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IMPROVED ZIRCONIUM DECONTAMINATION
IN PUREX PROCESS

By

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IMPROVED ZIRCONIUM DECONTAMINATION IN PUREX PROCESS

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ABSTRACT

Studies showed that the improved Zirconium decontamination observed in the Hanford Purex Plant when processing Zircaloy-clad fuel elements (which were decontaminated in ammonium fluoride solutions - The Zirflex Process) was due to the presence of fluoride in the feed solutions. Related studies identified uranium carbide as a source of material that forms highly extractable zirconium complexes which limit the attainable decontamination. The effects of complexants other than fluoride were also evaluated.

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INTRODUCTION

When Zircaloy-clad fuels were first processed through the Hanford Purex plant, decontamination from $^{95}\text{Zr} + ^{95}\text{Nb}$ in the first solvent extraction column (HA column) was much greater than normally obtained with aluminum-clad fuels. Because the Zirflex Process⁽¹⁾ was used to declad the Zircaloy fuels, this effect was labeled the "Zirflex Tonic Effect." The first proposed explanation of this effect was that fluoride carried over from the decladding step to the core dissolution step and lowered the extraction of Zr and/or Nb, thereby giving increased decontamination, even though aluminum had been added to complex the fluoride and reduce corrosion. This hypothesis was tested in the plant by adding fluoride (and aluminum) to feed solution obtained from aluminum-clad fuels; only a minor improvement in the Zr + Nb decontamination factor was obtained. At this point a laboratory program was initiated to learn the reason for this effect. The results of that program are the subject of this report.

SUMMARY

- The "Zirflex Tonic Effect" was shown to be due to the presence of fluoride in the feeds prepared from the Zircaloy-clad fuels. Improved decontamination from Zr was found to increase with increasing fluoride concentration and to decrease with increasing aluminum-to-fluoride mole ratio. Fluoride was effective in improving Zr decontamination when added to the scrub as well as when added to the feed.
- Products of the reaction of nitric acid with uranium carbide were shown to affect the solvent extraction behavior of zirconium. One such product forms an inextractable complex with zirconium while another forms a highly extractable zirconium complex which limits the decontamination that can be achieved (in the absence of fluoride). These products can be destroyed by treatment of the feed solution with various oxidants.
- Decontamination from zirconium was also improved by addition of oxalic acid or ethylenediaminetetraacetic acid (EDTA) to the feed solution. The complexants were not effective when added to the scrub solution, however.
- The addition of inactive zirconium to the feed solution was also found to increase the decontamination from zirconium.
- These observations yield a fairly simple picture of the solvent extraction behavior of zirconium from Purex feed. At least three zirconium complexes of varying extractability are present in the feed. These complexes equilibrate rapidly in the aqueous phase, but the highly extractable complex equilibrates slowly in the organic phase and thus is not removed in the scrub steps.

EXPERIMENTAL PROCEDURE

The solvent extraction procedure employed in this study involved adjustment of the feed solution to $0.72 \text{ M U} + 4.2 \text{ M HNO}_3$, extracting the bulk of the uranium and portions of the nitric acid and zirconium into an tributyl phosphate-normal paraffin hydrocarbon organic phase, and scrubbing the organic extract four times with fresh aqueous solution. Samples of plant organic (30% tributyl phosphate (TBP) in normal paraffin hydrocarbon (NPH) diluent) were used. In the extraction contact, the volume of the organic phase was three times as large as the aqueous volume while equal phase volumes were used in the scrub contacts. The composition of the scrub solution was the same as that of the initial aqueous phase after extraction ($0.015 \text{ M U} + 3.1 \text{ M HNO}_3$) so that the concentrations of uranium and nitric acid would be the same in all five contacts. Changes in zirconium distribution were thus due to changes in the relative amounts of zirconium species present. Each contact was a period of three minutes at room temperature (about 23°C). Following each contact, centrifugation was employed to assure good separation of the phases.

The zirconium contents of the phases were determined by gamma counting, using a lithium-drifted germanium detector that allowed the amounts of ^{95}Zr and ^{95}Nb to be measured separately.

