained 26 normal-enrichment UO₂ pellets and was the north-east corner rod of a total of 120 in Bundle 0320, the middle bundle in a vertical

stack of 7 in Blanket Assembly K-10.²⁻³ The assembly was inserted into Blanket Region 2 for the initial critical experiments of PWR-1 (December 18, 1957), and was removed at the end of life of Core 1 on February 9, 1964, after 27,780 effective full power hours (EFPH) of operation.⁴ The rod was sent to ORNL by the Westinghouse Expanded Core Facility for use in fuel reprocessing studies.

This report represents the combined efforts of individuals from several divisions of the Oak Ridge National Laboratory. Members of Chemical Development Section B of the Chemical Technology Division (J. H. Goode, V. C. A. Vaughen, and technicians L. A. Byrd, G. D. Davis, and O. L. Kirkland) planned and carried out the experimental work and the primary tritium separations. Operations Division and Metals and Ceramics Division personnel (R. L. Lines, G. A. Moore) punctured the rod to release fission gases and sectioned the fuel rod. Analytical Chemistry Division members (C. E. Lamb, F. L. Layton, J. H. Moneyhun, J. E. Morton, W. T. Mullins, H. A. Parker, J. R. Sites, R. R. Rickard, and E. I. Wyatt) performed the chemical, radiochemical, and mass spectrometric analyses. C. M. Cox, of the Fuels Evaluation Group of the Metals and Ceramics Division, performed the reactor and physics calculations.

PROCEDURES

The intact fuel rod was weighed, measured to determine the diametric dimensions at 0° and 90° in the mid-plane, punctured to release fission gases, and cut into 10 segments (Fig. 1). About 3.25% of the fuel and cladding was "consumed" by the alumina cut-off wheel. The segments were individually canned and weighed to determine the weight of material lost during cutting (Table 1).

We leached or dissolved weighed portions of the fuel, cladding, or both (Table 2) in a closed system, using an argon-4% hydrogen purge gas to carry vapors or gases from the dissolver to the sampling points (Fig. 2). In-line absolute filters prevented entrained particles from entering the gas handling equipment. Hot copper oxide (~700°C) was used to convert hydrogen and tritium to water, and the water was sorbed onto dry Linde Type 5A molecular sieve. The water was later desorbed at

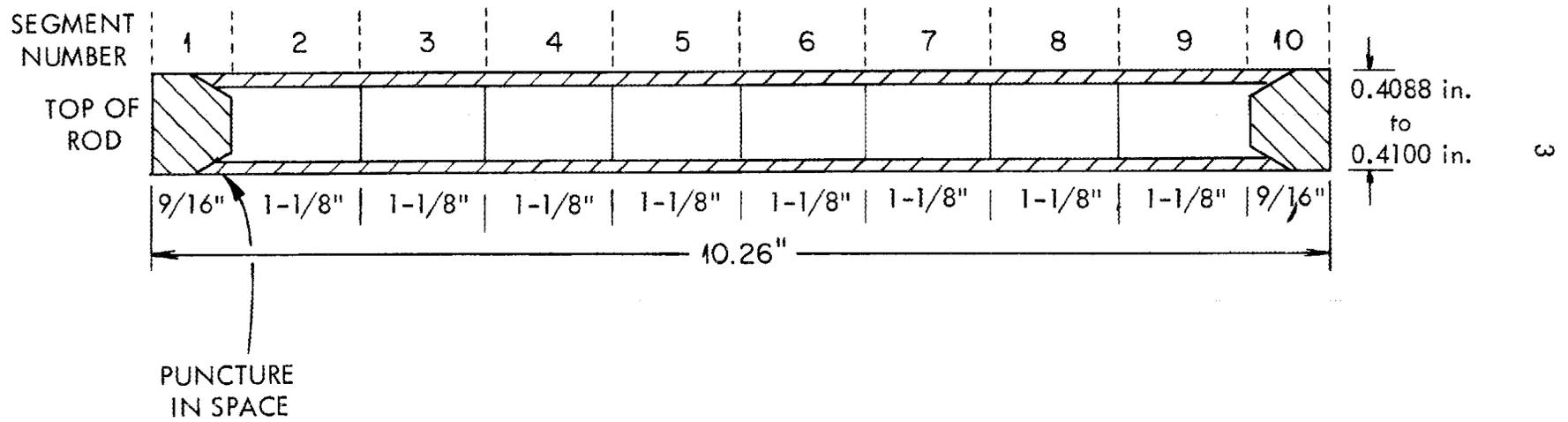


Fig. 1. Sectioning Diagram, Rod 10, Bundle 0320, Blanket Assembly K-10, PWR Core 1.

Table 2. Analytical Scheme for PWR Fuel Rod

Segment Number	Portion of Segment	Treatment	Leach No.	Analyzed ^a		
				Solution	Off-Gas	Mol. Sieve
1, 10	All	Leached, 12 <u>M</u> HNO ₃	L-1	X	X	X
2, 3, 4	Cladding	Leached, 12 <u>M</u> HNO ₃	L-2	X	X	X
2, 3, 4	Cladding ^b	Dissolved, Zirflex ^c	L-5, L-7, L-8	X	X	X
2, 3, 4	Fuel	Leached, 12 <u>M</u> HNO ₃	L-3	X	X	X
6, 7, 8, 9	All	Leached, 12 <u>M</u> HNO ₃	L-4	X	X	X
6, 7, 8, 9	Cladding ^b	Dissolved, Zirflex ^c	L-6	X	X	X
5	All	Archive Sample	-	-	-	-

^aAnalyzed for uranium, plutonium, ³H₂, ⁸⁵Kr, gamma-emitting fission products as applicable.

