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HOT-CELL EVALUATION OF THE RELEASE OF TRITIUM AND ^{85}KRYPTON DURING PROCESSING OF ThO_2-UO_2 FUELS

J. H. Goode

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ORNL-3956

Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION

Chemical Development Section B

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JUNE 1966

OAK RIDGE NATIONAL LABORATORY
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CONTENTS

Page

Abstract 1

1. Introduction 1

2. Experimental 3

2.1 Irradiation Data and Theoretical Tritium Yield 3

2.2 Apparatus and Procedures 4

3. Results and Discussion 9

3.1 System Calibration 9

3.2 Gas Evolution During Shearing 9

3.3 Fuel Dissolution 12

3.4 Feed-Adjustment Distillation 12

4. Discussion of Results 12

5. References 16

Appendix A 17

Appendix B 18

HOT-CELL EVALUATION OF THE RELEASE OF TRITIUM AND ^{85}KRYPTON DURING PROCESSING OF ThO_2-UO_2 FUELS

J. H. Goode

ABSTRACT

Hot-cell experiments with prototype ThO_2-UO_2 power reactor fuel specimens irradiated to 24,600 Mwd/metric ton (Th + U) showed that up to 0.2% of the fission-produced tritium and 1% of the ^{85}Kr were released when the fuel was sheared into short lengths. In our apparatus, another 0.2% of the tritium and the balance of the krypton were released to the atmosphere when the fuel was dissolved to make solvent extraction feed. This quantity of tritium is less than 0.5% of the estimated 1200 curies of ^{3}H that may be safely discharged to the atmosphere from a 6-ton-per-day processing plant.

In the preparation of solvent extraction feed, the acidic dissolver solution, containing about 430 curies of tritium per ton of fuel, is evaporated and the excess acid recovered in a rectification column. The solvent extraction feed would contain an estimated 30% of the tritium, the rectifier overhead about 47%, and the concentrated acid about 23%. This rectifier overhead, containing 200 curies of tritium per ton of fuel, must be diluted with 18 million gallons of environmental water to permit discharge at concentrations less than 1/10 the MPC_w for tritium (3×10^{-3} μc/cc). Further dilution to 1×10^{-3} μc/cc would be required at the plant boundary to meet the MPC_w for tritium for the general population. Alternative tritium control measures would be volatilization of the tritiated water to disperse it into the atmosphere or recycle of the water within the plant.

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1. INTRODUCTION

The formation of tritium, 12.26-year ^{3}H, during the fission of uranium[1-3] presents a unique problem in the management of wastes from a radiochemical processing plant because tritium is neither separated nor concentrated by conventional procedures for waste treatment. Blomeke[4] has calculated from predictions of nuclear power growth in the U.S. that the annual production of tritium may be expected to rise from about 15,000 curies in 1968 to about 6 million in the year 2000. Since a fuel-

processing plant has only its immediate environs for dispersal, the capacity of the plant and its environs depends on the irradiation level of the fuel, the rate of tritium release, its chemical state, and on factors that are peculiar to the plant site, such as the availability of ground water for dilution. To learn more about some of the problems, we have taken irradiated prototype Consolidated Edison ThO_2-UO_2 fuel and traced the path of tritium through the processing steps of: (1) mechanical disassembly and shearing, (2) chemical dissolution of the sheared fuel, and (3) solvent extraction feed preparation.

Albenesius and Ondrejcin[2] measured a ratio of $(1.05 \pm 0.09) \times 10^4$ fissions per tritium atom for natural-uranium fuel rods irradiated in a heavy-water-moderated and cooled reactor, and Sloth et al.[3] measured a ratio of $(1.25 \pm 0.15) \times 10^4$ fissions per atom for normal uranium irradiated as U_3O_8. Horrocks proposed the use of tritium to measure burn-up of fissionable nuclides[5] and has measured ratios of 1.25×10^4 fissions per tritium atom for ^{235}U[6] and 1.41×10^4 for ^{233}U.[7] Albenesius and Ondrejcin[2] stated that about 25% of the yield of tritium appeared in the dissolver off-gas, indicating that 75% remained with the dissolver solution.

Blomeke estimated that a hypothetical power-reactor fuel-processing plant operating at a capacity of 6 tons per day on fuel irradiated to 16,000 Mwd/metric ton at a power of 16 Mw could release to the atmosphere about 1200 curies of tritium per day through a 100-m-high stack under very unfavorable conditions without exceeding 1/10 the maximum permissible concentration in air (MPC_a) for continuous occupational exposure at any point near the ground (2×10^{-7} μc/cc).

