# $D R=0048-5$ I. 25836 

OAK RIDGE NATIONAL LABORATORY

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Solvent Extraction Studies of Coprocessing Flowsheets Results from Campaign 6 of the Solvent Extraction Test Facility (SETF)
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## Consolidated Fuel Reprocessing Program

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Date Issued: November, 1986

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Prepared by the<br>$\checkmark$ OAK RIDGE NATIONAL LABORATORY<br>Oak Ridge, Tennessee 37831<br>operated by<br>MARTIN MARIETTA ENERGY SYSTEMS, INC.<br>for the<br>U.S. DEPARTMENT OF ENERGY<br>under<br>Contract No. DE-AC05-840R21400

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SOLVENT EXTRACTION STUDIES OF COPROCESSING FLOWSHEETS results from campaign 6 OF the solvent extraction test facility (setf)
D. E. Benker, J. E. Bigelow, E. D. Collins, F. R. Chattin, L. J. King, H. C. Savage, R. G. Ross, R. G. Stacy


#### Abstract

A series of five solvent extraction tests were made in the Solvent Extraction Test Facility (SETF) during Campaign 6. Each test used a coprocessing flowsheet that included coextractioncoscrubbing of the heavy metals followed by partial partitioning of the uranium and plutonium into separate uranium and uraniumplutonium products. The separation of the uranium and plutonium was aided by the addition of $\mathrm{HNO}_{2}$ to the organic backscrub stream. Two of these tests compared the performance of the traditional Purex solvent, tri-n-butyl phosphate (TBP), with a potential replacement, tri-2-ethylhexyl phosphate (TEHP). The remaining three tests were made with a chemically-degraded TBP solvent to compare the effectiveness of two solvent cleanup methods - treatment with silica gel or scrubbing with sodium carbonate and water.


## 1. INTRODUCTION

The Solvent Extraction Test Facility is located within one of the heavily shielded hot cells of the Transuranium Processing Plant at the Oak Ridge National Laboratory. ${ }^{1}$ In the SETF, flowsheets for the reprocessing of irradiated nuclear reactor fuels are evaluated in mixer-settler contactors that have a processing capability of $\sim 1 \mathrm{~kg} / \mathrm{d}$ of heavy metals. The results from these tests provide information on uranium and plutonium recoveries, fission product removal, and the general operability of the system.

The broad objectives of studying coprocessing flowsheets, which was begun in the previous work, ${ }^{1-3}$ was continued in Campaign 6. For each of the tests, the solvent extraction flowsheet included coextraction and coscrubbing of the heavy metals (uranium and plutonfum) in the first contactor, followed by back-extraction of the plutonium with part of the uranium (partial partitioning) in the second contactor. An organic scrub was used in the partition bank to control the amount of uranium that was recovered with the plutonium and to add excess nitrous acid ( $\mathrm{HNO}_{2}$ ) to improve the uranium-plutonium separation by reducing some of the plutonium to Pu(III).

The five tests that were made in Campaign 6 were similar except for the type of organic extractant that was used. The first two tests were made to compare the performance of an alternative extractant, tri-2ethylhexyl phosphate (TEHP), with that of the standard Purex extractant, tri-n-butyl phosphate (TBP). The other three tests were made using a TBP-based solvent that had been chemically degraded to simulate a recycled solvent. These latter tests compared the performance of two solvent cleanup methods, (1) treatment with base-treated silica gel or (2) conventional scrubbing with sodium carbonate and water.

Although the program emphasis was on the reprocessing of fast breeder reactor (FBR) fuel, no such fuel was readily available when Campaign 6 was made in the fall of 1981 . Approximately 10 kg of light water reactor (LWR) fuel from the H. B. Robinson Reactor was used for these tests which had been irradiated to a burnup of $\sim 31 \mathrm{MWd} / \mathrm{kg}$ and discharged from the reactor in May 1974.

## 2. EQUIPMENT AND OPERATIONAL PROCEDURES

The SETF equipment items and most of the operational procedures used during Campaign 6 for fuel dissolution, feed clarification and adjustment; and solvent extraction were similar to those described for previous campaigns, ${ }^{1-3}$ except as noted below.

### 2.1 FUEL DISSOLUTION AND ADDITION OF ${ }^{95} \mathrm{Zr}$ TRACER

The basic procedure for dissolving LWR fuels has remained unchanged since the first dissolution in Campaign 1 and includes: (1) adding the sheared fuel pieces to 3 M nitric acid ( $\mathrm{HNO}_{3}$ ), (2) heating to $50^{\circ} \mathrm{C}$, (3) adding $11 \mathrm{M}_{\mathrm{HNO}}^{3}$ at a controlled rate, and (4) heating and digesting 2 h at $90^{\circ} \mathrm{C}$. The objective of this multi-step procedure was to maintain a slow, controlled dissolution rate. The final solution concentrations were typically $3 \mathrm{M} \mathrm{HNO}_{3}$ and $350 \mathrm{~g} / \mathrm{L}$ of heavy metals. Since short-cooled fuel was not available for Campaign 6 , a small amount of ${ }^{95} \mathrm{Zr}$ tracer was added during the dissolution procedure. Although it is unknown whether the tracer is yielding quantitative data on the behavior of fission product zirconfum, the tracer was found to be useful in the previous campaign ${ }^{3}$ for comparisons of decontamination factor values (DFs) with flowsheet changes. In Campaign 6, the tracer solution was added to the $3 \mathrm{M}^{\mathrm{HNO}} 3$ solution in the dissolver prior to the dissolution, instead of after the fuel dissolution as was done in the previous campaign. It was hoped that this method of adding the ${ }^{95} \mathrm{Zr}$ tracer would provide a better opportunity for isotopic exchange of zirconium between the tracer and the fuel.

### 2.2 FEED CLARIFICATION AND ADJUSTMENT

The clarification of the dissolver solution was accomplished in two filtration steps. The first (primary) filtration was made after the fuel dissolution to remove the acid insoluble residues; the second (polishing) filtration was made after the feed adjustment as the feed was in transit to the mixer-settlers in order to remove solids that might have formed during the feed adjustment or by feed aging mechanisms during the period preceding mixer-settler start-up. Both filtrations were made with a deepbed type of filter that contains a bed of diatomaceous-earth filter aid as
the filtering medium. Although this deep-bed filter was used for the primary filtration in Campaign 4 and for the polishing filtration in Campaign 5, this was the first time deep-bed filters have been used for both filtrations.

