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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AIEN 304

TEES | TEXAS A&M ENGINEERING EXPERIMENT STATION

NUCLEAR SECURITY SCIENCE & POLICY INSTITUTE

Outline

Introduction
- Motivation
- Context
- Background

Objectives

Methodology
- Mathematical methodology
- Experimental Procedure

Current and Expected Results
- Experiment
- Mass Spectrometry
- Gamma Spectroscopy Results
- Future Work

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]

Wanted to take some time to talk about the motivation behind this project. There are a lot of stuff going on in the world. Here I've listed a few that pertain to nuclear weapons.

- Iran limiting capabilities for 8 years (give more background -liftin sanctions and unfreezing accounts
- India non safeguarded
- non-safeguarded reactors, in India for example
- North Korea, detonating a nuclear device October 2006
- ISIS, if they could, would probably want nuclear weapons, and they would use them on us

As 911 indicated, we have enemies, who don't like us. To quote William Perry, "Our greatest threat is a terrorist nuclear strike". Lukily, aquiring nuclear weapons is no easy task. And thanks to organizations like the international atomic energy agency, or the treaty on the non proliferation of nuclear weapons, the international community is generally on board with nonproliferation. Nontheless, all sources of special nuclear material are not safeguarded, and the "awful arithmetic of the atomic bomb", to quote Eisenhower, doesn't leave room for calculational errors.

- Forensic capabilities need to improve, which we will talk about in a minute, but first, some definitions

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

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

At this point I wanted to take a moment to define some terms. To help narrow our nonproliferation scope. First,

- **Say something about PUREX**
- Special nuclear material, this is the material that is traditionally safeguarded. One of three building blocks for nuclear weapons, which are the material, design information, and manufacturing skills
- Nuclear forensics - investigative activity, determining attributes of SNM. This could include composition, material form, or date since processing. Involves a broad range of scientific destructive and nondestructive techniques. The time scale for full nuclear forensic analysis for samples can vary widely, depending on the prepardness of whatever laboratory is receiving the sample. In Moody's book, nuclear forensic analysis, he cites several examples that took a couple of months to analyze
- For my project I will be determining several attributes or indicators for a sample. I wanted to note that these indicators are clues or evidence towards a conclusion. And the examples listed are indicators that I would like to determine.

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)**

According to a report from the committee on nuclear forensics released in 2010, the United States forensic capability has been demonstrated in real world scenarios, but is at risk unless certain developmental requirements are met. The committee listed 4 areas of concern, where improvment is needed.

- In terms of organization, nuclear forensics responsibility is shared by several agencies without central authority and with no consensus on strateic requirements to guide the program.
- For sustainability, our current capabilities are the fruit of the nuclear weapons program and our laboratory infrastructure, funding for both have been decling
- Skilled personnel in these areas are few, and key facilities are in need of replacement because they are old, outdated, or do not met modern environmental, health, or safety standards.
- In recent years, nuclear forensic techniques methodologies have been on the rise, some from presented from our own department, but according to this source, a large fraction of techniques are remnants of the cold war era, with less restrictions. Also, forensic excersises usually take months to complete, a time scale which is too long

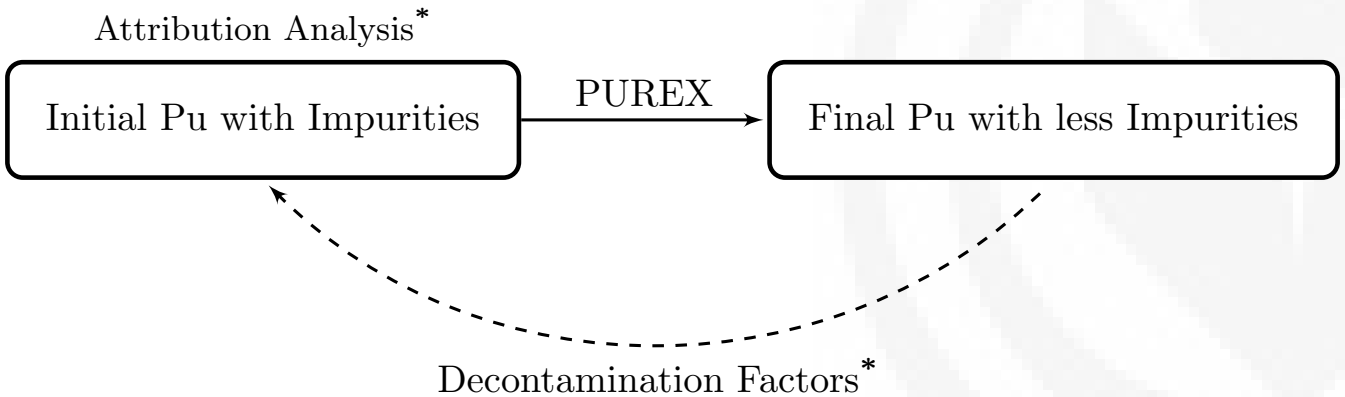
Problem must be met on an administrative level, but this project will help with two of these important goals. Developing the workforce, by example of yours truly, and the procedures we'll talk about in a moment.

Forensic Context

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

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

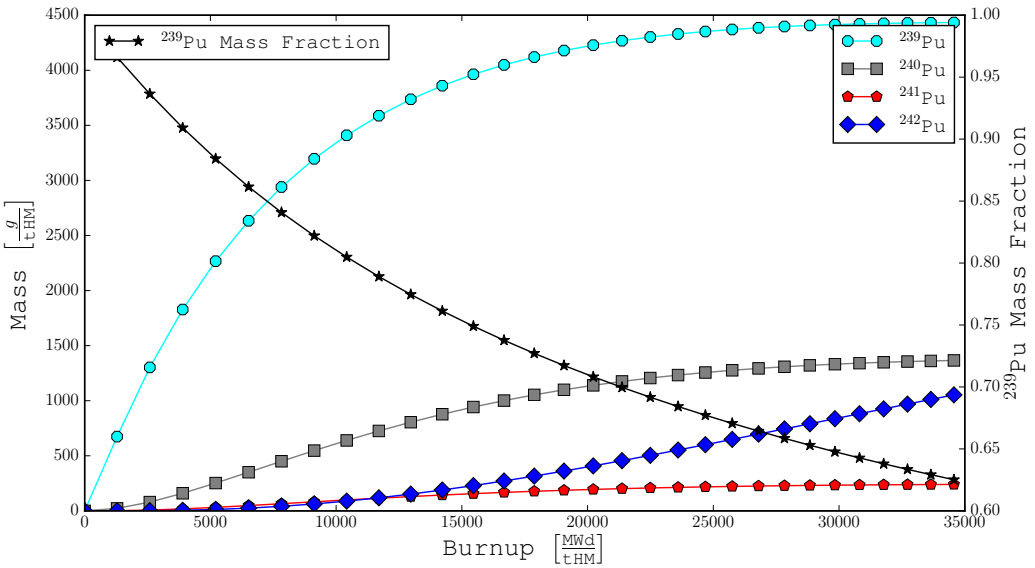
- Attribution for used or spent fuel has been previously studied [5, 6, 7]



- Inverse problems, naturally have many solutions. Where many paths could be taken to arrive at an end state
- our study will analyze plutonium production, which muddies waters more in its production than uranium
- Purification necessariy because there is a chance for fission along any point of production
- This complicates attribution by a considerable amount due to unknown DF values
- Studies have looked at isotopic compositions of irradiated fuel and came to conclusions about origins
- Describe the picture, and how it shows your project, also talk about the assumption...
- This requires that the specifics of the PUREX process used for plutonium separation are known, so that DFs are applied appropriately.
- If this information were not readily available, this same analysis would have to ensue with best estimate PUREX processes and experimentally determined Distribution Coefficients

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

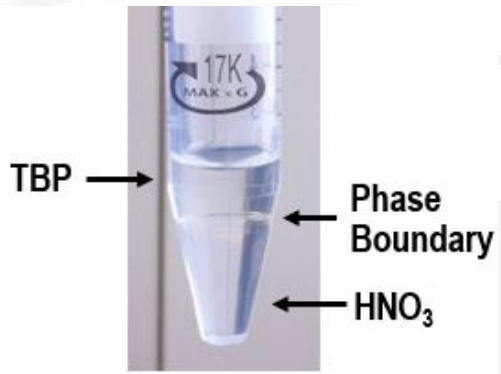


- **Explain graph, walk through graph**
- Lower burnup fuel produces weapons
- Plutonium isotopics depend on
  - Burnup (irradiation history)
  - Reactor neutron spectrum (core design)
- Fast Breeder Reactor
  - Madras Atomic Power Station Kalpakkam, India
  - Expected criticality in Jan 2017
  - Cost from 450 million euros to 750 euros
  - Sodium-cooled reactor design - U238 for breeding
  - 100 GWd/t for core, 40 year life, 1750 tonnes of sodium about 75% of olympic sized swimming pool.
  - liquid sodium has a density a little less than water
  - MOX fuel (UO2 and PuO2) fuel
  - Fuel discharged at 100GWd/t, but I just mentioned that we are worried about 1GWd/t, mistake?

