

## Experimental Characterization of Pu Separation by PUREX Process on a Low-Burnup, Pseudo-Fast-Neutron Irradiated $\text{DUO}_2$ for Product Decontamination Factors and Nuclear Forensics

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**TEXAS A&M ENGINEERING  
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AIEN 304

**NUCLEAR SECURITY  
SCIENCE & POLICY INSTITUTE**

# Outline

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- Motivation

- Contexts

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## Procedure

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- Mass Spectrometry

- Gamma Spectroscopy Results

- Gamma Spectroscopy Results

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# Motivation

## ❖ Current Events

- Joint Comprehensive Plan of Action
- Non-safeguarded reactors
- Islamic State of Iraq and Syria

## ❖ Past Events

- Septemer 11, 2001

## ❖ Limited scope of IAEA safeguards

## ❖ “the awful arithmetic of the atomic bomb”<sup>[1]</sup>

## ❖ Need for improved forensic capabilities<sup>[2, 3, 4]</sup>

## Definitions

- ❖ Special Nuclear Material (SNM)
  - Plutonium,  $^{233}\text{U}$ , or  $^{235}\text{U}$
- ❖ Nuclear Forensics
  - The investigative activity that surrounds the search for attributes of undetermined radioactive specimens for the purpose of attribution.
- ❖ SNM origin attributes/indicators
  - Indicators or clues for SNM origin attribution. Examples include burnup, fluence rate, initial fuel enrichment, fuel age, and fast-to-thermal irradiation ratios
- ❖ Decontamination Factors (DF)
  - A measure of the effectiveness with which a product is decontaminated from a contaminant

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

## National Context

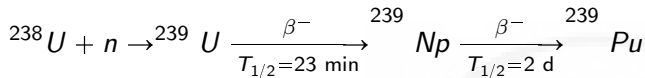
“The United States has developed a nuclear forensics capability that has been demonstrated in real-world incidents of **interdicted materials** and in exercises of actions required after a nuclear detonation. The committee, however, has concerns about the program and finds that without strong leadership,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

Major areas of concern include:

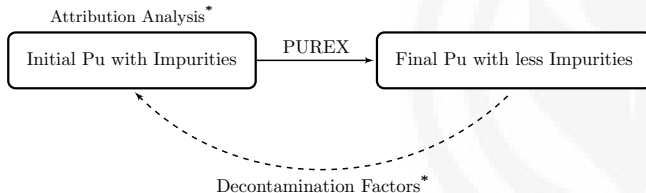
- ❖ Organization
- ❖ Sustainability
- ❖ **Workforce and Infrastructure**
- ❖ **Procedures and Tools**

## Forensic Context

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

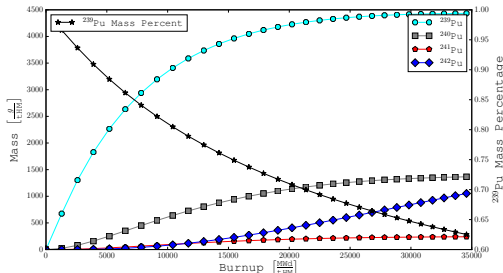


- ❖ Attribution for unpurified Pu has been previously studied [5, 6, 7]



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

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

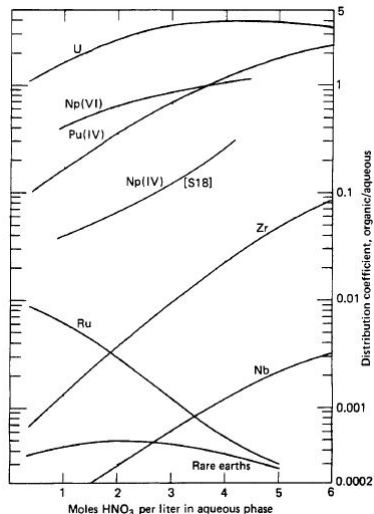
- Specific element to element
- Vary widely<sup>[8]</sup>
- The fraction of mass,  $f_o$  deposited in the organic phase, assuming a volume ratio between the aqueous and organic phases,  $V_R$ , is:

