NUCLEAR ENGINEERING
TEXAS A&M UNIVERSITY

Experimental Characterization of Pu Separation by PUREX Process on a Low-Burnup, Pseudo-Fast-Neutron Irradiated DUO_2 for Product Decontamination Factors and Nuclear Forensics

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Dr. Craig Marianno
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Monday, February 27, 2017, 10:00 am
AIEN 304

TEES | TEXAS A&M ENGINEERING EXPERIMENT STATION

NUCLEAR SECURITY SCIENCE & POLICY INSTITUTE

Outline

Motivation

- Current Events
  - Joint Comprehensive Plan of Action
  - Non-safeguarded reactors (Example: India)
  - Islamic State of Iraq and Syria
- Past Events
  - September 11, 2001
- Limited scope of IAEA safeguards
- "the awful arithmetic of the atomic bomb" [1]
- Need for improved forensic capabilities [2, 3, 4]

Definitions

- Special Nuclear Material (SNM)
  - Plutonium, ^{233}U, or ^{235}U
- What is Nuclear Forensics
  - Developing forensics evidence to identify the source of material and potentially the identity of the actor in a nuclear event (either using an RDD or a nuclear explosive)
- SNM origin attributes/indicators
  - Indicators or clues for SNM origin attribution. Examples include burnup, fluence rate, initial fuel enrichment, decay time, and fast-to-thermal irradiation ratios
- PUREX Decontamination Factors (DF)
  - A measure of the effectiveness with which separated Pu from irradiated fuel is decontaminated

$$DF_j = \frac{\frac{c_j}{c_{Pu}}\Big|_{\text{initial}}}{\frac{c_j}{c_{Pu}}\Big|_{\text{final}}}$$

National Context

"The United States has developed a nuclear forensics capability that has been demonstrated in real-world incidents of **interdicted materials** and in exercises of actions required after a nuclear detonation. The committee, however, has concerns about the program and finds that without strong leadership, careful planning, and additional funds, these capabilities will decline" [2]

Major areas of concern include:

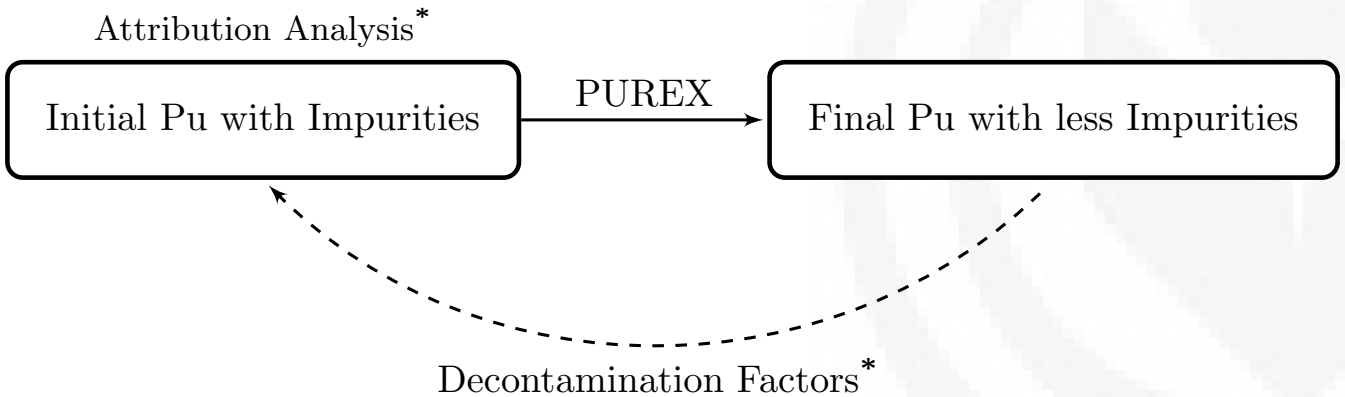
- Organization
- Sustainability
- **Workforce and Infrastructure**
- **Procedures and Tools (predetonation)**

Forensic Context

- Nature of inverse problems
- Plutonium purification necessary for weapons production

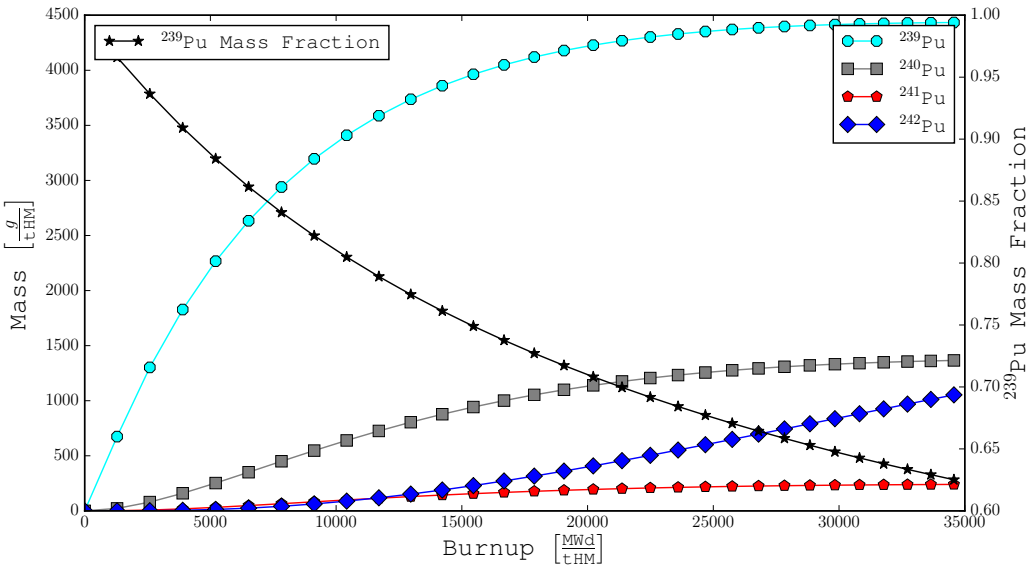
$$^{238}U + n \rightarrow ^{239}U \xrightarrow[T_{1/2}=23\text{ min}]{\beta^-} ^{239}Np \xrightarrow[T_{1/2}=2\text{ d}]{\beta^-} ^{239}Pu$$

- Attribution for used or spent fuel has been previously studied [5, 6, 7]



Nuclear Context

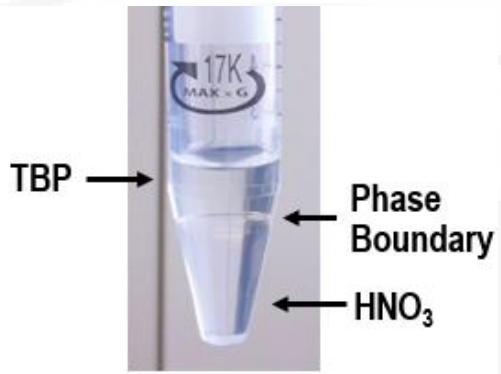
- Weapons-grade Pu can be extracted from reactor discharged fuel with a burnup of about 1 (GWD/tU)
- Pu isotopes produced in irradiated fuel can vary
- Two examples of reactors which can intentionally discharge low burned fuel for extracting weapon-grade Pu are:
  - Fast Breeder Reactor, CANDU Reactor



Chemical Context

- Plutonium Uranium Redox EXtraction (PUREX)
  - Liquid-liquid solvent extraction
  - Many contacts:
- Distribution Coefficient (D): The ratio between the organic and aqueous phases (aka: D-values)

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



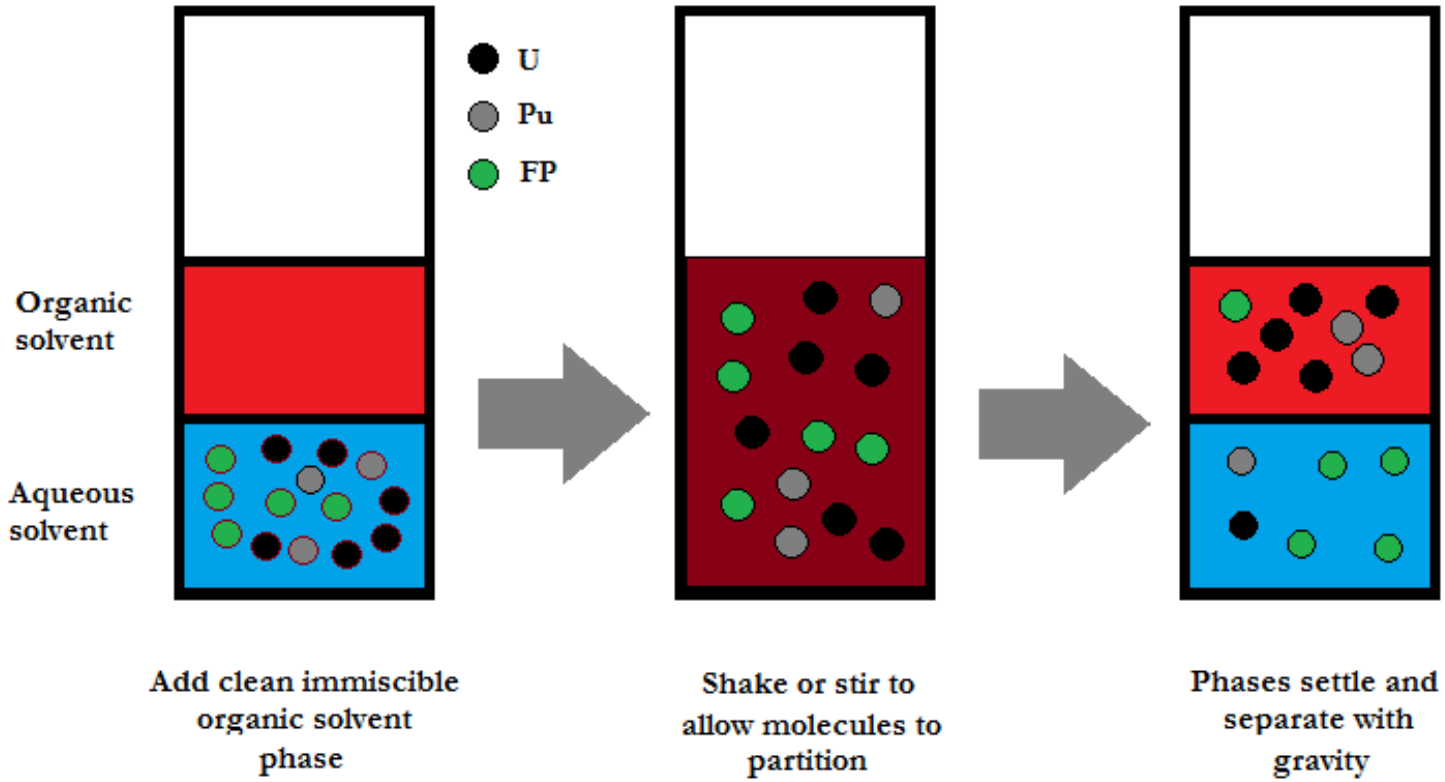
  - Specific element to element
  - Vary widely[8]
  - The fraction of mass, f_o deposited in the organic phase, assuming a volume ratio between the aqueous and organic phases, V_R, is:

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

Extraction

$$UO_{2(aq)}^{2+} + 2NO_{3(aq)}^{-} + 2TBP_{(o)} \leftrightarrow UO_2(NO_3)_2 \cdot 2TBP_{(o)}$$ [9]

$$Pu_{(aq)}^{4+} + 4NO_{3(aq)}^{-} + 2TBP_{(o)} \leftrightarrow Pu(NO_3)_4 \cdot 2TBP_{(o)}$$



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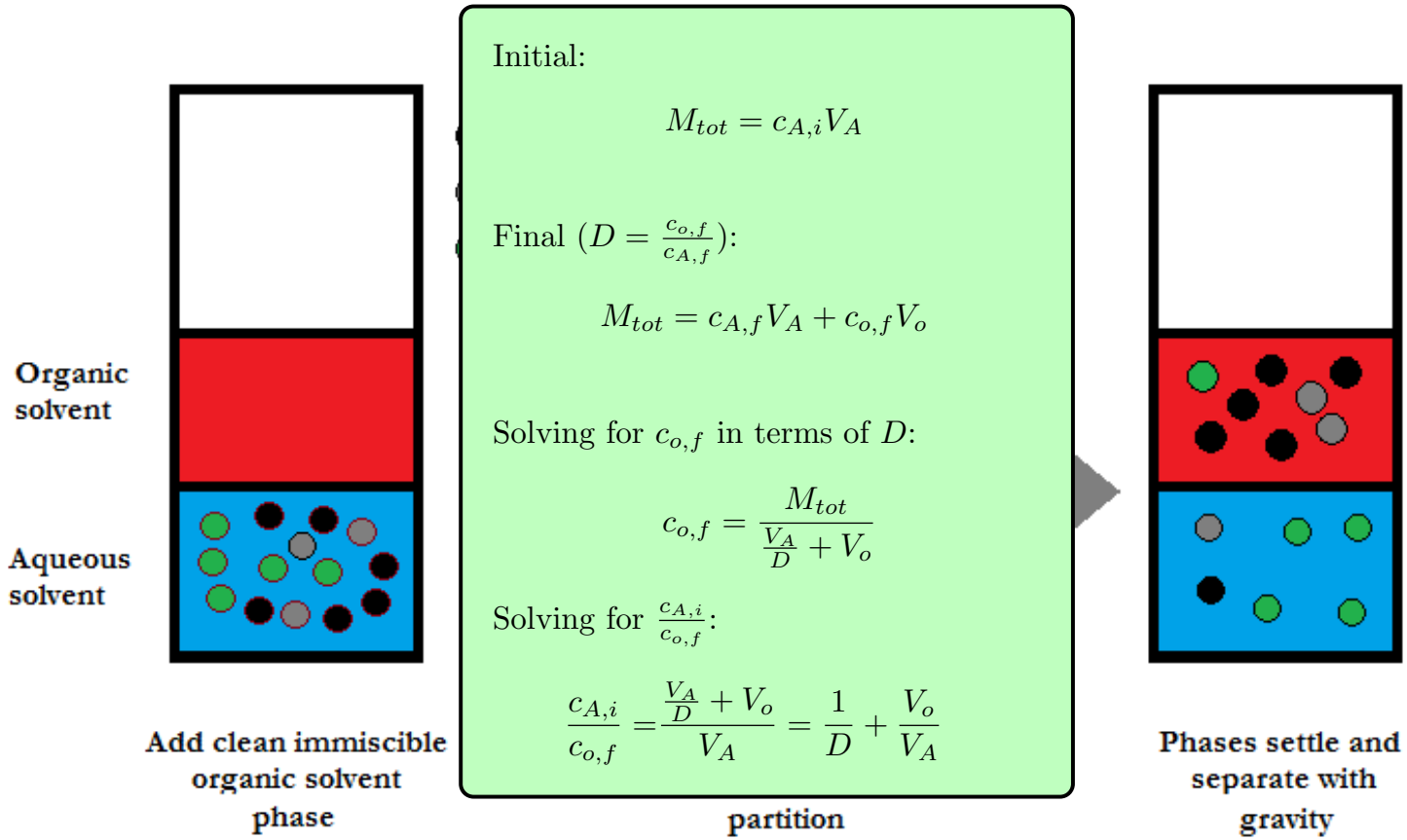

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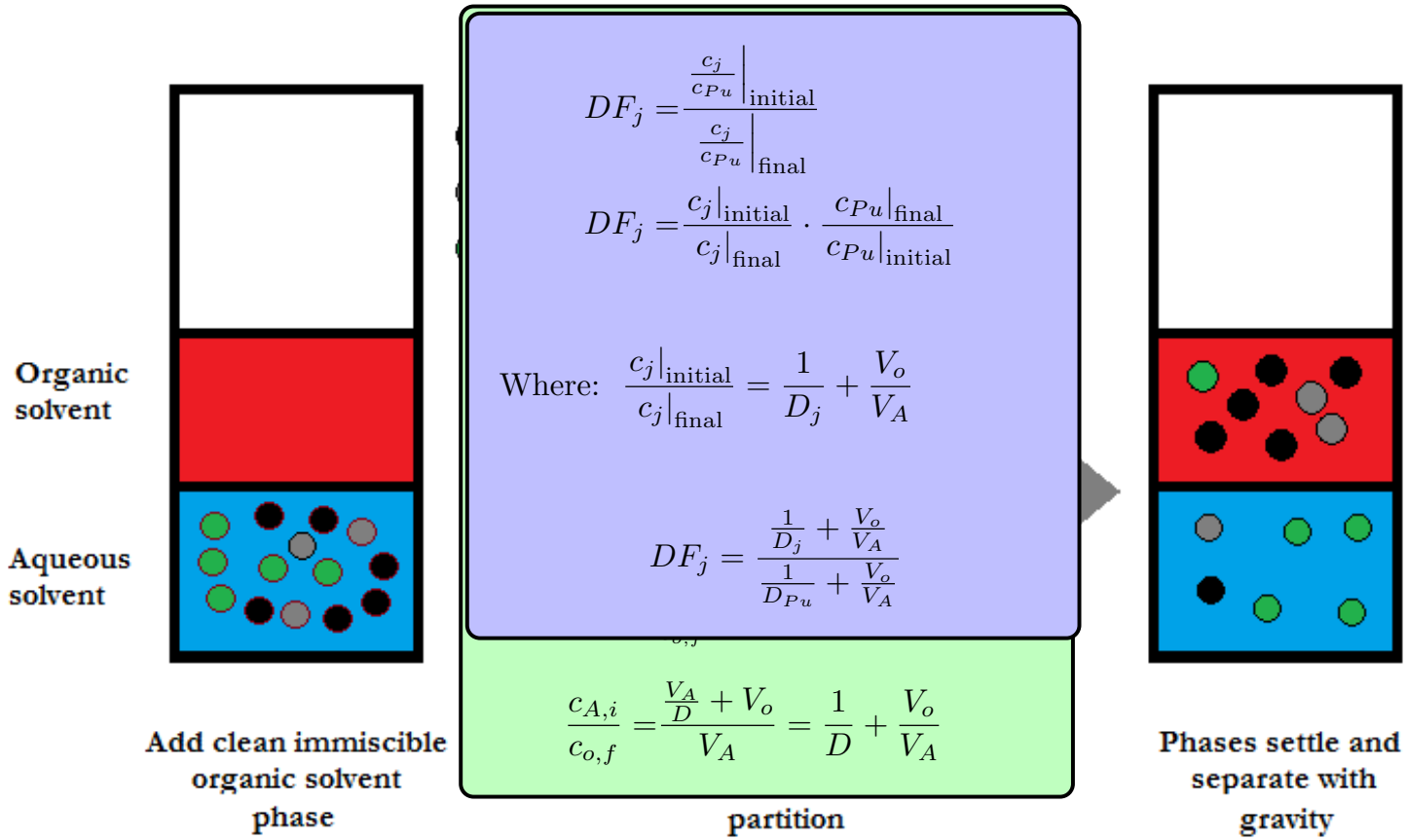
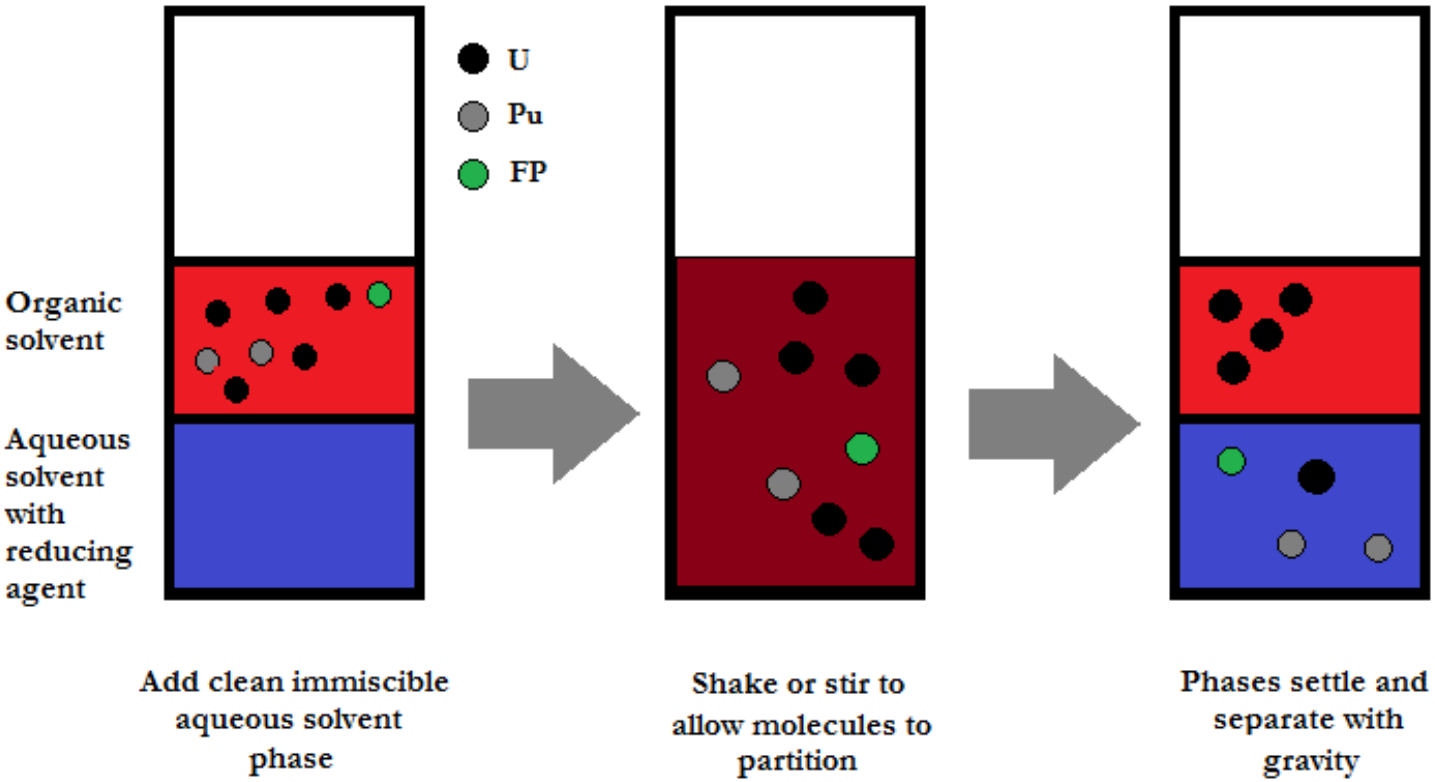