^bRandom 1 to 2-g samples of sidewall cladding.

^c6 M NH₄F--1 M NH₄NO₃.

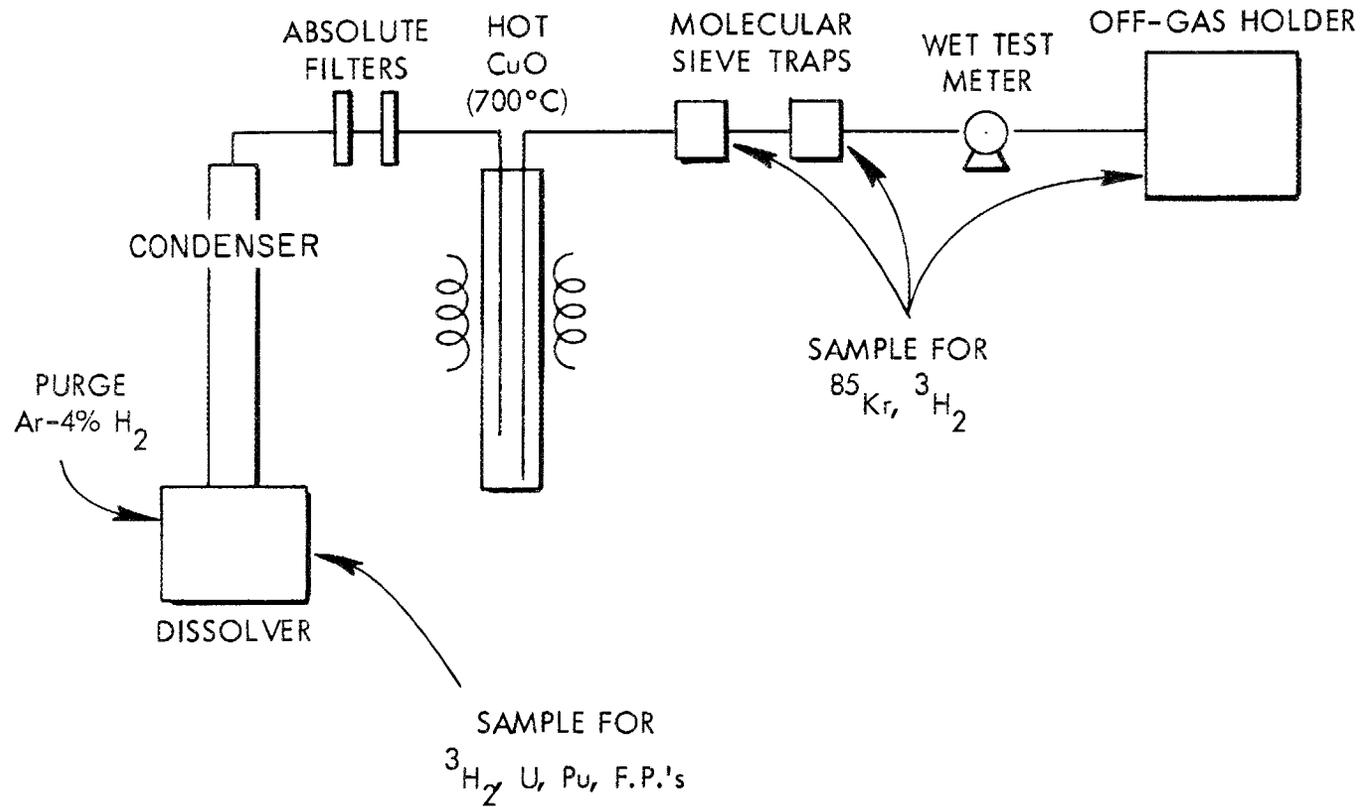


Fig. 2. Sample Points for Heavy Elements, Tritium, and Fission Products in PWR Blanket Rod.

500–600 °C, under vacuum, and collected in a cold trap at liquid nitrogen temperatures, using established techniques.^{1,5} The ice was dissolved in a standard POPOP-PPO scintillation liquid for tritium counting in a Packard Tri-Carb scintillation spectrometer. Large (approximately 300 cm³) gas samples were taken from the gas holder, mixed with hydrogen carrier, and oxidized with hot copper oxide; the resulting water was concentrated by sorption onto molecular sieve. This water was also desorbed and counted for tritium. Small (5 cm³) aliquots of gas were analyzed for ⁸⁵Kr in a gamma spectrometer, as was the molecular sieve prior to desorption of the water.