In this study, we assumed that bundles of stainless steel-clad ThO_2-UO_2 fuel rods would be mechanically disassembled, sheared into short lengths for dissolving the core in 13 M HNO_3--0.05 M HF--0.1 M $Al(NO_3)_3$, and that the thorium and uranium would be recovered by solvent extraction using the Acid Thorex process.[8] One important step is the feed adjustment treatment (FAT), in which the solvent extraction feed is made acid deficient[a] by

[a]That is, contains less than the stoichiometric amount of nitrate for $Th(NO_3)_4$, e.g., $Th(OH)(NO_3)_3$.

distilling nitric acid from the solution. This acid condensate is normally rectified to recover the nitric acid for reuse, and the overhead dilute acid is sent to a low or intermediate radioactivity-level waste stream.

We acknowledge the assistance of C. E. Boyd and A. S. Meyer, Jr., ORNL Analytical Chemistry Division, for the design and testing of the apparatus for conversion of tritium to tritiated water and the adsorption of ^{85}Kr. J. S. Eldridge, Analytical Chemistry Division, provided technical assistance, the liquid scintillation spectrometer for tritium counting, and the gamma spectrometer for krypton measurement. Technicians L. A. Byrd and G. E. Woodall performed the hot-cell manipulations. E. D. Arnold and J. W. Ullmann furnished reactor-operating data and aided in calculating the fission yields.

2. EXPERIMENTAL

2.1 Irradiation Data and Theoretical Tritium Yield

Sixty prototype Consolidated Edison fuel specimens were irradiated in positions J-21 and L-15 of the NRX reactor for 505 days (February 25, 1960, to November 20, 1961) at unperturbed neutron fluxes of 4.3 to 5.6 x 10^{13} to a calculated peak burnup of 25,600 Mwd/metric ton (Th + U). Each type 304 stainless-steel-clad specimen (4.5-in. long and 5/16 in. in diameter) contained seven pressed and fired pellets of 96% theoretical density. The composition of the unirradiated pellets was 95.55% ThO_2 and 4.45% UO_2 (93.24% enriched in ^{235}U). Table 1 summarizes the weight of thorium and uranium in each of the eight specimens, and the calculated perturbed flux of each.

The average burnup for 17 specimens in the irradiation holder was calculated from ^{137}Cs analysis to be 24,600 Mwd/metric ton (Th + U). This converts (Appendix A) to 7.50 x 10^{20} fissions per cc, or a total of 2.29 x 10^{21} fissions for the 3.05 cc of fuel, 20% due to ^{233}U and 80% due to ^{235}U. Using Horrocks' ratios of 1 tritium atom per 1.25 x 10^4 fissions of ^{235}U and 1.41 x 10^4 fissions of ^{233}U, we calculated that

Table 1. ThO_2-UO_2 Specimens for Tritium Experiments

Specimen Number	Th (g)	U (g)	^{235}U (g)	Perturbed Flux (x 10^{13})
672	23.6	1.1	1.0	4.00
677	23.7	1.1	1.0	4.44
699	23.7	1.1	1.0	4.41
704	23.7	1.1	1.0	4.01
719	23.8	1.1	1.1	4.18
720	23.7	1.1	1.1	4.33
725	23.7	1.1	1.1	4.20
826	23.7	1.1	1.1	4.01

1.793×10^{17} tritium atoms were formed in each specimen. In-pile decay of 4% left 1.720×10^{17} atoms at discharge, and 20% decay before use in 1966 left 1.375×10^{17} atoms, or a theoretical total of 6.70 mc of ^3H per specimen, based on a specific activity of 9.78×10^3 curies per gram of ^3H.[9]

2.2 Apparatus and Procedures

Figure 1 shows the in-cell hydraulic shear, tilting McLeod gage, thermocouple gage, and manometer used for cutting the irradiated specimens and measuring the volume of gas released. Total volume of the shear and gages was 300 cc; the inleakage rate of this part of the system was 0.45 μ/min after evacuating the shear to about 15 μ pressure.

A welded stainless steel tube connected the shear with the analysis system (Fig. 2) located in a glove box outside of the hot cell. The volume of tubing to the analytical apparatus was 453 cc. A Toepler pump moved the fission gases from the shear into an evacuated storage bulb. The shear and transfer lines were flushed and evacuated four or five times with 3-cc volumes of hydrogen to isotopically exchange with tritium