The feed adjustment was effected by the usual procedure; (1) sparging with $\mathrm{N}_{2} \mathrm{O}_{3}$ and air at ambient conditions to adjust the plutonium valence, and (2) addition of the appropriate $\mathrm{HNO}_{3}$ solution to make the final concentrations $\sim 3 \mathrm{M}_{\mathrm{HNO}}^{3}$ and $\sim 150 \mathrm{~g} / \mathrm{L}$ of heavy metals. The $\mathrm{H}_{2} \mathrm{O}_{2}$ addition and digest step that was used in Campaigns 2 through 5 was omitted because it appeared to have had a detrimental effect on the ruthenium DFs in the strip contactor.

## 3. DESCRIPTION OF THE SOLVENT EXTRACTION FLOWSHEETS

A diagram of the basic flowsheet used during Campaign 6 is shown in Fig. 1; the operating conditions and detailed stream analyses for each test are tabulated in the Appendix.

In each test, the first contactor was used to coextract the uranium and plutonium from the aqueous feed and coscrub the pregnant solvent with $\mathrm{HNO}_{3}$ to remove impurities. Because the use of two aqueous scrub streams had provided better zirconium removal during the previous campaign, it was included in Campaign 6. Also, the number of low-acid scrub stages was increased from three to six, with a corresponding decrease in extraction stages from 10 to 7 , in order to determine whether the additional stages would provide additional zirconium removal.

The second contactor was used to partially partition the uranium and plutonium. The two products desired from this type of flowsheet were, (1) a uranium stream (HBU) that was relatively free of plutonium, and (2) a mixed uranium-plutonium stream (HBP) that was enriched in plutonium. The amount of uranium that was stripped with the plutonium in the HBP was controlled in part by the organic backscrub stream (HBS). This stream was also used to add $\mathrm{HNO}_{2}$. At the $\mathrm{H}^{+}$concentrations used in these runs, $\mathrm{HNO}_{2}$

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FLOWSHEET FOR CAMPAIGN 6
Fig. 1. Solvent extraction flowsheet used in Campaign 6.
reduces $\mathrm{Pu}(\mathrm{IV})$ to $\mathrm{Pu}(I I I)$ which improves the efficiency of plutonium stripping and improves uranium-plutonium separation.

In a reprocessing plant the third contactor would be used to strip the uranium into an aqueous product stream. In these tests, the third contactor was used as a backup strip contactor for plutonium; uranium stripping was not studied. This mode was used to allow more freedom in testing partitioning flowsheets while ensuring that the waste solvent would meet disposal limits for plutonium. In addition, hydroxylamine nitrate (HAN) was added to the aqueous strip solution to reduce plutonium to $P u(I I I)$ and ensure complete stripping of plutonium from the solvent. The relatively high concentration ( 1.5 M ) of HAN acted as an inextractable nitrate salt that helped lessen the amount of uranium that would be stripped along with the plutonium. This simplified the subsequent recovery of plutonium from this stream.

The primary difference among the five runs was in the organic extractant that was used for each test. The first two runs (6-1 and 6-2) compared an alternative extractant, tri-2-ethyhexyl phosphate (TEHP), with the more common Purex extractant, tri-n-butyl phosphate (TBP). Laboratory and glove box studies with TEHP ${ }^{4,5}$ had proceeded to the point that further testing with irradiated fuel was desirable. Some of the possible advantages of TEHP over TBP, which had been indicated by the laboratory studies, include: (1) lower aqueous solubility, (2) higher extraction coefficients for heavy metals, (3) greater chemical stability, and, when processing thorium, (4) no third phase formation at high metal loadings. The typical extractant concentration for fuel reprocessing is 1.1 M ( 30 vol \%) TBP; however, because the phase separation characteristics of TEHP are poor at this concentration, the extractant concentration was lowered to 0.6 M for these comparison runs; 0.6 M corresponds to 28 and $16.5 \mathrm{vol} \%$ for TEHP and TBP, respectively. The diluent for these extractants was normal paraffin hydrocarbon.

The first two runs were further subdivided into two parts, "A" and " $B$ ", in order to determine the effect on the partitioning of adding different amounts of $\mathrm{HNO}_{2}$. In the " A " runs ( $6-1 \mathrm{~A}$ and $6-2 \mathrm{~A}$ ), $\sim 2$ mol of $\mathrm{HNO}_{2}$ per mol of plutonium was added to the partitioning contactor, and in the
" $B$ " runs (6-1B and 6-2B) five times as much, $\sim 10$ mol of $\mathrm{HNO}_{2}$ per mol of plutonium, was added. The $\mathrm{HNO}_{2}$ was added with the HBS stream, which contained $\sim 0.008 \mathrm{M} \mathrm{HNO}_{2}$ in the " A " runs and $0.04 \mathrm{M} \mathrm{HNO}_{2}$ in the " B " runs.

The other three runs were made using 30 vol \% TBP (the diluent was again normal paraffin hydrocarbon) that had been chemically degraded (described below) in order to simulate a recycled solvent. Two solvent cleanup methods were evaluated - (1) scrubbing with sodium carbonate and water and (2) treatment with base-treated silica gel. Sodium carbonate scrubbing is the traditional method for solvent treatment in a reprocessing plant; the silica gel method is a relatively new method developed by Tallent et al. ${ }^{6}$ for possible application in fuel reprocessing. The chemical degradation step was necessary because the SETF has no means for recycling solvent or of using alpha-contaminated solvents; all previous runs have been made with new solvent that was used once and then discarded.