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:

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

Different from DF PUREX Process

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

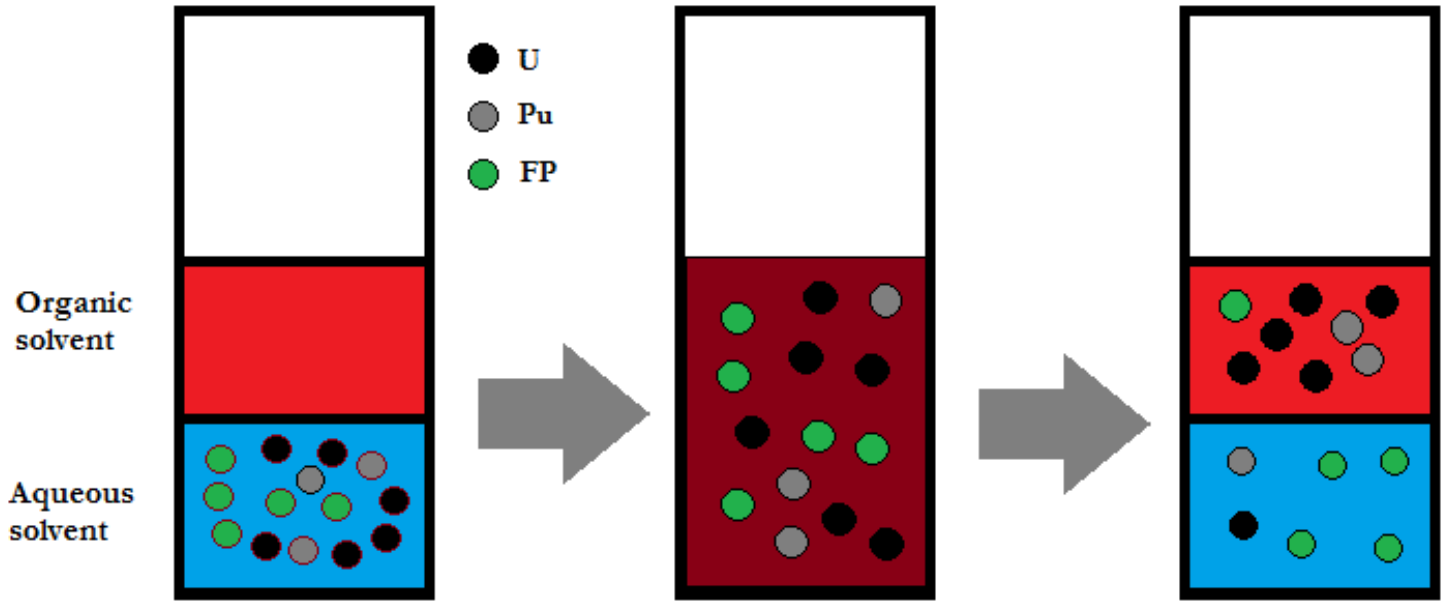
Distribution Coefficients (process depends on this)

1. Distribution coefficients can be reported in terms of volume basis (weight per unit volume), or a mass basis (mass of solute per unit mass of solute free solvent)- usually reported on volume basis
2. Vary Widely with: Composition of phases, solution saturation, temperature of the solvent
3. Note: Not a function of desnity, even though the two solutions have different densities, when solving for this value, it cancels out
4. Volume matters, Barnwell process has different volumes, more intuitive sense of things

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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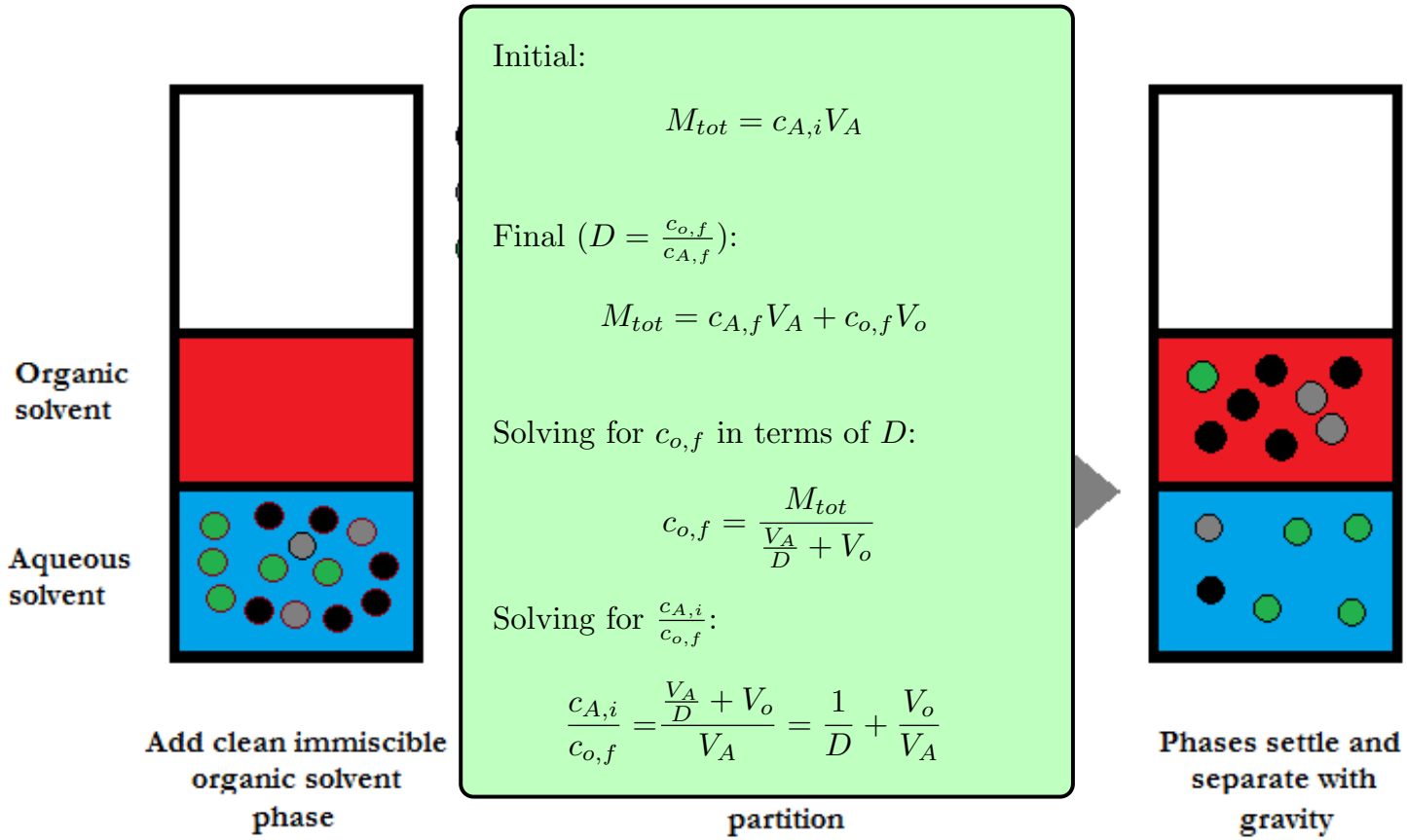

