NUCLEAR ENGINEERING
TEXAS A&M UNIVERSITY

Your Presentation

Author: Me

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Date of Presentation
Presented at All Hands Meeting

TEXAS A&M ENGINEERING EXPERIMENT STATION

NUCLEAR SECURITY SCIENCE & POLICY INSTITUTE

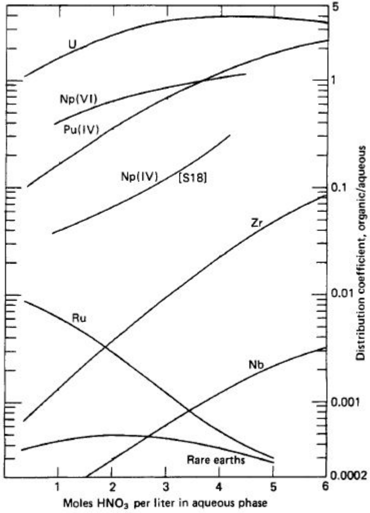
Outline

Big Picture

- Weapons-grade Pu can be extracted from reactor discharged fuel with a burnup of about 1 (GWD/tU)
- Pu isotopes produced in irradiated fuel can vary
- Two examples of reactors which can intentionally discharge low burned fuel for extracting weapon-grade Pu are:
  - Fast Breeder Reactor
  - CANDU Reactor

Smaller Picture

- Attribution for unpurified Pu has been previously studied [2, 6, 3]
- Interdicted Pu would likely have been processed
- Lack of literature on decontamination factors and distribution coefficients for useful forensic elements (Cs, Sb, Eu, Rb, Sr, Nd, Pm, and Sm)



Adapted from Stoller[7]

Background

What is PUREX - A type of laundry detergent?

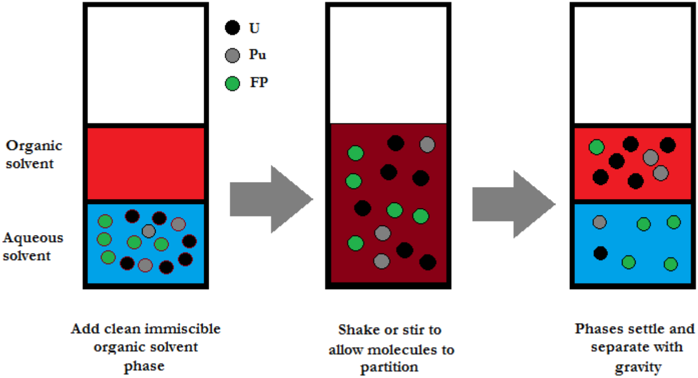
- Plutonium Uranium Redox EXtraction
  - Liquid-liquid solvent extraction
  - Many stages:
    1. Preparation for Dissolution
    2. Dissolution
    3. Preparation of Dissolved Feed
    4. Primary Decontamination - Extraction to organic*
    5. Scrubbing
    6. Plutonium Partition - Back-Extraction to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

$\ast$ - Discussing Next

Extraction

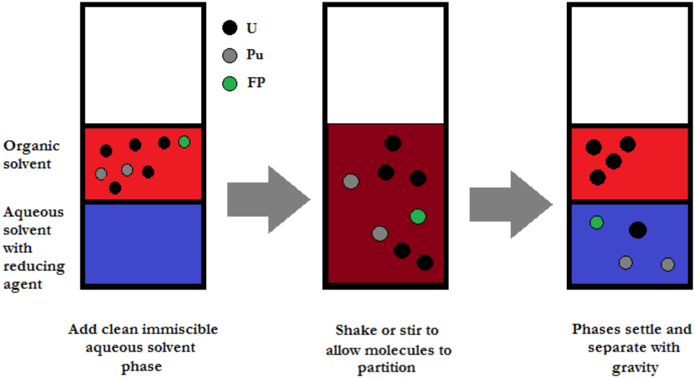
$$UO^{2+}_{2(aq)} + 2NO^{-}_{3(aq)} + 2TBP_{(o)} \leftrightarrow UO_2(NO_3)_2 \cdot 2TBP_{(o)}$$ [1]

$$Pu^{4+}_{(aq)} + 4NO^{-}_{3(aq)} + 2TBP_{(o)} \leftrightarrow Pu(NO_3)_4 \cdot 2TBP_{(o)}$$



Back-Extraction

$$Pu(NO_3)_4(TBP)_{2(o)} + Fe^{2+}_{(aq)} \leftrightarrow Pu^{3+}_{(aq)} + 4NO^{-}_{3(aq)} + 2TBP_{(o)}$$ [5]



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):