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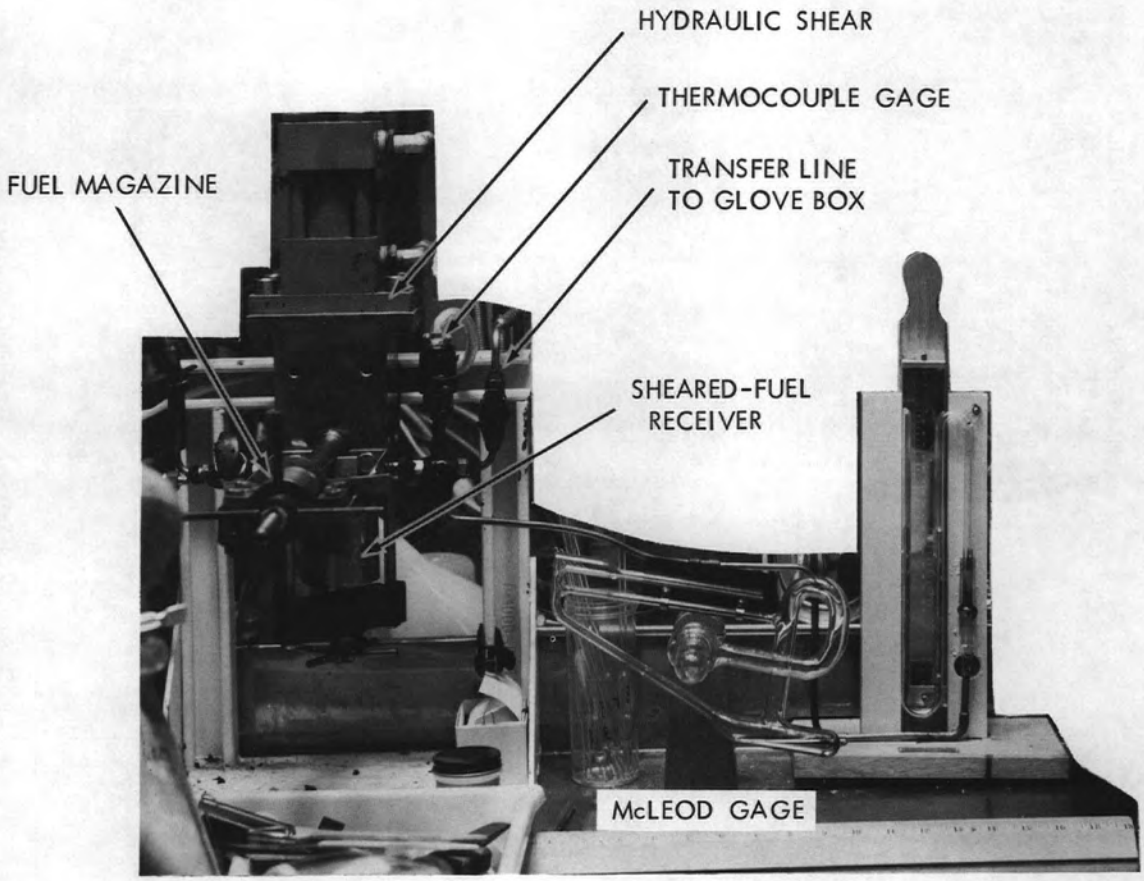

Fig. 1. Hot-Cell Shear and Pressure Measuring Devices.

