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DEMONSTRATION OF AN IRRADIATED UO_2 FUEL RECYCLE:
FIRST RECYCLE

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INTRODUCTION

In the operation of nuclear power reactors the fuel elements are removed after only a fraction of the fissile atoms are consumed. This is necessary because the reactivity of the fuel is lowered by consumption of fissile atoms and build up of nuclear poisons (fission products). There is economic incentive for reprocessing of the spent fuel to remove nuclear poisons and to recover the unused fissile material for use. The reprocessing strategy most generally assumed is one in which spent fuel is shipped to a central reprocessing plant of high throughput and low unit cost. A plant of this kind, employing highly developed solvent extraction or ion exchange technology will produce a highly decontaminated final product. An alternative strategy is to employ reprocessing facilities closely coupled with the reactor(s) served. Advantages of this approach are low fuel cycle inventories and the elimination of shipping charges which may be very high on highly radioactive discharged fuel. Although the complications of processing after a short cooling period and in a low throughput on-site chemical plant must be reckoned with, it is assumed that close-coupled systems may have an economic future if simple processes can be developed.

The Salt Cycle Process is a promising close-coupled approach which is currently under development in the Hanford Laboratories. In this process a UO_2 end product is produced by electrolytic reduction of uranyl chloride formed by dissolution of irradiated UO_2 in a molten chloride salt phase. The process is inherently simple and allows concentration of plutonium and depletion of high cross-section fission products in the UO_2 end product. However, the process, like all nonaqueous processes, does not produce a plutonium-uranium product which is well decontaminated by

traditional solvent extraction process standards. Therefore, the Salt Cycle Process must be coupled with a fuel refabrication process designed to tolerate considerable gamma activity.

This document describes the results of the first integrated test of a fuel cycle based on processes of this kind. Irradiated UO_2 was cleaned up by the Salt Cycle Process, this fuel was made into a fuel element by a remotized fabrication process, the element was irradiated without incident and, finally, postirradiation examination was performed. This work comprises the first successful demonstration of a close-coupled UO_2 fuel recycle concept.

SUMMARY

Irradiated UO_2 was processed for recycle by one version of the Salt Cycle Process. The irradiated fuel was dissolved in fused NaCl-KCl and the bulk of the uranium and plutonium were recovered as UO_2 and PuO_2 by electrodeposition. A reduction in the concentration of fission product poisons in the fuel was also achieved in the electrodeposition step.

Fabrication of the partially decontaminated, irradiated UO_2 into a fuel rod and subsequent incorporation of the fuel rod into a four-rod cluster fuel element was accomplished by remote handling techniques. Two rods of the cluster contained nonrecycled, electrodeposited UO_2 , one contained sintered UO_2 pellets, and the fourth contained the recycled, electrodeposited UO_2 .

The element was irradiated in the MTR with a maximum surface heat flux of approximately 300,000 Btu/ (hr)(sq ft) to an exposure of approximately 450 MWD/ T.

The fuel rods containing electrodeposited UO_2 (both recycled and nonrecycled) suffered no adverse effects during irradiation.

RESULTS AND DISCUSSION

UO₂ Reprocessing

The recycle UO₂ was prepared by one version of the Salt Cycle Process in the first test of this process concept with irradiated UO₂. The flow sheet, shown in Figure 1, included cladding removal and oxide dissolution operations, along with electrolysis steps designed to yield product for recycle having a higher plutonium-to-uranium ratio than the feed. This was to be done by (a) performing a partition deposition to remove a significant portion of the uranium from the melt along with an insignificant portion of the plutonium, thus increasing the plutonium-to-uranium ratio in the melt; and (b) codepositing PuO₂ and UO₂ at a ratio approximately equal to the plutonium-to-uranium ratio in the melt. In the work reported herein, the partition deposition was accomplished successfully, but the codeposition electrolyses yielded deposits having low plutonium-to-uranium ratios. Minor alterations of conditions in subsequent work did result in satisfactory codeposition of PuO₂ with UO₂.

To remove the feed UO₂ from the cladding, the UO₂ was oxidized to finely divided U₃O₈ powder which fell from the cladding. During this operation fuel element sections contained in an oxidizer cartridge were suspended above the surface of the fused salt so the U₃O₈ fell directly onto the salt and no further transfer of this material was required. The cartridge was vibrated to facilitate removal of the U₃O₈ from the cladding. The oxidation, which was performed in an air atmosphere at about 450 C, required about four hours for completion with one inch long fuel element sections. A total of 445 grams of UO₂ was processed in this manner.

