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Voloxidation and Dissolution of Irradiated (Th,U)O₂

J. H. Goode
R. G. Stacy

~~APPLIED TECHNOLOGY~~

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VOLOXIDATION AND DISSOLUTION OF IRRADIATED (Th,U)O₂

J. H. Goode
R. G. Stacy

Chemical Technology Division

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VOLOXIDATION AND DISSOLUTION OF IRRADIATED (Th,U)O₂

J. H. Goode
R. G. Stacy

ABSTRACT

Exploratory hot-cell tests were made to determine the amounts of fission products released by high-temperature oxidation (voloxidation) of irradiated fuel, initially (Th_{0.96}U_{0.04})O₂, and whether the treatment would affect the subsequent dissolution of the oxide. Fifteen-year-decayed fuel rods were sheared to dislodge the (Th,U)O₂ from its cladding, sieved to determine the size range, and an aliquot was oxidized in flowing air at 600°C for ~2 hr while tumbling at 12 rpm. The voloxidation released only 0.2% of the ⁸⁵Kr and 6% of the ³H contained in the (Th,U)O₂. The oxidized and unoxidized portions were leached three times in a 95°C Thorex reagent to dissolve the fuel. The voloxidation apparently had no effect on the dissolution of the mixed oxide. The presence of cladding slowed the dissolution by limiting the amount of fuel exposed to the acid.

1. INTRODUCTION

An exploratory hot-cell test was made to determine the amount of fission products released by high-temperature oxidation (voloxidation) of irradiated fuel, initially (Th_{0.96}U_{0.04})O₂. The unclad mixed-oxide fragments were heated at 600°C in a stream of air for 2 hr while being tumbled at 12 rpm; these conditions are known to release significant portions of ³H, ¹⁴C, and ⁸⁵Kr from irradiated UO₂ and (U,Pu)O₂.¹⁻³ The fuel fragments used in these experiments had been irradiated in the Chalk River, Ontario NRX reactor, and discharged in 1963. Consequently, only the relatively long-lived fission products were present. The 4.5-in.-long rods (from holder No. 4) were clad in stainless steel and were sheared into short lengths to dislodge the mixed-oxide fuel from the cladding. One loose fraction was oxidized, while two other samples were used as controls. The fuel was dissolved in 13 M HNO₃--0.1 M Al(NO₃)₃--0.05 M KF to complete the material balances for heavy metals and fission products. A single dissolution experiment, conducted with a similar Zircaloy-clad (Th,U)O₂ rod from NRX holder No. 2, was designed to look for possible interference with the dissolution from a reaction between the zirconium and the fluoride in the dissolvent that was used.

2. EXPERIMENTAL

The specimens of NRX holders 2 and 4 (Fig. 1) were fabricated by the ORNL Metals and Ceramics Division using reactor grade Zircaloy-2 or type 316 stainless steel cladding materials and $(\text{Th}_{0.96}\text{U}_{0.04})\text{O}_2$ pellets purchased from the Nuclear Materials and Equipment Corporation (NUMEC). Each of the fuel rods contained seven mixed-oxide pellets, having a total weight of ~ 27.2 g ThO_2 and ~ 1.2 g UO_2 ; the uranium used in the pellets had been enriched to $\sim 93\%$ ^{235}U . Holder No. 4 was inserted into position N-11 of the NRX reactor on May 24, 1962, and irradiated with water cooling (peak cladding temperature $\leq 125^\circ\text{C}$) to a nominal peak burnup of 20,000 MWd/ton; it was discharged in April 1963 and was shipped to Oak Ridge after a short decay period. Holder No. 2 was irradiated in position M-12 from February 24, 1960, until April 3, 1961 (20,000 MWd/ton peak). These rods were stored at ORNL until the conduction of the test in the early summer of 1978.

The rods were sheared in a single-rod hydraulic shear and oxidized in a heated, stainless steel rotary voloxidizer; the equipment has been described in other reports.¹⁻³ Off-gases from the voloxidizer or the dissolver were passed through a series of traps or scrubbers, and a gamma spectrometer was used for an analysis of the fission product contents. The entire off-gas from either step was collected in a large bag for the composite samples.