Back-Extraction

$$Pu(NO_3)_4(TBP)_{2(o)} + Fe^{2+}_{(aq)} \leftrightarrow Pu^{3+}_{(aq)} + 4NO^{-}_{3(aq)} + 2TBP_{(o)}$$ [10]



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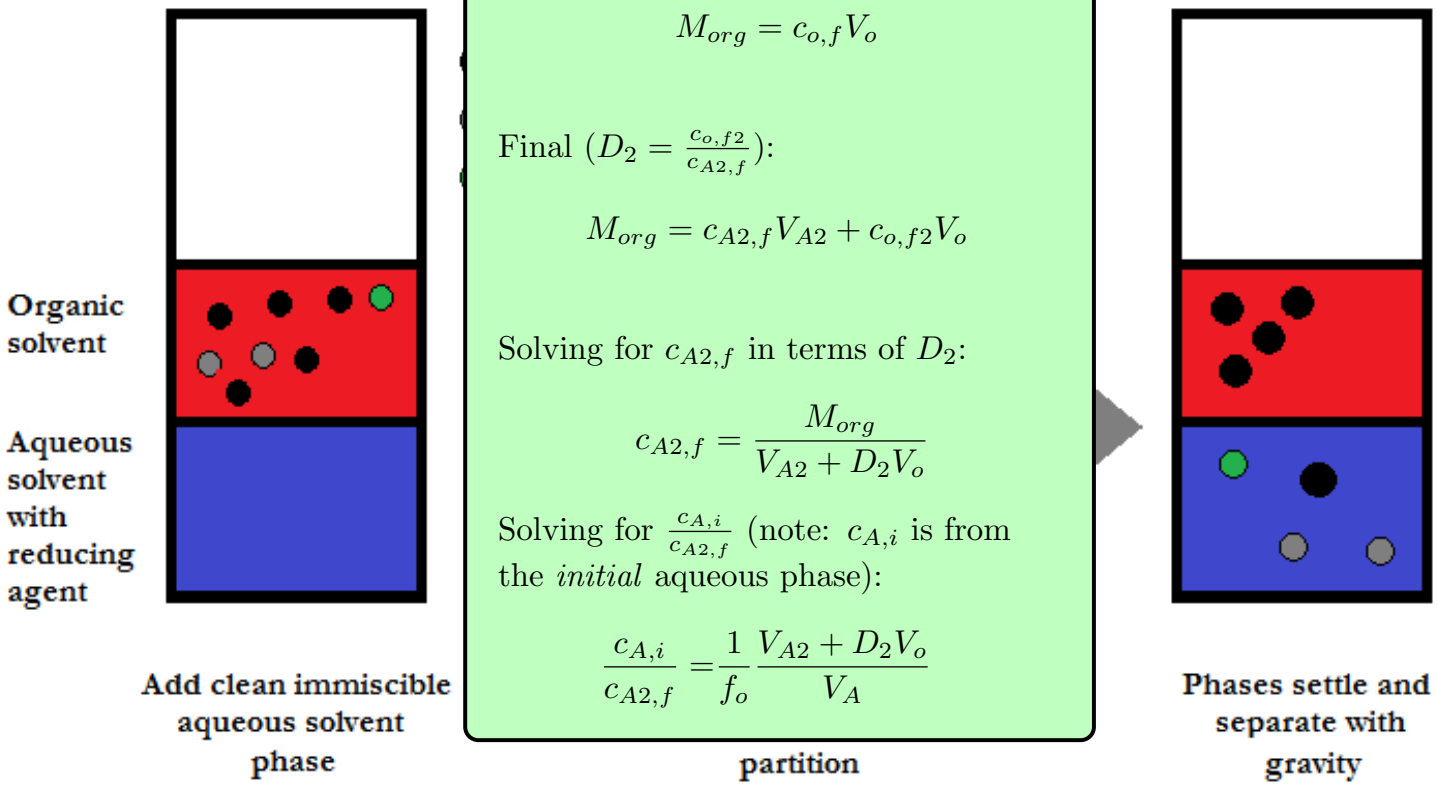

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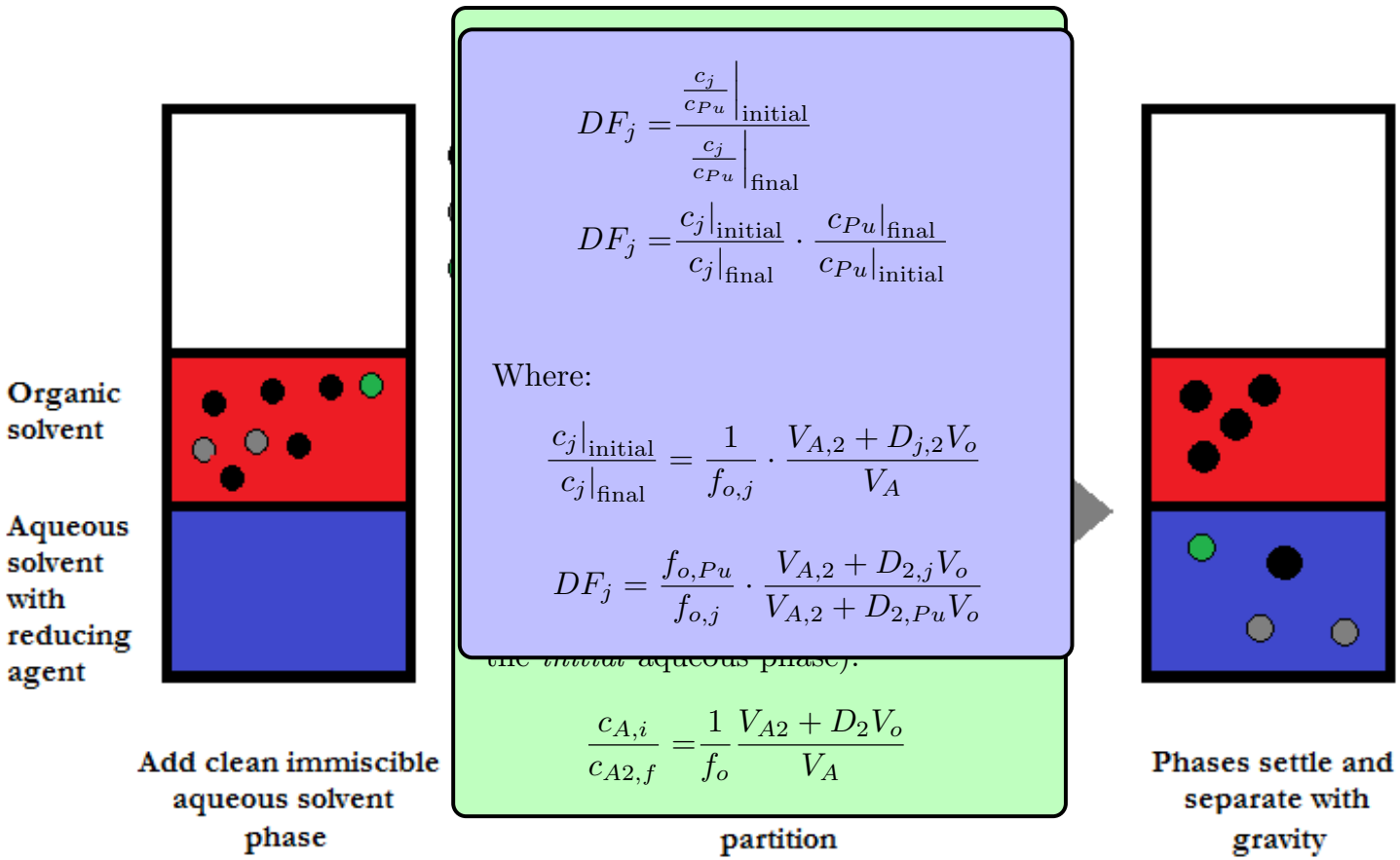


$$DF_j = \frac{\frac{c_j}{c_{Pu}}\Big|_{\text{initial}}}{\frac{c_j}{c_{Pu}}\Big|_{\text{final}}}$$

$$DF_j = \frac{c_j|_{\text{initial}}}{c_j|_{\text{final}}} \cdot \frac{c_{Pu}|_{\text{final}}}{c_{Pu}|_{\text{initial}}}$$