The U₃O₈ and PuO₂ were dissolved in three kilograms of equimolar NaCl-KCl with an HCl-Cl₂ sparge at 750 to 800 C to give a solution of uranyl chloride and plutonium(III) chloride. Dissolution required about three hours for a 200-gram batch of U₃O₈ with a gas rate of about 250 ml/min each of HCl and Cl₂.

FIGURE 1
Flow Sheet for UO₂ Reprocessing

The partition deposition was made at 750 C with a current of 20 amps and a current density of about 0.1 amp/cm^2 . About 20 per cent of the uranium and only about 0.1 per cent of the plutonium were removed from the melt in this deposit, which was smooth, dense, and adhered tenaciously to the graphite electrode. To prevent codeposition of PuO_2 with UO_2 , the plutonium was maintained in the melt as plutonium(III) chloride by excluding air from the system during deposition. This was done by allowing the chlorine produced at the anode to blanket the melt. The uranium(IV) chloride which was formed during the electrolysis by reaction of the graphite electrodes with chlorine and uranyl chloride also aided in maintaining an oxygen-free and water-free melt.

The UO_2 for recycle was prepared in three codeposition electrolyses. These were made at 750 C with a current of 15 amps and a current density of about 0.1 amp/cm^2 . A platinum-clad cathode was employed and the (graphite) anode was shrouded with a quartz sleeve to limit access of chlorine to the cathode compartment. Air was swept over the surface of the melt in the cathode compartment to prevent build up of uranium(IV) chloride during electrolysis.

The first codeposition electrolysis was made after the melt had been air-sparged for 0.5 hour at a rate of about 500 ml/min following the partition deposition. This was done to oxidize the uranium(IV) chloride to uranyl chloride and the plutonium(III) chloride to PuO_2 , which would be incorporated in the UO_2 as it was deposited. The plutonium-to-uranium in this deposit was only about 16 per cent as high as the plutonium-to-uranium ratio in the melt prior to the air sparge. This low ratio was due to insufficient air sparging; subsequent work involving a two-hour air sparge between partition and codeposition electrolyses yielded deposits having plutonium-to-uranium ratios about 130 per cent as high as the ratio in the melt prior to air sparge.

The deposit obtained in the first codeposition electrolysis is shown in Figure 2. As is typical of deposits obtained with an air blanket over the melt, the deposit was made up of loosely packed, coarse, crystalline aggregates and was easily removed from the cathode after soaking in dilute acid.

After the first codeposition the salt was allowed to freeze. After remelting, the salt was sparged with HCl-Cl_2 and then with air for 0.5 hour and a second codeposition electrolysis made. The plutonium content of the deposit was more representative of those later obtained with more prolonged air sparging directly after a partition electrolysis.

To increase the amount of product available for fuel element fabrication, a third codeposition was made. Most of the UO_2 which had been removed in the partition deposition was redissolved in the melt with an HCl-Cl_2 sparge (after oxidation to U_3O_8). No air sparge was used prior to electrolysis but the other conditions were the same as in the two previous codeposition electrolyses.

The UO_2 produced for recycle had an average oxygen-to-uranium ratio of 2.04. Treatment of this material at 800 C for an average time of ten hours with four per cent H_2 in N_2 reduced the oxygen-to-uranium ratio to 2.02.

The pour density of the combined UO_2 after hydrogen reduction was only 3.8 g/cm^3 . After the aggregates were crushed the pour density was 5.2 g/cm^3 .

Of 320 grams of UO_2 deposited in the three codeposition electrolyses, 230 grams were recycled to the reactor and 90 grams were retained in various samples for analysis and study.

FIGURE 2
Electrodeposited UO_2 for Recycle

Analyses of the various deposits and of the material combined for recycle are shown in Table I. The poor decontamination from cerium is of little concern because of its low neutron capture cross section. Of greatest concern is the low decontamination from the rare earths other than cerium, which as a group account for greater than 50 per cent of the fission product poisoning. Means of improving decontamination from these rare earths are under study.

