

Fission Product Decontamination Factors for Plutonium Separated by PUREX from above Low Burn-up, fast-neutron irradiated Depleted UO_2

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Abstract

Experimental investigations to determine fission product (FP) separation from actinides (U and Pu) while employing the Plutonium Uranium Redox Extraction (PUREX) process to purify plutonium produced in a fast neutron irradiated depleted uranium dioxide (DUO_2) target were conducted. The sample was a DUO_2 surrogate pellet (0.2562 wt.% *initial* ^{235}U) irradiated to a low-burn-up (4.43 ± 0.31 GWd/tHM) that was PUREX process 538 days after neutron irradiation. Decontamination factors (DF) for elements U, Mo, Ru, Ce, Sm, Sr, Pm, Eu, Nd, Pd, Cd, Ba and Sn for two PUREX experiments using 30 vol.% tri-n-butyl phosphate in a kerosene diluent with less than 0.3% uranium saturation in 4 M nitric acid were determined. The first experiment characterized Pu DFs for a single contact extraction and back-extraction while the second had multiple contacts for larger product recovery. The bench-top scale PUREX process had an overall 76% and 94% Pu recovery and an overall activity decontamination factor of 20 and 5 for the first and second experiments respectfully.

Keywords: PUREX, Decontamination Factor, Depleted Uranium

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1. Introduction

Background. In a recent publication, our group suggested that investigation of PUREX-processed plutonium for trace contaminants could give indication of

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material origins, but that a broad study of many elements would be necessary

5 [1]. Descriptions of various PUREX processes are provided in many sources [2, 3, 4] with explanations of chemistry including flow sheets and DF values, while additional sources provide other DFs for PUREX [5, 6, 7]. These sources generally report overall beta or gamma DFs of up to 10^8 with Pu recoveries of 99.7% for industrial-scale reprocessing facilities.

10 While distribution coefficients (DC) for the various process separation steps of PUREX have been previously reported, details about elemental DFs for PUREX cycles have been largely limited to the major activity contributors, such as ^{106}Ru and ^{95}Zr [3]. A compilation of distribution data for PUREX extraction processes provide DC information for U, Th, and Pu in a variety of
15 concentrations [8]. DCs for Zr, rare earth metals, Pu, and Th are also available [9, 10, 11, 12, 13, 14, 15]

Although a DC (coupled with process information) can be used to calculate a reasonable estimate of DF [16, 17, 18, 19], variability of DCs under different system conditions give rise to uncertainty in calculated results. For example,
20 DCs between tri-n-butyl phosphate (TBP) and nitric acid (HNO_3) have been reported for U, Pu, Zr, Nb, Ru, and the rare earth elements, but vary with nitric acid concentration and uranium saturation in TBP [17, 3]. These sources also derive mathematical correlations between DC and DF, but experimental PUREX DFs for a large number of individual elements were not provided. Ad-
25 ditionally, gallium has been studied for separation [20] because it is a common contaminate in weapons grade Pu.

In the current work, a 12.9 mg of DUO_2 was irradiated in a pseudo-fast neutron spectrum at the High Flux isotope Reactor at Oak Ridge National Laboratory. The DUO_2 pellet, containing FP and weapons-grade Pu, was dissolved
30 into a nitric acid solution and subjected to two different PUREX experiments

for DF characterization and Pu product recovery. Solutions were analyzed at each step with Inductively Coupled Plasma-Mass Spectrometry (ICP-MS). The experimental work used bench-top-scale methods to isolate a large fraction of Pu, measure DFs for fission products, and measure gamma DFs as part of a
35 larger project to develop forensic radio-analytical capabilities at Texas A&M University.