Where:

$$\frac{c_j|_{\text{initial}}}{c_j|_{\text{final}}} = \frac{1}{f_{o,j}} \cdot \frac{V_{A,2} + D_{j,2}V_o}{V_A}$$

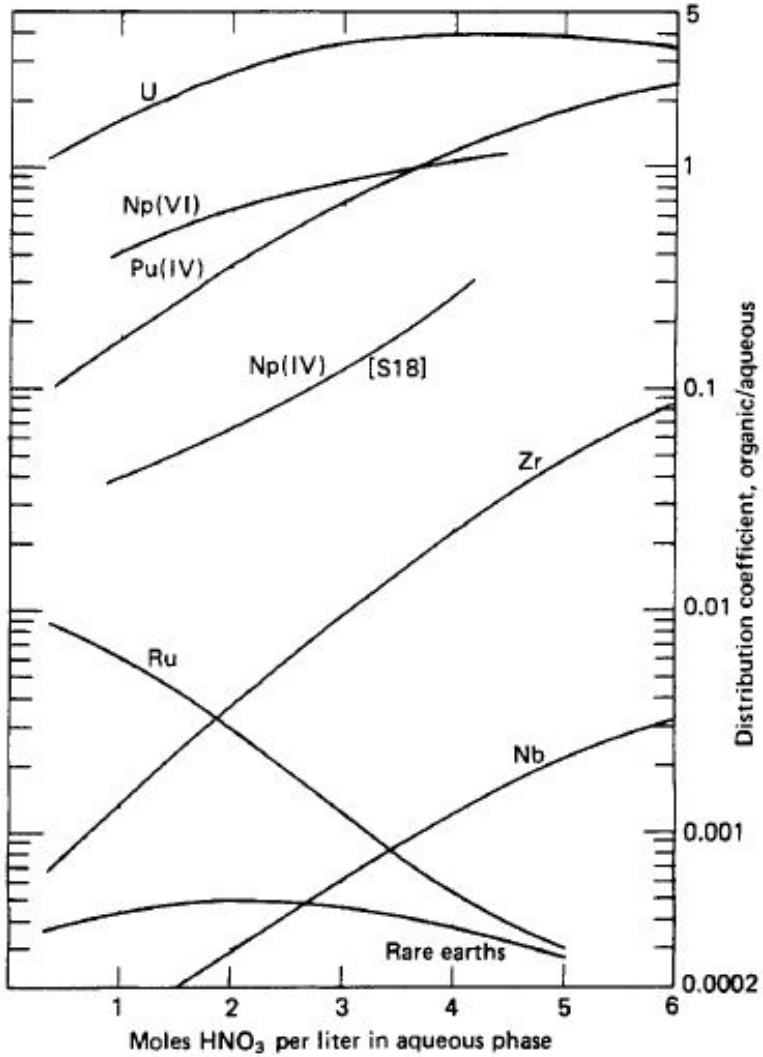
$$DF_j = \frac{f_{o,Pu}}{f_{o,j}} \cdot \frac{V_{A,2} + D_{2,j}V_o}{V_{A,2} + D_{2,Pu}V_o}$$

the *initial* aqueous phase):

$$\frac{c_{A,i}}{c_{A2,f}} = \frac{1}{f_o}\frac{V_{A2} + D_2V_o}{V_A}$$

Chemical Context

- Lack of literature on decontamination factors and distribution coefficients for useful forensic elements (Cs, Sb, Eu, Rb, Sr, Nd, Pm, and Sm)
- With a known process and D-values, DF values for individual elements can be determined



Adapted from Stoller[8]

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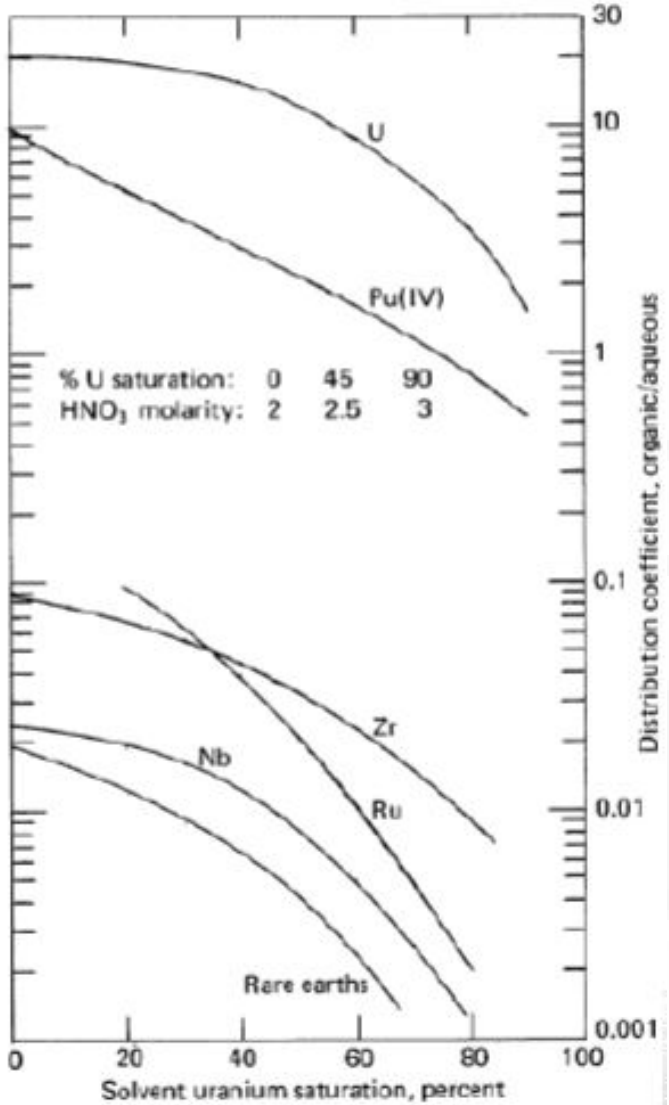
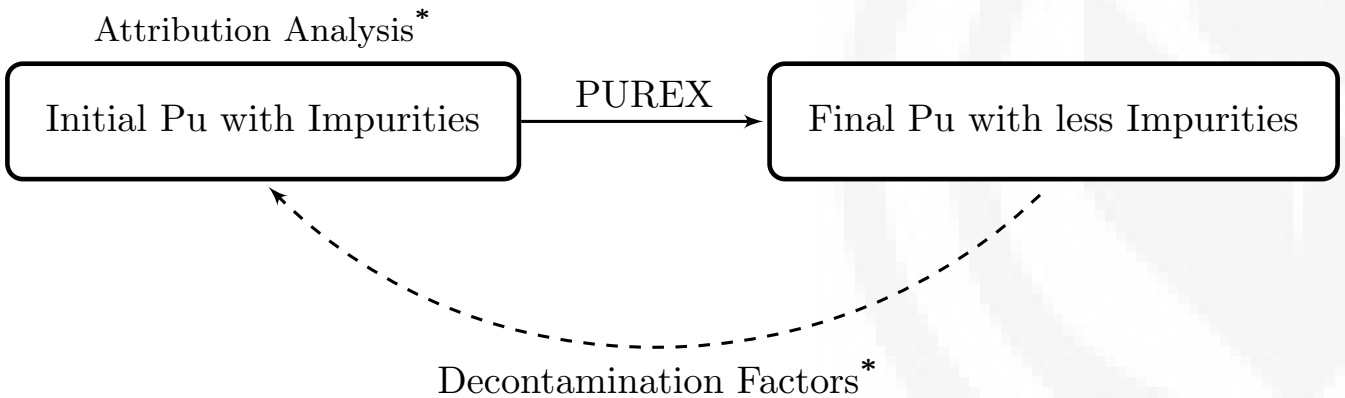


Adapted from Stoller[8]

Decontamination Factors and their use

- After several cycles of Pu extraction/scrubbing/back-extraction are completed, the effectiveness of a PUREX cycle is described by the decontamination factor (DF):
- DFs are characteristic of different process cycles
- Larger values (10^7) for industrial scale PUREX (compared to benchtop)[8, 9]



Objectives I

Characterize a 4 extraction 3 back-extraction PUREX process

- Collect D-values for each step
  - ✓ ^{144}Ce,$^{155}Eu^*$,$^{154}Eu^*$,^{125}Sb, ^{106}Ru, ^{134}Cs, ^{137}Cs (Measured Gamma in triplicate)[5]
  - ✓ Convert Gamma Spectrum to D-values
  - ✓ $^{85}Rb^*$,$^{90}Sr^*$,$^{97,98,100}Mo$,$^{101,102,104}Ru$, ^{110}Pd,^{112}Cd,$^{133}Cs^*$,$^{140,142}Ce$,$^{143}Nd^*$, $^{147}Pm^*$,$^{151}Sm^*$,$^{154}Eu^*$,U^*,Pu^* (Mass Spec)
    - Convert all mass spec data to D-values
- Collect DF-values for the process (and the two steps)
  - ✓ Prepare alpha samples for each step (triplicate)
    - Analyze alpha samples for each step
    - Convert alpha spec + gamma spec data to DF values
  - ✓ Convert Mass spec data to DF values (published)[11]
- Mathematically connect D-values to DFs
  - Derive equations with uncertainty propagation
  - Analyze connection with uncertainty

Objectives II

Determine attribution indicators:

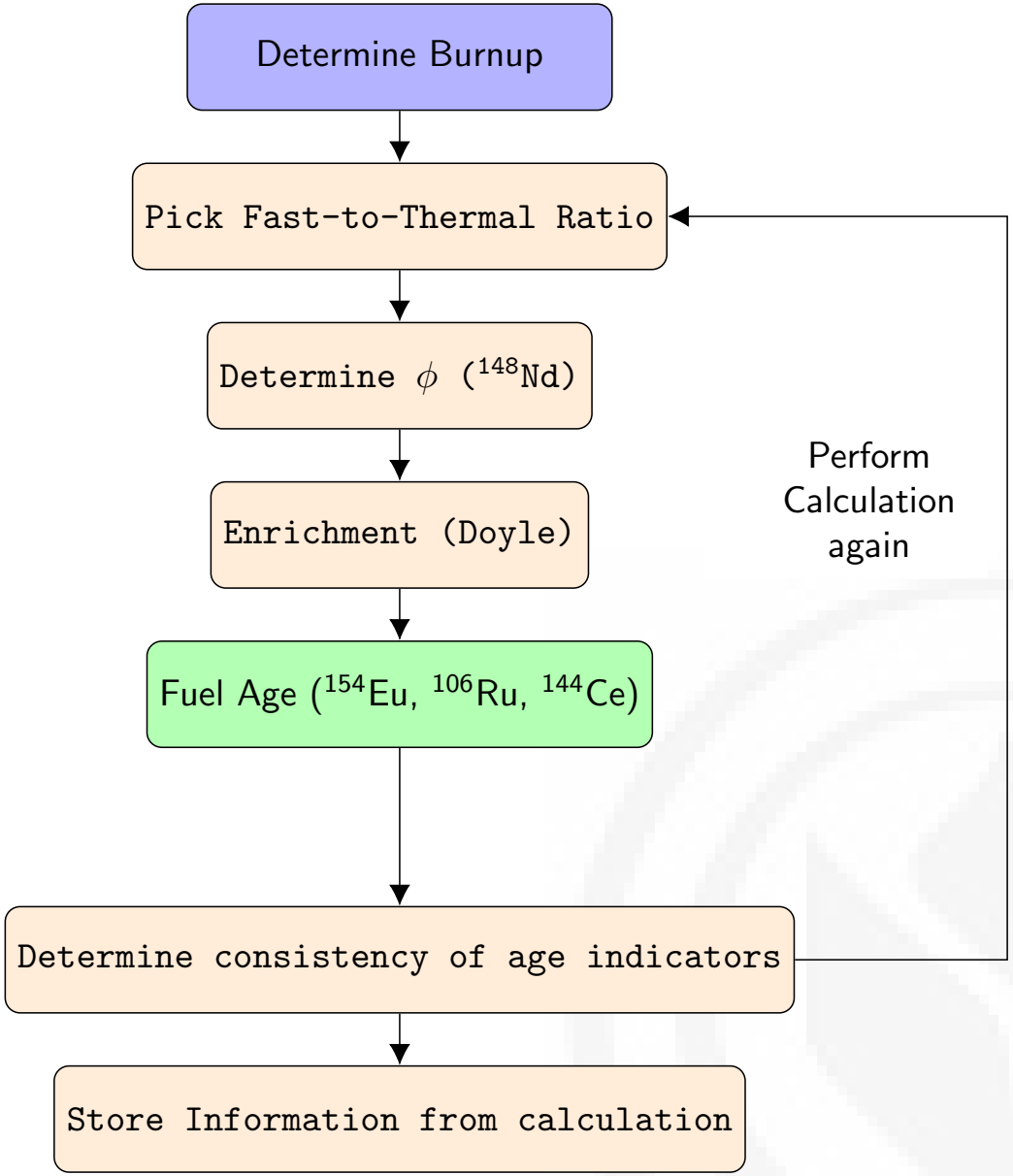
- Mathematically derive equations for above indicators with respect to one of the isotopes determined above
  - ✓ Burnup
  - ✓ Fluence Rate
  - ✓ Initial Enrichment
  - ✓ Decay Time
  - Fast-to-thermal ratios (requires iteration)
- Program a system to iteratively solve for these parameters given heavy metal concentration ratios
  - Make a program that can read ENDF files for x-sections
  - ✓ Create/Use a Bateman solver with automated x-section modifications
  - ✓ Create program to calculate single group x-sections from ENDF data and an assumed fast-to-thermal ratio
  - Couple all programs together in a single program
- Use above information to determine indicators for three sets of data

Present Status of the Question

- Stable noble fission gases as burnup verification[12]
- Determine burnup, enrichment, and decay time from used fuel in a RDD[13]
- Analysis of purified plutonium isotopics for reactor type[14]
- PUREX co-processing DF values for U and Pu[15]
- PUREX D-values and DF values under numerous circumstances[9, 16, 17, 18, 19, 8]
  - DF values for ^{106}Ru and ^{95}Zr[8]
  - Compilation of D-values for U, Th, and Pu[20]
  - D-values for rare earths, Pu, Th[18, 21, 22, 23, 24, 25, 26, 27, 28, 29]
  - Gd D-values[29]

Subsection 1

Mathematical methodology


Determine Burnup
Pick Fast-to-Thermal Ratio
Determine ϕ (^{148}Nd)
Enrichment (Doyle)
Fuel Age (^{154}Eu, ^{106}Ru, ^{144}Ce)
Determine consistency of age indicators
Store Information from calculation
Perform Calculation again

Analytical Procedure

- 12.9±0.1 mg of DUO_2 irradiated at HFIR

$$\frac{dn_i}{dt} = -\lambda_i^{eff} n_i + \sum_{j=1}^{N} b_{j\to i}^{eff} n_j$$

$$\lambda_i^{eff} = \lambda_i + \phi \sum_{j=1}^{N} \sigma_{i\to j}$$

$$b_{j\to i}^{eff} = b_{j\to i}\lambda_j + \sigma_{j\to i}\phi + \gamma_{j\to i}\sigma_{j,f}\phi$$

$$\frac{d\vec{n}}{dt} = \mathbf{A}\vec{n}(t) \to \vec{n} = e^{\mathbf{A}t}\vec{n}_0$$

Analytical Procedure - Burnup

- Operational Parameter
- Calculated:
  - Specific heat calculation on working fluid
  - Number and isotopes that fission in the system
  - **burnup isotope indicator**

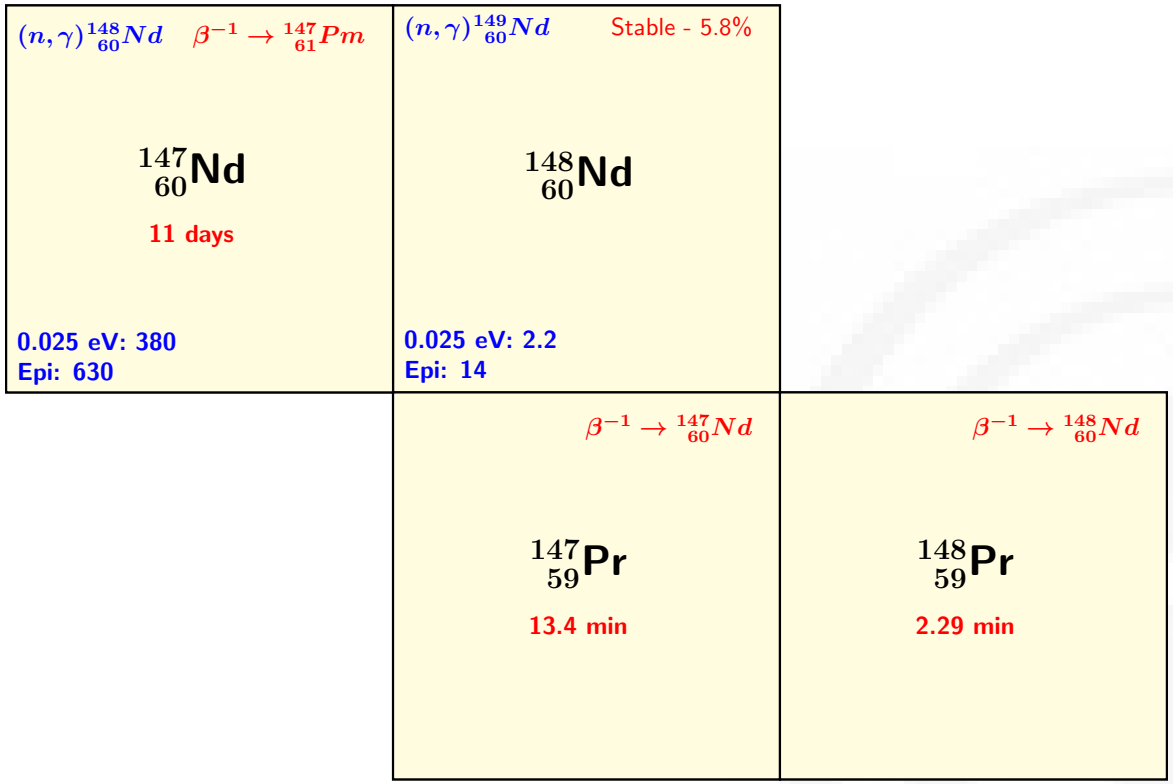
$$BU = \frac{\text{Power[MW]} \cdot \text{days}}{m[HM]}$$

$$= \left[\frac{N^B}{N_0^{HM}} \right] \frac{N_A E_R}{\gamma_B} \cdot \frac{1}{M_0^{HM}}$$

Analytical Procedure - Burnup

$(n,\gamma)^{137}_{55}Cs$ $\beta^{-1} \rightarrow {}^{136}_{56}Ba$ $^{136}_{55}$Cs 13 days 0.025 eV: 11 Epi: 57	$(n,\gamma)^{138}_{55}Cs$ $\beta^{-1} \rightarrow {}^{147}_{56}Ba$ $^{137}_{55}$Cs 0.025 eV: 0.2 Epi: 0.3	
	$(n,\gamma)^{137}_{54}Xe$ Stable 8.9% $^{136}_{54}$Xe 0.025 eV: 0.2 Epi: 0.1	$\beta^{-1} \rightarrow {}^{137}_{55}Cs$ $^{137}_{54}$Xe 3.8 min

Analytical Procedure - Burnup



Analytical Procedure - Fluence Rate

- Burnup Indicator assumptions
  - Short lived precursors of same mass bin with small cross sections
  - **Small contributions from other mass bins**
  - *Stable*

$$\phi \approx \frac{\lambda_7}{\sigma_7 \left(\frac{\gamma_7}{\gamma_8^* - \gamma_8} - 1 \right)}$$

- Assumptions:
  - Build in can be described by constant ϕ, single group x-sections
  - Constant fissionable material, σ_a for ^{148}Nd≈ 0
  - Cumulative yield instantly arrives at ^{147}Nd
  - Utilizes a Taylor series expansion