The end plugs, cladding segments from which the fuel had been mechanically removed, the broken fuel fragments, and complete segments were leached and/or dissolved in boiling 12 M HNO₃. Aliquots of the solutions were purified by distillation, and the tritium content of the distillate was determined by counting in a liquid scintillation spectrometer. Selected segments of the Zircaloy-2 cladding were dissolved in 6 M NH₄F--1 M NH₄NO₃ (Zirflex solution) to release tritium and hydrogen contained within the metal; both the solutions and the off-gases were analyzed for tritium. Finally, portions of the cladding that had been leached in 12 M HNO₃ were heated to release contained gases; these gases were analyzed by mass spectrometry.

Material balances, based on actual weights and analyses of solutions for uranium, plutonium, and fission products, were made and compared with the calculated quantities.

RESULTS

Zircaloy-2 and UO₂ Material Balance

The overall material balance indicated that a 3.25% weight loss (approx. 1.0 g of Zircaloy-2 and 5.5 g UO₂) occurred during sectioning of the rod with a cut-off saw (Table 1). We accounted for 99.4% of the UO₂ and Zircaloy tubing from Segments #2, 3, 4, 6, 7, 8, and 9 (Appendix A) and 99.6% of the fuel and cladding for the entire rod (Appendix B).

Burnup

The nitric acid leach solutions, containing fuel and fission products, were analyzed for uranium and ^{137}Cs (Table 3).

Table 3. Uranium and ^{137}Cs in Rod 10, Bundle 0320

Leach No.	Volume (ml)	Uranium		^{137}Cs	
		mg/ml	mg	dpm/ml	dpm
1	98	0.022	2.2	7.86×10^6	7.86×10^8
2	100	0.639	63.9	1.68×10^8	1.68×10^{10}
3	250	190.0	47,500.0	3.93×10^{10}	9.825×10^{12}
4	250	259.0	64,750.0	5.21×10^{10}	13.025×10^{12}
	Total		112,316.1		22.87×10^{12}

We then calculated the amount of ^{137}Cs in the entire rod:

$$\frac{1.123 \times 10^5 \text{ mg U}}{0.881} = 127.47 \text{ g UO}_2 \text{ in L-1, -2, -3, -4.}$$

$$2.287 \times 10^{13} \text{ dpm } ^{137}\text{Cs} \times \frac{152.06 \text{ g UO}_2}{127.47 \text{ g UO}_2} = 2.728 \times 10^{13} \text{ dpm } ^{137}\text{Cs in rod}$$

(See Appendix C).

The fuel burnup, based on the ^{137}Cs activity, was calculated. The fuel pin, containing 0.357-in.-diam solid UO_2 pellets (10.08 g/cm^3) operated through four seed loadings, as summarized in Table 4. The burnup calculation assumed the idealized operating history shown in Fig. 3, used the fission yields in Table 5, and used spectrum-averaged neutron cross-sections that were determined by a previously described technique.⁸ The fuel burnup was calculated as 7.67×10^{20} fissions/ cm^3 or 3.41% FIMA. The corresponding fuel isotopic and tritium concentrations, along with average heat rates at various stages of the fuel pin lifetime, are given in Table 6.

Table 4. Operating History of Shippingport Core 1⁴

	Seed 1	Seed 2	Seed 3	Seed 4
Date of full power startup	12-23-57	5-7-60	10-24-61	1-20-63
Date of shutdown	11-2-59	8-16-61	11-26-62	2-9-64
Equivalent full power days	241.9	329.2	305.4	281.0

Table 5. Fission Yields for Thermal Neutrons

Fissionable Isotope	atoms/fission	
	¹³⁷ Cs ⁶	Tritium ⁷
²³⁵ U	0.062	0.00013
²³⁶ U	(0.062) ^a	(0.00013)
²³⁸ U	(0.062)	(0.00013)
²³⁹ Np	(0.065)	(0.0002)
²³⁹ Pu	0.0656	0.00023
²⁴⁰ Pu	(0.0656)	(0.00023)
²⁴¹ Pu	0.0646	(0.00023)

^aValues in parentheses are rough estimates, but gross errors should not significantly affect the results, as indicated in Table 8.