The degraded solvent was prepared by refluxing a small batch of $30 \%$ TBP in $8 \mathrm{M}_{\mathrm{HNO}}^{3}$ while sparging with $\mathrm{NO}_{2}$, and then diluting this concentrate with new $30 \%$ TBP to make a final solution that was 0.003 M dibutyl phosphate (DBP) and $\mathbf{\sim} 0.8 \%$ diluent degradation products.
4. EXPERIMENTAL RESULTS AND DISCUSSIONS OF SOLVENT EXTRACTION TESTS

### 4.1. RESULTS USING TRI-2-ETHYLHEXYL PHOSPHATE

The initial use of TEHP in the SETF equipment caused problems that required some minor operational changes. During the solvent cleanup step (sodium carbonate and water scrubs), a slower agitator speed was needed to prevent the water scrubs from forming a stable emulsion. More batch contacts were used to compensate for the less vigorous mixing. Also, in order to achieve proper hydraulic operation in the SETF mixer-settlers, minor adjustments of the agitator positions were required to improve the pumping action from the mixers. On the other hand, the TEHP run did seem qualitatively to have less interfacial crud than did the later run with TBP (6-2) 。

### 4.1.1 Coextraction-Coscrub

The overall results from these runs indicated similar results for both solvents, except for uranium losses and zirconium DFs (Table l). For the run with TEHP solvent (6-1), uranium was not detected in the aqueous raffinate (HAW); the results shown are the limits of detection for the given samples. Because the uranium loss is so much lower than the plutonium loss in Run 6-1, it may be the result of sampling or analysis problems rather than from using TEHP. Concentration profiles for uranium, plutonium, and free $\mathrm{H}^{+}$are shown in Fig. 2.

The concentration profile for the ${ }^{95} \mathrm{Zr}$ tracer is shown in Fig. 3. The higher ${ }^{95} \mathrm{Zr}$ DF in the TBP run was not only the result of less ${ }^{95} \mathrm{Zr}$ extracting but also of better ${ }^{95} \mathrm{Zr}$ removal in the scrub section. The ${ }^{95} \mathrm{Zr}$ concentration in Run $6-2$ was $2.6 \times 10^{7} \mathrm{~Bq} / \mathrm{L}$ in the solvent at stage 10 (aqueous feed inlet) and dropped by a factor of $\sim 1000$ across the scrub section. In the run with TEHP, the feed stage had twice as much ${ }^{9}{ }^{5} \mathrm{Zr}, 6.4$ x $10^{7} \mathrm{~Bq} / \mathrm{L}$, and the concentration only dropped by a factor of 200 across the scrub section. No significant difference was noted in the behavior of ${ }^{106} \mathrm{Ru},{ }^{137} \mathrm{Cs},{ }^{144} \mathrm{Ce}$, or ${ }^{154} \mathrm{Eu}$ for either solvent.

The ${ }^{95} \mathrm{Zr}$ DFs measured for Run 6-2 (2 x $10^{4}$ ) is significantly higher than the DF measured in Run 5-1 (750) during Campaign 5 and may be the result of using additional low-acid scrub stages for Campaign 6 (six instead of three). Other differences that might have also influenced the ${ }^{95} \mathbf{Z r}$ behavior include: the fuel (Campaign 5 used EBR-II fuel), the addition of the tracer at a different point in the dissolution, and the lower TBP concentration (although Run 6-3, which used $30 \%$ TBP and is discussed below in section 4.2 .1 , had the same DF as Run 6-2).

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Fig. 2. Concentration profiles for the coextraction-coscrub contactor for the runs with TEHP and TBP.


Fig. 3. Concentration profiles for ${ }^{95} \mathrm{Zr}$ (tracer) in the coextraction-coscrub contactor for the runs with TEHP and TBP.

Table l. Uranium and plutonium losses and fission product decontamination results in coextraction-coscrubbing for the runs with TEHP and TBP.

|  | Run No. |  |
| :---: | :---: | :---: |
|  | 6-1 (TEHP) | 6-2 (TBP) |
| Extraction losses, \% |  |  |
| Uranium | $<0.001$ | 0.040 |
| Plutonium | 0.031 | 0.038 |
| Fission product DFs |  |  |
| ${ }^{9} 5 \mathrm{Zr}$ | 1E3 | 2E4 |
| $9{ }^{5} \mathrm{Nb}$ | 7E3 | 5E3 |
| ${ }^{106} \mathrm{Ru}$ | <9E3 | <4E3 |
| ${ }^{137} \mathrm{Cs}$ | $>2 \mathrm{E} 7$ | >2E7 |
| $14^{4} \mathrm{Ce}$ | $\sim 2 \mathrm{E} 4$ | $\sim 1 E 4$ |
| $1{ }^{4} \mathrm{Eu}$ | $>3 \mathrm{E} 5$ | >2E5 |

### 4.1.2 Partitioning

Although a slightly larger flow rate for the aqueous strip was used in the runs with TEHP in order to compensate for its higher extraction coefficients, the losses of plutonium into the uranium product (HBU) were still excessive, 1 to $2 \%$ of the total plutonium (Table 2). On the other hand, slightly less uranium was stripped into the plutonium product (HBP). The net result was poorer $U-P u$ separation with TEHP; the overall $U-P u$ separation factors (Table 2) were lower by a factor of $\sim 10$.

Increasing the $\mathrm{HNO}_{2} / \mathrm{Pu}$ mol ratio from $\sim 2$ (Runs $6-1 \mathrm{~A}$ and $6-2 \mathrm{~A}$ ) to a mol ratio of $\sim 10$ (Runs $6-1 B$ and $6-2 B$ ) decreased the plutonium losses to the uranium product by a factor of about 2 ; but, at the same time, increased the amount of uranium stripped to the plutonium product by factors of 1.2 and 1.4 .

Concentration profiles for uranium, plutonium, nitric acid, and nitrous acid are shown in Fig. 4.


Fig. 4. Concentration profiles for the partition contactor for the runs with TEHP and TBP.

Table 2. Results from partial partitioning contactor for comparison runs using TEHP and TBP solvents.

|  | Run No. |  |  |  |
| :---: | :---: | :---: | :---: | :---: |
|  | $6-1 A^{a}$ | $6-18^{\text {b }}$ | $6-2 A^{c}$ | $6-2 B^{\text {d }}$ |
| Feed solution (HAF) |  |  |  |  |
| $\mathrm{Pu}, \mathrm{g} / \mathrm{g}$ of U | $\sim 0.008$ | $\sim 0.008$ | $\sim 0.008$ | $\sim 0.008$ |
| Phase ratio ( $0 / A$ ) ${ }^{\text {e }}$ |  |  |  |  |
| Scrub | 1.9 | 1.9 | 2.1 | 2.1 |
| Strip | 6.8 | 6.9 | 7.2 | 7.2 |
| \% of Pu in |  |  |  |  |
| $\mathrm{U}-\mathrm{Pu}$ product ( HBP ) | 97.4 | 98.5 | 99.72 | 99.88 |
| U product ( HBU ) | 2.6 | 1.5 | 0.28 | 0.12 |
| Uranium product (HBU) |  |  |  |  |
| $\mathrm{Pu}, \mathrm{ug} / \mathrm{g}$ of U | 238 | 154 | 26 | 12 |
| Pu DF | 34 | 52 | 310 | 670 |
| U-Pu product (HBP) |  |  |  |  |
| $\mathrm{Pu}, \mathrm{g} / \mathrm{g}$ of U | 0.081 | 0.074 | 0.071 | 0.049 |
| U DF | 10 | 9.2 | 8.9 | 6.1 |
| $\mathrm{U}-\mathrm{Pu}$ separation factor ${ }^{\text {f }}$ | 340 | 480 | 2700 | 4100 |