3. RESULTS AND DISCUSSION

3.1 Shearing $(\text{Th,U})\text{O}_2$ Fuel

The single-rod hydraulic shear was used to prepare the 4.5-in.-long rods for voloxidation and dissolution studies. One Zircaloy-clad NRX-2 rod (No. B227) was cut into 0.5-in.-long pieces. Six stainless-steel-clad NRX-4 rods (Nos. 825, 826, 827, 851, 855, and 859) were sheared into ~ 0.25 -in. lengths to provide an inventory of unclad $(\text{Th,U})\text{O}_2$. Two other NRX-4 stainless-steel-clad rods (Nos. 823 and 860) were also sheared into short lengths to provide unclad fuel for a separate series of reprocessing studies.⁴

Fig. 1. Schematic NRX irradiation specimen.

An average shear force of about 570 ± 30 kg was sufficient to cut through the Zircaloy-clad rod. About 22% of the fuel was dislodged from the rod during shearing even though nearly half of the cut pieces had deformed ends with up to 50% closure. On a per-cut basis, fuel dislodgement amounted to 0.8 g (0.1-in.) or $\sim 3\%$ of the fuel with each blade pass.

Shear forces varied considerably from cut to cut during shearing of the stainless-steel-clad rods. The average cutting force required was in the range of 700 ± 200 kg. Apparently this variation was due to the compressing and mashing of the relatively soft, nonembrittled steel rather than smooth shearing of the rod. The resultant short pieces were actually rough-ended and jagged fragments, many with closed ends. About 85% of the $(\text{Th,U})\text{O}_2$ was effectively dislodged from the steel cladding pieces during the shearing process. This amounted to ~ 1.7 g (0.21 in.) or 6.1% of the fuel in each rod being dislodged with each cut.

Dislodged fuel from six of the NRX-4 rods was sieved; a size distribution is listed in Table 1. The fuel comprising the fractions between 18 and 200 mesh was selected as the voloxidation test standard. The larger fuel fragments and smaller fines, totaling ~ 43 g, were dissolved together to determine fission product inventories of the NRX-4 fuel. The stainless steel cladding pieces containing residual fuel ($\sim 15\%$ of the total mixed oxide) were also leached and subsequently dissolved.

Table 1. The size distribution of $(\text{U,Th})\text{O}_2$ fuel dislodged by shearing

Size fraction		Weight (g)	Percent
Mesh	Opening (μm)		
+ 18	1000	11.5	8.1
+ 35	500	33.0	23.2
+100	149	49.1	34.5
+200	74	16.7	11.7
+325	44	8.6	6.0
-325	<44	<u>23.6</u>	<u>16.5</u>
Total		142.5	100.0

Dissolution of the voloxidized and unvoloxidized fragmented fuel is discussed in Sect. 3.3. The 0.5-in.-long sheared segments of NRX-2 Zircaloy-clad (Th,U)O₂ were dissolved in a separate experiment.

3.2 Voloxidation

3.2.1 Voloxidation - general

A batch rotary voloxidation was made with unclad (Th,U)O₂ to identify gaseous release totals and release rates, fission product distribution, and oxygen consumption. The voloxidation apparatus, the gas-flow system, and the operating and sampling procedures employed in this study were the same as those used during similar experiments with other fuel types. Descriptions of equipment and procedures appear in reports outlining work with UO₂ and (U,Pu)O₂ fuels.^{1,2}

Voloxidation conditions were the following: 600°C in air flowing at 300 cm³/min (STP); rotation at 12 rpm; duration of test, 2 hr. The temperature of 600°C was selected because previous studies with irradiated uranium-plutonium mixed oxides indicated improved oxidation and tritium release at that particular level.³ To minimize cross-contamination of the voloxidizer charge by fuel and fission product residues left from earlier experiments, the (Th,U)O₂ was oxidized in a new voloxidizer. Once the fuel was loaded, the system was assembled and leak-tested. At this point, the temperature was increased at the rate of 15°C/min over a 40-min period until the voloxidizer reached 600°C, where it was maintained for the duration of the oxidation. Although the multichannel analyzer was not used for off-gas monitoring during this run, samples of flowing gas were collected at 10-min intervals to determine ⁸⁵Kr concentration. Traps containing silica gel were sequentially switched into the off-gas line at 15-min intervals to collect the evolved tritium (as ³H₂O). To assure completeness of the reaction at the above conditions, the voloxidation was continued for an hour beyond the point at which oxygen consumption had ended. Following the run, the voloxidizer was given a 2-hr air purge during cooling to sweep any remaining off-gases to the traps and collection system.