Terminology. A DCs is defined as the concentration ratio between the organic (org) and aqueous (aq) phases as shown in Equation 1, and describes the steady-state distribution of any species in the system during PUREX separation pro-
40 cesses [4]:

$$DC = \frac{c_{org}}{c_{aq}} \quad (1)$$

where c is the concentration of the specific species in the indicated phase. DCs are specific to an element and vary widely with the concentration and temperature of the solvents. They are also affected by saturation of uranium and plutonium in the system and time since preparation of the solution [3, 21].
45 For PUREX, the fraction of mass f_{org} deposited in the organic (TBP) phase for a single element (assuming equal contact volumes) is given by Equation 2.

$$f_{org} = (1 + DC^{-1})^{-1} \quad (2)$$

After several cycles of plutonium extraction/decontamination are complete, the measured effectiveness of a PUREX cycle is described by the DF, which is fundamentally determined by DCs and measure the effectiveness with which a
50 contaminant, j , is removed from a product. In this work, the product of interest is plutonium, and the DF is defined by Equation 3.

$$DF_j = \frac{\left. \frac{c_j}{c_{Pu}} \right|_{initial}}{\left. \frac{c_j}{c_{Pu}} \right|_{final}} \quad (3)$$

Initial and *final* refer to the values before and after purification, respectively. DFs are also characteristic of different process cycles, and may have larger values ($> 10^7$) for industrial scale PUREX compared to the bench-top
 55 scale version presented here [3, 4].

Industrial processes report either an overall DF value, or a DF value for a single isotope. What is needed for forensics purposes is DFs for individual FP contaminants, which is why elemental DF values were obtained for bench-top-scale PUREX process performed on a DUO₂ surrogate sample.

60 2. Experiment

A commercially acquired pellet containing 12.9 ± 0.1 mg of DUO₂ was irradiated over the course of three months with two shut down periods in the HFIR flux spectrum to 4.43 ± 0.31 GWd/tHM [22]. The burn-up was determined by measuring the ¹³⁷Cs activity. This produced 0.237 ± 0.008 mg of Pu.
 65 After the short lived radioisotopes had opportunity to decay, the irradiated pellet was shipped to Texas A&M University, counted with a standard Canberra electrode coaxial Ge detector to determine relative activity, and transferred to a round-bottom flask.

Samples were prepared as shown in Figure 1, and described below. 5.0 ml of 8 M HNO₃ was added to the flask, which was heated to 50 °C with constant 100 rpm stirring for two hours. This solution will be referred to as the “dissolution solution”. The flask was connected to a cold trap with the help of a Schlenk line. The fission product gases such as H₂, CO₂, Kr, Br, I and N₂O were captured in a cold trap containing molecular sieves that were chilled by liquid

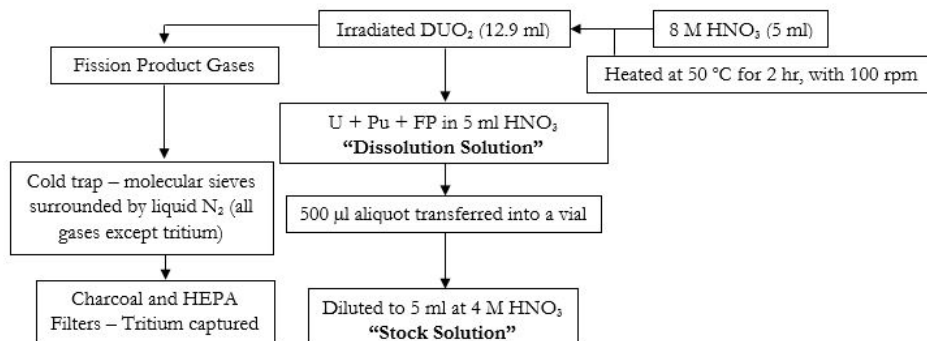


Figure 1: Flow chart for dissolution.

nitrogen. In order to reduce the amount of activity per sample, 500 μl from the dissolution solution was diluted to 5.0 ml and the concentration was changed to 4 M HNO_3 . From this solution, referred to as the “stock solution”, 0.5 ml aliquotes, containing $\sim 1\%$ of the pellet, were used in bench-top-scale PUREX experiments described in the two subsections below. The activity concentration of the stock solution was approximately 80 $\mu\text{Ci}/\text{ml}$.