The results are generally presented in terms of decontamination factor (DF), which is defined as the ratio of zirconium to uranium in the feed divided by the same ratio in the product. Thus, for a given uranium distribution coefficient, a low zirconium distribution coefficient (the concentration of zirconium in the organic phase divided by that in the aqueous phase) gives a high zirconium decontamination factor.

The dissolutions of specimens of unirradiated uranium were performed in a flask fitted with an updraft condenser. The acid concentration was ten molar initially and near zero molar when the reaction was stopped. Reaction time was about seven hours with the low-carbon specimen and about four hours with the others.

Zirconium "tracer" solutions were obtained by scrubbing zirconium from organic extracts of plant feed solutions and then washing with dodecane to remove dissolved TBP. Some of the experiments used tracer purchased from ORNL; the oxalate solution of this tracer was added to concentrated nitric acid to destroy the oxalate at least a week prior to use.

A specific ion electrode was used to measure fluoride concentrations when fluoride distribution coefficients were determined. The fluoride which extracted was stripped from the organic for analysis; the calculated distribution coefficients assume complete stripping of the fluoride.

RESULTS

EFFECT OF FLUORIDE (LABORATORY TESTS)

Initial experiments with samples of plant dissolver solutions showed that the procedure used in the laboratory gave results comparable to those observed in the plant - a much lower ZrNb DF with feed derived from only aluminum-clad fuel than with feed derived from Zircaloy-clad fuels or with a blend of the two feeds. These data are given in Table 1.

While subsequent work explained the increase in Zr DF, the reason for the increased Nb DF obtained in these experiments was never ascertained. The effect was reproducible with this first sample of Zircaloy-clad dissolver solution but was not observed with a second sample. The search for an explanation

TABLE 1. Laboratory Comparison of Plant Dissolver Solutions

<u>Source of Dissolver Solution</u>	<u>Decontamination Factor⁽¹⁾</u>		
	<u>Zr</u>	<u>Nb</u>	<u>ZrNb</u>
Aluminum-clad uranium	10,000	35,000	15,000
Zircaloy-clad uranium	120,000	360,000	200,000
Blend of the two ⁽²⁾ (15% Zircaloy-clad)	57,000	400,000	110,000

1. *Through extraction and four scrubs.*

2. *Digested for about seven hours at about 50 °C.*

of the Nb behavior was dropped when analysis of plant samples showed the Nb content of first-column product (HAP) during processing of aluminum-clad fuels to be low enough that the decrease in Zr + Nb content observed when processing Zircaloy-clad fuels could be due solely to decreases in Zr content. These analyses showed ^{95}Nb to ^{95}Zr ratios of 0.027, 0.061, and 0.044 at total Zr-Nb concentrations of 1.29×10^4 , 4.23×10^4 , and 6.13×10^4 $\mu\text{Ci/gal}$ respectively in samples taken 3/10/69, 6/6/69, and 11/3/69.

In Figure 1 are presented the Zr DF's through each step of the experiments summarized in Table 1. Of special interest is the fact that the increased Zr DF resulted entirely from the scrub steps, which were of identical composition in all the experiments. It appears that additional scrubbing of the extracts of the feeds containing Zircaloy-clad uranium would have given even greater Zr DF's, while additional scrubbing would have had little effect on the extract of the aluminum-clad uranium dissolver solution. The zirconium remaining in the organic phase after exhaustive scrubbing is referred to as unscrubbable zirconium.

The contaminants known to be present in dissolver solutions obtained from Zircaloy-clad uranium are inactive zirconium, fluoride, and aluminum (which is added to complex the fluoride and reduce corrosion of the stainless steel equipment). Results of experiments to test the effect on Zr DF

FIGURE 1. Comparison of Plant Dissolver Solutions

of additions of these materials to the dissolver solution obtained from aluminum-clad uranium are given in Figure 2. Both fluoride and inactive zirconium are effective in increasing the Zr DF, the effect of fluoride being much greater. In these experiments the contaminants were digested with the feed for about 7 hours at about 50 °C prior to the solvent extraction steps; later experiments gave similar results when the digestion was omitted.