3.2.2 Oxygen consumption profile

During the heat-up period, oxygen consumption began when the temperature of the charge reached $\sim 500^\circ\text{C}$. The consumption reached a maximum at approximately the same time as the voloxidation temperature of 600°C was attained. Utilization of O_2 from the feed air stream was nearly finished after 1.0 hr. At the point of peak consumption, $\sim 50\%$ of the oxygen in the air was being consumed. (Combined profiles for oxygen consumption and gaseous fission product release are presented in Sect. 3.2.4.)

3.2.3 Oxidation of $(\text{Th},\text{U})\text{O}_2$

A material balance for the voloxidation of unclad NRX-4 $(\text{Th},\text{U})\text{O}_2$ is shown in Table 2. The small weight gain indicates that very little oxidation of the bulk fuel took place. Actually, complete oxidation of the UO_2 in nominal $(\text{Th}_{0.96},\text{U}_{0.04})\text{O}_2$ would be expected to result in a weight gain of only 0.16%. Comminution in the voloxidizer

Table 2. Material balance for $(\text{Th},\text{U})\text{O}_2$ voloxidation

	Run TH-2 (600°C , air)
Input wt., ^a g	98.7
Output wt., ^a g	99.0
Measured wt. gain, g	0.3

^aIncludes weight of $(\text{Th},\text{U})\text{O}_2$ + weight of fission products and insoluble residue.

should not occur because of the low uranium concentration. According to Peterson and Curtis,⁵ the dioxides of thorium and uranium form a continuous series of cubic-structured solid solutions over the entire range of composition. When heated in air or other oxidizing media, the mixed dioxides take up oxygen to an extent that depends on temperature and oxygen activity. Oxidation increases with uranium content but occurs without a phase change unless the UO_2 content exceeds 50 or 60%. In some cases, the cubic-structured U_4O_9 is formed. Highly oxidized uranium-rich $(\text{Th},\text{U})\text{O}_2$ could contain a second phase, the orthorhombic U_3O_8 .

In more recent studies aimed at the demonstration of a practical means of fracturing the structure of ceramic-oxide fuels, such as $\text{Th}_x\text{U}_{1-x}\text{O}_2$, Tennery and Federer⁶ measured considerable uranium oxidation of $(\text{Th}_{0.2}\text{U}_{0.8})\text{O}_2$ samples starting at $\sim 450^\circ\text{C}$ in flowing air. Those samples disintegrated to powder, but samples with the composition $(\text{Th}_{0.4}\text{U}_{0.6})\text{O}_2$ were only partially fragmented. Their preliminary conclusion was that solid solutions containing <0.6 mole of uranium oxide probably would have to be fragmented by methods other than oxidation.

In the present case, with $<4\%$ UO_2 in the NRX fuel,* only limited oxidation is likely to have resulted, with no phase change taking place. In the absence of any phase restructuring, powdering of the fuel fragments would also be greatly limited. This appears to have been the case as shown by the sieve size distributions on the fuel before and after voloxidation (see Table 3). The lifting and tumbling action (at 12 rpm) was of little significance in promoting oxidation or in aiding pulverization.

Table 3. Size distribution of voloxidized and unvoloxidized $(\text{Th,U})\text{O}_2$

Mesh size	Opening (μm)	Before voloxidation		After voloxidation	
		wt (g)	%	wt (g)	%
+ 35	500	33.0	33.4	30.9	31.3
+100	149	49.1	49.8	49.2	50.0
+200	74	16.6	16.8	15.9	16.1
+325	44	-	-	1.5	1.5
-325	<44	-	-	1.1	1.1
Total		98.7	100.0	98.6 ^a	100.0

^aLost 0.4 g during sieving.

3.2.4 Gaseous release profiles

Curves showing the concentration of tritium and krypton in the off-gas during voloxidation are plotted as a function of run time in Fig. 2 and are related to the corresponding oxygen content of the

*Irradiation resulted in UO_2 concentrations in fuel of ~ 2.8 to 3.3% ; see dissolution discussion Sect. 3.3.

Fig. 2. Concentrations of ^3H , ^{85}Kr , and O_2 in the off-gas during voloxidation of $(\text{Th,U})\text{O}_2$.

voloxidizer off-gas. Cumulative oxygen consumption and fission gas release profiles for the run are shown in Fig. 3.