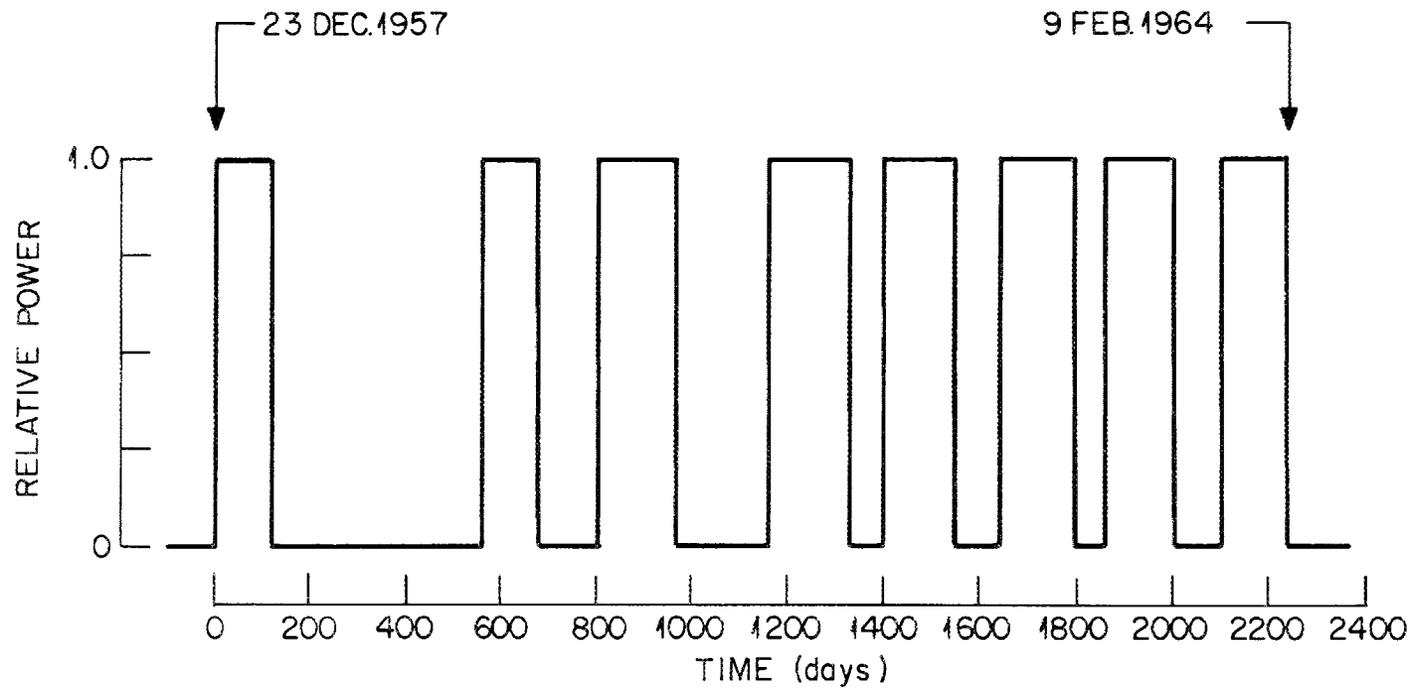


Fig. 3. Idealized Power History for PWR Core 1.

Table 6. Summary of Calculated Operating Conditions and Isotopic Concentrations for Shippingport Fuel Rod 0320-10^a

	Exposure Period					
	Initial Startup	Seed				April 1, 1970
		1	2	3	4	
Average heat rate, kw/ft	0.37	5.07	4.61	4.59	4.58	-
Concentrations, atoms/cm ³ UO ₂ × 10 ⁻²⁴						
²³⁵ U	1.7-4 ^b	4.28-5	6.57-6	1.15-6	2.33-7	2.33-7
²³⁶ U	0	1.88-5	2.35-5	2.36-5	2.31-5	2.31-5
²³⁷ U	0	1.68-8	2.15-8	2.16-8	2.12-8	~ 0
²³⁸ U	2.426-2	2.41-2	2.39-2	2.37-2	2.35-2	2.35-2
²³⁹ U	0	1.61-8	1.60-8	1.58-8	1.57-8	~ 0
²³⁹ Np	0	2.32-6	2.29-6	2.27-6	2.26-6	~ 0
²³⁹ Pu	0	6.29-5	6.87-5	6.84-5	6.78-5	7.01-5
²⁴⁰ Pu	0	2.11-5	4.23-5	4.84-5	5.00-5	5.00-5
²⁴¹ Pu	0	4.10-6	1.18-5	1.45-5	1.52-5	1.10-5
¹³⁷ Cs	0	1.16-5	2.51-5	3.70-5	4.75-5	4.12-5
³ H	0	3.02-8	7.19-8	1.10-7	1.42-7	1.00-7
FPP ^c	0	1.85-4	3.99-4	5.91-4	7.67-4	7.67-4
% FIMA	0	0.82	1.77	2.63	3.41	3.41

^aBased on ¹³⁷Cs activity = 1.7965×10^{11} dpm/g UO₂ on April 1, 1970, and a UO₂ density of 10.18 g/cm³.

^b1.7-4 = 1.7×10^{-4} , etc.

^cFPP = total fission product pairs.

For convenience, the sources of the tritium existing on April 1, 1970, are summarized in Table 7.

Table 7. Calculated Sources of Tritium Present on April 1, 1970

Fissionable Isotope	Fraction of Total Tritium
^{235}U	0.100
^{236}U	3×10^{-5}
^{238}U	0.017
^{239}Np	9×10^{-6}
^{239}Pu	0.723
^{240}Pu	2.2×10^{-4}
^{241}Pu	0.160

The burnups in the mirror image bundle (Bundle 0202, Blanket Assembly J-11) were first estimated to average 6.97×10^{20} fissions/cm^{3,2}, although a later document reported the average burnup as 5.7×10^{20} fissions/cm^{3,9}. The other rods in the mirror image Bundle 0202 had burnups ranging from 4.9 to 7.5×10^{20} fissions/cm^{3,9}. Stachew reported that the ratio of fissions of ^{239}Pu to fissions of ^{235}U in some of the high-burnup blanket rods at the end of the third seed (21,035 EFPH) was slightly greater than 3:1;³ our calculations indicated a ratio of 2.2:1 at the end of the third seed life and 3.0:1 at the end of the fourth for rod 10 in Bundle 0320. We expect that these calculations provide a good estimate although firm data are not available on other rods examined after the fourth seed.¹⁰

Fission Product Concentrations

There appears to be only slight differences between the top and bottom halves of the rod, as indicated by radiochemical analyses (Table 8).