Analytical Procedure - Initial Enrichment and Decay Time

Initial Enrichment

- Heavy metal converts to heavier metal and to Fission products
- Account for in Burnup, and mass in heavy metals
- Solution assumes all higher actinides are built into the system from capture in ^{238}U

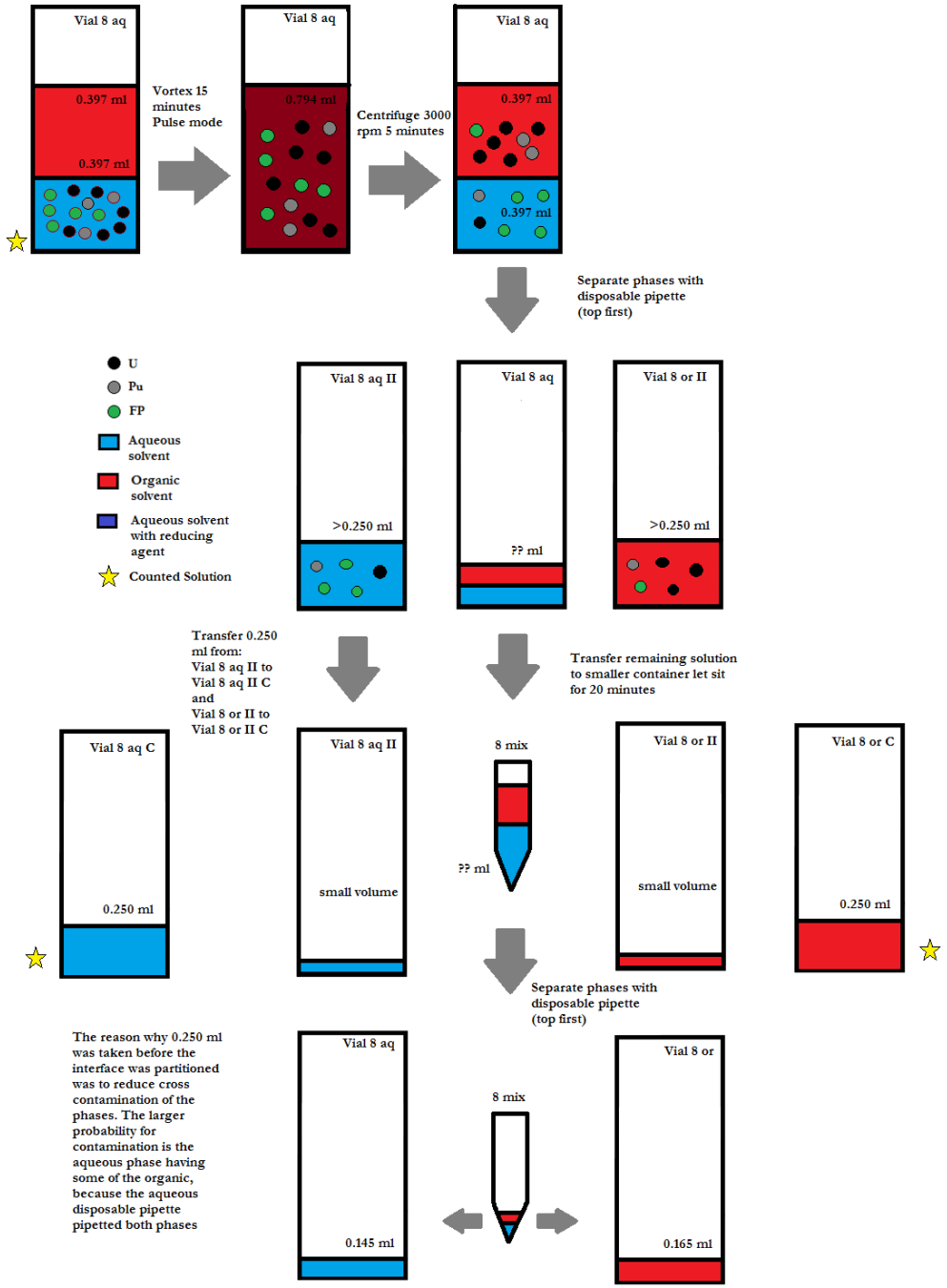
$$\epsilon_0 = \frac{N^{U238}(T)}{N_0^U}\left[\frac{N^{U235}(T)}{N^{U238}(T)} + \frac{N^{U236}(T)}{N^{U238}(T)}\right] + \frac{M_0^U}{N_A E_R} BU(T) - G^{238} - G^{239} - G^{240} - G^{241}$$

Decay Time

$$t_d = -\frac{1}{\lambda} ln\left(\frac{N_{\text{measured}}}{N_{EOI}}\right)$$

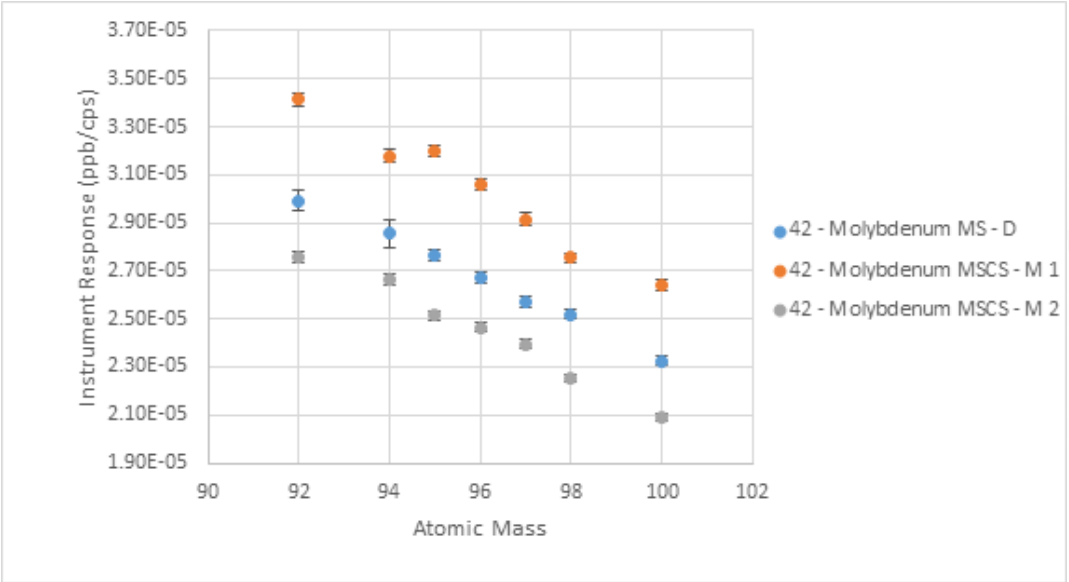
Subsection 2

Experimental Procedure


Vial 8 aq
0.397 ml
0.397 ml
Vortex 15 minutes Pulse mode
Vial 8 aq
0.794 ml
Centrifuge 3000 rpm 5 minutes
Vial 8 aq
0.397 ml
0.397 ml
Separate phases with disposable pipette (top first)
U
Pu
FP
Aqueous solvent
Organic solvent
Aqueous solvent with reducing agent
Counted Solution
Vial 8 aq II
>0.250 ml
Vial 8 aq
?? ml
Vial 8 or II
>0.250 ml
Transfer 0.250 ml from: Vial 8 aq II to Vial 8 aq II C and Vial 8 or II to Vial 8 or II C
Transfer remaining solution to smaller container let sit for 20 minutes
Vial 8 aq C
0.250 ml
Vial 8 aq II
small volume
8 mix
?? ml
Vial 8 or II
small volume
Vial 8 or C
0.250 ml
Separate phases with disposable pipette (top first)
The reason why 0.250 ml was taken before the interface was partitioned was to reduce cross contamination of the phases. The larger probability for contamination is the aqueous phase having some of the organic, because the aqueous disposable pipette pipetted both phases
Vial 8 aq
0.145 ml
8 mix
Vial 8 or
0.165 ml

Experimental Procedure - Mass Spectrometry

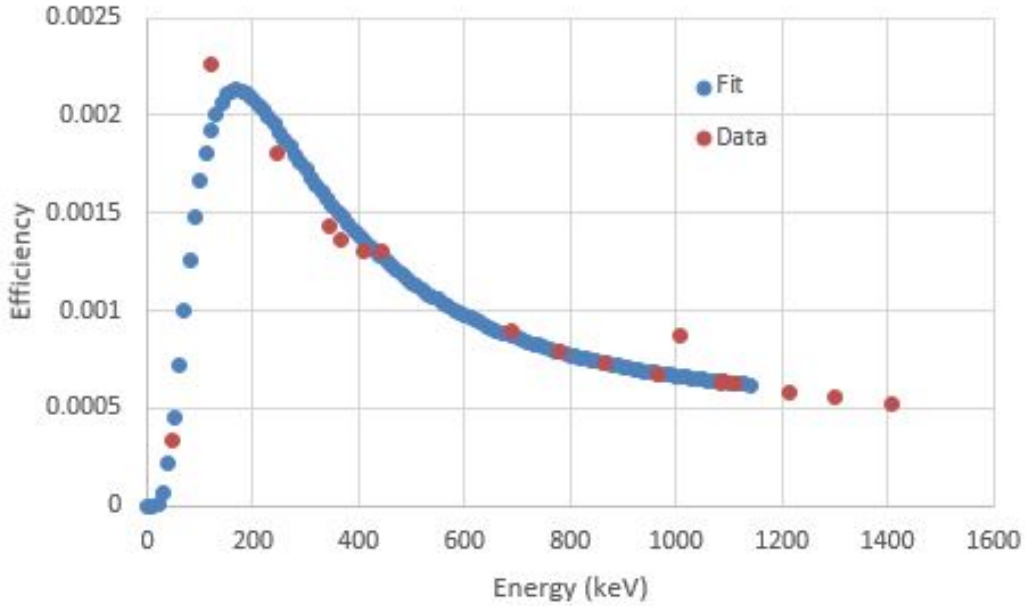
- NexION 300X quadrupole
- 7 different concoction standards (including Pu and U standards)
- Two point calibration curves
- Extrapolation for radioactive species



Experimental Procedure - Gamma Spectroscopy

- Canberra electrode coaxial HPGe
- ^{152}Eu liquid calibration source
- Program written to extract and analyze 26 gamma peaks