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



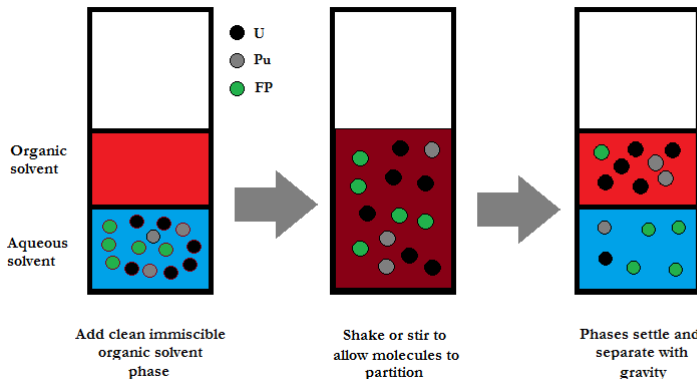
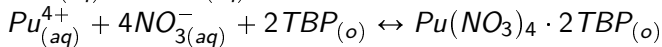
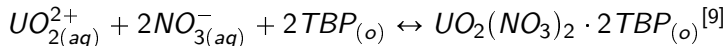
# Chemical Context

- ❖ 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

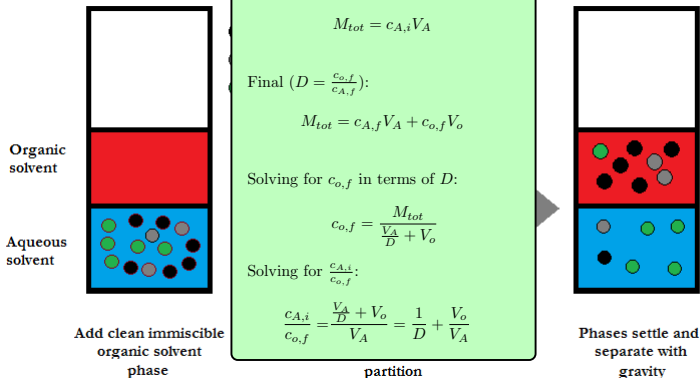
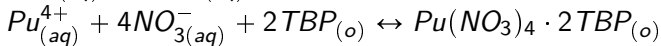
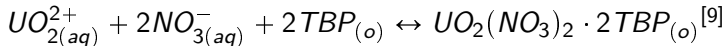


Adapted from Stoller<sup>[8]</sup>

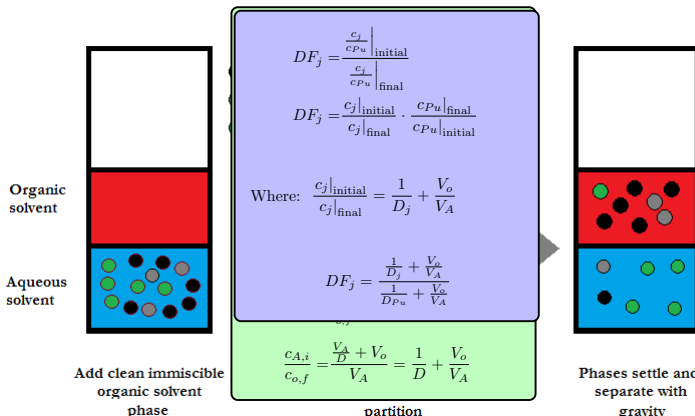
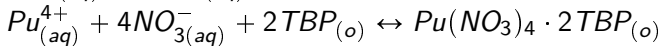
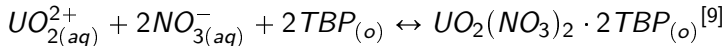
# Extraction



