

Uncertainty quantification of depletion calculations for specific isotopes using ORIGEN2

Paul Mendoza

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SCIENCE & POLICY INSTITUTE**

Outline

Background

- Introduction

- Current Problem

Future

- The PUREX Process

- Distribution Coefficients

- Decontamination Factors

Previous Work

- Experiment

- Recovery of Pu and U

- Experimental Decontamination Factors

Future Work

- ❖ Determining composition of irradiated fuel importance
 - Flux calculations
 - Reprocessing
 - Irradiation history verification
- ❖ Usually determined with a type of Bateman solver
- ❖ Uncertainties rarely reported
 - Flux Shape
 - Fission yield
 - **Cross Sections**
 - Half-lives

Current Problem

- ❖ Use of depletion code ORIGEN2
 - Solves with exponential method
 - Requires library
 - ◆ Decay information
 - ◆ Fission yield data
 - ◆ Single group cross sections
- ❖ PWR system with 3 wt% enriched uranium
- ❖ Varied ^{235}U , ^{238}U , ^{239}Pu , ^{240}Pu , and ^{241}Pu
 - Originally, σ_γ and γ for the FP were considered

Isotope solve list

^{133}Cs	^{136}Ba	^{153}Eu
^{134}Cs	^{138}Ba	^{154}Eu
^{135}Cs	^{149}Sm	^{239}Pu
^{137}Cs	^{150}Sm	^{242}Pu
^{148}Nd	^{106}Rh	^{125}Sb

Future

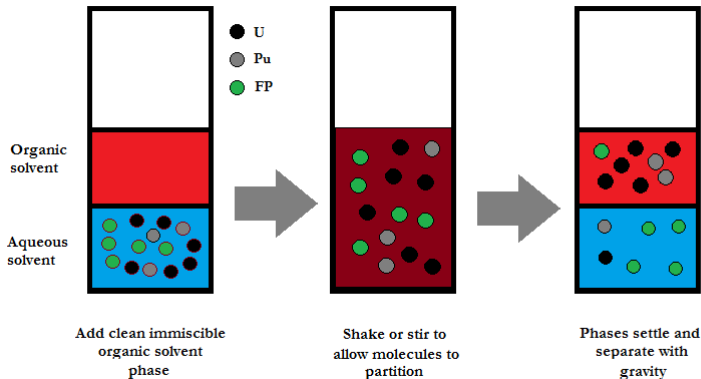
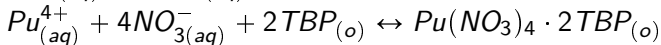
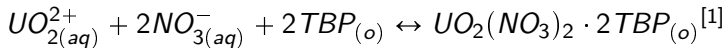
What is PUREX - A type of laundry detergent?

❖ Plutonium Uranium Redox EXtraction

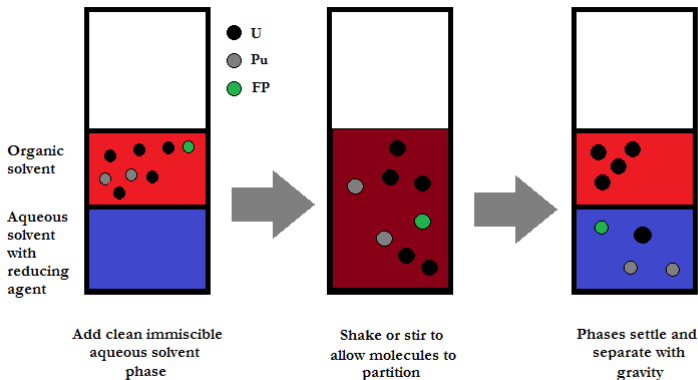
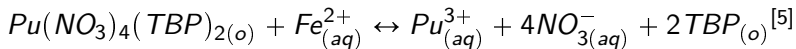
- Liquid-liquid solvent extraction
- Many stages:
 1. Preparation for Dissolution
 2. Dissolution
 3. Preparation of Dissolved Feed
 4. Primary Decontamination - Extraction to organic^{*}
 5. Scrubbing
 6. Plutonium Partition - Back-Extraction to aqueous^{*}
 7. Plutonium Purification