${ }^{\text {a }}$ Solvent was $0.6 \underline{\mathrm{M}}$ (28\%) TEHP with a $\mathrm{HNO}_{2} / \mathrm{Pu}$ mol ratio of $\sim 2$.
bSolvent was 0.6 M (28\%) TEHP with a $\mathrm{HNO}_{2} / \mathrm{Pu}$ mol ratio of $\sim 10$.
${ }^{\text {c Solvent was }} 0.6 \mathrm{M}$ ( $16.5 \%$ ) TBP with a $\mathrm{HNO}_{2} / \mathrm{Pu}$ mol ratio of $\sim 2$. ${ }^{\text {d }}$ Solvent was 0.6 M ( $16.5 \%$ ) TBP with a $\mathrm{HNO}_{2} / \mathrm{Pu}$ mol ratio of $\sim 10$.
${ }^{e}$ (organic flow rate)/(aqueous flow rate)
$\mathrm{f}_{\text {Product of }}$ of U and Pu DFs.

### 4.2 RESULTS USING DEGRADED SOLVENT

### 4.2.1 Coextraction-Coscrub

The flowsheet conditions were initially set to achieve a higher solvent loading than was used in previous SETF runs in an attempt to improve the efficiency of the coextraction-coscrub contactor. Unfortunately, the conditions were set too close to the maximum loading, and minor uncertainties in the feed composition and minor variations in the flow rates allowed the solvent loading to reach about $115 \mathrm{~g} / \mathrm{L}$ ( $90 \%$ saturation) at stages 10 and 12 for Runs 6-3 and 6-4, respectively. This led to unacceptable losses of heavy metals to the aqueous raffinate (the very low uranium number in Table 3 for Run 6-3 is probably caused by an analysis or sampling problem). The flowsheet conditions were corrected in Run 6-5. The peak solvent loading was lowered to about $70 \%$ of saturation, and the loss of heavy metals was reduced to about $0.04 \%$, which is similar to previous SETF runs. As a result, there were significant variations in the solvent loading during these runs, as shown in Fig. 5.

The DFs for ${ }^{137} \mathrm{Cs},{ }^{144} \mathrm{Ce}$, and ${ }^{154} \mathrm{Eu}$ were similar for each solvent and are apparently unaffected by the presence of solvent degradation products or by the type of cleanup method used. However, the DFs for ${ }^{95} \mathrm{Zr},{ }^{95} \mathrm{Nb}$, and ${ }^{106} \mathrm{Ru}$ were noticeably worse (by factors of about 1000,100 , and 10 , respectively) when using the unpurified degraded solvent (Run 6-5). The two treatment methods (sodium carbonate or silica gel) yielded similar DFs for ${ }^{95} \mathrm{Nb}$ and ${ }^{106} \mathrm{Ru}$, which were also similar to the DFs measured for new solvent. However, the ${ }^{95} \mathrm{Zr}$ DF was a factor of 10 lower for the solvent that was treated with silica gel. After reviewing the purification procedure, it was determined that the poorer $D F$ was probably the result of inadequate water scrubbing of the silica gel product, which would leave some DBP in the solvent. If better water scrubbing had been used for the silica gel treatment, it might have yielded ${ }^{95} \mathrm{Zr}$ results similar to the sodium carbonate treatment. Concentration profiles for ${ }^{95} \mathrm{Zr},{ }^{95} \mathrm{Nb}$, and ${ }^{106} \mathrm{Ru}$ are shown in Figs. 6 through 8.


Fig. 5. Concentration profiles for the coextraction-coscrub contactor for the runs with chemically degraded solvent.

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Fig. 6. Concentration profiles for ${ }^{95} \mathrm{Zr}$ (tracer) in the coextraction-coscrub contactor for the runs with chemically degraded solvent.


Fig. 7. Concentration profiles for ${ }^{95} \mathrm{Nb}$ (tracer) in the coextraction-coscrub contactor for the runs with chemically degraded solvent.


Fig. 8. Concentration profiles for ${ }^{106} \mathrm{Ru}$ in the coextraction-coscrub contactor for the runs with chemically degraded solvent.

Table 3. Uranium and plutonium losses and fission product decontamination results in coextraction-coscrub contactor for runs with degraded solvent and previous runs with new solvent.

|  | New solvent ${ }^{\text {a }}$ | Run No. |  |  |
| :---: | :---: | :---: | :---: | :---: |
|  |  | $\overline{6-3}$ | $6-4{ }^{\text {c }}$ | 6-5d |
| Extraction losses, \% |  |  |  |  |
| Uranium | $\sim 0.01$ | <8E-4 | 4.6 | 0.03 |
| Plutonium | $\sim 0.01$ | 0.3 | 4.2 | 0.04 |
| Fission product DFs |  |  |  |  |
| ${ }^{95} \mathrm{Zr}$ | - | 2E4 | 2E3 | 4 |
| $95^{\mathrm{Nb}}$ | - | 7E3 | 3E3 | 50 |
| ${ }^{106} \mathrm{Ru}$ | 1E3-1E4 | <2E3 | 1 E 4 | 5E2 |
| ${ }^{137} \mathrm{Cs}$ | 1E6-1E7 | $1 E 7$ | 3E7 | >1E6 |
| ${ }^{144} \mathrm{Ce}$ | 1E4-1E5 | $\sim 2 \mathrm{E} 5$ | $\sim 2 \mathrm{E} 4$ | $\sim 4 \mathrm{E} 3$ |
| $1^{154} \mathrm{Eu}$ | 1E4-1E5 | >3E4 | $>5 \mathrm{E} 5$ | >9E4 |

a Used new solvent that was purified with sodium carbonate and water scrubs.
$b_{\text {Used }}$ degraded solvent that was purified with sodium carbonate and water scrubs.
${ }^{c}$ Used degraded solvent that was purified by base-treated silica gel.
Used degraded solvent without purification.