TABLE I

ANALYSES OF ELECTROLYTIC UO_2 PREPARED FOR REACTOR RECYCLE

Sample	Pu		Ce		Rare Earths Other Than Ce	Sr	O/U Ratio
	d/m/mgU	c/m/mgU	c/m/mgU	c/m/mgU	c/m/mgU	c/m/mgU	
Partition	7.4×10^2	1.4×10^5	5.4×10^3	8.8×10^4	--		
1 Codeposit	4×10^4	4.5×10^5	5.5×10^4	1.2×10^5	2.017		
2 Codeposit	2.4×10^5	1.6×10^7	6.8×10^5	3.1×10^5	2.054		
3 Codeposit	8.3×10^4	8.4×10^6	2.8×10^5	7.4×10^5	2.044		
Combination of 1, 2, & 3	1.1×10^5	1.1×10^7	3.7×10^5	2.6×10^5	2.02 ⁽²⁾		
Feed UO_2 ⁽¹⁾	1.6×10^5	2.0×10^7	5.7×10^6	8.3×10^6			

(1) Feed UO_2 was 1.6 per cent enriched (U^{235} in total U), irradiated to approximately 1000 MWD/T and cooled 22 months.

(2) After reduction.

Fuel Rod Fabrication

The recycle UO_2 was fabricated into a Zircaloy clad fuel rod which was incorporated into a four rod cluster test element. The fabrication process was patterned after the vibrational compaction method evolved by Hauth.* Although the UO_2 powder was far from ideal with respect to particle size or particle size distribution--two requirements for obtaining high density by vibrational compaction--this method was believed to be the one most adaptable to remote fabrication techniques. The density obtained in the recycle rod was very low (61 per cent theoretical). There was insufficient recycle UO_2 available in this test but subsequent swaging and vibrational compaction studies using electrodeposited UO_2 with proper particle sizes and size distribution control have produced densities of 89 per cent theoretical.

The three rods containing the nonrecycle material were fabricated first, two of them by vibrational compaction. The third contained sintered pellets, as indicated in Table II which also gives the UO_2 fuel data.

Figure 3 shows side and end views of the test element with the fuel rod designations. The element was 12-1/8 inches in over-all length with a fuel length of about 10-1/2 inches. The rods were 0.565-inch OD with a cladding thickness of 0.030 inch and a fuel diameter of 0.505 inch.

The recycle rod was fabricated in a steel cell lined with 4 inches of lead brick. The various steps in the process are shown in Figure 4. The UO_2 was transferred into the loading tool shown in Figure 5. With the tool in position 1, the powder was fed into the vibrated fuel rod at a controlled rate. The weighted shaft "B" was used as a tamping rod (position 2) and was calibrated to show when the fuel rod was full. Upon completion of compaction, the shaft "B" was locked (position 3) so the end of the shaft was flush with the loading tool tip. The spread of contamination during tool removal was thus minimized.

* Hauth, J. J. Vibrationally Compacted Ceramic Fuels, HW-67777.
January 10, 1961.

TABLE II
PREIRRADIATION UO₂ DATA

Property	Rod "R" (1)	Rod 3 (2)	Rod 6 (2)	Rod 9 (3)
<u>O/U Ratio</u>	2.02	2.013	2.013	2.013
Tap Density (g/cm ³)	5.2	7.74	6.21	3.5 (4)
Enrichment (U235 in total U)	1.6	1.6	1.6	1.6
Particle Density (g/cm ³)	-	10.76	10.41	-
Surface Area (m ² /g)	-	0.103	0.096	-
<u>Sieve Analysis (Per Cent)</u>				
-20 mesh	100			
-16 + 35		50	38	(5)
-35 + 48		12.5	23.8	
-48 + 65		-	3.9	
-65 + 100		17.5	10.5	
-100 + 200		-	12.7	
-200		20.	11.1	
<u>Gas Analysis (ppm)</u>				
N ₂ O		120	10	6
CO ₂		340	4	4
O ₂		0.4	0.05	0.6
N ₂		-	10	1
CO		85	0.5	5
H ₂		4	7	-
A		--	0.3	0.2
C ₃ H _x		0.5	0.02	0.4
HCl		0.4	0.25	0.4
C ₂ H _x		0.3	0.18	0.5
CH ₄		-	0.1	0.9
<u>Chemical Analysis (ppm)</u>				
C		550	100	-
Cl		100	30	-
Na		4	240	200
K		17	1500	-

- (1) Recycle, vibrationally compacted.
 (2) Electrodeposited (nonrecycle) vibrationally compacted.
 (3) Sintered Pellets
 (4) Tap density of powder prior to pelletizing.
 (5) 0.502" ± 0.001 diameter pellets from 1/2" to 3/4" long.

FIGURE 3
Test Element

FIGURE 4
Recycled Fuel Element

FIGURE 5

Loading Tool Operation

Compaction frequencies between 200 and 5000 cps were used with accelerations up to approximately 100 g. The fuel rod and loading tool were held during compaction by an air operated chuck which left no scars, deformations or cladding imperfections. After compaction the fuel rod surface was essentially contamination-free. A swab test showed even the threads on the fuel rod cap to be clean of UO_2 powder.

