Characterizaiton of Pu Separation by PUREX

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TEXAS A&M ENGINEERING EXPERIMENT STATION
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Outline

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Big Picture

- Pu isotopes produced in irradiated fuel can vary depending on
  - Burnup (irradiation history)
  - Reactor neutron spectrum (core design)
- Weapons-grade Pu can be extracted from reactor discharged fuel with a burnup of about 1 (GWD/tU)
- Two examples of non-safeguarded reactors which can intentionally discharge low burned fuel for extracting weapon-grade Pu are:
  - Indian Prototype Fast Breeder Reactor (PFBR-500MWe)
  - Indian Pressureized Heavy Water Reactor (PHWR-CANDU type 220 MWe)
- In accord with the Indo-US 123 agreement, these reactors were not required to be kept under IAEA safeguards

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Smaller Picture

- Attribution for unpurified Pu has been previously studied [2, 6, 3]
- Intercepted Pu would likely have been processed via PUREX
- Due to a lack of literature on decontamination factors and distribution coefficients for useful forensic elements (Cs, Sb, Eu, Rb, Sr, Nd, Pm, and Sm) this research was pursued.

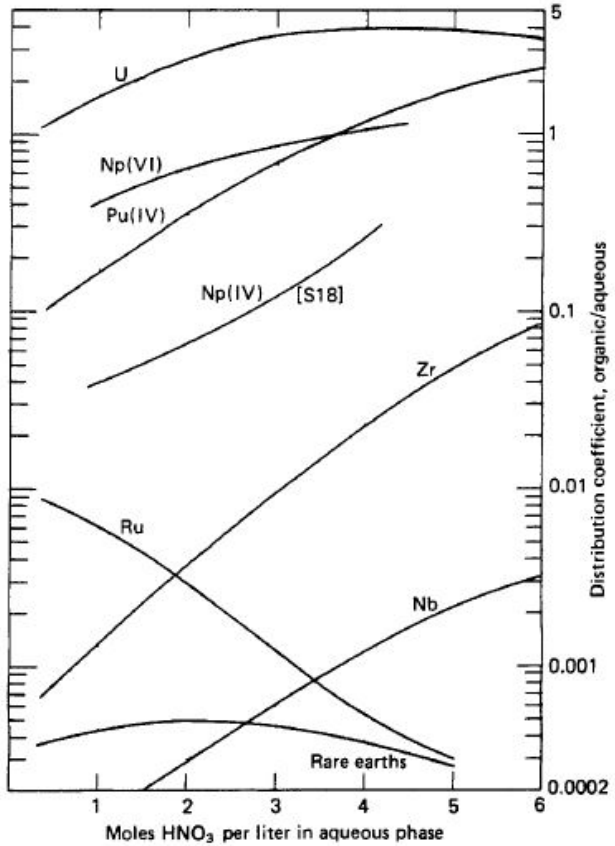

Figure 1: Effect of nitric acid concentration on distribution coefficients [7]

Background

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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