### 4.2.2 Partitioning

Concentration profiles for the partitioning bank are shown in Fig. 9. The plutonium losses to the uranium product during the runs with degraded solvent were larger than those measured during a previous run with new solvent (Table 4). Using degraded solvent without purification yielded the largest loss, $\sim 7 \%$. When the degraded solvent was purified by one of the treatment methods (sodium carbonate or silica gel), the plutonium loss was $\sim 0.5 \%$, which is only slightly poorer than the $0.1 \%$ that was measured for the run with new solvent.


Fig. 9. Concentration profiles for the partition contactor for the runs with new solvent and chemically degraded solvent.

Although the plutonium content in the degraded solvent was larger, the plutonium was not irreversibly lost. It was successfully stripped in the backup strip contactor ( $0.2 \mathrm{M} \mathrm{HNO}_{3}$ and 1.5 M hydroxylamine nitrate aqueous strip). The plutonium content of the final waste solvent from this contactor was $<1 \mathrm{mg} / \mathrm{L}$, which is similar to the plutonium concentrations achieved when using new solvent.

Table 4. Results from partial partitioning contactor for comparison runs using degraded and new solvents.

|  | Run No. |  |  |  |
| :---: | :---: | :---: | :---: | :---: |
|  | $3-2 c^{\text {a }}$ | 6-3b | 6-4c | 6-5d |
| Phase ratio (0/A) ${ }^{\text {e }}$ |  |  |  |  |
| Scrub section | 1.1 | 0.99 | 0.93 | 0.99 |
| Strip section | 4.0 | 3.5 | 2.8 | 3.0 |
| $\%$ of Pu in |  |  |  |  |
| $\mathrm{U}-\mathrm{Pu}$ product ( HBP ) | 99.9 | 99.6 | 99.5 | 93.1 |
| U product (HBU) | 0.1 | 0.4 | 0.5 | 6.9 |
| \% of $U$ in |  |  |  |  |
| U-Pu product (HBP) | 10.6 | 10.8 | 16.9 | 10.7 |
| U product (HBU) | 89.4 | 89.2 | 83.1 | 89.2 |
| Uranium product |  |  |  |  |
| $\mathrm{Pu}, \mu \mathrm{g} / \mathrm{g}$ of U | 9 | 37 | 55 | 589 |
| U-Pu product |  |  |  |  |
| $\mathrm{Pu}, \mathrm{g} / \mathrm{g}$ of U | 0.082 | 0.070 | 0.051 | 0.066 |

a Used new solvent purified with sodium carbonate and water scrubs.
$b_{\text {Used }}$ degraded solvent purified with sodium carbonate and water scrubs.
CUsed degraded solvent purified by base-treated silica gel.
dused degraded solvent without purification.
${ }^{e}$ (Organic flow rate)/aqueous flow rate).

### 4.3 TECHNETIUM DISTRIBUTION

The distribution of technetium was measured in the outlet streams for Run 6-2 (Table 5). This had not been done before because technetium is not detectable in routine gamma-scans and special analytical techniques are required to measure it. In the coextraction-coscrub bank, about 86\% of the technetium was measured in the aqueous waste which corresponds to a technetium DF of $\sim 7$ for the coextraction-coscrub bank. In the partitioning contactor, no significant separation of technetium and uranium was noted.

Table 5. Distribution of technetium and uranium in outlet streams for Run 6-2.

|  | Tc <br> $(\%)$ | U <br> $(\%)$ |
| :--- | :---: | :---: |
| Coextraction-coscrub bank |  |  |
| Aqueous waste (HAW) | 86 | 0.04 |
| U-Pu product (HAP) | 14 | 99.96 |
| Partial partitioning bank |  |  |
| U-Pu product (HBP) | 19 | 12 |
| Uranium product (HBU) | 81 | 88 |

5. SUMMARY AND CONCLUSIONS

The most significant results and conclusions from SETF Campaign 6 are as follows:

The extractant TEHP was tested and compared with the traditional TBP extractant for fuel reprocessing. Its use yielded no major advantages compared with TBP. The raffinate losses and fission product removals were similar; and although TEHP did initially cause some hydraulic problems,
they were corrected by minor changes in equipment and operational procedures. The only significant disadvantage was the poorer U-Pu separation with TEHP, which was primarily the result of poorer plutonium stripping. Using a different plutonium reductant or different flowsheet conditions might mitigate this difference with TBP.

Several tests were made to compare the solvent extraction behavior of (1) new solvent, (2) chemically degraded solvent ( 0.003 M DBP and $0.8 \%$ diluent degradation products), and (3) chemically degraded solvent that had been purified by either sodium carbonate or base-treated silica gel. When the degraded solvent was left unpurified, it yielded poorer DFs for ${ }^{95} \mathrm{Zr},{ }^{95} \mathrm{Nb}$, and ${ }^{106} \mathrm{Ru}$, in addition to much higher plutonium retention in the solvent from the partitioning bank ( $7 \%$ plutonium loss versus $0.1 \%$ for new solvent). Treating the solvent with either of the above purification methods, restored the fission product DFs to values similar to those obtained with new solvent, but still left a small amount of plutonium in the solvent from the partitioning contactor ( $0.5 \%$ ). Little difference was noted between the runs that used sodium carbonate or silica gel treated solvent, except for the ${ }^{95} \mathrm{Zr}$ DF; the lower ${ }^{95} \mathrm{Zr}$ DF for the silica-treated solvent may have resulted from inadequate water scrubbing following the treatment.

## 6. ACKNOWLEDGMENTS

The experimental work done in the SETF was performed by the Chemical Technology Division staff of the Transuranium Processing Plant. The preparation of the degraded solvent and the purification by silica gel was performed by 0. K. Tallent and J. C. Mailen of the Chemical Technology Division. The extensive analytical work was carried out by Analytical Chemistry Division personnel, primarily under the supervision of J. H. Cooper and J. M. Peele. Engineering and maintenance services were provided by personnel from the ORNL Engineering Division, the Instrumentation and Controls Division, and Plant and Equipment Division, under the supervision of S. O. Lewis, A. A. Shourbaji, E. M. Shuford, and B. R. Scarborough.