Table 8. Radiochemical Analysis of Top and Bottom Halves of PWR Blanket Rod

Fission Product	Top Half of Rod	Bottom Half of Rod
Pu α	3.06×10^6 cpm/mg U	3.17×10^6 cpm/mg U
$^3\text{H}_2$	1.12×10^6 dpm/mg U	1.08×10^6 dpm/mg U
^{106}Ru	1.94×10^7 dpm/mg U	1.87×10^7 dpm/mg U
^{137}Cs	2.07×10^8 dpm/mg U	2.01×10^8 dpm/mg U
^{144}Ce	1.88×10^7 dpm/mg U	1.61×10^7 dpm/mg U
^{154}Eu	5.42×10^7 dpm/mg U	5.44×10^7 dpm/mg U

Linear Heat Rating

Lynam¹¹ examined rods from a similar blanket assembly (F-5) at the end of the second refueling (Seed 3). He reported no microstructural changes in the UO_2 from a rod calculated to have experienced peak heat ratings, during operation of Core 1, of $333,000 \text{ Btu/hr-ft}^2$,¹¹ or about 10.3 kw/ft . The lifetime average heat rating for the given bundle was reported to be $155,000 \text{ Btu/hr-ft}^2$,¹⁰ or about 5 kw/ft . Rubin and Lynam calculated that the peak center temperature of the pellets in the high-rated rod was 1440°C and that the pellet surface temperature was 400°C .¹² Westinghouse's best estimate of the surface temperature of the cladding was 531°F (277°C), with an average of 520°F (271°C).

The rod we examined was from a bundle having slightly lower temperatures.^{9,11} Based on a total operation of 1157.5 equivalent full power days and an energy release of 200 Mev/fission , the time averaged linear heat rate of rod 10 is 4.84 kw/ft , corresponding to a cladding surface heat flux of $154,000 \text{ Btu/hr-ft}^2$. By comparison with Table 6, this is seen to be representative of the heat rate throughout the pin lifetime; however, short periods of higher power can be expected due to seed changes and control rod positioning. Using the time-averaged heat rate, the fuel center temperature was calculated as 660°C . This calculation was made with the PROFIL

code¹³ assuming a cladding surface temperature of 259°C and using the Asamoto et al.¹⁴ correlation for UO₂ thermal conductivity. The fuel surface temperature was calculated as 350°C.

⁸⁵Kr Content of Rod 10, Bundle 0320

The rod was punctured in a high vacuum apparatus above the pellet column by laser and the released gas was analyzed for ⁸⁵Kr, ³H, and mass distribution by mass spectrometry. The total volume of released gas was 1.232 cc.¹⁵ Mass analysis showed the following composition:¹⁶

H ₂	7%	A	1%	¹³¹ Xe	7%
He	43	CO ₂	< 1	¹³² Xe	23
H ₂ O	4	Xe	37	¹³⁴ Xe	26
N ₂ + CO	5	Kr	2	¹³⁶ Xe	44
O ₂	1				

The puncture gas contained a total of 1.85×10^9 dpm of ⁸⁵Kr, representing a release of 0.16% of the 1.16×10^{12} dpm found in the rod (Table 9). By comparison, Lynam reported fission gas releases of 0.26 to 0.93% in the blanket rods.¹¹ We actually accounted for 88% of the calculated yield of ⁸⁵Kr for Rod 10, Bundle 0320 (Appendix C), but recovered slightly more than the calculated yield for the mirror image rod. We did not count the molecular sieve traps used during the leaches of the end plugs and the unfueled cladding; however, based on the ratio of dpm ⁸⁵Kr per mg U in the rod, the quantity of ⁸⁵Kr would be insignificant.

Tritium in Cladding

Samples of leached cladding were heated to 1400°C in a high vacuum apparatus to release gases contained within the metal. A known "spike" of deuterium in neon was added to determine the degree of recovery of sample after mass spectrometer analysis for H₂, D₂ and T₂. The average analyses were:^{16,17}

Table 9. Release of ^{85}Kr From Rod 10

Sample ^a	Volume (ml)	^{85}Kr		% Total Released ^b
		dpm/ml	Total dpm	
L-1 Off-gas	4,600	$\leq 8.5 \times 10^3$	$\leq 3.91 \times 10^7$	<0.01
L-2 Off-gas	4,900	1.34×10^5	6.57×10^8	0.06
L-3 Off-gas	16,500	2.18×10^7	3.60×10^{11}	31.03
L-3 Mol. Sieve	-	-	2.23×10^9	0.19
L-4 Off-gas	17,000	2.73×10^7	4.65×10^{11}	40.09
L-4 Mol. Sieve	-	-	1.24×10^9	0.11
L-5 Off-gas	9,500	3.59×10^5	3.41×10^9	0.29
L-6 Off-gas	6,500	5.40×10^4	3.51×10^8	0.03
Puncture Gas	1.232	1.50×10^9	1.85×10^9	0.16
			8.34×10^{11}	71.96 ^b

^aSee Table 2 for identification of leaches (L-).