Evolution of gas from the fuel was greatest during the heat-up period and for the first few minutes at 600°C. Both concentration curves peaked after less than 30 min at the voloxidation temperature, indicating that initial degassing due to oxidation and diffusion had subsided. At the end of the 2-hr voloxidation, the ^{85}Kr concentration in the off-gas had returned to near system background levels. Tritium concentrations in the off-gas were still about $10^2 \text{ dis sec}^{-1} \text{ cm}^{-3}$. The release profile curve after 2 hr at temperature indicates that tritium was still slowly diffusing from the fuel.

3.2.5 Release of gaseous fission products

Voloxidation release of gaseous radionuclides from NRX (Th,U) O_2 was determined from samples of collected voloxidizer off-gas and from residual amounts in the voloxidized fuel. These are presented in Table 4 as percentages of the total fuel inventories (fission product inventory determination is discussed in Sect. 3.3). All gaseous releases were quite low for voloxidation under this set of conditions.

Table 4. Release of gaseous radionuclides during voloxidation of unclad (Th,U) O_2

Nuclide	Fuel inventory [dis sec $^{-1}$ g (U+Th) $^{-1}$]	Released to voloxidizer off-gas ^a (% of fuel inventory)
^3H	2.39×10^6	6.30
^{14}C	2.16×10^3	4.63
^{85}Kr	2.03×10^8	0.20

^aThe releases of ^3H and ^{85}Kr were determined from collected voloxidizer off-gas; ^{14}C release was based on residual levels determined from dissolution of voloxidized oxide.

Campbell⁴ has achieved >99% tritium releases from unclad specimens of NRX (Th,U) O_2 by heating in an inert atmosphere for 24 hr at 1000°C in

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Fig. 3. Oxygen consumption and gaseous release profiles for voloxidation of $(\text{Th,U})\text{O}_2$.

flowing argon. No measurable oxidation or communitation to the fuel was noted. The main force during the release was probably diffusion. The 6.3% tritium release in 2 hr at 600°C, seen in our oxidation experiment, is consistent with Campbell's results.

3.2.6 Fission product distribution during voloxidation

Recoveries of selected fission products on voloxidizer system surfaces and traps following the oxidation of (Th,U)O₂ fuel are presented in Table 5 (as percentages of the total fuel inventories). In several cases, activity levels were only slightly above the limits of detection by gamma spectrometry.

Table 5. Fission product recoveries from the voloxidizer and off-gas system following oxidations of unclad (Th,U)O₂

Nuclide	Fuel inventory ^a [dis sec ⁻¹ g (U+Th) ⁻¹]	Distribution (% of fuel inventory)	
		Voloxidizer walls	Off-gas system
¹⁰⁶ Ru	1.86 x 10 ⁶	1.11	0.582
¹²⁵ Sb	9.63 x 10 ⁵	<0.001	0.004
¹²⁹ I	b	c	b
¹³⁴ Cs	2.18 x 10 ⁶	0.249	0.007
¹³⁷ Cs	1.20 x 10 ⁹	0.035	<0.001
¹⁴⁴ Ce	b	b	c

^aDetermined from dissolution of voloxidized and unvoloxidized fuels.

^bTrace quantities.

^cNot detected.

Due to the long cooling period, ¹³⁷Cs was the only gamma-emitting fission product oxide present in the fuel in any appreciable quantity. The limited oxidation and fracturing received by the fuel in this experiment did very little to release the cesium from the oxide matrix (<0.25%). A larger percentage (~1%) of the ¹⁰⁶Ru was found on the system surfaces after the run. This is consistent with the higher temperature operation. As in earlier (U,Pu)O₂ voloxidations,³ most of the deposited activity was found on sheet samples contacting the voloxidizer walls.

These fuel samples contained trace quantities of ^{129}I ; during voloxidation, most of that was transferred to charcoal traps in the off-gas line.

3.3 Dissolution of Clad and Unclad $(\text{Th,U})\text{O}_2$

Clad segments and loose fragments of the irradiated $(\text{Th,U})\text{O}_2$ were leached three times at 95°C with Thorex reagent (13 M HNO_3 -- $0.1\text{ M Al}(\text{NO}_3)_3$ -- 0.05 M KF) in Pyrex flasks fitted with reflux condensers. The off-gases from the first leaches were scrubbed with 3 M NaOH and then collected for determination of the ^{85}Kr content. Four dissolution tests were made.