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## APPENDIX A

Table A.l. Campaign 6 first cycle tests - extraction/scrub bank conditions and results

|  | Run No. |  |  |  |  |
| :---: | :---: | :---: | :---: | :---: | :---: |
|  | 6-1 | 6-2 | 6-3 | 6-4 | 6-5 |
| Dates 1 | 10/19-21/81 | 10/29-31/81 | 11/9-10/81 | 11/17-18/81 | 12/7-8/81 |
| Bank temperature, ${ }^{\circ} \mathrm{C}$ | 51 | 51 | 51 | 52 | 51 |
| Number of stages final scrub/intermediate scrub/extraction | 6/3/7 | 6/3/7 | 6/3/7 | 6/3/7 | 6/3/7 |
| HAX stream flow rate, L/h | 1.50 | 1.56 | 1.52 | 1.45 | 1.54 |
| Flow ratios |  |  |  |  |  |
| has/HAX | 0.118 | 0.118 | 0.121 | 0.126 | 0.118 |
| hais/hax | 0.0609 | 0.060 | 0.060 | 0.064 | 0.064 |
| HAF/HAX | 0.259 | 0.230 | 0.564 | 0.591 | 0.525 |
| Inlet stream compositions |  |  |  |  |  |
| HAS Stream, $\mathrm{HNO}_{3}$, mol/L | L 0.51 | 0.49 | 0.48 | 0.48 | 0.52 |
| HAIS Stream, $\mathrm{HNO}_{3}$, mol/L HAX Stream | L 3.0 | 3.0 | 3.0 | 3.0 | 3.0 |
| $\begin{aligned} & \text { \% TBP } \\ & \% \text { TEHP } \end{aligned}$ | $28+0.5$ | 16.5+0.5 | $30 \pm 0.5$ | $30 \pm 0.5$ | $30 \pm 0.5$ |
|  |  |  |  |  |  |
|  | 3.08 | 3.04 | 2.84 | 2.88 | 2.76 |
|  | 149 | 153 | 163 | 158 | 144 |
|  | 1.31 | 1.18 | 1.17 | 1.31 | 1.21 |
|  | 49 | 48 | 56 | 52 | 53 |
|  | 2.77 | 2.53 | 2.59 | 2.68 | 2.44 |
|  | 1.48 | 2.18 | 1.32 | 0.895 | 0.87 |
|  | 1.34 | 1.05 | 1.57 | 1.23 | 0.90 |
|  | $<13$ | $<10$ | $<5$ | 7.14 | 7.7 |
|  |  | <6 | <3 | 3.8 | $\stackrel{6}{348}$ |
|  | 411 | 375 | 389 | 403 | 348 |
|  | <6 | <6 | <3 | <4 | <6 |
|  | 15.2 | 13.0 | 12.4 | 14.8 | 13 |

Table A-1 (continued)

|  | Run No. |  |  |  |  |
| :---: | :---: | :---: | :---: | :---: | :---: |
|  | 6-1 | 6-2 | 6-3 | 6-4 | 6-5 |
| Outlet stream compositions |  |  |  |  |  |
| HAW stream |  |  |  |  |  |
| $\mathrm{HNO}_{3}$, mol/L | 2.5 | 2.35 | 2.6 | 2.7 | 2.8 |
| U, mg/L | $<1$ | 31 | <1 | 4990 | 30 |
| Pu, mg/L | 0.23 | 0.24 | 2.25 | 39 | 0.35 |
| $241 \mathrm{Am}, \mathrm{mg} / \mathrm{L}$ | 35 | 34 | 44 | 49 | 44 |
| $244 \mathrm{Cm}, \mathrm{mg} / \mathrm{L}$ | 1.6 | 1.6 | 1.9 | 2.1 | 2.1 |
| ${ }^{95} \mathrm{Zr}, \mathrm{GBq} / \mathrm{L}$ | 0.919 | 0.57 | 0.74 | 1.14 | 0.418 |
| $95 \mathrm{Nb}, \mathrm{GBq} / \mathrm{L}$ | 0.703 | 0.55 | 0.97 | 1.25 | 0.698 |
| ${ }^{106} \mathrm{Ru}, \mathrm{GBq} / \mathrm{L}$ | <4 | $<4$ | 5.95 | 5.28 | 5.7 |
| $125 \mathrm{Sb}, \mathrm{GBq} / \mathrm{L}$ | <2 | <2 | 3.67 | 3.01 | <2 |
| 137 Cs , GBq/L | 240 | 224 | 304 | 296 | 281 |
| $144 \mathrm{Ce}, \mathrm{GBq} / \mathrm{L}$ | <2 | $<2$ | <2 | 5.09 | $<2$ |
| $154 \mathrm{Eu}, \mathrm{GBq} / \mathrm{L}$ | 8.4 | 7.52 | 10.2 | 11.8 | 10.8 |
| HAP stream. |  |  |  |  |  |
| $\mathrm{HNO}_{3}, \mathrm{~mol} / \mathrm{L}$ | 0.01 0.00045 | 0.01 | 0.0219 | 0.03 0.0030 | 0.05 0.0028 |
| HNO 2; mol/L $\mathrm{U}, \mathrm{~g} / \mathrm{L}$ | 0.00045 37.5 | 0.00078 31.4 | 0.0019 86.9 | 0.0030 80.3 | 73.5 |
| Pu, g/L | 0.327 | 0.262 | 0.63 | 0.69 | 0.60 |
| $95 \mathrm{Zr}, \mathrm{MBq} / \mathrm{L}$ | 0.294 | 0.020 | 0.035 | 0.251 | 109 |
| $95 \mathrm{Nb}, \mathrm{MBq} / \mathrm{L}$ | 0.048 | 0.050 | 0.12 | 0.211 | 9.47 |
| ${ }^{106} \mathrm{Ru}, \mathrm{MBq} / \mathrm{L}$ | 0.358 | 0.729 | 1.56 | 0.387 | 7.0 |
| $125 \mathrm{Sb}, \mathrm{MBq} / \mathrm{L}$ | $<0.02$ | $<0.02$ | $<0.03$ | $<0.03$ | $<0.5$ |
| 137 Cs , MBq/L | <0.005 | $<0.005$ | 0.016 | 0.0076 | $<0.1$ |
| $144 \mathrm{Ce}, \mathrm{MBq} / \mathrm{L}$ | $<0.06$ | <0.1 | $<0.1$ | $<0.09$ | $<0.6$ |
| $154 \mathrm{Eu}, \mathrm{MBq} / \mathrm{L}$ | <0.01 | $<0.02$ | $<0.02$ | <0.02 | $<0.07$ |