^bRemaining fraction in undissolved Segment #5 and cutting losses. The total quantity of ^{85}Kr was calculated to be:

$$8.34 \times 10^{11} \text{ dpm } ^{85}\text{Kr} \times \frac{152.06 \text{ g UO}_2}{127.47 \text{ g UO}_2} = 1.16 \times 10^{12} \text{ dpm } ^{85}\text{Kr in rod.}$$

Cladding Segment Numbers	T_2 ppm	H_2 ppm
2, 3, 4	< 0.02	64-130
6, 7, 8, 9	< 0.02	9

These analyses for segments 2, 3, and 4 generally confirm those of Westinghouse for the hydrogen content of the cladding; their analyses ranged from 52 to 93 ppm after Seed 4.⁴ The tritium content was too low to be measured by mass spectrometry.

We dissolved four samples of Zircaloy-2 cladding in Zirflex Reagent, 6 M NH_4F --1 M NH_4NO_3 . Three of these samples had been leached with 12 M HNO_3 and one had not been leached. Two samples (L-5 and L-6) were dissolved in the hot cell using the usual apparatus shown in Fig. 2, and two (L-7 and L-8) were removed from the hot cell and dissolved in the laboratory. In the laboratory, we used two gas bubbler-scrubbers containing 1 N H_2SO_4 to remove ammonia from the gas stream, and a dry-ice-cooled cold trap was used to remove the water vapor. The scrubbers were inserted between the reflux condenser and the copper oxide unit. All solutions and gases were analyzed for tritium after each dissolution (Table 10).

Table 10. Tritium Content of Zircaloy-2 Cladding

Leach No.	Sample from Segment No.	Weight, g	3H_2 dpm $\times 10^{-8}$				3H_2 dpm/g Zr-2
			Solution	Scrubbers	Mol. Sieve	Gas	
L-5 ^a	2,3,4	1.35	0.32	none	6.15	0.02	4.81×10^8
L-6 ^b	2,3,4	2.10	0.68	none	11.00	0.01	5.48×10^8
L-7 ^a	2,3,4	1.46	0.36	0.01	3.64	<0.01	2.75×10^8
L-8 ^a	2,3,4	1.02	0.33	<0.01	3.23	<0.01	3.49×10^8

^aLeached with nitric acid.

^bNot leached with nitric acid.

A negligible amount of tritium was found in the sulfuric acid scrubbers and, since 90-95% was found as water on the molecular sieve, the results suggest that the tritium was probably in the metal as zirconium tritide. Metallographic examination of Zircaloy cladding at Westinghouse showed that the hydrogen was present as hydride platelets distributed near the outer (cooler) portion of the cladding.⁴ The tritide-hydride apparently reacted with the water in the Zirflex reagent, releasing T_2-H_2 rather than being evolved as NH_3 or NT_3 . Greater than 80% of the yield of ammonia from the Zircaloy was found in the scrubber. Another possibility is that the tritium may have been held as a gas in the Zircaloy lattice; however, the metallographic evidence of hydride platelets suggests that this is not the case. Additional laboratory work to answer this question is indicated.

Assuming an average tritium content of 3.68×10^8 dpm/g (L-5, L-7, L-8) for the Zircaloy, the 29.75 g of sidewall cladding would contain 1.13×10^{10} dpm of tritium. (The two end plugs, on the basis of total tritium per unit area of interior surface, would contain only about 2×10^7 dpm.) This quantity of tritium is equivalent to about 17 parts per billion by weight, thus confirming the mass spectrometer analyses.

Tritium in the UO_2

Leach solutions 1 through 4 were analyzed for tritium, other fission products, and uranium and plutonium (Table II). The dissolved fuel, as a whole, contained 1.10×10^6 dpm/mg U. Thus, we recovered 1.237×10^{11} dpm of 3H_2 from 127.49 g of UO_2 in L-1 through L-4, or a total of 1.483×10^{11} dpm from all of the fuel in the rod.

Tritium in Puncture Gas

After determination of the ^{85}Kr content, we analyzed 0.546 cc of the puncture gas for tritium, and found 7.53×10^4 dpm. The total volume, 1.232 cc, therefore contained 1.70×10^5 dpm, or about 0.001% of the tritium in the rod.

Table 11. Tritium Content of UO₂ Fuel

Leach No.	Vol., ml	³ H ₂ in Solution, dpm Σ	Other ³ H ₂ ^a dpm Σ	U, Σ mg	³ H ₂ dpm/mg U
1	98	2.10 × 10 ⁶	0.37 × 10 ⁶	2.2	1.12 × 10 ⁶
2	100	5.16 × 10 ⁷	0.02 × 10 ⁷	63.9	8.11 × 10 ⁵
3	250	5.32 × 10 ¹⁰	0.02 × 10 ¹⁰	4.75 × 10 ⁴	1.12 × 10 ⁶
4	250	6.98 × 10 ¹⁰	0.03 × 10 ¹⁰	6.47 × 10 ⁴	1.08 × 10 ⁶

^aMolecular sieve traps, off-gases, etc.