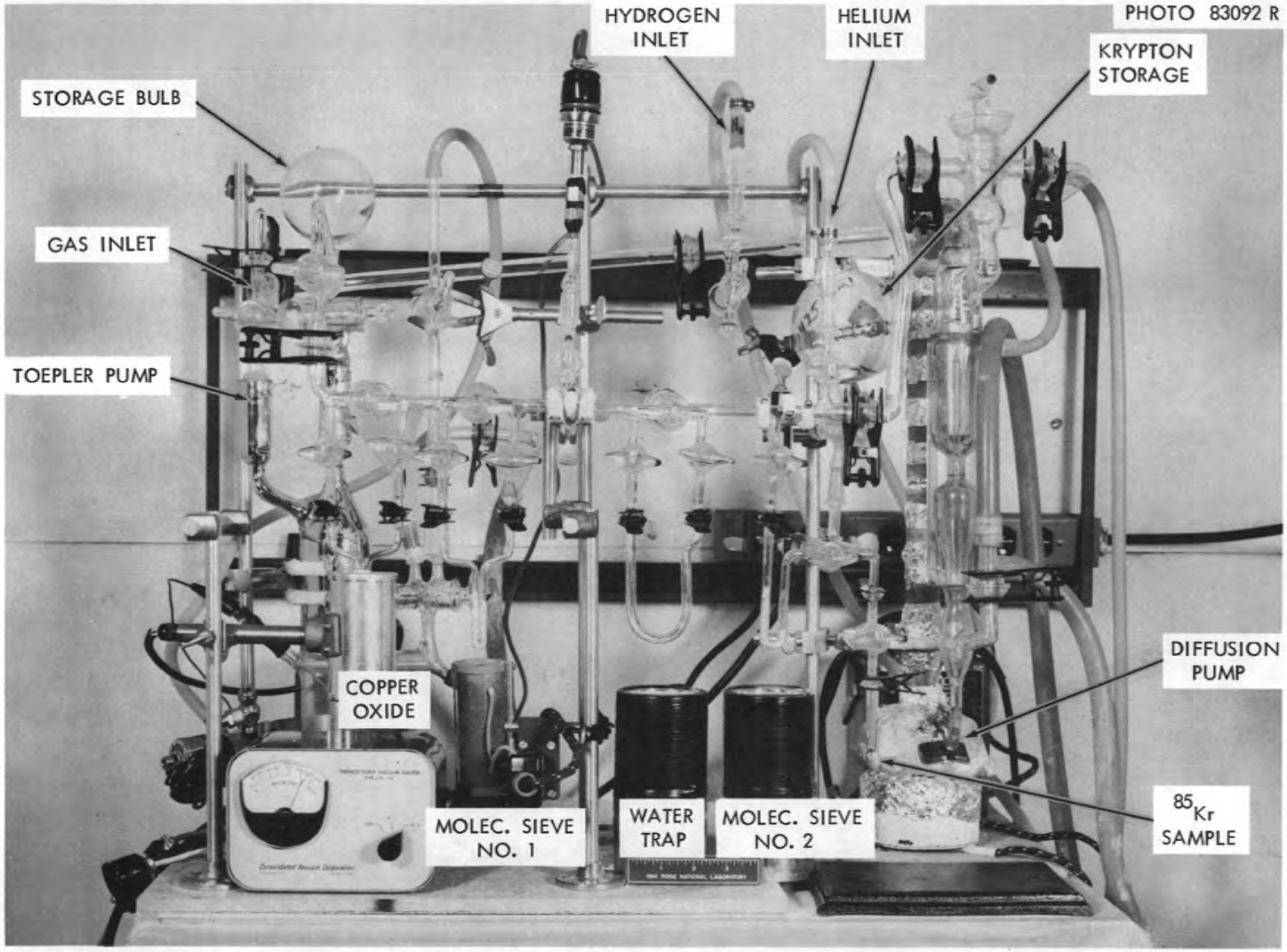

Fig. 2. Tritium-Krypton Analyzer.

adsorbed in the system and to provide a carrier gas. These mixed gases were cryogenically pumped from the storage bulb through CuO at 800°C to convert tritium to tritiated water; and the THO, xenon, and krypton were adsorbed on 13X molecular sieve No. 1 at liquid-nitrogen temperature.

Unadsorbed helium, from back-filling of the specimens prior to irradiation, was pumped off, and the molecular sieve was heated to 400°C for 30 min to drive off the THO, xenon, and krypton. The water was frozen out of the gases in a liquid-nitrogen-cooled trap and the xenon and krypton adsorbed on molecular sieve No. 2 at liquid-nitrogen temperature. The frozen THO was melted and refrozen three or four times to ensure release of the ^{85}Kr. This ^{85}Kr was then adsorbed on the second molecular sieve.

The THO was taken up in 15 ml of dioxane-modified PPO-POPOP scintillation liquid (Appendix B), and counted at 9.85% efficiency in a "Tri-Carb" liquid scintillation spectrometer manufactured by the Packard Instrument Company. The counts were compared with those for a tritiated water standard to determine counting efficiency. The purity of the experimental THO was determined by both the dual-channel scintillation ratio[7] and by scanning for fission product contamination in a gamma spectrometer. Typical counting rates were about 1.81×10^5 counts/min for the THO samples.

The adsorbed krypton and xenon were flushed from molecular sieve No. 2 with helium, after warming, into a 440-cc storage bulb, from which krypton-counting aliquots were taken.

Fission gases evolved from dissolving ThO_2-UO_2 fuel were collected in the apparatus shown in Fig. 3. A carrier gas, 0.2% hydrogen in helium, moved the dissolution vapors through the condenser and 10 M NaOH scrubber located in the hot cell, to a glove box system which included a tube furnace containing CuO at 800°C and a trap for THO at -77°C.