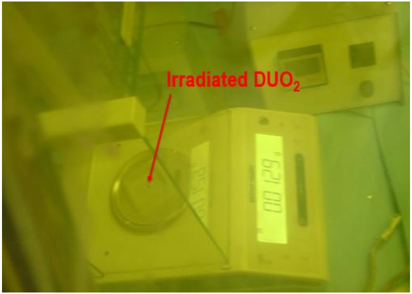
$$DF_j = \frac{\left|\frac{c_j}{c_{Pu}}\right|_{initial}}{\left|\frac{c_j}{c_{Pu}}\right|_{final}}$$

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

Previous Work

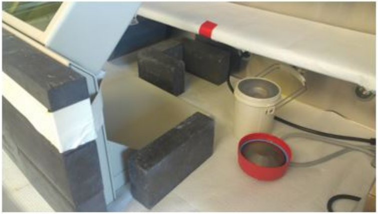
Irradiation

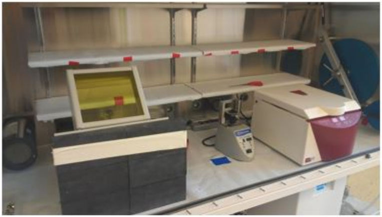
- 12.9 ± 0.1 mg of DUO_2 was irradiated
  - High Flux Isotope Reactor at Oak Ridge National Laboratory
- Burnup was 4.43 ± 0.31 GWd/tHM[8]
- $0.196 \pm$ mg of total Pu was produced as measured by ICP-MS



Dissolution of the spent fuel pellet

Glovebox





Experiments

- 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	$(83.4\pm9.5)\%$	$(11.2\pm1.3)\%$
Multi-contact Cycle 1	$(99.7\pm4.2)\%$	$(6.8\pm0.3)\%$
Multi-contact Cycle 2	$(93.0\pm4.6)\%$	$(6.6\pm0.3)\%$
Overall Experiment 2	$(92.7\pm6.0)\%$	$(0.45\pm0.03)\%$

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.

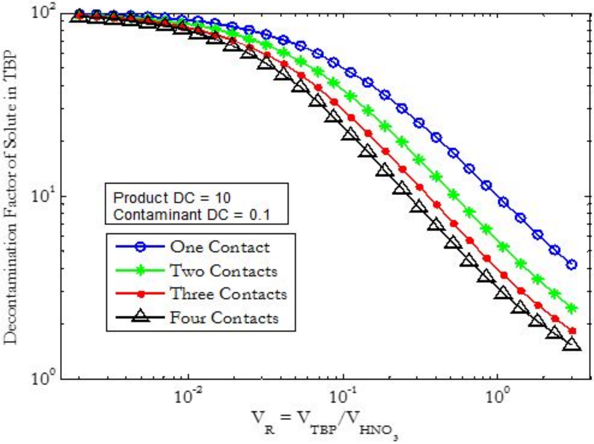
Future Work

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.

References I

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[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.

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

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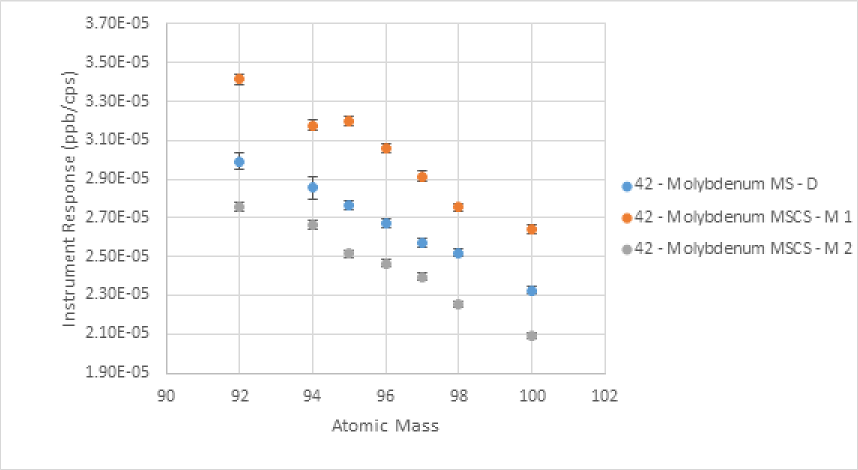
[8] Mathew Wayne Swinney. *Experimental and Computational Assessment of Trace Nuclide Ratios in Weapons Grade Plutonium for Nuclear Forensics Analysis*. PhD thesis, 2015.

Mass Spec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3.50E-05
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3.10E-05
2.90E-05
2.70E-05
2.50E-05
2.30E-05
2.10E-05
1.90E-05
Instrument Response (ppb/cps)
90
92
94
96
98
100
102
Atomic Mass
42 - Molybdenum MS - D
42 - Molybdenum MSCS - M 1
42 - Molybdenum MSCS - M 2