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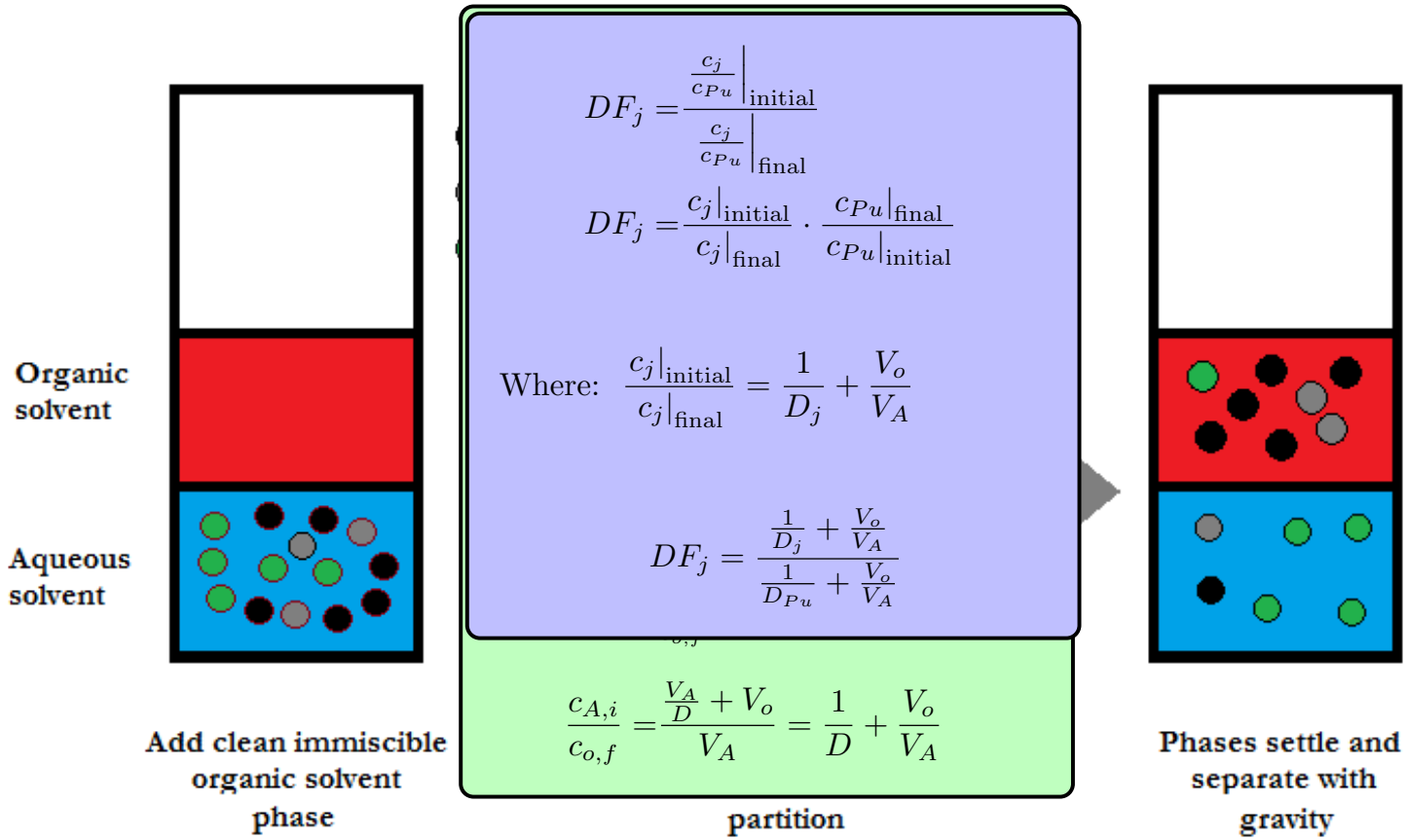
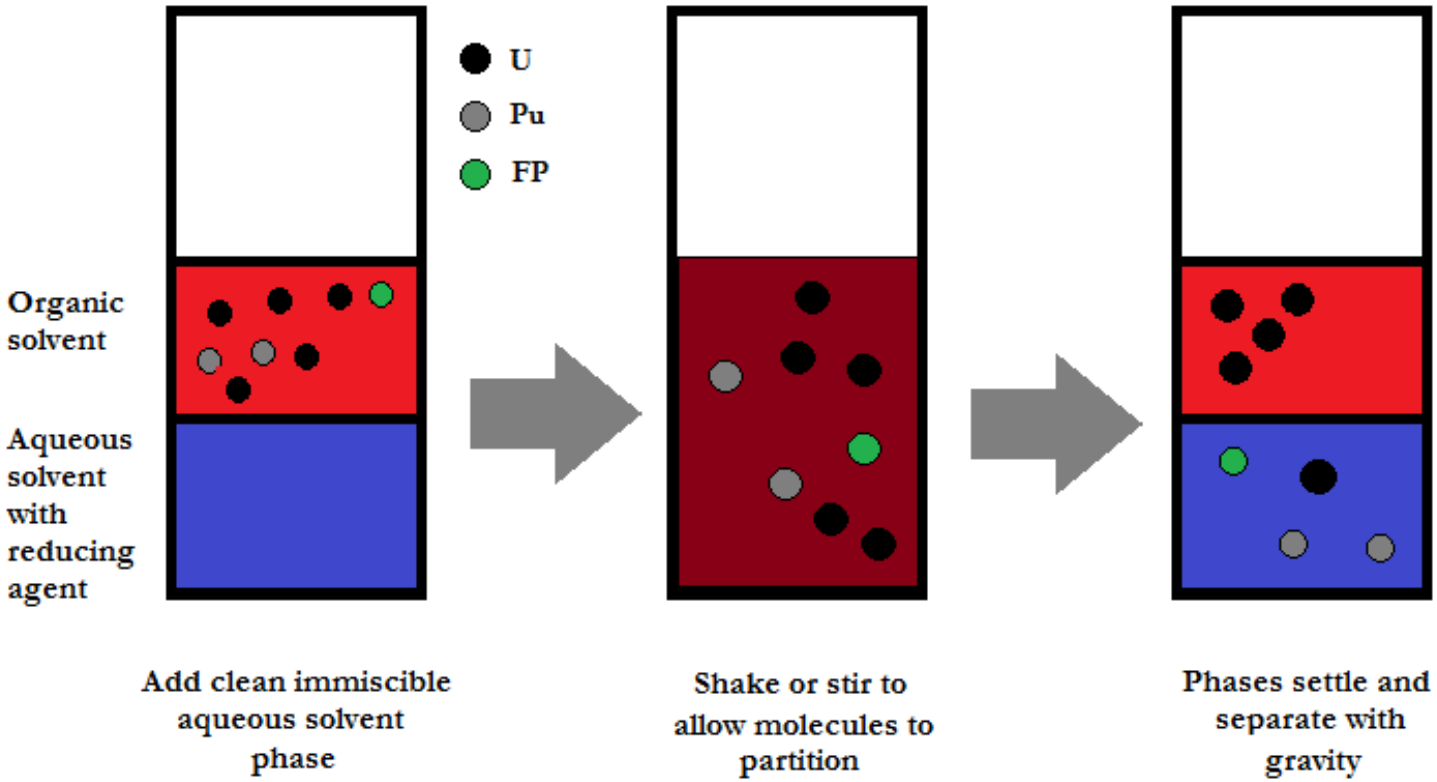


- For the PUREX process, there are two main separation steps, for which we will look at more closely
- Most of the fission products are left in the aqueous solution at valence III and V states[10]

Back-Extraction

$$Pu(NO_3)_4(TBP)_{2(o)} + Fe^{2+}_{(aq)} \leftrightarrow Pu^{3+}_{(aq)} + 4NO^{-}_{3(aq)} + 2TBP_{(o)}$$ [11]



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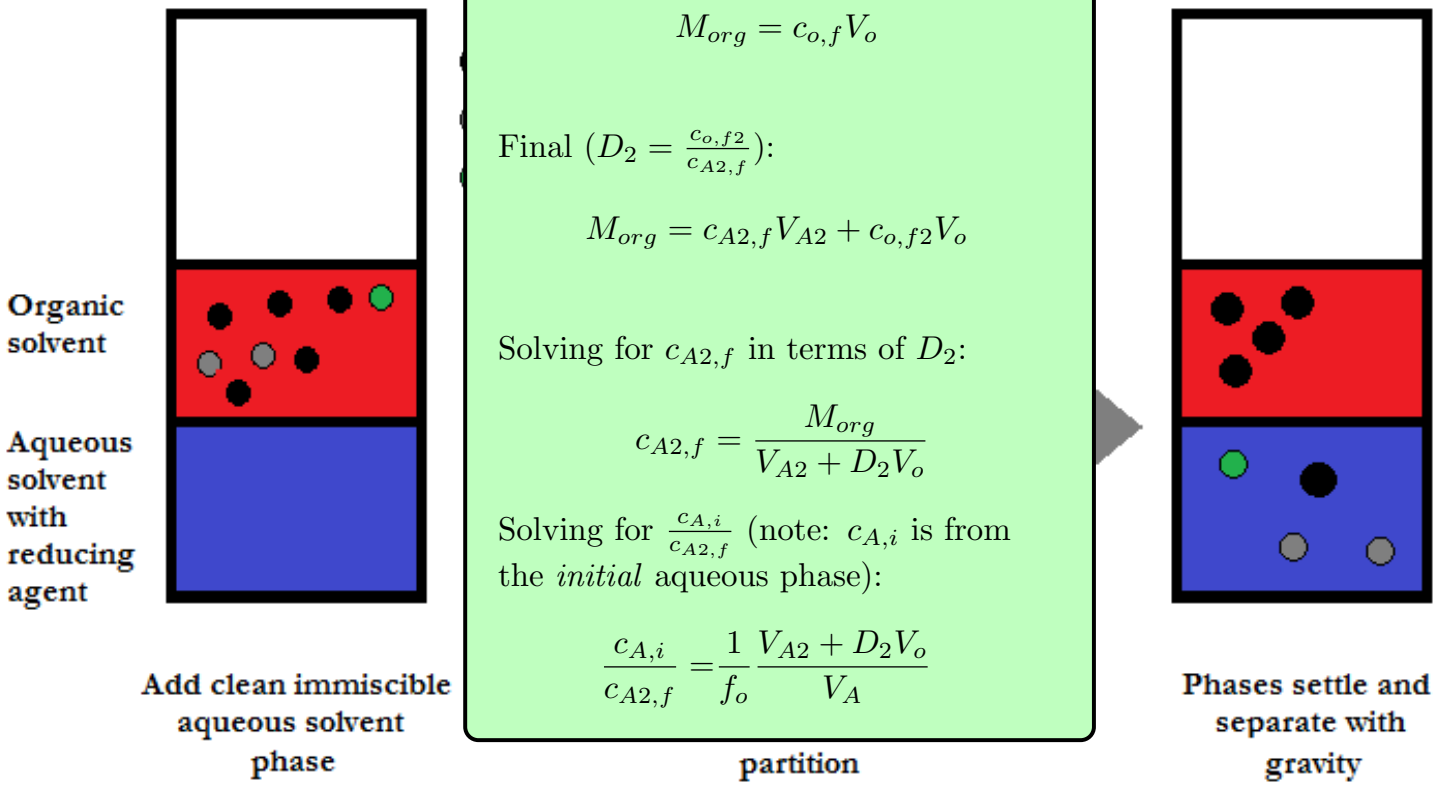