The tritium (as THO) content of the dissolved fuel and caustic scrubber solutions was determined by distillation and scintillation counting. The fuel solutions were distilled without neutralization (to prevent bumping) until the solution reached >136°C, the temperature

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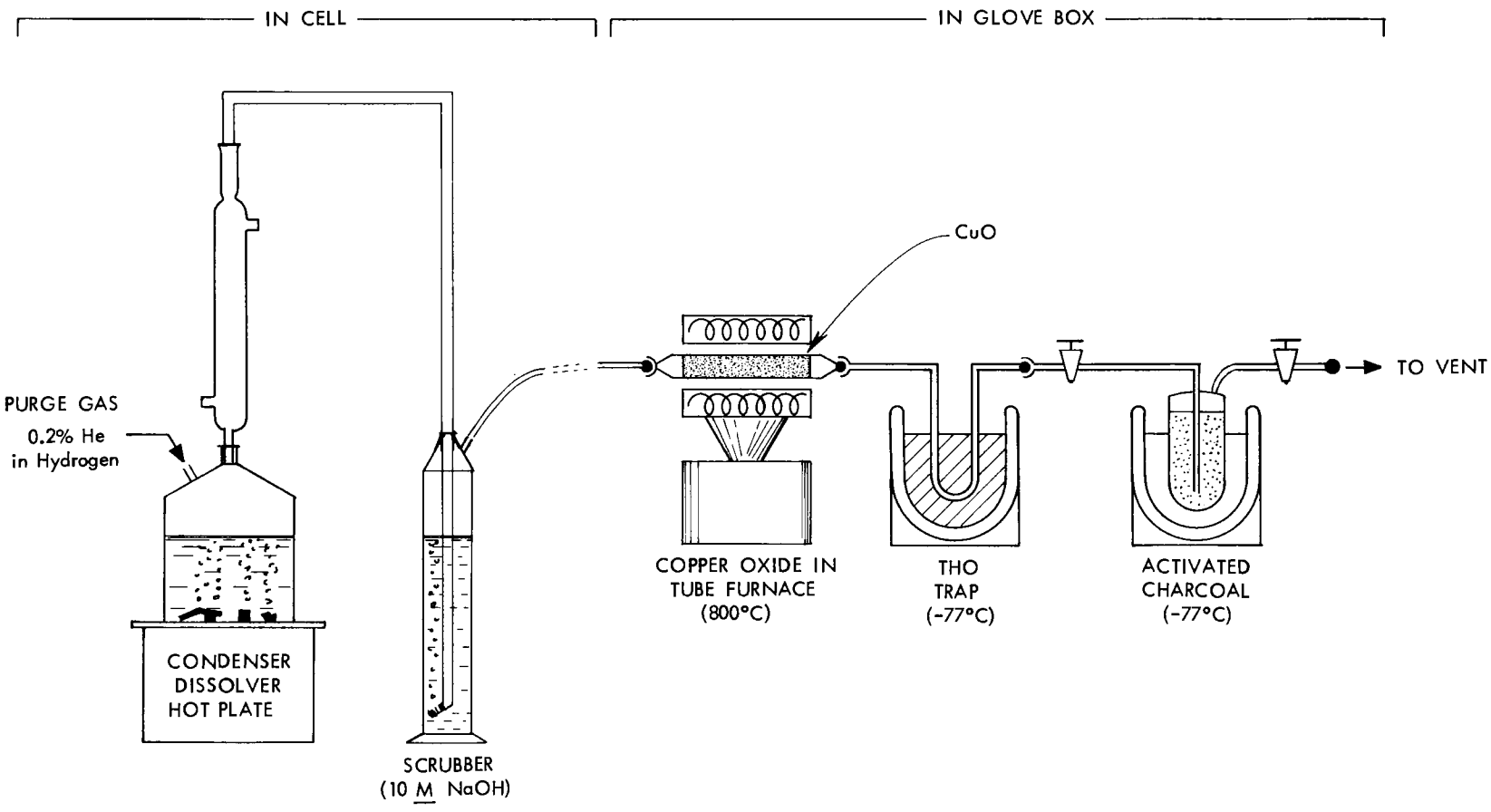

Fig. 3. Equipment for Determination for Tritium in Dissolution Vapors.

necessary to make a solvent extraction feed about 0.1 M acid deficient. The acid condensates were removed from the hot cell, diluted with an equal volume of 10 M NaOH, and redistilled to decontaminate them from $^{106}RuO_4$. Two distillations were necessary to achieve adequate purity. The caustic scrubber solutions required only one purification. Counting rates for 50 λ of the final condensate were about 2.88×10^5 counts per min.

3. RESULTS AND DISCUSSION

3.1 System Calibration

A glass capsule containing 0.721 cc of a T_2-H_2 standard was crushed in the shear, and the released gases analyzed. A 100% yield (2400 counts per min) was obtained when the tritiated water was counted in the scintillation spectrometer.