Additional data showing the Zr DF to increase with increasing fluoride concentration and decreasing aluminum-to-fluoride mole ratio are given in Tables 2 and 3 and Figure 3. The results in Table 2 (and Figure 3) were obtained with plant solutions while those in Table 3 were obtained with a solution prepared by dissolving non-irradiated uranium metal and then adding Zr "tracer" just before the solvent extraction steps were performed. The Zr "tracer" used here was scrubbed from organic extracts of plant aluminum-clad dissolver solutions.

Fluoride was also found to be effective in increasing the Zr DF when added to the scrub solutions, as shown in Figure 4. The same increase in Zr DF can be achieved with less fluoride when fluoride is added to the scrub solution instead of to the feed.

Addition of fluoride to the scrub solutions was also found to overcome the decreased Zr DF arising from the presence of dibutyl phosphate (DBP), as shown in Figure 5.

EFFECT OF FLUORIDE (PLANT TESTS)

As a result of the experimental work, two plant tests were performed. The results of these tests verified that fluoride is responsible for the "Zirflex Tonic Effect." The first test, performed during a period of co-product processing when the

FIGURE 2. Effect of Additives on Zr DF

TABLE 2. Effect of Fluoride on Zr DF Using Plant Solutions

Solution ⁽¹⁾	[F]	[Al]	Digest ⁽²⁾	Zr DF (cumulative through indicated step)				
				Extract.	1S	2S	3S	4S
2	—	—	Yes	10	120	930	2,700	3,500
2	0.01	0.03	Yes	11	150	1,900	18,000	54,000
2	0.01	—	Yes	25	480	5,600	71,000	410,000
4	—	—	Yes	22	270	1,200	1,700	2,000
4	0.01	0.03	Yes	18	270	3,200	16,000	24,000
4	0.017	0.03	Yes	19	270	3,700	42,000	150,000
6	—	—	No	11	150	1,900	8,100	12,000
6	0.01	0.03	No	13	200	3,300	37,000	125,000
6	0.01	0.03	Yes	13	240	3,900	35,000	150,000
6	0.03	0.03	No	34	800	12,000	170,000	700,000

(1) Dissolver solutions from aluminum-clad uranium (No. 2 taken 4-24-68, No. 4 taken 7-22-68, No. 6 taken 5-8-69)

(2) For ~7 hr at ~50 °C with $[HNO_3] \sim 1.5 M$

TABLE 3. Effect of Fluoride on Zr DF

Using Zr "tracer" spiked into solutions prepared by dissolution of unirradiated uranium

Relative ⁽¹⁾ Zr Concentration	[F]	[Al]	Zr DF (cumulative through indicated step)				
			Extract.	1S	2S	3S	4S
0.05	—	—	41			2,900	4,700
0.05	0.0033	0.01	22			12,000	21,000
0.05	0.01	0.03	26			21,000	47,000
0.05	0.01	0.06	15			8,000	15,000
0.1	—	—	29	290	1,100	1,800	2,400
0.1	0.01	0.03	14	200	1,600	8,700	20,000
0.1	0.01	—	47	1,300	11,000	61,000	
0.1	0.003	—	23	410	4,200	29,000	
0.1	0.002	—	19	290	3,000	15,000	22,000
0.1	0.001	—	13	160	1,300	5,700	8,600

(1) For $[Zr]$ in plant solution = 1.0

FIGURE 3. Effect of Fluoride on Zr DF

FIGURE 4. Fluoride in Scrub Solutions

ZR DF was exceptionally high, involved increasing the aluminum concentration of the feed.* Based on the laboratory data, this should result in an increase in the HAP Zr concentration due to a lowering of the concentration of free fluoride in the feed. As shown in Figure 6, the Zr behavior was as predicted.

* The feed is a blend of Zircaloy-clad and aluminum-clad dissolver solutions

FIGURE 5. Effects of Addition of DBP and DBP Plus Fluoride

FIGURE 6. Changes in HAP Activity with Al Concentration of Feed
(Fluoride in HAF $\sim 0.05 \text{ M}$)

The second test involved the addition of fluoride (and aluminum) to the feed during processing of only aluminum-clad fuel. As seen in Figure 7, making the HAF 0.02 M in fluoride and 0.06 M in aluminum resulted in a 100-fold decrease in the concentration of Zr in the HAP.