Experiments started with transferring a 500 μl aliquot of stock solution and
 70 0.5 mg of NaNO_2 to a 15 ml centrifuge tube. The tube was subsequently stirred and covered to retain the resulting NO_2 gas. The solution was left overnight so that NO_2 gas completely oxidized Pu(III) to Pu(IV) . During extraction and back-extraction, both experiments had the aqueous and organic phases mixed on a vortex mixer for 15 minutes at 1500 rpm, after which the two phases were
 75 allowed to settle and separate. The phases were physically separated into two different vials through careful pipetting.

Each extraction and back-extraction mixed organic and aqueous mixtures with unequal volumes. The solution being added always contained an extra 200 μl to reduce the chance of accidentally pipetting HNO_3 . For example, if TBP
 80 were being added to the stock solution described above, 700 μl would be added

initially and 500 μl removed. This excess volume will be later referenced as hold-up volume.

The kerosene (100%) and sodium nitrite (100%) used for these experiments were acquired from sciencelab.com [23], 69% nitric acid was acquired from
85 Mallinckrodt Chemicals [24], Tri-n-butyl phosphate (>99%) was acquired from Fisher Scientific [25], and iron sulfamate (40.26%) was acquired from Strem Chemicals Incorporation [26].

The pellet, both prior to dissolution and after, was counted with a Canberra HPGe detector model number CC4018 which was connected to a Canberra Lynx
90 MCA [27, 28]. Canberra’s software package GENIE-2000 version 3.2.1 [29] was used to collect spectra with samples inside a lead tomb. The same detector was used to count the various process solutions. Inductively coupled plasma mass spectrometry (ICP-MS) was also conducted for aqueous samples using a PerkinElmer NexION 300X quadrupole ICP-MS [30].

95 *First Experiment.* The purpose of the first experiment was to quantify product recovery and DF values for a single contact extraction and back-extraction of Pu. U(VI) and Pu(IV) were extracted and decontaminated by contacting the prepared stock solution with a solution of 30 vol.% TBP with a kerosene diluent. After mixing, and separation of the two phases, Pu(IV) was reduced to Pu(III)
100 and back-extracted by contacting the removed TBP solution with 0.75 M HNO_3 in a 0.024 M ferrous sulfamate solution via oxidation of Fe(II). The stock solution both before and after TBP contact, as well as the final solution containing back-extracted Pu, were analyzed with ICP-MS.

Second Experiment. The purpose of the second experiment was to extract a
105 large fraction of Pu. Utilizing the results from the first experiment, it was determined that contacting the prepared stock solution four times with TBP would extract over 90% of the Pu. Therefore, this experiment had four TBP

contacts with the prepared stock solution. The TBP was then collected into a single vial, and contacted three times with the ferrous sulfamate solution.

110 In order to ensure minimal U back-extraction, the HNO_3 concentration for this experiment was increased to 4 M because NO_3^- concentrations affect the distribution ratio for U. Higher HNO_3 concentrations reduce the degree to which U is back-extracted [3]. Three contacts of the ferrous sulfamate solution ensured complete back-extraction of Pu, while the higher nitric acid concentration min-
115 imized back-extraction of U. The same solutions as described in the first experiment were analyzed with ICP-MS. The final back-extracted Pu solution underwent the 4 extraction 3 back-extraction process once more to verify the repeatability of the process and for comparison with the first extraction/back-extraction cycle. The final solution Pu was reset with the addition of 0.5 mg of
120 NaNO_2 to convert all the Pu(III) to Pu(IV).

3. Results

The Pu recovery for the first and second experiments are shown in Table 1. For experiment 1, the amount of U recovery was much higher than for experiment 2, even with multiple contacts. This is due to the different HNO_3
125 concentrations for the back-extraction solutions between the two experiments, as described earlier. The second experiment second cycle had a 90% Pu recovery with an 95% of the remaining U left in the organic phase. The reason the second cycle did not perform as well as the first was due to Fe(II) catalytically oxidizing to Fe(III) with NO_2^- [3].