3.2 Gas Evolution During Shearing

Table 2 summarizes chronologically the shearing experiments, including refinements based on experience, and shows that as much as 20.4 μc of tritium gas was released from the specimens. The increase in tritium content from 0.64 to 1.80 μc during the first four experiments suggested that tritium had diffused from fuel segments held in the shear. A blank experiment (No. 5) was made in which the shear and connecting lines were equilibrated several times with hydrogen and the gases analyzed; we found 1.17 μc of ^3H, which indicated that diffusion had taken place. The shear was therefore emptied after each experiment.

Specimens 720 and 725 were each cut eight times (about 0.5-in. lengths) and the released fissions gases analyzed immediately and again after diffusion overnight in the evacuated shear. An additional 0.58 to 1.20 μc of ^3H were found after the diffusion period, and, in the case of specimen 725, the system was purged with hydrogen a third time to remove residual tritium; 0.40 μc was recovered.

Table 2. Release of Tritium and Krypton from Sheared ThO_2-UO_2 Fuel Rods Irradiated to 24,600 Mwd/metric ton (Th + U)

Experiment Number	Specimen Number	No. Cuts	Gas Vol. (cc)	^3H Found (μc)	^3H Released, (%)	^{85}Kr Found (mc)	^{85}Kr Released (%)
1	704	1	0.50	0.64	0.006	1.96	0.50
2	672	1	0.45	0.85	0.008	--	--
3	719	1	0.45	1.44	0.018	1.83	0.47
4	726	1	0.48	1.80	0.017	2.16	0.55
5	Blank[a]	0	--	1.17	--	--	--
6A	677[b]	1	0.50	neg.	--	--	--
6B	677[c]	5	--	1.95	0.018	--	--
7A	720[d]	8	0.75	2.40	0.022	4.26	1.08
7B	720[e]	--	--	0.58	0.005	--	--
8A	725[d]	8	0.68	18.80	0.202	3.32	0.84
8B	725[e]	--	--	1.20	0.013	0.11	0.03
8C	725[f]	--	--	0.40	0.004	--	--
9	699[d,e]	17	2.0[d]	11.50	0.108	--	--

[a] H_2 flush of shear and analysis system. Cut segments from four specimens left inside shear.

[b] Determination of $^3H_2$ or ^3H_2O. Bypassed CuO furnace; passed gases through water trap.

[c] Cut remainder of specimen into 0.5-in. lengths.

[d] Prompt release of fission gases.

[e] Overnight diffusion from cut segments inside shear plus H_2 flushing.

[f] Purged system with H_2 to flush lines.

The fission gas volume increased from about 0.5 cc for a single cut to 0.68-0.75 cc for eight cuts, indicating that shearing to short lengths releases more gas. Specimen 699, cut 17 times into 0.25-in.-long pieces (with considerable powdering of the fuel) released 2.0 cc of fission gas. Table 3 summarizes the amount of gas released by multiple shearing of the fuel specimens and shows that about twice as much gas was released from 0.25-in. cuts as from 0.5-in. cuts.

Table 3. Incremental Release of Fission Gas Upon Shearing ThO_2-UO_2 Fuel

		Specimen Number			
		726	720	725	699
Length of cut, in.		0.5	0.5	0.5	0.25
Volume released, cc		0.48	0.75	0.68	2.0
		Incremental Release, %			
Cut No.	1	100.0	58.8	64.7	41.4
	2	--	64.7	70.6	44.8
	3	--	70.6	76.5	48.4
	4	--	76.5	76.5	55.2
	5	--	82.5	82.5	62.2
	6	--	88.4	88.3	65.6
	7	--	94.1	94.1	65.6
	8	--	100.0	100.0	69.2
	9	--	--	--	69.2
	10	--	--	--	72.5
	11	--	--	--	72.5
	12	--	--	--	75.9
	13	--	--	--	82.8
	14	--	--	--	86.3
	15	--	--	--	93.3
	16	--	--	--	96.6
	17	--	--	--	100.0

Specimen 677 was cut once (experiment 6A), and the 0.5 cc of fission gases was flushed with hydrogen directly through the water trap, bypassing the CuO, to determine whether the tritium was present in the oxide fuel as $^3H_2$ or as ^3H_2O. Almost no tritium (about 780 counts/min) was found, indicating that only $^3H_2$ was present. The remainder of the specimen was

cut five more times (6B), releasing an additional 1.95 μc of tritium, which was converted to THO and counted.

3.3 Fuel Dissolution

The sheared segments from specimen 725 were refluxed for 6 hr in 100 ml of 13 M HNO_3--0.05 M HF--0.1 M $Al(NO_3)_3$, and the caustic-scrubbed off-gas was passed through the out-of-cell analyzer. We found that a total of 5.08 μc of tritium had passed through the scrubber, the balance remaining in the fuel solution. Specimen 719 was similarly dissolved, and we found that 4.12 μc had passed through the scrubber; 15.75 μc of tritium was found in the 200 ml of scrubber solution. A total of 0.2% of the theoretical yield, therefore, escaped the dissolver. We were not equipped for ^{85}Kr trapping during these experiments.