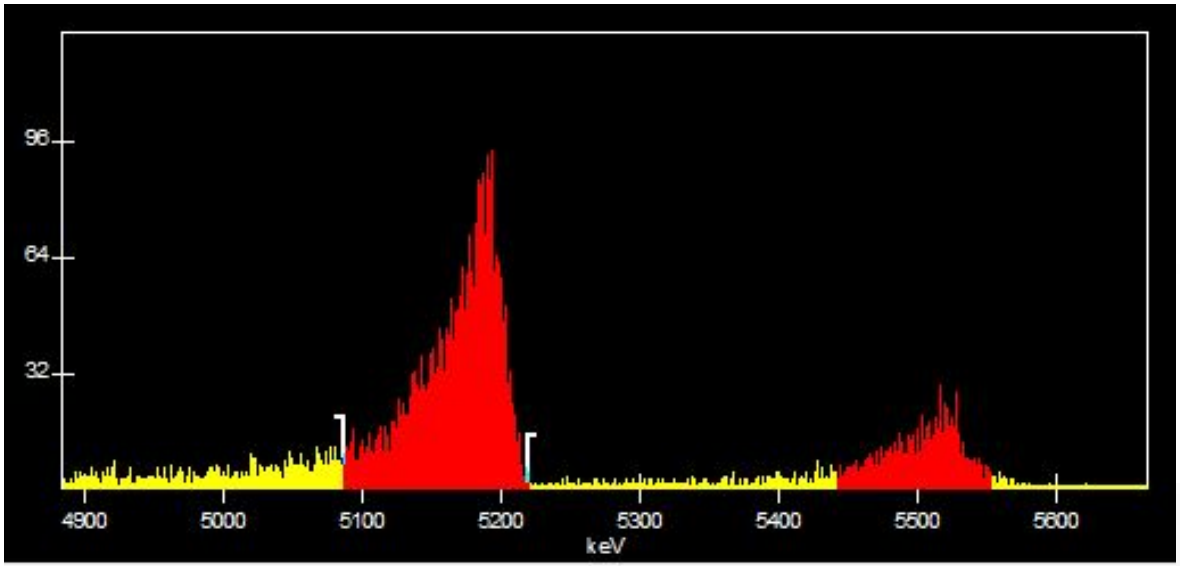
^{155}Eu,^{154}Eu,^{144}Ce,^{125}Sb,^{106}Rh,^{134}Cs,^{137}Cs



Experimental Procedure - Alpha Spectroscopy

- Passivated implanted planar silicon (PIPS) detector
- 4-peak Pu, Am, Gd, and Cm source from Eckert and Ziegler
- Samples prepared by evaporating 10-20 μl on aluminium surface

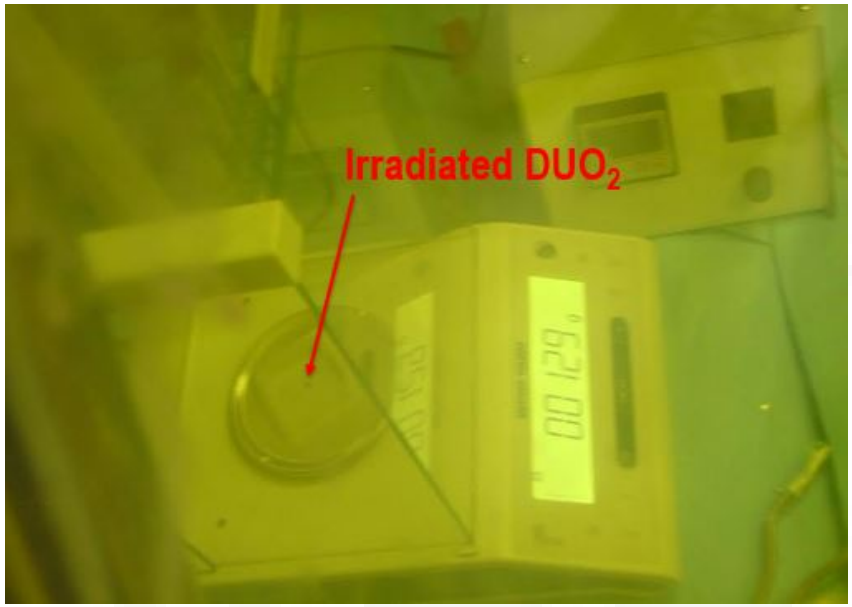
^{239}Pu,^{240}Pu,^{241}Am



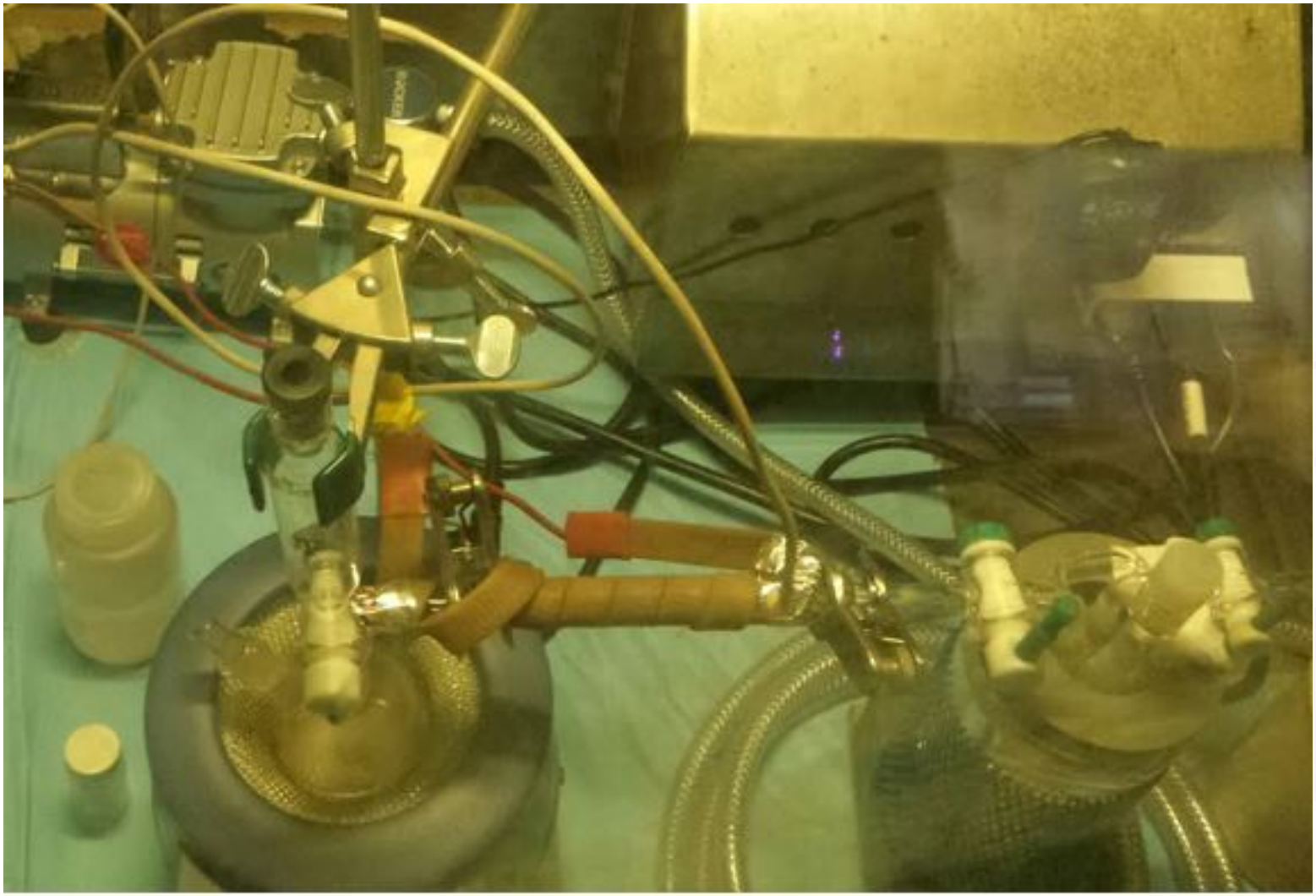
Current and Expected Results

Irradiation

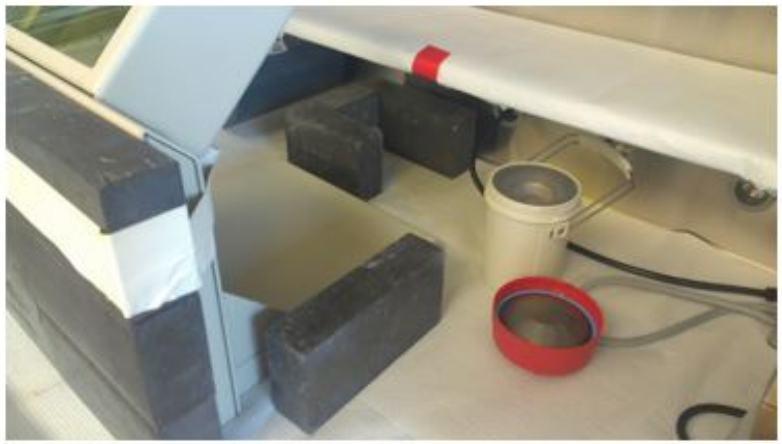
- 12.9 $\pm$ 0.1 mg of DUO_2 was irradiated
  - High Flux Isotope Reactor at Oak Ridge National Laboratory
- Burnup was 4.43 $\pm$ 0.31 GWd/tHM[30] from ^{137}Cs
- 0.196 $\pm$ mg of total Pu was produced as measured by ICP-MS