130 The DCs for U and Pu were 37.2 ± 5.3 and 16.2 ± 2.3 , respectfully, which indicate that large fractions of the U and Pu were removed from stock solutions. Using Equation 2, which assumes equal volumes, with these two DC values, over 99.99% of the U and Pu should be extracted, with nearly all of the Pu being

back-extracted. The reason for lower recovery values is due to hold-up volumes
left in the extraction/back-extraction vials [18]. This is be further explained in
the next section.

Table 1: Recoveries of U and Pu for the different experiments

	Pu Recovery	\pm	U Recovery	\pm
Experiment 1	76%	0.30%	25%	0.03%
Experiment 2 Cycle 1	94%	0.90%	7%	0.06%
Experiment 2 Cycle 2	90%	0.60%	5%	0.10%
Experiment 2 Cycles 1&2	85%	0.99%	0.35%	0.08%

The DF calculations utilized concentration ratios between contaminants that
were normalized to the Pu so that volume changes due to processing were
negated, as shown in Equation 3. Both the first and second experiment first
cycle DF values are shown in Table 2. It should be noted that the Ba calculation
utilized ^{138}Ba with background Ba subtracted; the background was determined
with ^{134}Ba , and is subject to very high errors due to the low amounts of ^{134}Ba
in the system.

Table 2: Decontamination factors for single and multiple contact PUREX

Element	Exp 1	\pm	Exp 2	\pm	Isotopes Used
Rb	32	1.55	1.84	0.26	^{85}Rb
Sr	233.5	12.74	38.26	2.23	^{90}Sr
Mo	20.67	2.03	1.19	0.25	^{97}Mo
Ru	49	1.90	2.84	0.11	$^{101,102,104}\text{Ru}$
Pd	65	14.3	3.62	0.94	$^{108,110}\text{Pd}$
Cd	61	6.60	3.50	0.98	^{112}Cd
Sn	7.45	0.43	13.85	1.29	^{119}Sn
Cs	146	7.58	11.92	0.96	^{133}Cs
Ba	344	200	0.39	50	$^{134,138}\text{Ba}$
Ce	35.24	1.68	3.2	0.67	$^{140,142}\text{Ce}$
Nd	16.37	0.65	5.94	2.01	^{143}Nd
Pm	10.70	0.66	3.3	0.50	^{147}Pm
Sm	9.94	0.25	2.5	0.19	^{151}Sm
Eu	8.40	0.49	2.6	0.23	^{154}Eu
U	6.85	0.46	15.08	0.60	^{238}U

The common trend is that the DF gets worse from the first experiment to

145 the second. The major exception is U, which has a better DF value. This is expected due to the change in HNO_3 concentration in the iron sulfamate solution, and has been explained previously. The rest of the elements have worse DF values because of multiple contacts, and are low in general because of the volume difference between the phases and hold-up volumes.

150 *Volumes Differences and Multiple Contacts.* An excellent description for how hold-up volumes decrease DF values is given in [18], in the differential extraction section. In short, if TBP is used to extract Pu, and if some TBP is left in contact with the nitric acid as hold-up, then some Pu will be lost.

Another reason why the DF values were lower than industrially reported
155 values is due to the fact that the volume ratio of aqueous and organic phase was less than unity. If Equation 2 were rewritten to include the volume ratio between the aqueous and organic phase, V_R , then the fraction of mass that passes to the organic phase, f_{org} , would be given by Equation 4.

$$f_{org} = (1 + DC^{-1}V_R)^{-1} \quad (4)$$

If the aqueous phase volume is less than the organic phase volume, then a
160 larger percentage of contaminant will pass into the organic phase. More Pu(IV) and U(VI) will also be extracted, but since their DC values are large, the effect is not as tangible as for the fission products, with DC values below one. This effect is shown in Figure 2, where theoretical DFs for the extraction step is shown as a function of volume ratio and number of contacts.

165 Figure 2 shows how DF decreases with increasing volume of TBP and with the number contacts. The number of contacts decreases DF because less and less product is removed with each extraction, while the amount of contaminant removed is about the same.