The fuel solutions from these two specimens were distilled and the condensates decontaminated of fission products. Specimen 725 contained 9.34 mc of ^{3}H; specimen 719, 8.01 mc. These unexpectedly large quantities of tritium (nearly 1.5 times the calculated amount: 6.70 mc) were confirmed by dissolving the six remaining specimens, combining the fuel solutions, and performing two feed-adjustment distillations on batches of 250 and 280 ml. The condensates were analyzed and gave an average value of 10.65 mc of ^{3}H per specimen, or about 430 curies per ton.

3.4 Feed-Adjustment Distillation

Feed-adjustment distillations were made on the fuel solution from specimen 719 and on two batches of solutions combined from specimens 672, 726, 677, 699, 704, and 720. Table 4 shows the results of the tests and confirms that the transfer of tritium to the condensate is directly proportional to the volume distilled.

4. DISCUSSION OF RESULTS

Hot-cell experiments on the release of tritium and krypton during reprocessing of prototype Consolidated Edison ThO_2-UO_2 power reactor fuel

Table 4. Transfer of Tritium to Condensate During Acid Thorex FAT Step

	Specimen Number		
	719	Comb. 1	Comb. 2
Tritium in fuel solution, mc/ml	0.089	0.107	0.106
Initial volume, ml	80	250	280
Evaporated volume, ml	24	100	100
Condensate volume, ml	56	150	180
Condensate volume, %	70	60.3	64.3
Tritium in condensate, %	69.3	60.5	64.3
Feed volume, ml	80	260	290
Feed acidity, M	0.09 AD	0.56	0.55

showed the following:

1. Shearing the fuel rods to very short lengths, that is, to less than 0.5 in., released the maximum amount of tritium and krypton to the stack for dispersal.

2. Diffusion of small amounts of the fission gases occurred during overnight storage of the newly segmented fuel at ambient temperature. Extended storage, especially at the calculated high temperatures in the storage baskets for the 4- to 5-month-decayed, sheared fuel (790°F center line to 355°F at basket surface),[10] should increase the rate of diffusion and disperse greater quantities of tritium and krypton into the atmosphere via the plant stack.

3. Our experiments showed that up to 20.4 μc of ^{3}H were released when irradiated ThO_2-UO_2 specimens containing 24.8 g of thorium plus uranium were sheared into 0.5-in. pieces and stored overnight. Based on an actual tritium content of 10.65 mc per specimen, only 0.19% of the total tritium was released, along with about 1% of the ^{85}Kr. This extrapolates to 0.82 curie of tritium per metric ton of thorium plus uranium or about 0.41% of the permissible 1200 curies per day that may be released to the atmos-

phere from a 6-ton-per-day processing plant. Our results also point out the difference in the amount of "free" tritium in various irradiated specimens. Whereas others found from 1/4 to 1/2 of the tritium present as a gas, we found less than 0.2% not retained in the solid. This is probably due to differences between their experiments with irradiated uranium metal or low-density U_3O_8 and our use of high-density ThO_2-UO_2 pellets. In other tests we found 9.27% of the theoretical yield of tritium present as a gas when we sheared Zircaloy-clad specimens containing UO_2 pellets of 93% theoretical density.

4. The source of the tritium above and beyond the theoretical fission yield was investigated. Flame-photometer analysis of a solution from an unirradiated fuel specimen of the same batch indicated that less than 3 ppm of lithium was present in the ThO_2-UO_2 pellets. A more refined method, in which the thorium and uranium were removed from solution by ion exchange followed by concentration of the column effluent, showed that less than 0.1 ppm of lithium was present. The $^6Li(n,\alpha)^3H$ reaction, with a cross-section of 950 barns, could produce 3×10^{14} atoms of tritium from this concentration of natural lithium, or less than 0.2% of the total tritium atoms produced by fission at discharge (1.72×10^{17}). Formation of tritium from the helium used to back-fill the specimens by the $^3He(n,p)^3H$ reaction is improbable due to the low abundance of ^3He present.

Horrocks[6] suggested a possible dependence of the tritium yield on the flux spectrum since his measurements were based on irradiations of lithium-free (<5 ppb) material in a purely thermal neutron flux in Argonne's CP-5 reactor. Our specimens were irradiated under reactor conditions such that a broad spectrum of neutrons was present.

5. About 0.20% of the tritium in the fuel and the remainder of the ^{85}Kr is released to the off-gas and stack during normal chemical dissolution of the sheared pieces; the balance of the tritium, 99.6%, stays in the fuel solution.