- Th-1: segments of Zircaloy-2-clad $(\text{Th,U})\text{O}_2$ from a single rod from NRX-2 irradiation holder (20,000 MWd/ton)
- TH-2: voloxidized, loose fragments of $(\text{Th,U})\text{O}_2$ from six rods from NRX-4 holder ($\leq 20,000$ MWd/ton)
- TH-3: unvoloxidized, loose fragments of $(\text{Th,U})\text{O}_2$ from six rods from NRX-4 holder
- TH-4: residual $(\text{Th,U})\text{O}_2$ contained in the stainless steel cladding from eight rods from the NRX-4 holder.

Material balances. The recoveries of thorium, uranium, and fission products (Table 6) were generally within the limits of the accuracy of the remote electronic balance ($\pm 0.1\text{ g}$) and the chemical analysis for uranium ($\sim 5\%$) and thorium ($\sim 10\%$).

Dissolution rate. The quantities of uranium and thorium dissolved in the Thorex reagent in the series of three 2-hr leaches at 95°C are shown in Fig. 4 and Table 7. The presence of the Zircaloy or stainless steel cladding clearly slowed the rate of dissolution of the fuel but did not prevent its completion.

Uranium isotopic analyses. Mass spectrographic analyses of the first leach solution in runs TH-1 and TH-2 indicated that the rod from the NRX-2 holder had a higher burnup ($\sim 18,000$ MWd/ton) than that from the six blended rods from HRX holder No. 4 ($\sim 12,000$ MWd/ton):⁷

Table 6. Material balances for (Th,U)O₂ dissolution tests

	Run number			
	TH-1 ^a	TH-2 ^b	TH-3 ^c	TH-4 ^d
Input				
Weight, g	43.3	98.8	43.6	187.0
Recovery				
ThO ₂ , g	21.85	95.48	41.85	22.26
UO ₂ , g	0.74	2.72	1.18	0.86
Residue, g	0.65	0.33	0.06	0.01
Leached cladding, g	<u>17.7</u>	<u>0.00</u>	<u>0.00</u>	<u>155.4</u>
Output, g	40.94	98.53	43.09	178.53
Material balance, %	94.6	99.7	98.8	95.5
Ratio $\frac{U}{(Th+U)}$	0.033	0.028	0.028	0.037

^aZircaloy-clad, unvoloxidized (Th,U)O₂.^bUnclad, voloxidized (Th,U)O₂ only.^cUnclad (Th,U)O₂ only - not voloxidized.^dStainless steel cladding plus (Th,U)O₂ - not voloxidized.

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Fig. 4. Dissolution of clad and unclad $(\text{Th},\text{U})\text{O}_2$ in Thorex reagent.

Table 7. Dissolution of (Th,U)O₂ in Thorex reagent [13 M HNO₃--0.1 M Al(NO₃) --0.05 M KF]
for 2 hr each at 95°C

Leach number	TH-1 ^a		TH-2 ^b		TH-3 ^c		TH-4 ^d	
	Th (%)	U (%)	Th (%)	U (%)	Th (%)	U (%)	Th (%)	U (%)
1	83.16	83.01	98.77	98.94	98.22	98.55	89.21	83.69
2	9.88	10.81	0.58	<0.01	0.61	1.23	4.22	11.60
3	6.41	4.69	0.65	1.03	1.17	0.11	6.57	4.71
Residue	<0.01	<0.01	<0.01	0.03	<0.01	<0.01	<0.01	<0.01

^aNo voloxidation; 43.3 g of fuel and Zircaloy cladding as 37.7 g of 1/2-in.-long clad segments and 5.6 g of loose fuel.

^bVoloxidized 600°C, air at 200 to 300 cm³/min, 12 rpm; no cladding present (33.3 g + 35 mesh, 49.3 g + 100 mesh, 16.8 g + 200 mesh).

^cNo voloxidation; no cladding present (11.7 g + 18 mesh, 8.6 g + 325 mesh, 23.6 g - 325 mesh).

^dNo voloxidation; 187.0 g fuel + stainless steel cladding as 1/8-in. segments.

Nuclide	NRX-2	NRX-4
^{233}U	18.35	12.33
^{234}U	1.64	1.31
^{235}U	54.85	70.75
^{236}U	9.17	6.47
^{238}U	15.99	9.14

The lower ^{233}U content indicates that the six rods from NRX-4 probably were not from the peak-flux region, but rather from either the top or bottom (low-flux regions) of the holder.