Upon removal of the loading tool, a threaded end cap plug was screwed in place. Figure 6 shows the end cap placement and weld and illustrates the technique used to insure proper positioning and closure.

All welds were made with a TIG time-sequence welder. The welding sequence included an argon purge, a welding cycle, and an argon purged cooling period. Properly integrated turntable speed and welding sequence produced excellent welds with no discolorations, evidence of alloying, or embrittlement.

Final assembly and welding of the recycle rod to the four-rod cluster was accomplished with the aid of a copper positioning block. This block held the three nonrecycle rods in position while the recycle rod was positioned in the cruciform and welded in place with a TIG spot gun.

Irradiation

The test element was irradiated in the GEH-4 facility of the Materials Testing Reactor during cycle number 150. The coolant was 80 F, 300 psi water. The vertical and horizontal neutron flux profiles, as determined from 0.1 per cent Co-Al flux monitor wires attached to the fuel element basket assembly, are shown in Figure 7. Because it was necessary to handle the element by remote techniques prior to insertion into the reactor, no thermocouples were attached to the fuel basket assembly.

Each rod of the cluster generated a maximum surface heat flux of about 300,000 Btu/(hr)(sq ft). The element was irradiated for 10.5 effective full power days, to an exposure of approximately 450 MWD/T.

FIGURE 6
End Cap Closure

FIGURE 7
Neutron Flux Profiles

Postirradiation Examination

The fuel rods containing electrodeposited UO_2 suffered no adverse effects during irradiation.

A photograph of the test element after irradiation is shown in Figure 8. The scratches on the cladding surface resulted from handling subsequent to discharge. As shown in Table III, the rods containing electrodeposited fuel were dimensionally stable during irradiation. No evidence of warp or swelling was observed. In contrast, the rod containing sintered pellets warped 0.084-inch (single throw) during irradiation. The warp probably was due to insufficient diametral clearance between the pellets and the cladding.

TABLE III
FUEL CLADDING DIAMETERS

<u>Fuel Rod Number</u>	<u>Average Outside Diameter</u>	
	<u>Before Irradiation</u>	<u>After Irradiation</u>
R	~ 0.565	0.565
3	0.565	0.566
6	0.560	0.561
9	0.559	0.563

Internal pressure buildup during irradiation was not sufficient to damage the cladding although approximately 30 per cent of the fission gases formed were released from the fuel.

Cross sections of each rod were examined metallographically to determine the in-reactor sintering characteristics of each type of fuel material. Figure 9 shows the as-polished cross sections of each rod in the cluster. Extensive grain growth and sintering occurred in rods 3, 6, and "R", but the fuel in rod 9 (sintered pellets) experienced comparatively little change during irradiation. Photomicrographs of the irradiated

FIGURE 8
Element After Irradiation

electrodeposited UO_2 of rods 3, 6, and "R" are shown in Figures 10, 11, and 12, respectively. The structure observed in rods 3, 6, and "R", i. e. , large columnar grains surrounding a central cavity, smaller grains surrounding the columnar grains, and a region of nonsintered material adjacent to the cladding, has been observed in many other UO_2 specimens after irradiation at comparable core temperatures. The differences in the degree of sintering which occurred in the various fuel rods were due mainly to the differences in preirradiation bulk densities. Fuel having a low initial density has a low thermal conductivity and, therefore, attains a high core temperature during irradiation.

The number of voids in the irradiated, electrodeposited UO_2 was considerably greater than the number of voids observed in other irradiated specimens of vibrationally compacted UO_2 . However, the presence of these voids did not adversely affect the performance of the fuel element during irradiation.

The Zircaloy cladding on these fuel rods was unaffected by impurities present in the electrodeposited UO_2 . No evidence of interaction between cladding and the UO_2 was observed.

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This demonstration of a fuel recycle concept was accomplished through the joint efforts and cooperation of personnel in three components of Hanford Laboratories Operation; Chemical Research, Ceramic Fuels Development and Radiometallurgy.

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CROSS SECTION OF IRRADIATED FOUR-ROD CLUSTER: GEH-4-59

FIGURE 9

Cross Section of Irradiated Four-Rod Cluster: GEH-4-59

IRRADIATED ELECTRODEPOSITED UO_2 :
GEH 4-59, ROD NO.3

FIGURE 10

IRRADIATED ELECTRODEPOSITED UO_2 :
GEH 4-59, ROD NO.6

FIGURE 11

IRRADIATED RECYCLED UO_2 :
GEH 4-59, ROD R

FIGURE 12

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