Table A-2. Campaign 6 first-cycle tests - strip bank conditions and results

|  |  | Run No. |  |  |  |  |  |  |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  | 6-1A | 6-1B | 6-2A | 6-2B | 6-3 | 6-4 | 6-5 |
| Dates |  | 10/19-20/81 | 10/20-21/81 | 10/29-30/81 | 10/30-31/81 | 11/9-10/81 | 11/17-18/81 | 12/7-8/81 |
| Bank te | mperature, ${ }^{\circ} \mathrm{C}$ | 25 | 25 | 32 | 32 | 25-26 | 25 | 25 |
| Number of stages Strip/scrub |  |  |  |  |  |  |  |  |
|  |  | 13/3 | 13/3 | 13/3 | 13/3 | 13/3 | 13/3 | 13/3 |
| BX stream flow rate, $\mathrm{L} / \mathrm{h}$ Flow ratios |  | 0.307 | 0.303 | 0.304 | 0.304 | 0.616 | 0.760 | 0.752 |
|  |  |  |  |  |  |  |  |  |
| HAP/ | / HBX | 4.89 | 4.95 | 5.12 | 5.12 | 2.47 | 1.91 | 2.04 |
| HBS/ | / HBX | 1.93 | 1.95 | 2.07 | 2.07 | 0.989 | 0.933 | 0.989 |
| Inlet stream compositions |  |  |  |  |  |  |  |  |
| HBX stream, $\mathrm{HNO}_{3}$, mol/L HBS stream |  | L 0.10 | 0.10 | 0.10 | 0.10 | 0.10 | 0.10 | 0.10 |
|  |  |  |  | 16.5+0.5 | 16.5+0.5 | 30+0.5 | 30+0.5 | $30+0.5$ |
|  | \% TEHP | 28+0.5 | $28+0.5$ |  |  |  |  |  |
|  | $\mathrm{HNO}_{3}$, mol/L | $<0 . \overline{0} 1$ | $<0 . \overline{01}$ | $<0.01$ | $<0.01$ | 0.02 | 0.01 | 0.02 |
|  | $\mathrm{HNO}_{2}$, mol/L | 0.0085 | 0.043 | 0.0085 | 0.040 | 0.083 | 0.089 | 0.099 |
|  | Stream |  |  |  |  |  |  |  |
|  | \% TBP |  |  | $16.5 \pm 0.5$ | $16.5 \pm 0.5$ | $30 \pm 0.5$ | $30 \pm 0.5$ | $30 \pm 0.5$ |
|  | \% TRHP | $28+0.5$ | $28+0.5$ |  |  |  |  |  |
|  | $\mathrm{HNO}_{3}$, mol/L | $0 . \overline{0} 1$ |  | 0.01 |  | 0.02 | 0.03 | 0.05 |
|  | $\mathrm{HNO}_{2}, \mathrm{~mol} / \mathrm{L}$ | 0.00045 |  | 0.00078 |  | 0.0019 | 0.0030 | 0.0028 |
|  | $\mathrm{U}, \mathrm{g} / \mathrm{L}$ | 37.5 |  | 31.4 |  | 86.9 | 80.3 | 73.5 |
|  | ${ }_{9} \mathrm{Pu}, \mathrm{g} / \mathrm{L}$ | 0.327 |  | 0.262 |  | 0.63 | 0.69 | 0.60 |
|  | ${ }^{95} \mathrm{Zr}, \mathrm{MBq} / \mathrm{L}$ | 0.294 |  | 0.020 |  | 0.035 | 0.251 | 109 |
|  | ${ }^{95} \mathrm{Nb}, \mathrm{MBq} / \mathrm{L}$ | 0.048 |  | 0.050 |  | 0.12 | 0.211 | 9.47 |
|  | $106 \mathrm{Ru}, \mathrm{MBq} / \mathrm{L}$ $125 \mathrm{Sb}, \mathrm{MBa} / \mathrm{L}$ | 0.358 $<0.02$ |  | 0.729 $<0.02$ |  | 1.56 $<0.03$ | 0.387 $<0.03$ | 7.0 $<0.5$ |
|  | ${ }^{137} \mathrm{Cs}$, MBq/L | $<0.005$ |  | <0.005 |  | -0.016 | <0.03 0.0076 | $<0.5$ $<0.1$ |
|  | $1{ }^{144} \mathrm{Ce}, \mathrm{MBq} / \mathrm{L}$ | <0.06 |  | <0.1 |  | <0.1 | $<0.09$ | <0.6 |
|  | ${ }^{154} \mathrm{Eu}, \mathrm{MBq} / \mathrm{L}$ | <0.01 |  | $<0.02$ |  | <0.02 | <0.02 | <0.07 |

Table A-2 (continued)