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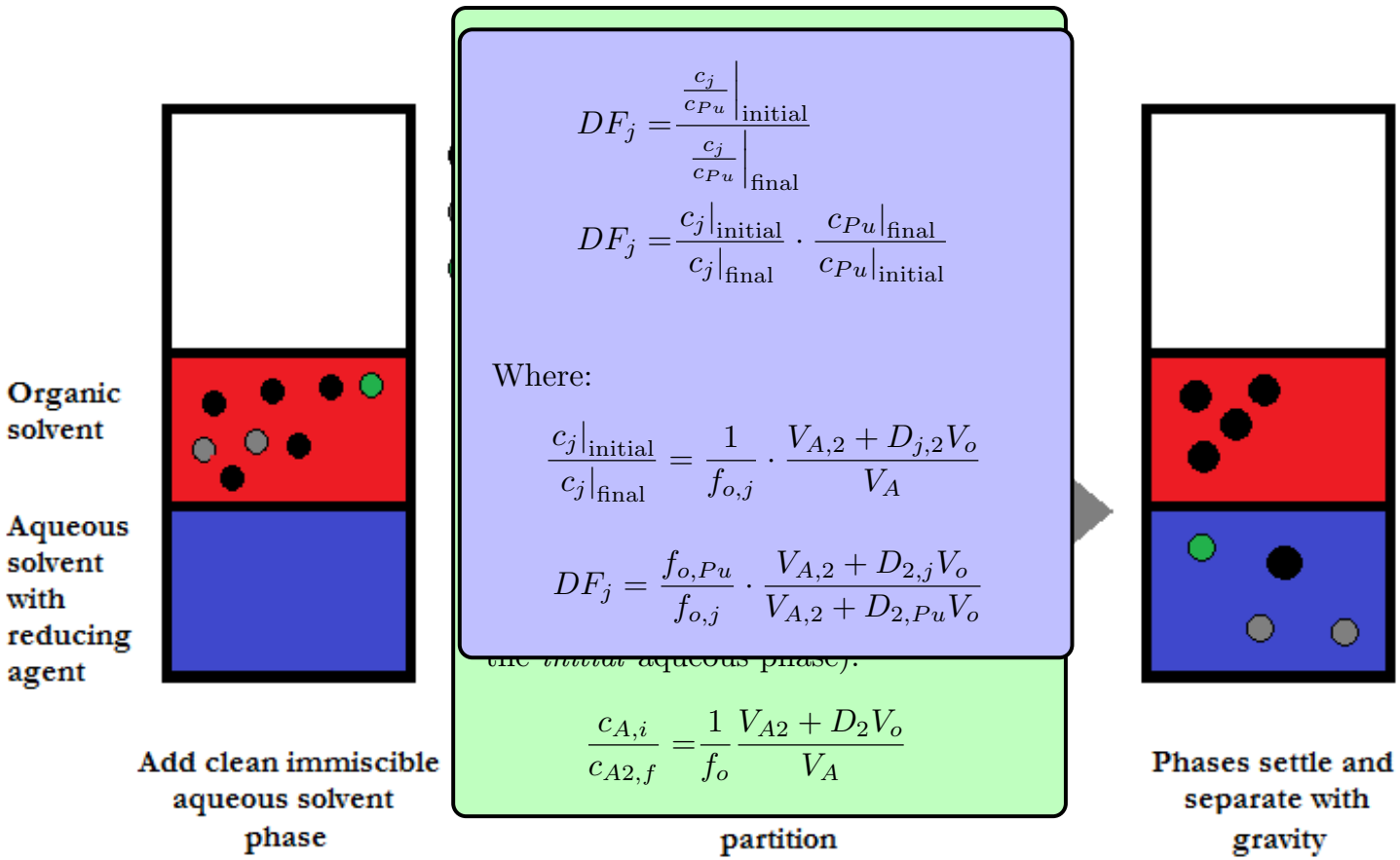


$$DF_j = \frac{\frac{c_j}{c_{Pu}}\Big|_{\text{initial}}}{\frac{c_j}{c_{Pu}}\Big|_{\text{final}}}$$

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

Where:

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

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

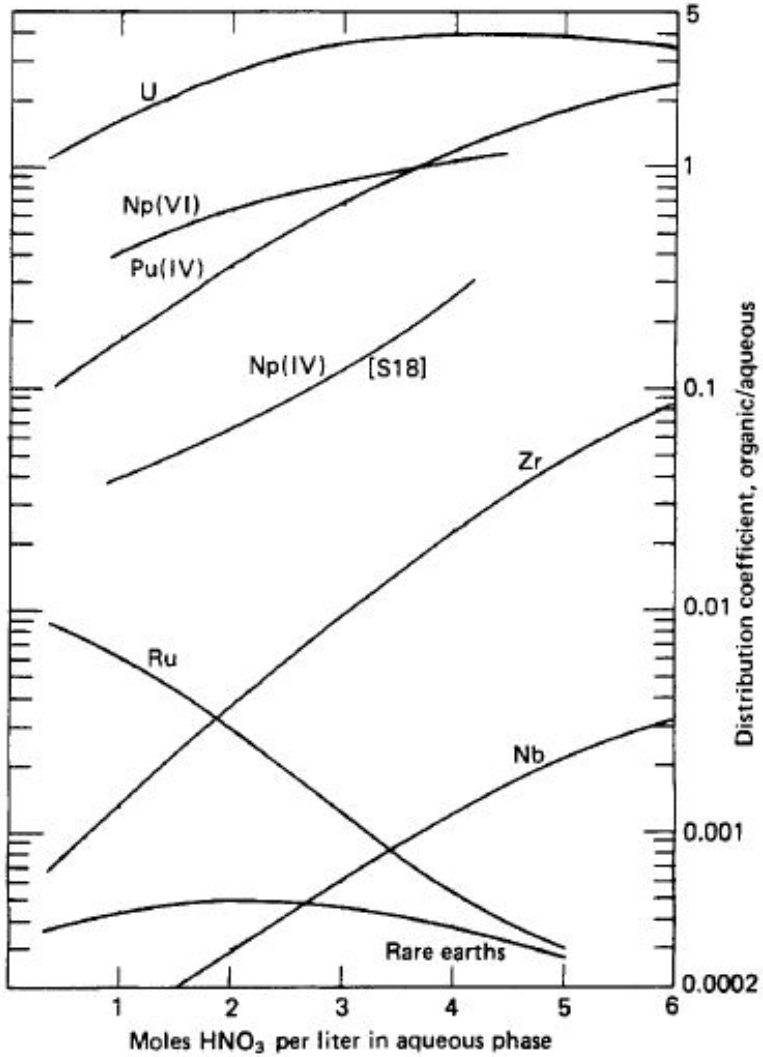
the *initial* aqueous phase):

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

- The fission products that contribute mostly to the radioactive contamination of product in PUREX are zirconium, niobium, and ruthenium - with multiple oxidation states.

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]

Talk about separation of Pu, and where our experiments are. Give an example from the plot

Chemical Context

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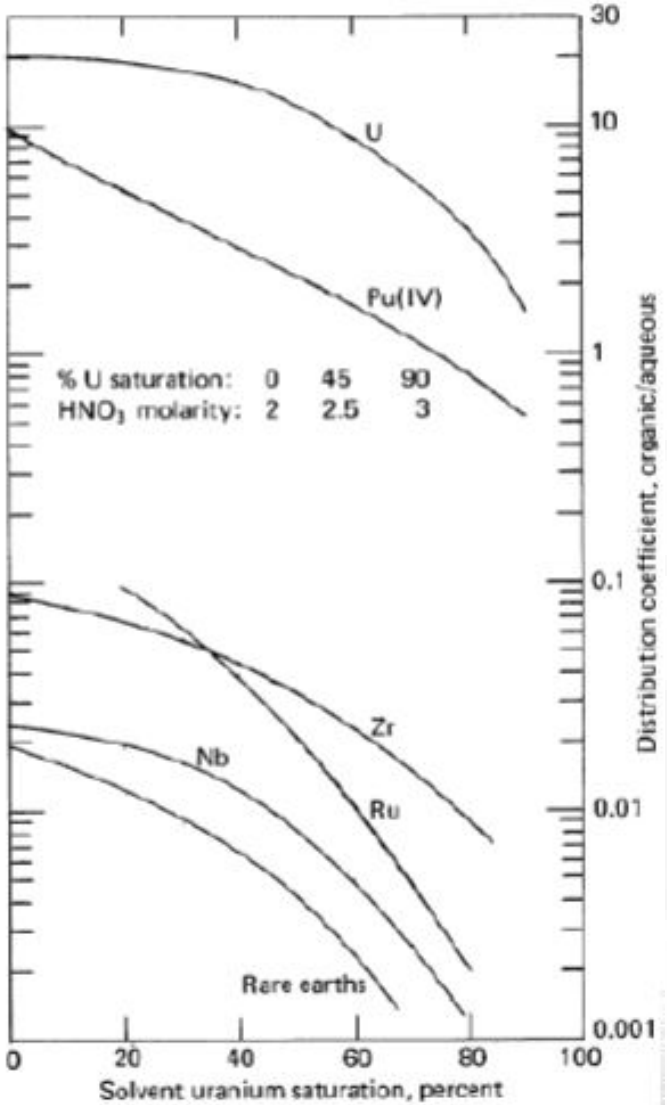
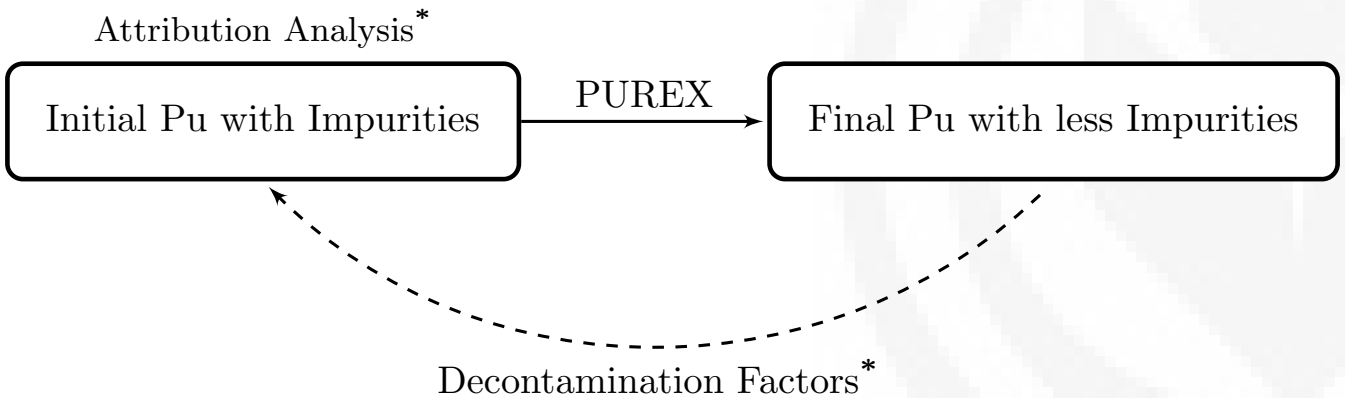