* - Discussing Next

Extraction



Back-Extraction



Distribution Coefficients - The Missing link

- ❖ Distribution Coefficient (D): The ratio between the organic and aqueous phases (aka: D-values)

$$D = \frac{C_o}{C_{aq}}$$

- ❖ Specific element to element
- ❖ Vary widely with:^[7]
 - Composition of phases
 - Solution saturation
 - Temperature of the solvent
- ❖ The fraction of mass, f_o deposited in the organic phase, assuming a volume ratio between the aqueous and organic phases, V_R , is:

$$f_o = (1 + D^{-1}V_R^{-1})^{-1}$$

Decontamination Factors - The Pot of gold

- ❖ After several cycles of Pu extraction/scrubbing/back-extraction are completed, the effectiveness of a PUREX cycle is described by the decontamination factor (DF):

$$DF_j = \frac{\left| \frac{c_j}{c_{Pu}} \right|_{initial}}{\left| \frac{c_j}{c_{Pu}} \right|_{final}}$$

- ❖ DFs are characteristic of different process cycles
- ❖ Larger values (10^7) for industrial scale PUREX (compared to benchtop)^[7, 1]

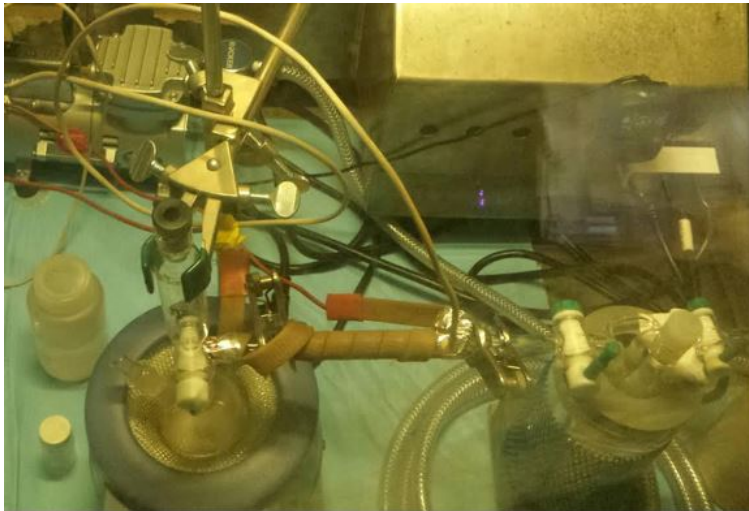
Previous Work

Irradiation

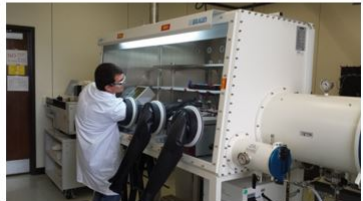
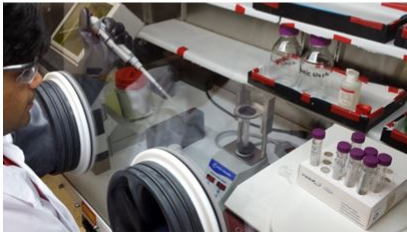
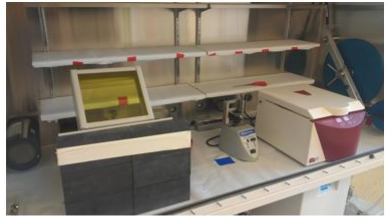
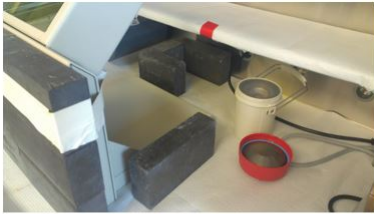
- ❖ 12.9 ± 0.1 mg of DUO_2 was irradiated
 - High Flux Isotope Reactor at Oak Ridge National Laboratory
- ❖ Burnup was 4.43 ± 0.31 GWd/tHM^[8]
- ❖ $0.196 \pm$ mg of total Pu was produced as measured by ICP-MS



Dissolution of the spent fuel pellet



Glovebox



❖ Single stage extraction and back-extraction

- Purpose: quantify product recovery, D-values and DF values for single stage extraction and back extraction
- Conditions:

Starting Solution	Extraction Solution	Back extraction solution
4 M nitric acid	30% vol.% TBP, 70 vol.% kerosene	0.024 M ferrous sulfamate in 0.75 M nitric acid

❖ Multi-contact extraction and back-extraction

- Purpose: Maximize recovery of Pu with 4 extractions, 3 back extractions
- Conditions:

Starting Solution	Extraction Solution	Back extraction solution
4 M nitric acid	30% vol.% TBP, 70 vol.% kerosene	0.024 M ferrous sulfamate in 4 M nitric acid

Previous Experiment Results

Recoveries of U and Pu

	Pu Recovery	U Recovery
Single stage	$(83.4 \pm 9.5)\%$	$(11.2 \pm 1.3)\%$
Multi-contact Cycle 1	$(99.7 \pm 4.2)\%$	$(6.8 \pm 0.3)\%$
Multi-contact Cycle 2	$(93.0 \pm 4.6)\%$	$(6.6 \pm 0.3)\%$
Overall Experiment 2	$(92.7 \pm 6.0)\%$	$(0.45 \pm 0.03)\%$

Previous Experiment Results

Decontamination Factors

Element (Z)	SS	Error	MC Cycle 1	Error	Isotopes Used
Rb(37)	39.0	5.9	11.8	0.8	⁸⁵ Rb
Sr(38)	283	43	84.6	5.9	⁹⁰ Sr
Mo(42)	5.7	0.8	1.9	0.2	^{97,98,100} Mo
Ru(44)	59.2	6.4	16.6	2.5	^{101,102,104} Ru
Pd(46)	65	14	8.9	1.2	¹¹⁰ Pd
Cd(48)	74	17	22.1	2.5	¹¹² Cd
Cs(55)	177	28	52.9	3.9	¹³³ Cs
Ce(58)	43	16	11.5	4.9	^{140,142} Ce
Nd(60)	19.2	2.1	5.9	0.4	¹⁴³ Nd
Pm(61)	12.8	1.9	3.9	0.3	¹⁴⁷ Pm
Sm(62)	11.5	1.5	3.6	0.3	¹⁵¹ Sm
Eu(63)	10.0	1.4	3.6	0.3	¹⁵⁴ Eu
U(92)	7.4	1.2	14.7	0.9	²³⁸ U

Conclusions

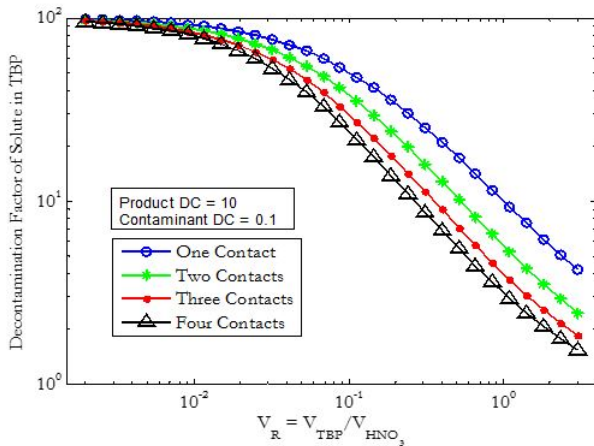
- ❖ Two PUREX experiments were conducted
 - Single stage: Determined DC values for Pu, U and several FP
 - Multi-contact: Utilized Experiment 1 to recover over 92% of Pu while leaving less than 1% of the U
- ❖ DF values were measured for 12 FP elements
- ❖ DF values were lower than those typically found in industrial scale PUREX plants due to multiple extraction and back-extraction steps without an intermittent scrubbing step.
- ❖ This work provide DF data that will be built upon for nuclear forensic investigations of interdicted Pu.

Future Work

- ❖ Modify Multi-contact extraction, to recover a larger fraction of Pu
- ❖ Investigation of how D-values for (Cs, Sb, Eu, Rb, Sr, Nd, Pm, and Sm) change as a function of nitric acid concentration
- ❖ Determine statistical uncertainty of D and DF values.
 - Repeat above experiments 3-5 times
- ❖ Connect D-values with process information to DF values

Questions?

Previous Experiment Results



Decontamination Factors for multi-contact extraction.

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Mass Spec