FIGURE 7. Lowering the Activity in HAP by Adding Fluoride to HAF

The plant test of fluoride addition performed before this research was undertaken had involved addition of 0.005 M F + 0.015 M Al to the HAF (this test gave only a two-fold improvement in Zr-Nb DF). It was later learned that HAF

obtained from aluminum-clad fuel "normally" contains about $0.05 \text{ M Al}^{(2)}$ so that the HAF during the early plant test probably contained about 0.065 M Al . Because of the high aluminum to fluoride ratio, it is not surprising that little improvement in DF was obtained.

EFFECT OF ZR CONCENTRATION

Results presented in Figure 2 showed the Zr DF to be increased by the addition of inactive Zr to plant dissolver solution. The results of a more detailed study of this effect are presented in Table 4. These experiments were done with a solution prepared by dissolving unirradiated uranium, to which was added varying amounts of Zr "tracer" and zirconyl nitrate. An increase in Zr DF with increasing Zr concentration is again observed.

TABLE 4. Effect of Zr Concentration on Zr DF

<u>[Zr]</u>	<u>Zr DF through:</u>	
	<u>Extraction</u>	<u>Extraction + 4 Scrubs</u>
$0.02 \times 10^{-4} \text{ M}^{(a)}$	24	1500
$0.07 \times 10^{-4} \text{ M}^{(a)}$	27	2500
$0.2 \times 10^{-4} \text{ M}^{(a)}$	23	4400
$3.5 \times 10^{-4} \text{ M}^{(b)}$	26	7000
$3.5 \times 10^{-4} \text{ M}^{(c)}$	21	6100
$17 \times 10^{-4} \text{ M}^{(b)}$	14	10,500

(a) Based on $4 \times 10^{-4} \text{ M}$ Zr in plant dissolver solutions.

(b) Added as zirconyl nitrate solution just prior to solvent extraction.

(c) Zirconyl nitrate solution digested with Zr "tracer" for 20 hr at 50°C in 2.5 M HNO_3 before use.

EFFECT OF URANIUM PURITY

As mentioned in a previous section, the solvent extraction behavior of Zr from plant dissolver solution was duplicated by adding "tracer" Zr to solutions prepared by dissolving unirradiated uranium metal of comparable purity. Further tests showed the Zr behavior to depend on the carbon content of the uranium, higher carbon contents giving lower Zr DF, as shown in Figure 8. The reaction of uranium carbide with HNO_3 is known to produce a variety of organic products; the material which forms the highly extractable (therefore, unscrubbable) Zr species is doubtless one of the products of this reaction. Oxalic acid and mellitic acid have been identified⁽³⁾ among the products of this reaction but test showed neither to give unscrubbable Zr species. Oxalic acid is probably responsible for the decreased extraction of Zr from the feed in the higher carbon case.

TREATMENT OF DISSOLVER SOLUTIONS TO IMPROVE Zr DF

The uranium carbide reaction products that affect the solvent extraction behavior of Zr can be destroyed by treatment with oxidizing agents. Data illustrating the improvements achieved by treatment with dichromate or cerium (IV) are shown in Figure 9. Hydrogen peroxide was also found to be effective, though to a lesser degree.

EXTRACTION OF FLUORIDE

Fluoride distribution coefficients were measured under the conditions used in the zirconium solvent extraction studies. In the absence of aluminum a distribution coefficient of 0.06 was obtained. With 0.01 $\underline{\text{M}}$ F + 0.03 $\underline{\text{M}}$ Al present in the feed, the value was 0.006 and, with 0.01 $\underline{\text{M}}$ F to 0.01 $\underline{\text{M}}$ Al, the value was 0.035.