|  | Run No. |  |  |  |  |  |  |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  | 6-1A | 6-18 | 6-2A | 6-2B | 6-3 | 6-4 | 6-5 |
| t stream composition |  |  |  |  |  |  |  |
| HBP Stream |  |  |  |  |  |  |  |
| $\mathrm{HNO}_{3}, \mathrm{~mol} / \mathrm{L}$ | 0.18 | 0.16 | 0.14 | 0.20 | 0.19 | 0.26 | 0.20 |
| $\mathrm{U}, \mathrm{g} / \mathrm{L}$ | 19.0 | 21.3 | 19.4 | 29.4 | 23.3 | 26.4 | 17.4 |
| Pu, g/L | 1.54 | 1.58 | 1.38 | 1.44 | 1.64 | 1.34 | 1.14 |
| \% Pu(IV) |  |  |  |  | 42 | 47 | 44 |
| ${ }^{95} \mathrm{Zr}, \mathrm{MBq} / \mathrm{L}$ | 1.84 | 1.56 | 0.0674 | 0.047 | 0.0365 | 0.398 | 188 |
| ${ }^{95} \mathrm{Nb}, \mathrm{MBq} / \mathrm{L}$ | 0.373 | 0.334 | 0.0399 | 0.0442 | 0.0581 | 0.128 | 5.8 |
| ${ }^{106} \mathrm{Ru}, \mathrm{MBq} / \mathrm{L}$ | <0.1 | $<0.1$ | <0.06 | <0.06 | <0.05 | <0.08 | $<0.9$ |
| $125 \mathrm{Sb}, \mathrm{MBq} / \mathrm{L}$ | <0.04 | <0.05 | $<0.02$ | <0.02 | <0.02 | <0.02 | <0.5 |
| ${ }^{137} \mathrm{Cs}$, MBq/L | 0.039 | 0.060 | 0.0208 | 0.0101 | 0.0156 | 0.070 | 0.28 |
| $1{ }^{144} \mathrm{Ce}, \mathrm{MBq} / \mathrm{L}$ | $<0.1$ | $<0.1$ | <0.1 | $<0.25$ | $<0.1$ | $<0.1$ | <0.5 |
| $154 \mathrm{Eu}, \mathrm{MBq} / \mathrm{L}$ | <0.02 |  | <0.01 | <0.02 | $<0.01$ | $<0.02$ | <0.08 |
| HBU stream |  |  |  |  |  |  |  |
| $\mathrm{HNO}_{3}$, mol/L | $<0.01$ | <0.01 |  |  | 0.01 | 0.02 | 0.03 |
| $\mathrm{HNO}_{2}, \mathrm{~mol} / \mathrm{L}$ | 0.0020 | 0.0083 | 0.0024 | 0.0080 | 0.018 | 0.024 | 0.024 |
| $\mathrm{U}, \mathrm{g} / \mathrm{L}$ | 25.6 | 21.8 | 20.2 | 20.5 | 55.2 | 45.8 | 47 |
| Pu, mg/L | 6.1 | 3.4 | 0.53 | 0.24 | 2.1 | 2.5 | 27.8 |
| ${ }^{95} \mathrm{Zr}$, MBq/L | 0.039 | 0.020 | 0.0164 | 0.0092 | 0.022 | 0.0507 | 20.9 |
| $95 \mathrm{Nb}, \mathrm{MBq} / \mathrm{L}$ | 0.012 | 0.012 | 0.0237 | 0.0284 | 0.0774 | 0.158 | 5.06 |
| 106 Ru , MBq/L | 0.320 | <0.24 | 0.536 | 0.554 | 1.42 | 0.266 | 5.2 |
| $125 \mathrm{Sb}, \mathrm{MBq} / \mathrm{L}$ | $<0.02$ | $<0.02$ | $<0.02$ | 0.02 | $<0.03$ | <0.02 | $<0.2$ |
| ${ }^{137} \mathrm{Cs}, \mathrm{MBq} / \mathrm{L}$ | <0.006 | $<0.003$ | $<0.004$ | 0.0037 | $<0.006$ | 0.0059 | <0.04 |
| $1{ }^{144} \mathrm{Ce}, \mathrm{MBq} / \mathrm{L}$ | <0.04 | $<0.03$ | $<0.04$ | $<0.03$ | <0.06 | $<0.05$ | $<0.2$ |
| $154 \mathrm{Eu}, \mathrm{MBq} / \mathrm{L}$ | <0.01 | <0.01 | <0.01 | <0.01 | <0.02 | <0.01 | <0.02 |

Table A-3. Campaign 6 first cycle tests - Pu scavenging bank
conditions and results

|  | Run No. |  |  |  |  |
| :---: | :---: | :---: | :---: | :---: | :---: |
|  | 6-1A | 6-2A | 6-3 | 6-4 | 6-5 |
| Dates | 10/19-20/81 | 10/29-30/81 | 11/9-10/81 | 11/17-18/81 | 12/7-8/81 |
| Bank temperature, ${ }^{\circ} \mathrm{C}$ | 40 | 31-32 | 41-42 | 41 | 41 |
| Number of stages | 16 | 16 | 16 | 16 | 16 |
| HCX stream flow rate, L/h | 0.195 | 0.762 | 0.205 | 0.203 | 0.203 |
| Flow ratios |  |  |  |  |  |
| HBU/CX | 10.7 | 2.87 | 10.4 | 10.6 | 11.2 |
| Inlet stream compositions HCX Stream |  |  |  |  |  |
|  |  |  |  |  |  |
| . $\mathrm{HNO}_{3}$, mol/L | 0.17 | 0.10 | 0.2 | 0.21 | 0.22 |
| HAN, mol/L | 1.7 |  | 1.6 | 1.5 | 1.5 |
| HBU Stream \% TBP |  | $16.5+0.5$ | 30+0.5 | 30+0.5 | 30+0.5 |
| \% TEHP | $28+0.5$ |  |  |  |  |
| $\mathrm{HNO}_{3}$, mol/L | $<0 . \overline{01}$ |  | 0.01 | 0.02 | 0.03 |
| $\mathrm{HNO}_{2}, \mathrm{~mol} / \mathrm{L}$ | 0.0020 | 0.002 | 0.018 | 0.024 | 0.024 |
| $\mathrm{U}, \mathrm{g} / \mathrm{L}$ | 25.6 | 20.2 | 55.2 | 46 | 47 |
| Pu, mg/L | 6.1 | 0.53 | 2.1 | 2.5 | 27.8 |
| ${ }^{95} \mathrm{Zr}, \mathrm{MBq} / \mathrm{L}$ | 0.039 | 0.016 | 0.022 | 0.051 | 20.9 |
| $95 \mathrm{Nb}, \mathrm{MBq} / \mathrm{L}$ | 0.012 | 0.024 | 0.0774 | 0.158 | 5.1 |
| $10{ }^{\text {R }}$ Ru, MBq/L | 0.320 | 0.54 | 1.42 | 0.266 | 5.2 |
| $125 \mathrm{Sb}, \mathrm{MBq} / \mathrm{L}$ | $<0.02$ | <0.02 | $<0.03$ | <0.02 | $<0.2$ |
| $137 \mathrm{Cs}, \mathrm{MBq} / \mathrm{L}$ | $<0.006$ | $<0.004$ | $<0.006$ | 0.0059 | $<0.04$ |
| $1{ }^{14} \mathrm{Ce}, \mathrm{MBq} / \mathrm{L}$ | $<0.04$ | $<0.04$ | $<0.06$ | $<0.05$ | $<0.2$ |
| $15{ }^{4} \mathrm{Eu}, \mathrm{MBq} / \mathrm{L}$ | <0.01 | <0.01 | $<0.02$ | $<0.01$ | <0.02 |

Table A-3 (continued)


ORNL/TM-9961
Dist. Category UC-86T
(Applied)

INTERNAL DISTRIBUTION

| 1-5. | D. E. Benker |
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83-128. Given distribution as shown in TIC-4500 under UC-86T, Consolidated Fuel Reprocessing Category (Applied)