Adapted from Stoller[8]

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)[12]
- 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

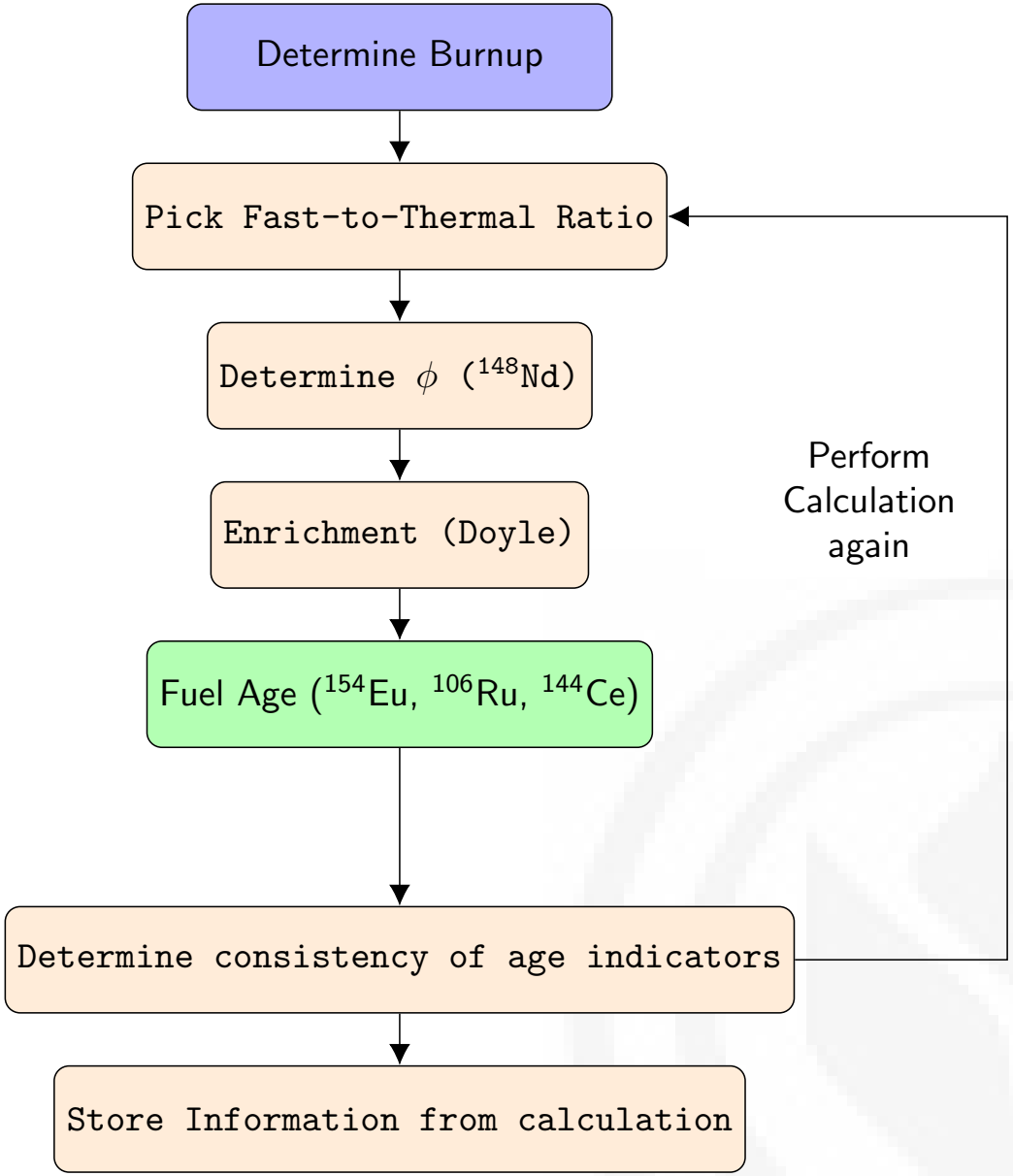
Present Status of the Question

- Stable noble fission gases as burnup verification[13]
- Determine burnup, enrichment, and decay time from used fuel in a RDD[14]
- Analysis of purified plutonium isotopics for reactor type[15]
- PUREX co-processing DF values for U and Pu[16]
- PUREX D-values and DF values under numerous circumstances[9, 17, 18, 19, 20, 8]
  - DF values for ^{106}Ru and ^{95}Zr[8]
  - Compilation of D-values for U, Th, and Pu[21]
  - D-values for rare earths, Pu, Th[19, 22, 23, 24, 25, 26, 27, 28, 29, 30]
  - Gd D-values[30]

1. verify burnup so no nefarious activities (we are advancing, because to purified material) also Cs is used for burnup
2. RDD not purified, no chemical processing
3. Chem processing, had some difficulty, but some sucess, only looked at differences between fast and thermal (not burnup)
4. limited in isotopes solved for
5. Not the isotopes we want
6. Work unique in that elemental DFs used for forensics

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

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

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

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

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

Production and loss depend on

- Neutron Spectra
- Fluence Rate
- Cross Section Data

Where N is the number of nuclides and blah
$\gamma_{j\rightarrow i}$ is the fission yield for isotope i from fission of isotope j. $b_{j\rightarrow i}$ is the fraction of radioactive disintegration by nuclide j, which leads to nuclide i. In the case of spontaneous fission $b_{j\rightarrow i}$ is the product of spontenous fission fraction for nuclide j and the yield for fission from nuclide j producing nuclide i. Where **A** is a matrix whose diagonal elements are $[-\lambda_1^{eff}, -\lambda_2^{eff}, ..., -\lambda_N^{eff}]$, all off diagonal elements are $b_{j\rightarrow i}^{eff}$ (i for the row, and j is for the column) and $\vec{n}(t) = [n_1, n_2, ..., n_N]$. **Single Group Cross Section and flux**