Fission product content. The experimentally determined values for the fission product content of the $(\text{Th},\text{U})\text{O}_2$ from the NRX holders are noted in Table 8.

Table 8. Fission product content of NRX holders 2 and 4

Nuclide	NRX-2	NRX-4	Ratio NRX-2/NRX-4
	dis sec ⁻¹ g (U+Th) ⁻¹	dis sec ⁻¹ g (Th+U) ⁻¹	
^3H	3.98×10^6	2.39×10^6	1.67
^{14}C	2.54×10^3	2.16×10^3	1.18
^{85}Kr	9.52×10^7	2.03×10^8	0.47
^{106}Ru	9.87×10^6	1.86×10^6	5.31
^{125}Sb	3.54×10^6	9.63×10^5	3.68
^{134}Cs	2.12×10^6	2.18×10^6	0.97
^{137}Cs	1.95×10^9	1.20×10^9	1.63
^{154}Eu	2.01×10^7	1.25×10^7	1.61

The ratios of some of the fission products in the two rods were also in the same ratio as the ^{233}U content, which generally confirms the difference in burnup.

About 12% of the tritium found in the Zircaloy-clad rod from NRX-2 was within the cladding and was released only when the cladding was dissolved in hot NH_4F -- NH_4NO_3 . Concentrations of various elements in the first leach solution from TH-1 are listed in Table 9.

Table 9. Nuclide concentrations in a first leach solution
(run TH-1, NRX-2)

Nuclide	Concentration ($\mu\text{g/ml}$)	Nuclide	Concentration ($\mu\text{g/ml}$)
Th	154.00 ^a	La	50
U	5.22 ^a	Mo	200
Zr	1.64 ^a	Ni	60
Gross α	5.98×10^7 ^b	Nd	350
Pu α	1.33×10^6 ^b	Pd	100
Ag	10	Pr	50
Ba	90	Rb	100
Ce	300	Rh	50
Cd	20	Ru	50
Cs	600	Sm	100
Co	5	Sr	70
Eu	30	Tc	30
Fe	30	Tb	10
Gd	20	Y	20

^aIn g/liter.

^bIn counts $\text{min}^{-1} \text{ml}^{-1}$.

Dissolution of cladding in Thorex reagent. The leach solutions were analyzed for zirconium and iron to indicate some measure of the attack on the cladding by the Thorex reagent (Table 10). In the experiment with the Zircaloy cladding (TH-1), the first leach apparently dissolved a readily soluble layer, while the second and third leaches dissolved the metal. In the experiment with the stainless steel cladding (TH-4), the dissolvent also attacked and removed a more-soluble component during the first two leaches and then attacked the metal more slowly.

Table 10. Dissolution of cladding in 95°C Thorex reagent

	TH-1 (Zircaloy)			TH-4 (stainless steel)		
	Zr (mg/ml)	Zr (%)	F/Zr ^a ratio ^a	Fe (mg/ml)	Fe (%)	F/Fe ^b ratio ^b
Leach 1	1.64	0.93	2.8	0.31	0.29	0.9
Leach 2	1.39	0.27	3.4	0.29	0.27	1.0
Leach 3	0.84	0.22	5.8	0.15	0.14	1.8

^aMoles fluoride present per mole dissolved zirconium.

^bMoles fluoride present per mole dissolved iron.

4. CONCLUSIONS

1. Voloxidation of unclad $(\text{Th}_{0.96}\text{U}_{0.04})\text{O}_2$ fragments at 600°C in air while tumbling at 12 rpm - conditions applicable to UO_2 and $(\text{U,Pu})\text{O}_2$ fuels - did not release significant amounts of gaseous fission products. The tritium release was only 6%, and the ^{85}Kr release was only 0.2%. Campbell et al. released >99% of the tritium by heating some of the same $(\text{Th,U})\text{O}_2$ for 24 hr at 1000°C in an argon atmosphere.⁶
2. The voloxidation did not affect the solubility of the $(\text{Th,U})\text{O}_2$.
3. The presence of cladding slowed the rate of dissolution. Exposure to fresh dissolvent, as in a countercurrent leacher, may permit complete dissolution in <8 hr.
4. From 1 to 3% of the stainless steel or Zircaloy cladding was dissolved by the 13 M HNO_3 --0.1 M $\text{Al}(\text{NO}_3)_3$ --0.05 M KF.

5. A significant fraction of the fission product tritium is retained by Zircaloy cladding.

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