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Extraction

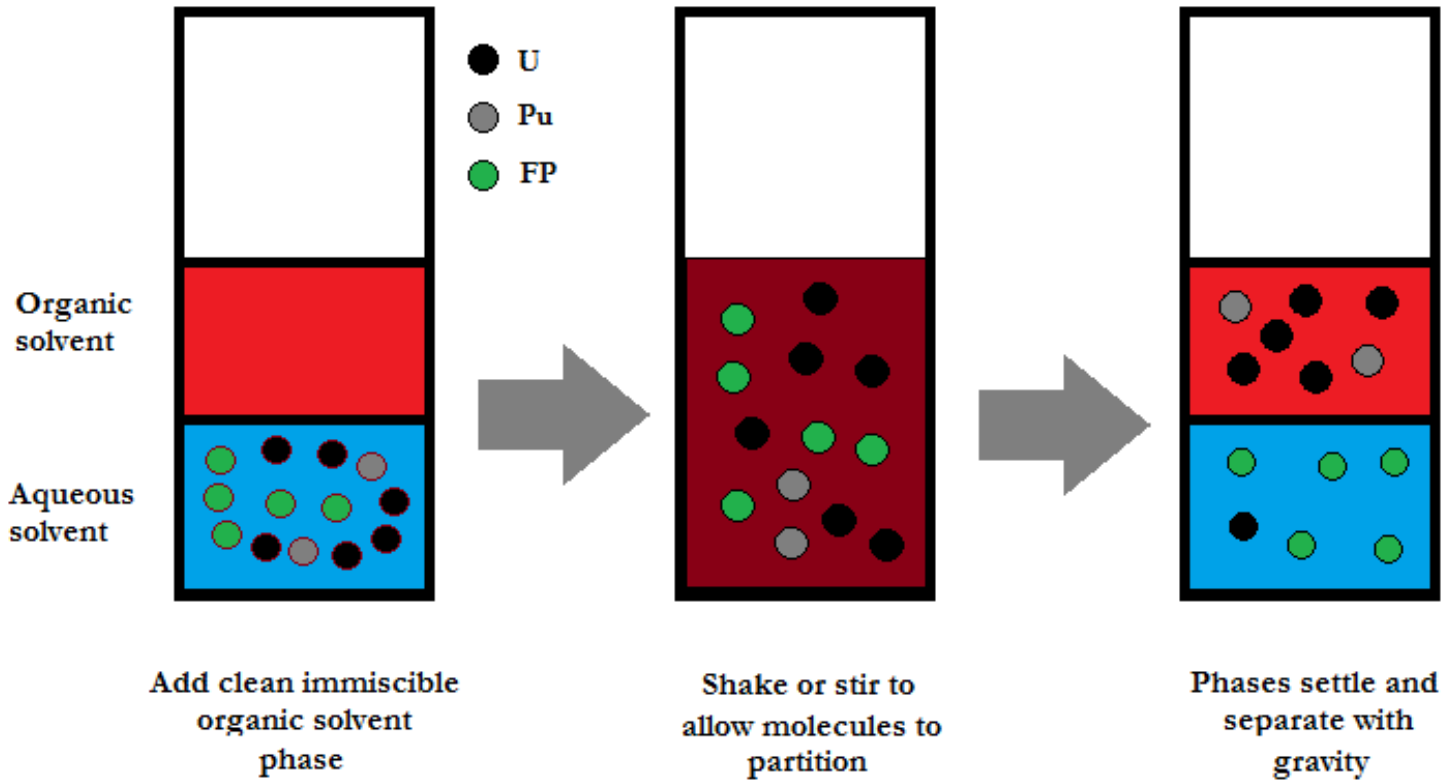

Figure 2: Extraction graphic

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Back-Extraction

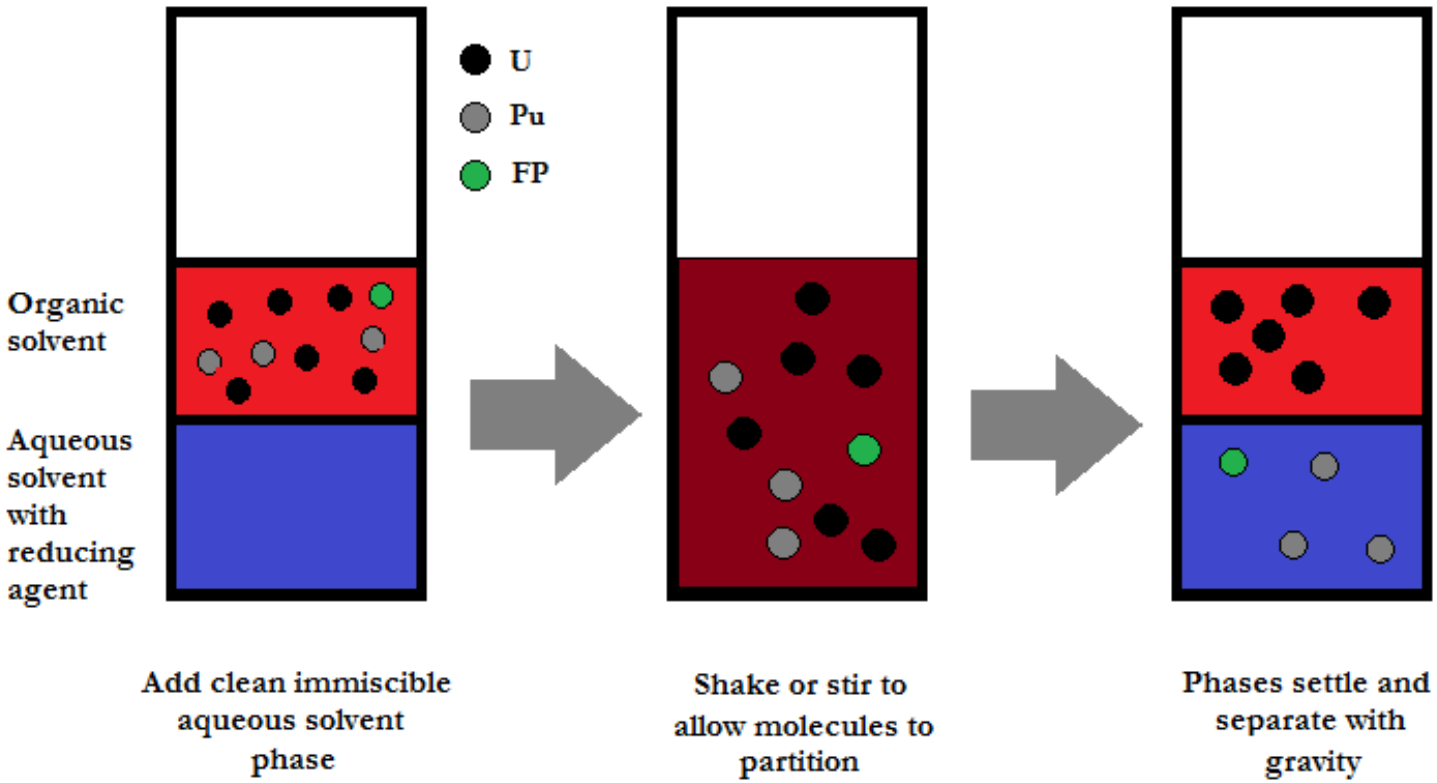

Figure 3: Back-extraction graphic

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Extraction and Back-extraction

- Extraction

$$UO_2^{2+}(aq) + 2NO_3^-(aq) + 2TBP(o) \leftrightarrow UO_2(NO_3)_2 \cdot 2TBP(o)$$ [1]