6. The solvent extraction feed-adjustment treatment by distillation resulted in the transfer of 70% of the tritium to the condensate. Since

normal radiochemical plant practice calls for rectification of this condensate to recover nitric acid for recycle, it follows that a proportionate amount of the tritium would report, as the overhead, to a normally low- or intermediate-level radioactive waste stream for disposal. Based on Thorex Pilot Plant experience at ORNL,[11] the volume of "lean acid" overhead was twice that of the concentrated acid; therefore, about 200 curies of tritium per ton would be discharged as a liquid waste.

As noted before, Blomeke[4] states that the quantity of tritium that can be released to surface waters (1) shall not exceed 1/10 the continuous, occupational exposure for water (MPC_w) of 3×10^{-3} μc/cc at the boundary of the controlled zone, and (2) it shall not subsequently exceed the ICRP-recommended level for the general population of 1/30 the MPC_w, or 1×10^{-3} μc/cc. If the acid-rectifier overhead, containing 200 curies of tritium from 1 ton of fuel, were to discharge to surface waters, about 18 million gallons of dilution water per ton of fuel would be required for it to be released into the controlled zone at 1/10 MPC_w. If a stream or small river emptying into a larger body of water for further dilution does not flow through the plant area, it thus appears that the tritium-bearing streams must be recycled within the processing plant to limit the release of tritium to the environment. When the tritium inventory of these streams reaches the level of that of the incoming fuel, they would be discharged for tank storage with high-level wastes. Other possible methods of tritium disposal could be through deep wells, concentration by electrolysis, or evaporation as steam into the atmosphere. Evaporation as steam may prove to be the best method of dispersal of the bulk of the tritium. Blomeke calculated that 150,000 cfm of stack gases, at 80°F and 65% relative humidity, can carry over 20,000 gallons of water per day without raising the relative humidity over 95%. If so, the overheads from waste evaporators could be picked up by the stack gases, instead of being condensed, and discharged to the atmosphere.

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APPENDIX A

Sample Calculation of Theoretical Tritium Yield

3.38×10^{16} fiss/sec/Mw $\times$ 8.64×10^{4} sec/da = 2.92×10^{21} fiss/Mwd

2.46×10^{4} Mwd/t $\times$ 10.06×10^{-6} t/cc = 2.463×10^{-1} Mwd/cc

2.92×10^{21} fiss/Mwd $\times$ 2.463×10^{-1} Mwd/cc = 7.50×10^{20} fiss/cc

7.50×10^{20} fiss/cc $\times$ 3.05 cc = 2.29×10^{21} fiss/specimen .

$2.29 \times 10^{21} \times 0.2 = 4.58 \times 10^{20}$ fissions due to ^{233}U

$2.29 \times 10^{21} \times 0.8 = 18.30 \times 10^{20}$ fissions due to ^{235}U

$$\frac{4.58 \times 10^{20} \text{ fiss } ^{233}U}{1.41 \times 10^{4} \text{ fiss/atom } ^{3}H} = 3.28 \times 10^{16} \text{ atoms } ^{3}H \text{ from } ^{233}U$$

$$\frac{18.30 \times 10^{20} \text{ fiss } ^{235}U}{1.25 \times 10^{4} \text{ fiss/atom } ^{3}H} = 14.65 \times 10^{16} \text{ atoms } ^{3}H \text{ from } ^{235}U$$

$3.28 \times 10^{16} + 14.65 \times 10^{16} = 1.793 \times 10^{17}$ total atoms ^{3}H formed

1.793×10^{17} atoms ^{3}H $\times$ 0.96 in-pile decay = 1.720×10^{17} atoms ^{3}H at discharge

1.720×10^{17} atoms ^{3}H $\times$ 0.80 out-of-pile decay = 1.375×10^{17} atoms ^{3}H at processing

$$\frac{1.375 \times 10^{17} \text{ atoms} \times 3 \text{ g/g-atom}}{6.025 \times 10^{23} \text{ atoms/at. wt.}} = 6.84 \times 10^{-7} \text{ g } ^{3}H \text{ present}$$

6.84×10^{-7} g ^{3}H $\times$ 9.78×10^{3} c/g = 6.70×10^{-3} c ^{3}H at processing .

APPENDIX B

Scintillation Liquid for Counting Samples of Tritiated Water

Place 80 g naphthalene, 5 g PPO (2,5 diphenyl oxazole), and 50 mg POPOP (1,4-bis-2(5 phenyl oxazolyl) benzene) in a 1-liter flask. Dissolve and dilute to 1 liter with a mixture of 385 ml xylene, 385 ml dioxane, and 230 ml ethyl alcohol.

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