Overall Tritium Distribution

Tritium distribution in the complete rod was:

Zircaloy cladding:	1.13 × 10 ¹⁰ dpm = 7.08%
UO ₂ fuel pellets:	1.483 × 10 ¹¹ dpm = 92.92%
Puncture gas:	1.70 × 10 ⁵ dpm = < 0.01%
Total	1.596 × 10 ¹¹ dpm = 100%

We recovered 99.4% of the theoretical yield of tritium (Appendix C), indicating that there was no significant loss of tritium from this blanket rod, which had operated at a time-averaged linear heat rating of 4.84 kw/ft and a clad temperature of 259°C. About 93% was in the UO₂ fuel and 7% in the Zircaloy-2 cladding.

REFERENCES

1. J. H. Goode and V. C. A. Vaughen, ORNL Experiments on the Behavior of Tritium During Head-End Processing of Irradiated Reactor Fuels, ORNL-TM-2793 (February 1970).
2. H. M. Roth, USAEC, Letter to J. A. Swartout, Deputy Director, Oak Ridge National Laboratory (April 1964).

3. J. C. Stachew, Isotopic Analysis of Natural UO_2 Fuel Irradiated to 22,000 Mwd/MTU: Theory vs Experiment, WAPD-TM-766 (August 1968).
4. A. J. Fiorelli and C. Santore, ed., PWR Core 1 Component Examination Program Summary Report, WAPD-320 (October 1969).
5. J. H. Goode, Hot Cell Evaluation of the Release of Tritium and Krypton-85 During Processing of ThO_2 - UO_2 Fuels, ORNL-3956 (April 1966).
6. M. E. Meek and B. F. Rider, Summary of Fission Product Yields for ^{235}U , ^{238}U , ^{239}Pu , and ^{241}Pu , APED-5398, Class I (March 1, 1968).
7. Dudey, et al., Chem. Eng. Highlights, May 1967 to April 1968, ANL-7450.
8. Fuels and Materials Development Program, Quarterly Progress Report for Period Ending March 3, 1969, ORNL-4420 (August 1969), pp. 25-28.
9. R. L. Henke, ed., Summary Design Report for Melba Test in PWR Core 2 Blanket, WAPD-TM-483 (March 1966).
10. M. Peterson, Bettis Atomic Power Laboratory, Personal Communication.
11. L. R. Lynam, Metallurgical Examination of PWR Core 1 Blanket Fuel Rods at the End of the Third Seed Life, WAPD-TM-433 (August 1964).
12. B. Rubin and L. R. Lynam, "Examination of Fuel and Cladding in PWR-1 Blanket Rods," Nucl. Appl. 2, 499-506 (1966).
13. C. M. Cox and F. J. Homan, PROFIL - A One-Dimensional FORTRAN IV Program for Computing Steady-State Temperature Distributions in Cylindrical Ceramic Fuels, ORNL-TM-2443 (March 1969).
14. R. R. Asamoto, F. Anselin, and A. E. Conti, The Effect of Density on the Thermal Conductivity of Uranium Dioxide, GEAP-5493 (April 1968).
15. G. A. Moore, ORNL, personal communication.
16. J. R. Sites, ORNL Analytical Chemistry Division, personal communication.
17. C. E. Lamb and F. L. Layton, ORNL Analytical Chemistry Division, personal communication.

APPENDIX A

Experimental Material Balance (Rod 10, Bundle 0320)

Starting wt. rod	197.90 g		
Our cut wt.	191.46 g	(1.034 factor to original rod)	
Wt. end plugs	<u>-13.73 g</u>		
Wt. fueled tubing	177.73 g		
Archive sample (#5)	<u>-23.37 g</u>	(3.78 g tubing, 19.59 g UO ₂)	
Fuel + clad for expts.	154.36 g	(Seg. #2, 3, 4, 6, 7, 8, 9)	
Seg. #2, 3, 4 Σ	66.61 g	Seg. #6, 7, 8, 9 Σ	87.73 g
Leached clad	<u>-17.33 g</u>	Leached clad	<u>-14.64 g</u>
UO ₂ wt.	55.28 g	UO ₂ wt.	73.09 g
UO ₂ by anal.	<u>-53.99 g</u>	UO ₂ by anal.	<u>-73.48 g</u>
UO ₂ Difference	1.29 g (-2.3%)	UO ₂ difference	0.89 g (+ 1.2%)
Total UO ₂ found (by anal.)		127.47 g	
Total cladding (by wt.)		<u>25.97 g</u>	
Fuel + clad found		153.44 g	
Fuel + clad input		<u>154.36 g</u>	
Difference		0.92 g (-0.60%)	
Recovery		99.40%	

APPENDIX B

Experimental Material Balance

UO ₂ in Seg. #2, 3, 4	53.99 g
UO ₂ in Seg. #5	19.59 g
UO ₂ in Seg. #6, 7, 8, 9	<u>73.48 g</u>
Total UO ₂	147.06 g x 1.034 cutting loss = 152.06 g

Zr-2 in end plugs	13.73 g
Zr-2 in Seg. #2, 3, 4	11.33 g
Zr-2 in Seg. #5	3.78 g
Zr-2 in Seg. #6, 7, 8, 9	<u>14.64 g</u>
Total Zr-2	43.48 g x 1.034 cutting loss = 44.96 g

152.06 g UO₂ + 44.96 g Zr-2 = 197.02 g total found.