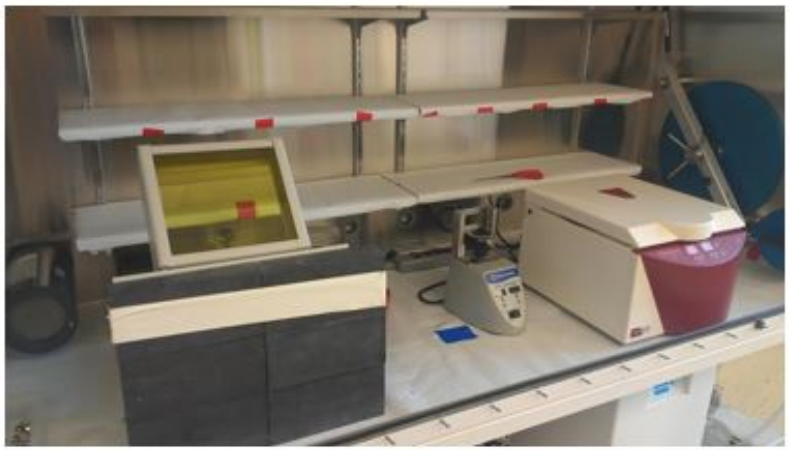


Dissolution of the spent fuel pellet



Glovebox





Experiments

Conditioned solutions with sodium nitrite ($Pu^{6+} \rightarrow Pu^{4+}$)

- Single contact extraction and back-extraction
  - Purpose: quantify product recovery, D-values and DF values for single contact 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: Quantify DF for a process 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

Mass spectrometry Results[11]

Recoveries of U and Pu

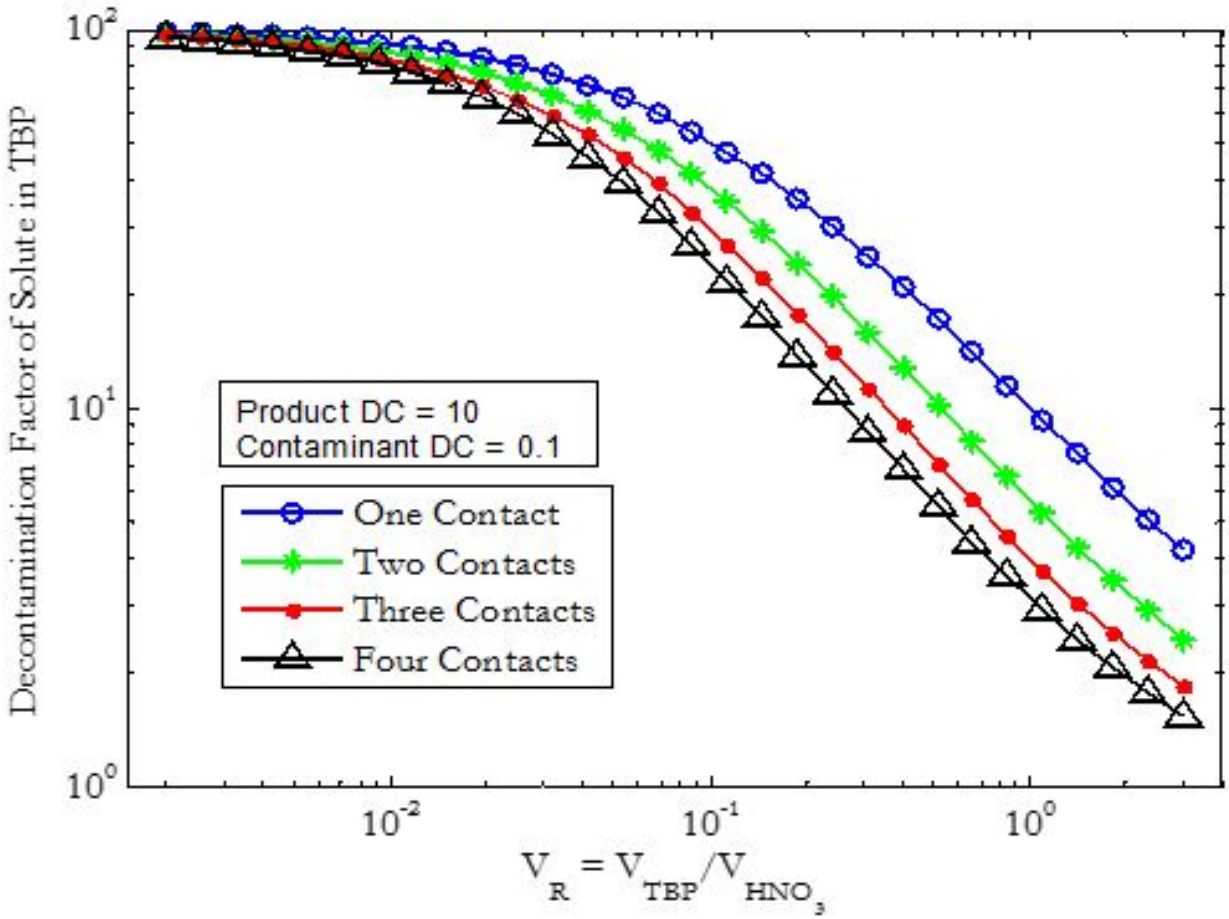
	Pu Recovery	U Recovery
Single contact	(83.4±9.5)%	(11.2±1.3)%
Multi-contact Process 1	(99.7±4.2)%	(6.8±0.3)%
Multi-contact Process 2	(93.0±4.6)%	(6.6±0.3)%
Overall Experiment 2	(92.7±6.0)%	(0.45±0.03)%

Mass Spectrometry Results[11]

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

Previous Experiment Results

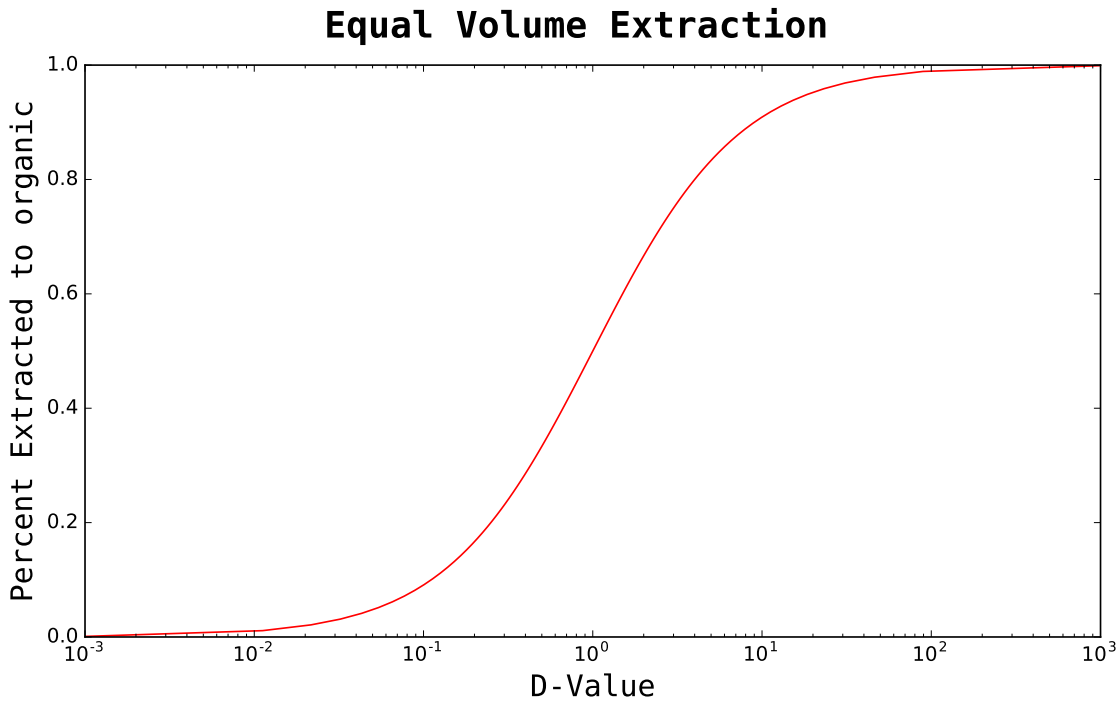


Decontamination Factors for multi-contact extraction.

Gamma Spectroscopy Results: 4M HNO_3, 2mM U, 30%vol TBP

D-Values:

Element (Z)	D-value	STD
Ce	0.04	0.01
Eu	0.08	0.02
Ru	0.04	0.02
Cs	3.9E-5	1.8E-5
Sb	0.005	0.007
Am	0.05	0.02



Initial vs Final solutions (process)

$c_{A,i}/c_{A,f}$ (Fission Products)

Element (Z)	Avg	Error
^{144}Ce (80 keV)	9.5	3.3
^{155}Eu (105 keV)	4.8	1.3
^{154}Eu	4.7	1.3
^{144}Ce (133 keV)	9.5	3.2
^{125}Sb	180	40
^{106}Ru	12.1	0.9
^{134}Cs	1200	800
^{137}Cs*	1070	560

Subsection 4

Future Work

Objectives I

Characterize a 4 extraction 3 back-extraction PUREX process

- Collect D-values for each step
  - ✓ ^{144}Ce,$^{155}Eu^*$,$^{154}Eu^*$,^{125}Sb, ^{106}Ru, ^{134}Cs, ^{137}Cs (Measured Gamma in triplicate)[5]
  - ✓ Convert Gamma Spectrum to D-values
  - ✓ $^{85}Rb^*$,$^{90}Sr^*$,$^{97,98,100}Mo$,$^{101,102,104}Ru$, ^{110}Pd,^{112}Cd,$^{133}Cs^*$,$^{140,142}Ce$,$^{143}Nd^*$, $^{147}Pm^*$,$^{151}Sm^*$,$^{154}Eu^*$,U^*,Pu^* (Mass Spec)
    - Convert all mass spec data to D-values
- Collect DF-values for the process (and the two steps)
  - ✓ Prepare alpha samples for each step (triplicate)
    - Analyze alpha samples for each step
    - Convert alpha spec + gamma spec data to DF values
  - ✓ Convert Mass spec data to DF values (published)[11]
- Mathematically connect D-values to DFs
  - Derive equations with uncertainty propagation
  - Analyze connection with uncertainty

Objectives II

Determine attribution indicators:

- Mathematically derive equations for above indicators with respect to one of the isotopes determined above
  - ✓ Burnup
  - ✓ Fluence Rate
  - ✓ Initial Enrichment
  - ✓ Decay Time
  - Fast-to-thermal ratios (requires iteration)
- Program a system to iteratively solve for these parameters given heavy metal concentration ratios
  - Make a program that can read ENDF files for x-sections
  - ✓ Create/Use a Bateman solver with automated x-section modifications
  - ✓ Create program to calculate single group x-sections from ENDF data and an assumed fast-to-thermal ratio
  - Couple all programs together in a single program
- Use above information to determine indicators for three sets of data

Questions?

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