Analytical Procedure - Burnup

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

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

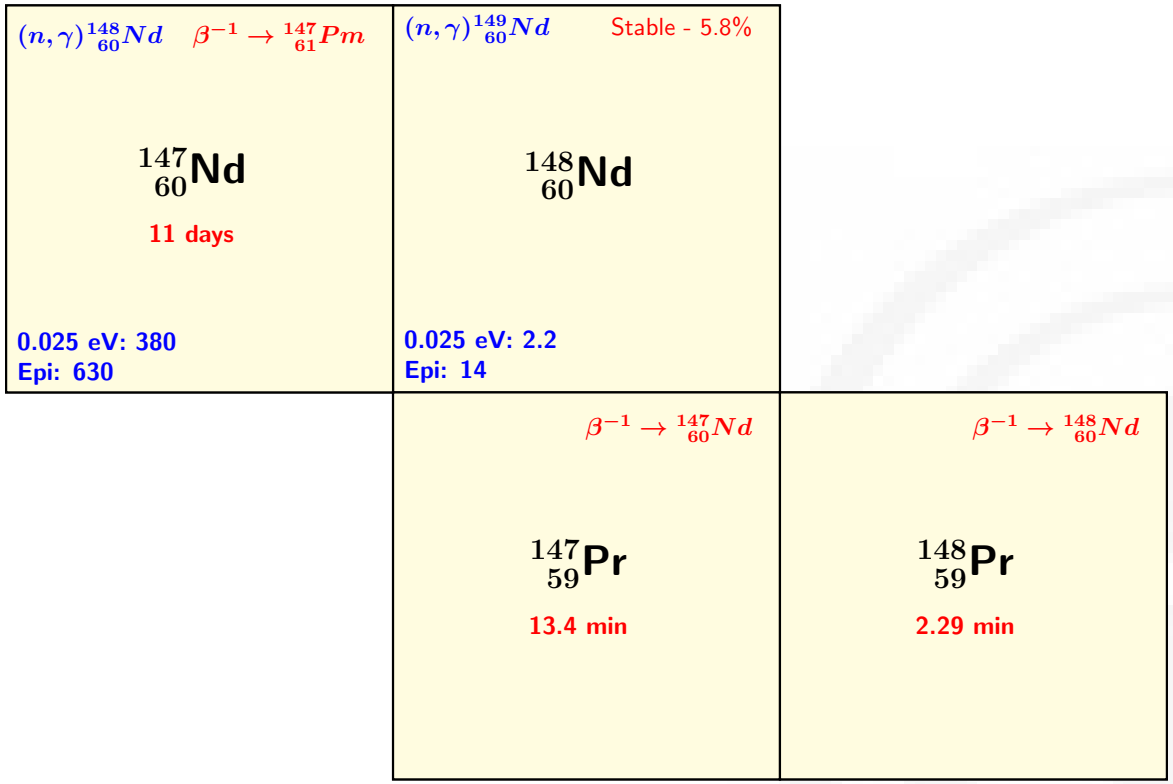
$$= \left[\frac{N^B}{N_0^{HM}} \right] \frac{N_A E_R}{\gamma_B} \cdot \frac{1}{M_0^{HM}}$$

Prepare to defend equation How do you know what the initial HM concentration is? How do you know Mo for the 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

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

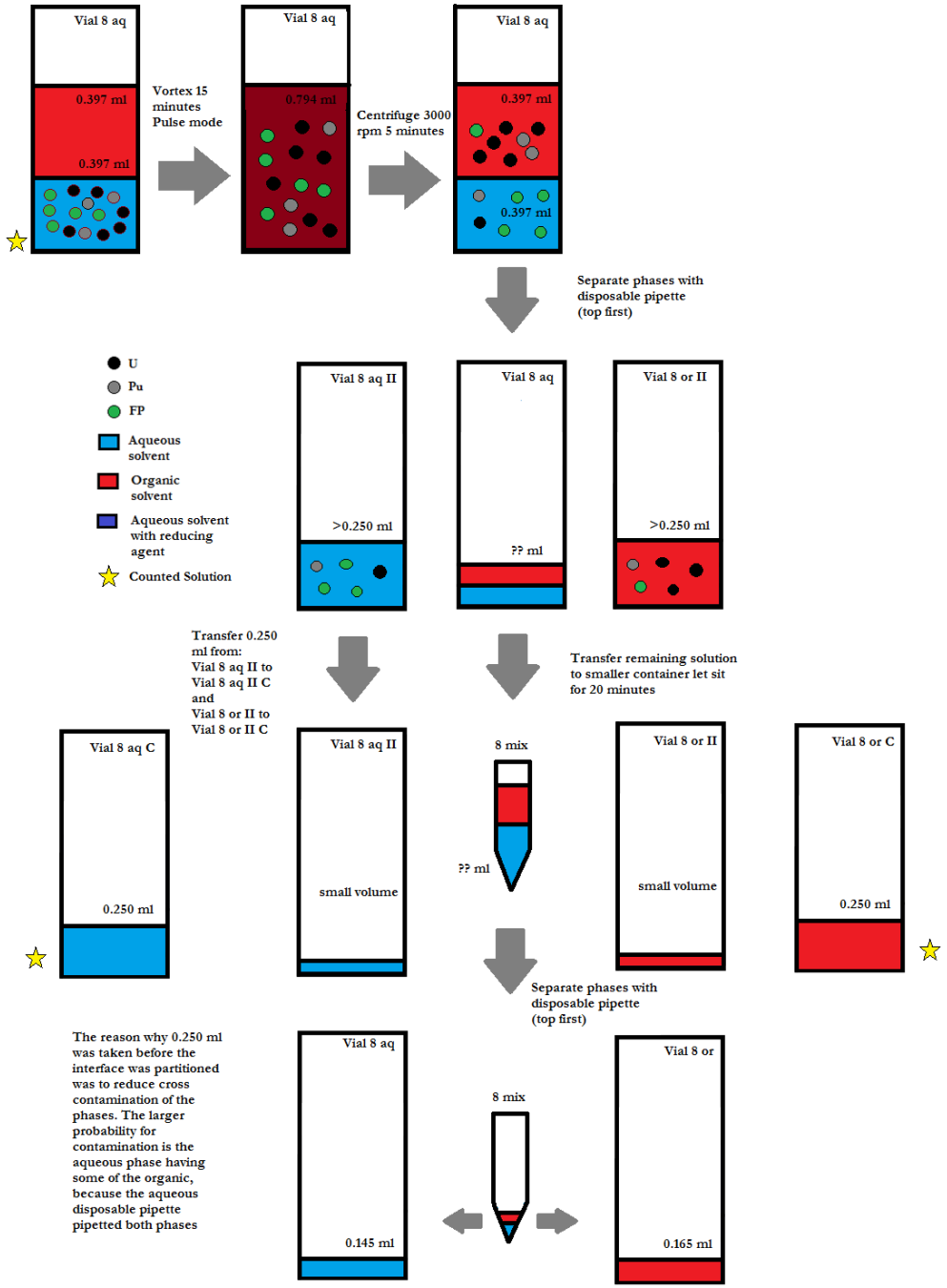
Decay Time

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

Initial Enrichment The first bracketed term corresponds to the final enrichment with reference to initial heavy metal – where initial heavy metal is previously solved for. The second term adds to the final enrichment the enrichment contribution from burn-up, where burn-up was previously solved for. The second term includes fissions from 238U, 239Pu, 240Pu, and 241Pu. In order to account for this, the additional terms are added to subtract out their individual contribution. **Decay Time** Solving Bateman equations, with assumptions on burnup, ϕ, initial enrichment, fast to thermal ratios (calculate 1000 x-sections)

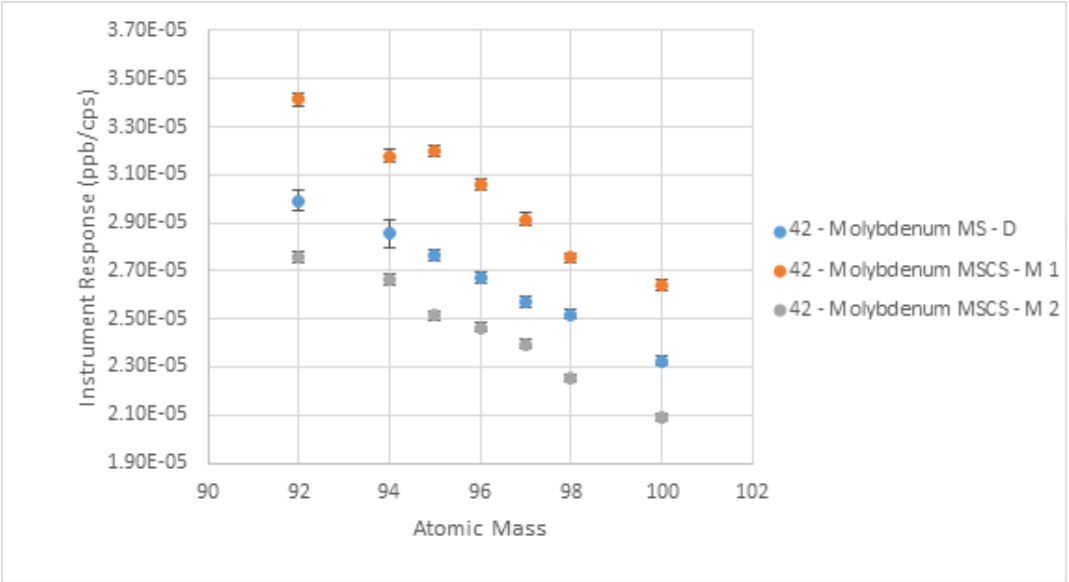
Subsection 2

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

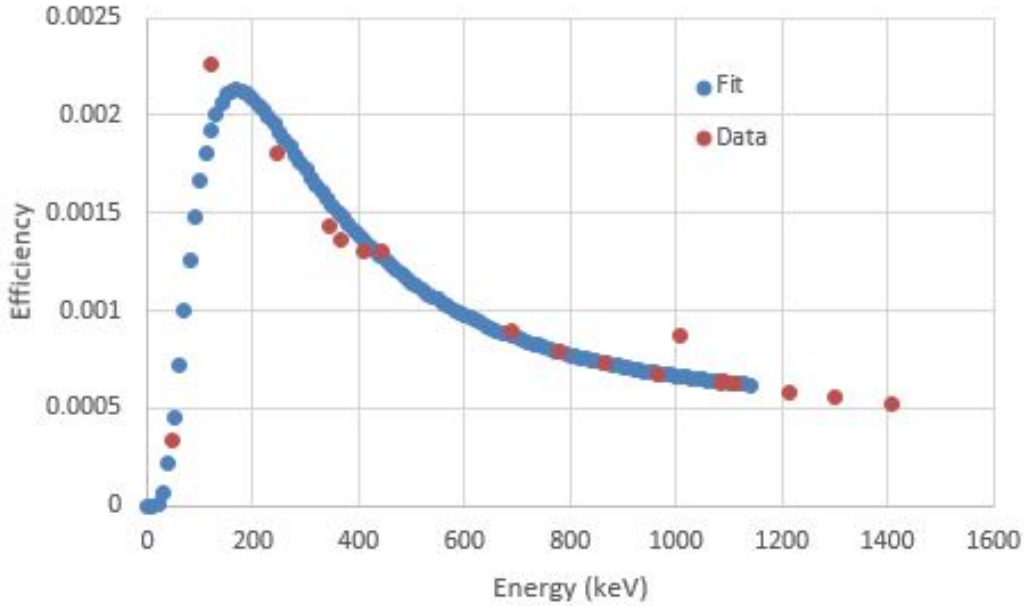


Talk about how we prepared samples, and analyzed data, they just ran it through the machine we conditioned the samples