$$Pu^{4+}(aq) + 4NO_3^-(aq) + 2TBP(o) \leftrightarrow Pu(NO_3)_4 \cdot 2TBP(o)$$

Most of the fission products are left in the aqueous solution at valence III and V states[4]

- Back-extraction

$$Pu(NO_3)_4(TBP)_{2_o} + Fe_{aq}^{2+} \leftrightarrow Pu_{aq}^{3+} + 4NO_{3_{aq}}^- + 2TBP_o$$ [5]

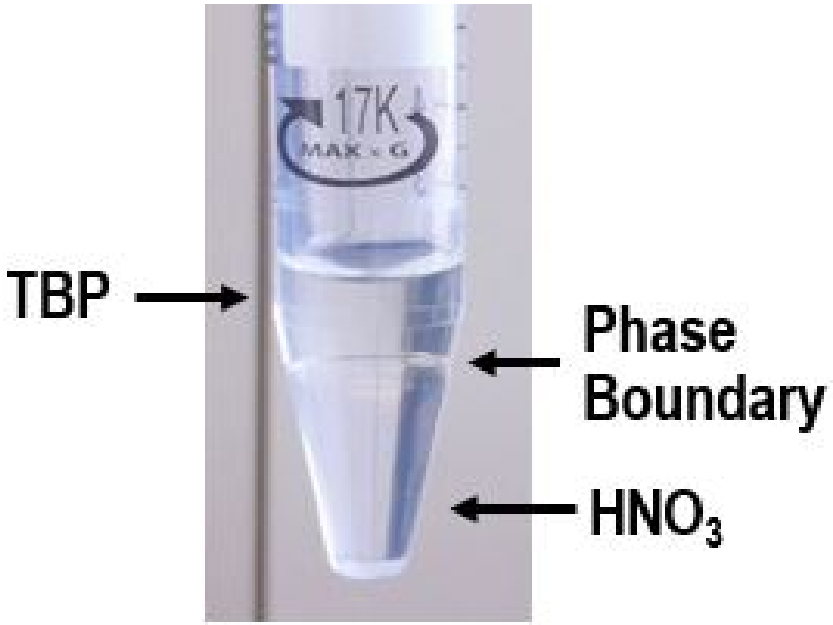

Figure 4: TBP and HNO_3

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Distribution Coefficients - The Missing link