Starting wt.	197.90 g
Calc. starting wt.	<u>-197.02 g</u>
Difference	0.88 g (-0.44%)
Recovery	99.56%

APPENDIX C

Calculated Fission Product Yields and Recoveries

^{137}Cs : From Table 6, there are 4.12×10^{19} atoms $^{137}\text{Cs}/\text{cm}^3$ present today.

$$\frac{4.12 \times 10^{19} \text{ atoms/cm}^3}{6.023 \times 10^{23} \text{ atoms/mole}} = 6.84 \times 10^{-5} \text{ moles } ^{137}\text{Cs/cm}^3.$$

$$6.84 \times 10^{-5} \text{ moles/cm}^3 \times 1.37 \times 10^2 \text{ g/mole} = 9.371 \times 10^{-3} \text{ g } ^{137}\text{Cs/cm}^3.$$

$$9.371 \times 10^{-3} \text{ g/cm}^3 \times 9.85 \times 10^4 \text{ mCi/g} = 9.230 \times 10^2 \text{ mCi/cm}^3.$$

$$9.230 \times 10^2 \text{ mCi/cm}^3 \times 2.22 \times 10^9 \text{ dpm/mCi} = 2.05 \times 10^{12} \text{ dpm } ^{137}\text{Cs/cm}^3.$$

$$1.494 \times 10^1 \text{ cm}^3 \times 2.05 \times 10^{12} \text{ dpm/cm}^3 = 3.06 \times 10^{13} \text{ dpm } ^{137}\text{Cs in rod today.}$$

$$\frac{2.73 \times 10^{13} \text{ dpm } ^{137}\text{Cs recovered}}{3.06 \times 10^{13} \text{ dpm } ^{137}\text{Cs yield}} = 89\% \text{ recovery of calculated yield.}$$

^{85}Kr : Calculations indicated 75% total fissions due to ^{239}Pu and 25% due to ^{235}U (p. 11, text). Yield of ^{85}Kr should therefore be:

$$(0.75)(1.215 \times 10^{-1}\% \text{ yield}) + (0.25)(3.049 \times 10^{-1}\% \text{ yield}) = 0.09112 + 0.07622$$

$$^{85}\text{Kr yield} = 0.1673\%.$$

From Table 6, 7.65×10^{20} fissions/ $\text{cm}^3 \times 1.494 \text{ cm}^3 = 1.146 \times 10^{22}$
total fissions in rod.

$$1.146 \times 10^{22} \text{ fissions} \times 1.673 \times 10^3 \text{ atoms/fission} = 1.917 \times 10^{19} \text{ atoms } ^{85}\text{Kr formed.}$$

$$\frac{1.917 \times 10^{19} \text{ atoms } ^{85}\text{Kr}}{6.023 \times 10^{23} \text{ atoms/mole}} = 3.182 \times 10^{-5} \text{ moles } ^{85}\text{Kr formed.}$$

$$3.182 \times 10^{-5} \text{ moles} \times 8.5 \times 10^1 \text{ g/mole} = 2.705 \times 10^{-3} \text{ g } ^{85}\text{Kr formed in rod.}$$

$$2.705 \times 10^{-3} \text{ g} \times 4.061 \times 10^2 \text{ Ci/g} = 1.098 \times 10^{-1} \text{ Ci } ^{85}\text{Kr formed in rod.}$$

$$1.098 \text{ Ci} \times 2.22 \times 10^{12} \text{ dpm/Ci} = 2.437 \times 10^{12} \text{ dpm } ^{85}\text{Kr formed.}$$

$$2.437 \times 10^{12} \text{ dpm} \times 0.54 \text{ decay factor} = 1.316 \times 10^{12} \text{ dpm } ^{85}\text{Kr today.}$$

$$\frac{1.16 \times 10^{12} \text{ dpm } ^{85}\text{Kr recovered}}{1.32 \times 10^{12} \text{ dpm } ^{85}\text{Kr formed}} = 88\% \text{ recovery of calculated yield.}$$

Tritium: From Table 6, there were

$$1.00 \times 10^{17} \text{ atoms/cm}^3 \times 1.494 \times 10^1 \text{ cm}^3 = 1.494 \times 10^{18} \text{ atoms tritium present today.}$$

$$\frac{1.494 \times 10^{18} \text{ atoms tritium}}{6.023 \times 10^{23} \text{ atoms/mole}} = 2.48 \times 10^{-6} \text{ moles tritium in rod.}$$

$$2.48 \times 10^{-6} \text{ moles} \times 3 \text{ g/mole} = 7.44 \times 10^{-6} \text{ g tritium present.}$$

$$7.44 \times 10^{-6} \text{ g} \times 9.73 \times 10^6 \text{ mCi/g} = 7.239 \times 10^1 \text{ mCi tritium present.}$$

$$7.239 \times 10^1 \text{ mCi} \times 2.22 \times 10^9 \text{ dpm/mCi} = 1.61 \times 10^{10} \text{ dpm tritium in rod on April 1, 1970.}$$

$$\frac{1.60 \times 10^{10} \text{ dpm } ^3\text{H}_2 \text{ recovered}}{1.61 \times 10^{10} \text{ dpm } ^3\text{H}_2 \text{ formed}} = 99.4\% \text{ recovery of calculated yield.}$$

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