Experimental Procedure - Gamma Spectroscopy

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

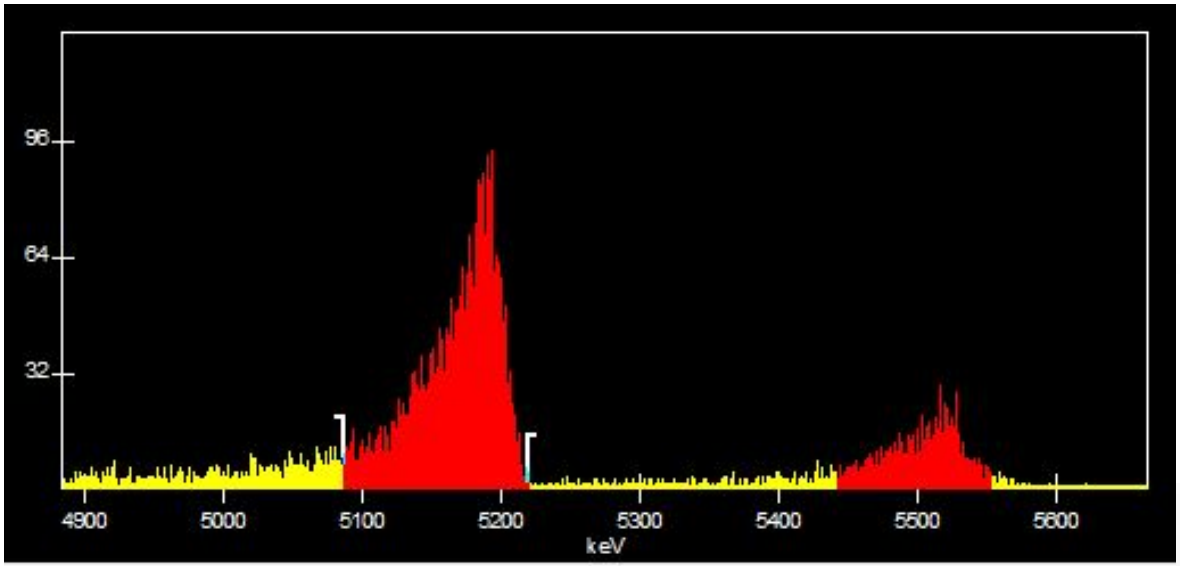
^{155}Eu, ^{154}Eu, ^{144}Ce, ^{125}Sb, ^{106}Rh, ^{134}Cs, ^{137}Cs