- Distribution Coefficient (DC): The ratio between the organic and aqueous phases

$$DR = \frac{c_o}{c_{aq}}$$

- Specific element to element
- Vary widely with:[7]
  - Concentrations of solutions
  - Saturation of U and Pu in the system
  - Temperature of the solvents
- The fraction of mass, f_o deposited in the organic phase, assuming a volume ratio between the aqueous and organic phases, V_R, is:

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

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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]

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Complexity of reprocessing schemes

Worry about dependancies of of DC Worry about non equilibrium flow rates, blah blah blah. Show mixer settler columns, show batch. What I want to do, get some distribution coefficients develop a reasonable process for isolating a large fraction of Pu, then, based on process, calculate what the decontamination factor should be, and then actually measure it

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Irradiation

- $12.9 \pm$

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Experiments

- Experiment 1
  - Purpose: quantify product recovery and DF values for single stage extraction and back extraction
  - Conditions: Latex Table
- Experiment 2
  - Purpose: recover a large fraction of Pu (4 extractions, 3 back extractions
  - Conditions Latex Table

Completed Work

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Previous Experiment Results

Show the first table here

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Previous Experiment Results

- Calculate Compton edge for each peak

$$E_c = E_{e-}|_{(\theta=\pi)} = E_\gamma \left(\frac{2E_\gamma}{m_e c^2 + 2E_\gamma} \right) \quad (1)$$

Compton edges for gamma-ray sources:

Element	E_γ (keV)	Compton Edge (keV)
^{133}Ba	356	207.25
^{137}Cs	662	477.65
^{54}Mn	835	639.36
^{22}Na (P.P.)	511	340.67
^{60}Co	1173	963.42
^{22}Na	1274	1061.18
^{60}Co	1332	1118.10

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Previous Experiment Results

Show pretty plot

Future Work

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Future Work

- Continue improving stilbene modeling parameters and capabilities in DRiFT
  - PSD using waveforms
  - Low-energy photon detection improvements

  2
- Begin modeling multiple stilbene detectors in different angle configurations
- Model full NEUANCE detector array
- Study detector cross-talk
- Compare MCNP6/DRiFT simulations using CGMF/FREYA against experimental measurements
  - ^{252}Cf: spontaneous fission
  - ^{239}Pu and ^{235}U: neutron-induced fission

Questions?

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References I

[1] M Benedict, H Levi, and T Pigford. Nuclear chemical engineering. *Nucl. Sci. Eng.;(United States)*, 82(4), 1982.

[2] Sunil S Chirayath, Jeremy M Osborn, and Taylor M Coles. Trace fission product ratios for nuclear forensics attribution of weapons-grade plutonium from fast and thermal reactors. *Science & Global Security*, 23(1):48–67, 2015.

[3] Alexander Glaser. Isotopic signatures of weapon-grade plutonium from dedicated natural uranium-fueled production reactors and their relevance for nuclear forensic analysis. *Nuclear Science and Engineering*, 163(1):26–33, 2009.

[4] Kenneth D Kok. *Nuclear engineering handbook*, volume 60. CRC Press, 2009.

[5] RJM Konings, LR Morss, J Fuger, LR Morss, NM Edelstein, and J Fuger. The chemistry of the actinide and transactinide elements. *Springer, Dordrecht*, 4:2113–224, 2006.

[6] Mark Robert Scott. *Nuclear forensics: attributing the source of spent fuel used in an RDD event*. PhD thesis, Texas A&M University, 2005.

[7] Sidney M Stoller, Walter Henry Zinn, Stuart MacLain, and Atomic Energy Commission USA. *Reactor handbook. 2. Fuel reprocessing*. Interscience Publ., 1961.