FIGURE 8. Effect of Carbon Content of Uranium Metal on Zr Solvent Extraction Behavior After Dissolution in Nitric Acid

FIGURE 9. Improve Zr DF by Treating Dissolver Solution with Oxidants

EFFECT OF OTHER COMPLEXANTS

Several complexing agents other than fluoride were tested for their effectiveness in increasing the Zr DF on solvent extraction. Those that showed little or no effect (at the 0.01 M level) were sulfuric acid, phosphoric acid, tartronic acid, mandelic acid, citric acid, barbituric acid, nitrilotriacetic acid (NTA), hydroxyethylethylenediaminetriacetic acid (HEDTA), and diethylenetriaminepentaacetic acid (DTPA). Those that were effective were oxalic acid and ethylenediaminetetraacetic acid (EDTA), when added to the feed but not when added to the scrub. All the improvement occurs in the extraction step with these complexants, in contrast to fluoride where the improvement occurs in the scrub steps. These data are given in Table 5.

TABLE 5. Effect of Oxalic Acid and EDTA on Zr DF

<u>Added to Feed</u>	<u>Zr DF in</u>		
	<u>Extraction</u>	<u>Scrubs</u>	<u>Total</u>
—	40	60	2,400
0.0067 <u>M</u> Oxalate	570	45	26,000
0.01 <u>M</u> EDTA	4,000	50	200,000
0.01 <u>M</u> Oxalate + 0.017 <u>M</u> Al	2,500	20	50,000
0.01 <u>M</u> EDTA + 0.017 <u>M</u> Al	1,500	15	17,000
<u>Added to Scrubs</u>			
0.001 <u>M</u> Oxalate		150	
0.001 <u>M</u> EDTA		50	
0.001 <u>M</u> Fluoride		1,500	

MISCELLANEOUS RESULTS

Among the results of early experiments on the solvent extraction behavior of Zr extracted from aluminum-clad

dissolver solutions, two were especially significant in developing the final picture. These were:

- 1) When zirconium was extracted, scrubbed out of the organic, and then re-extracted into plant solvent, little or no unscrubbable Zr was found. This demonstrated that the unscrubbable Zr observed in extracts of plant feed was not due to anything present in the solvent (and therefore had to be due to something present in the feed).
- 2) When three successive extractions were performed, the third extract contained about as much unscrubbable Zr as did the first. This demonstrated that the unscrubbable Zr was *not* present in the feed as an inert species of high extractability (or it would have been removed in the first extraction) and that the complexant present in the feed that forms unscrubbable Zr is itself extracted relatively little if at all. Later tests using feed prepared by dissolving unirradiated uranium did show extraction of some of this complexant.

DISCUSSION

The results of this work yield a fairly simple picture of the solvent extraction behavior of zirconium in the Purex HA column. In feed solutions prepared by nitric acid dissolution of irradiated uranium containing carbon impurity, the zirconium can be considered as being distributed among three forms - a highly extractable complex (ZrX), an inextractable complex (ZrY), and a nitrate complex which is moderately extractable. The ligands X and Y both result from the reaction of uranium carbide with nitric acid. The three complexes equilibrate rapidly in the aqueous phase but ZrX equilibrates slowly in the organic phase and thus is not removed in the scrub section.

Since the concentrations of X and Y are low, addition of small amounts of inactive zirconium lowers the concentrations of uncomplexed X and Y, thus decreasing the fraction of the zirconium present as ZrX and ZrY and increasing the decontamination from zirconium.

The addition of an appropriate complexant to the feed solution reduces the concentrations of all three forms of zirconium, thus increasing the decontamination achievable. With complexants such as EDTA or oxalate, whose complexes with zirconium appear to be inextractable, the increased decontamination occurs entirely in the extraction step and is immediately apparent. Fluoride, however, apparently forms an extractable zirconium complex under certain conditions, so that the increased decontamination is not apparent until the scrub steps have been performed. The conditions which give appreciable amounts of the extractable zirconium-fluoride complex are those normally used in the plant (e.g., 0.01 M F + 0.03 M Al); at higher fluoride (or lower aluminum) concentrations, inextractable zirconium-fluoride complexes predominate.

For use in the scrub section, fluoride is the only complexant shown to be of benefit. Were it not for the slow equilibria involving ZrX in the organic phase, other complexants should work equally as well. The extractability of fluoride may be the reason for its effectiveness.

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