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

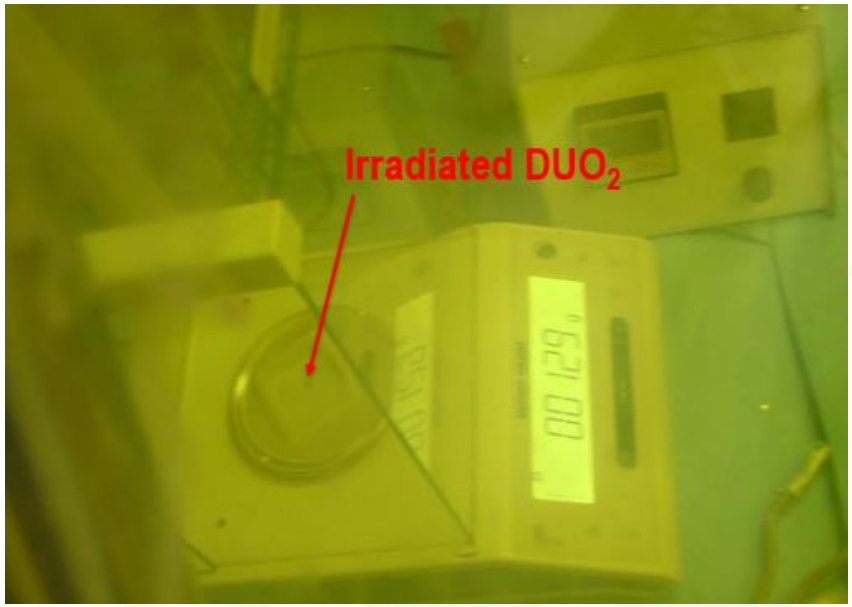
^{239}Pu,^{240}Pu,^{241}Am



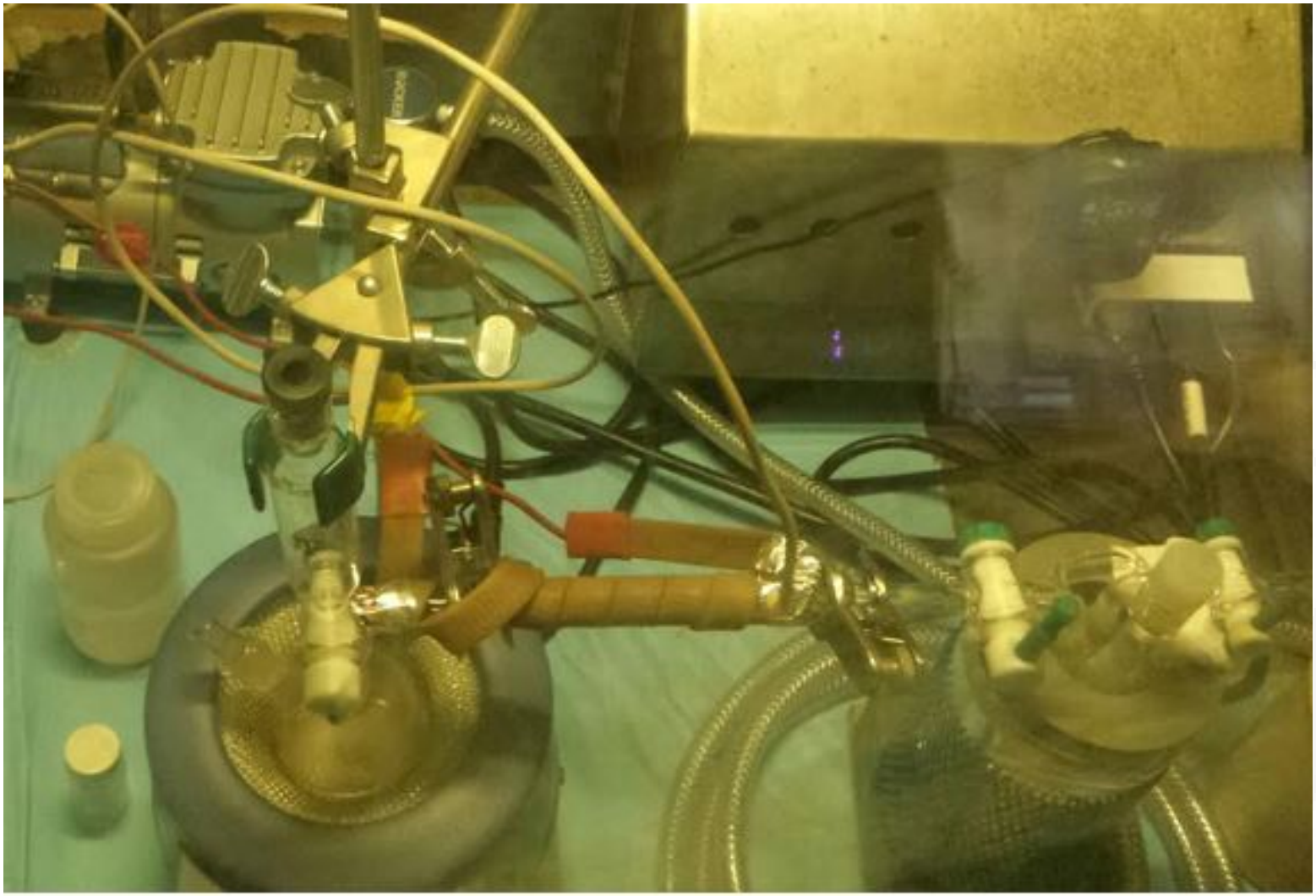
Current and Expected Results

Irradiation

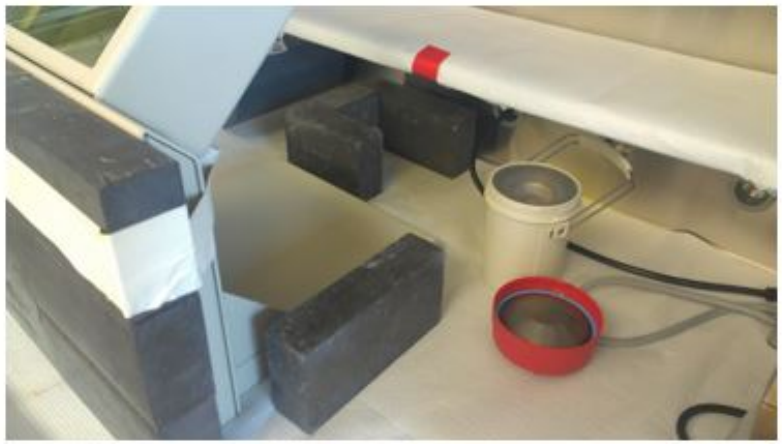
- 12.9 $\pm$ 0.1 mg of DUO_2 was irradiated
  - High Flux Isotope Reactor at Oak Ridge National Laboratory
- Burnup was 4.43 $\pm$ 0.31 GWd/tHM[31] from ^{137}Cs
- 0.196 $\pm$ mg of total Pu was produced as measured by ICP-MS

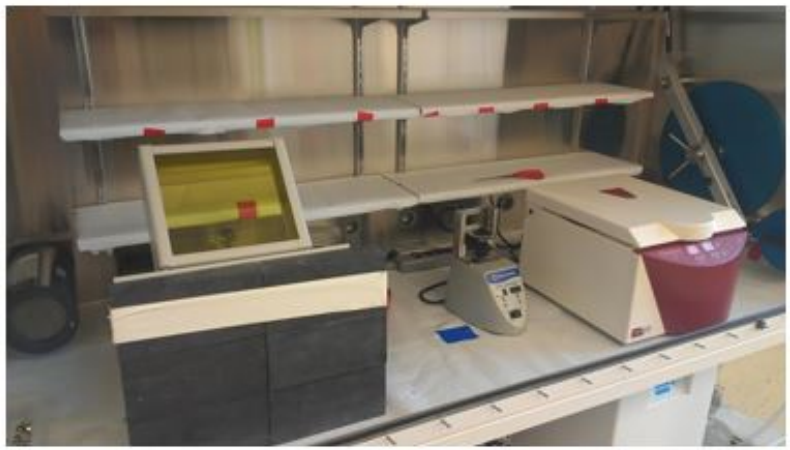


Dissolution of the spent fuel pellet



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[12]

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)%

Contact: One Extraction, one back-extraction

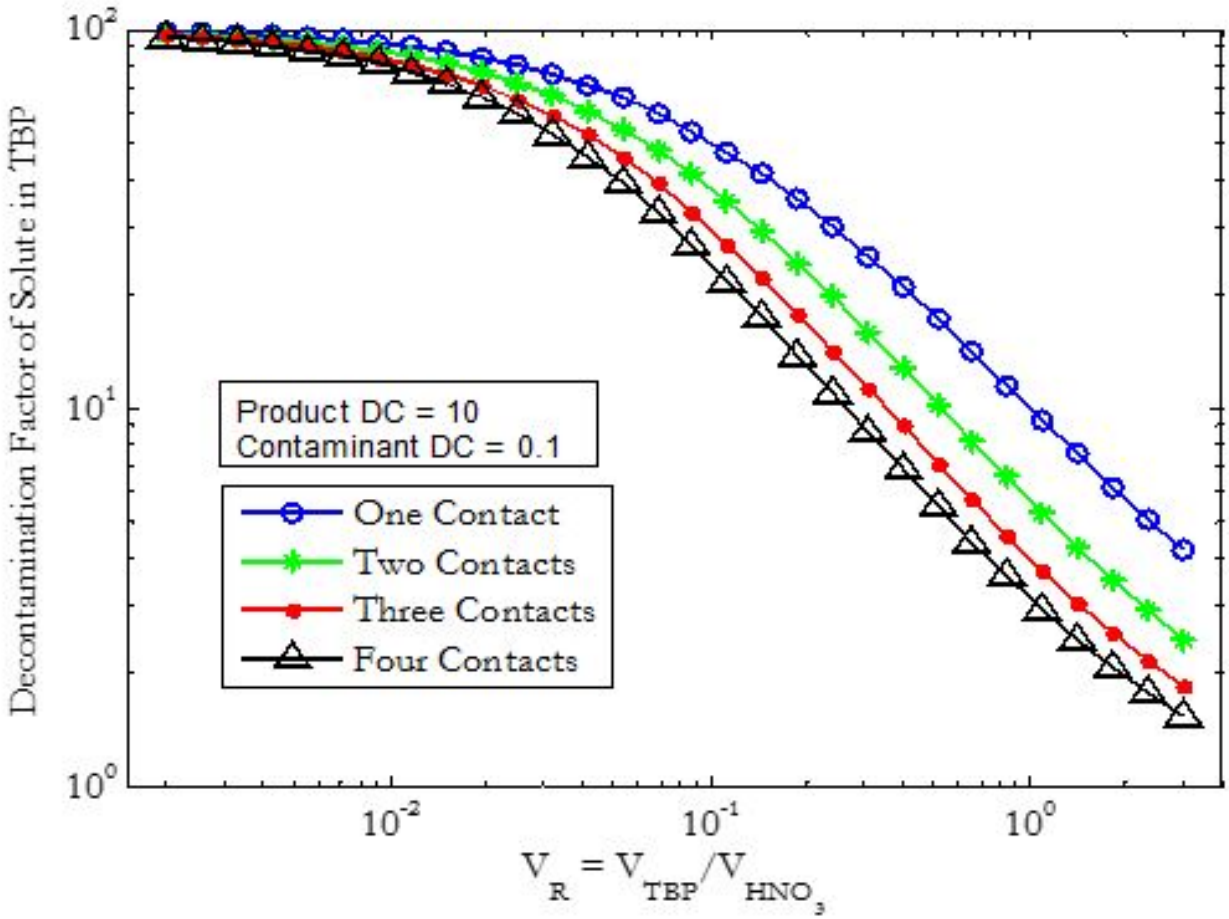
Mass Spectrometry Results[12]

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

Why 177 to 52.9 on board

Previous Experiment Results

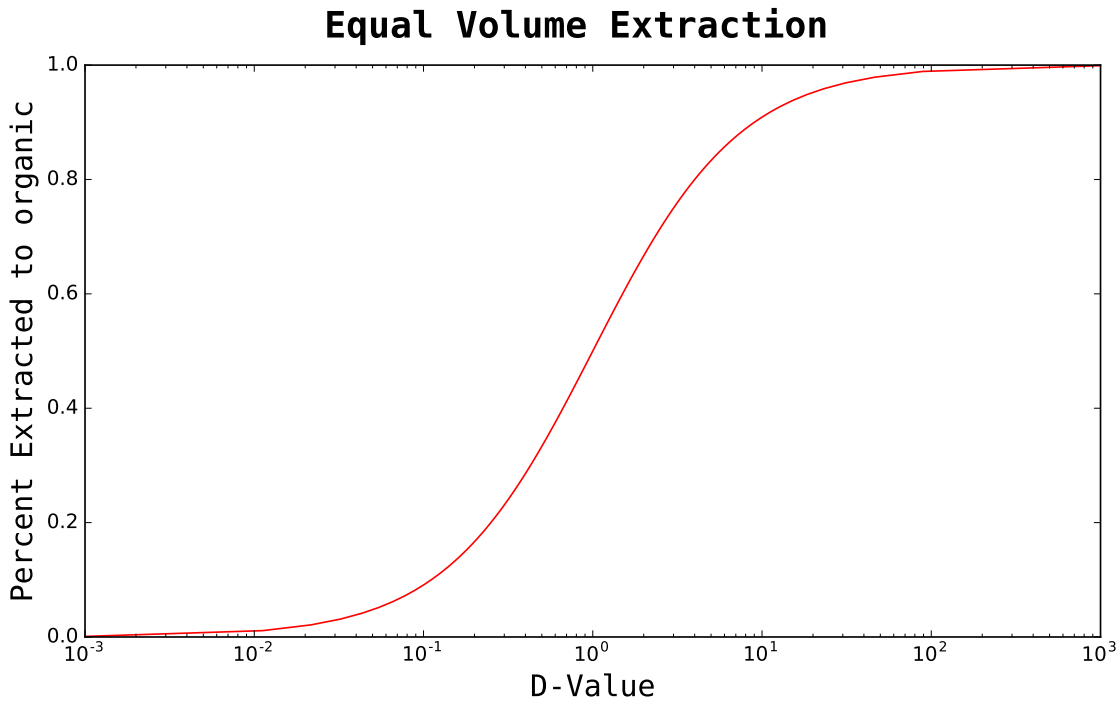


Decontamination Factors for multi-contact extraction.

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

Only used two experiments for Cs-137, encountered lower limit for one of the final solutions (order of background), the minimum value for the DF for this solution was 3000

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)[12]
- 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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