Using equations described previously, the DF for both experiments can be

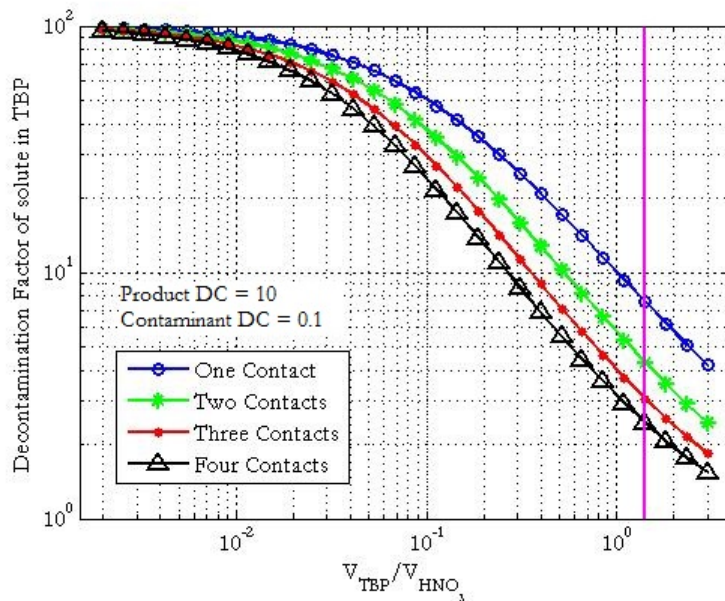


Figure 2: DFs as a function of volume ratios for the first to fourth contact in TBP.

170 calculated with the assumption that the DC for both the extraction and back-extraction are equal. Simulating these experiments and plotting DC versus DF is shown in Figure 3. The ratio of the DF for the second experiment and first is about 0.30. Which means that the second experiment DF value should be no better than 30% of the first experiment. Looking back to Table 2, the ratio of
 175 the DF values between the second and first experiments, excluding U, is 0.27, with values above and below.

4. Conclusions

Two experiments were conducted to quantify DF values for a variety of elements as well as extract a large fraction of Pu. It was determined that the
 180 volume ratio between organic and aqueous phases in extraction have an impact on DF values, and that multiple extraction steps lead to large product recovery,

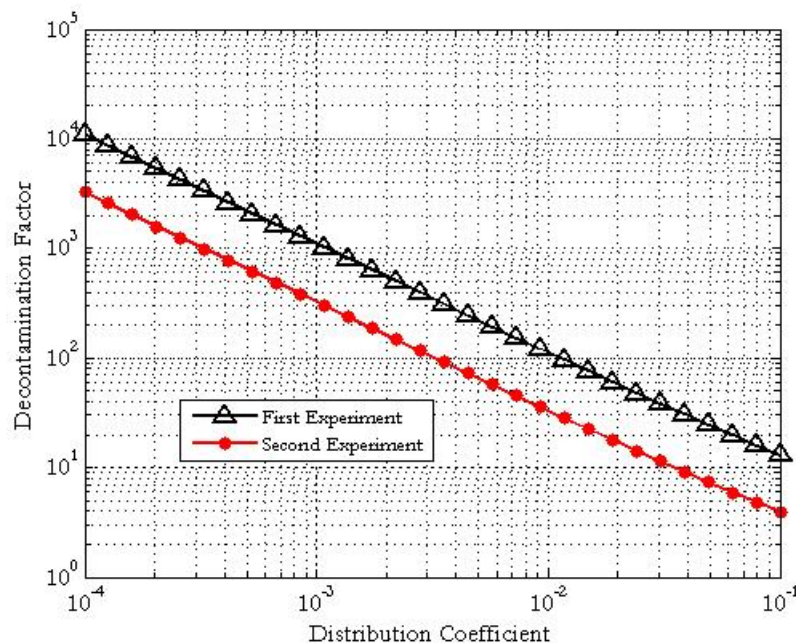


Figure 3: Decontamination factors as a function of Distribution Coefficient for the first and second experiment.

but can also decrease DF values. Future experiments will utilize a scrubbing stage in the PUREX process.

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