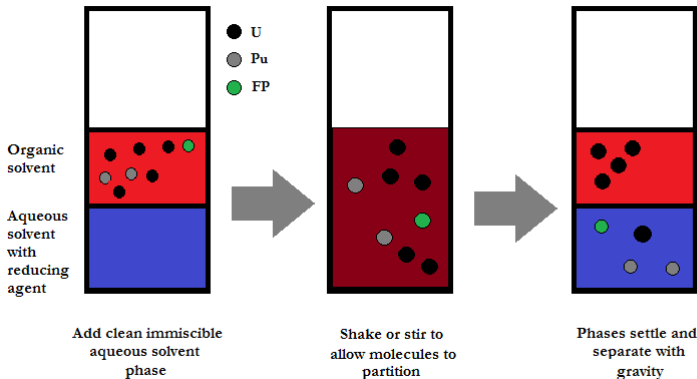
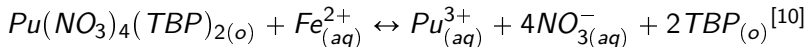
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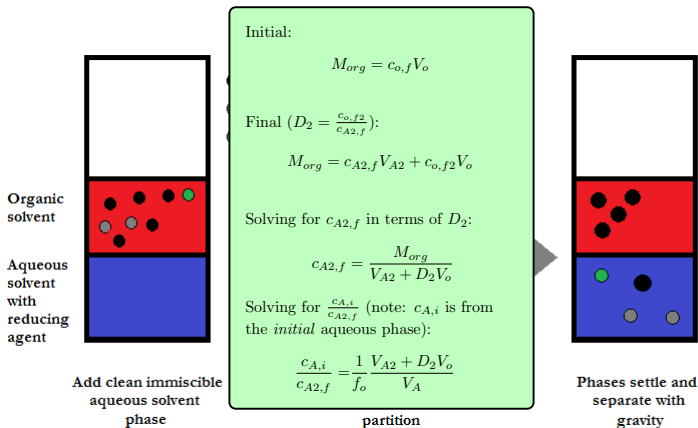
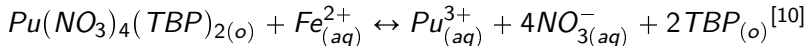
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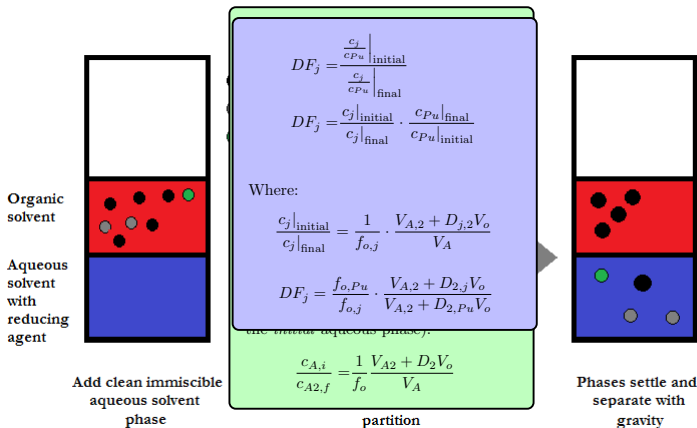
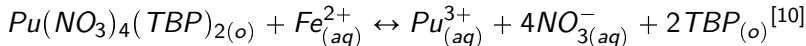
# Back-Extraction



# Back-Extraction

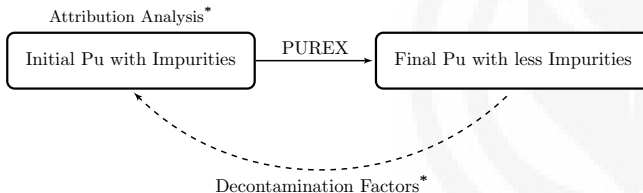


# Back-Extraction



## 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)<sup>[8, 9]</sup>





# Objectives I

## Characterize a 4 extraction 3 back-extraction PUREX process

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

## Objectives II

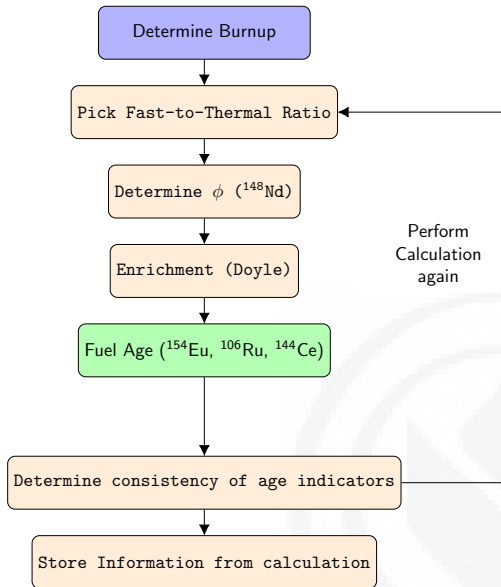
### Determine attribution indicators:

- Mathematically derive equations for above indicators with respect to one of the isotopes determined above
  - ✓  
□ Burnup
  - ✓  
□ Fluence Rate
  - ✓  
□ Initial Enrichment
  - ✓  
□ Fuel Age
    - 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<sup>[11]</sup>
- ❖ Determine burnup, enrichment, and fuel age from used fuel in a RDD<sup>[12]</sup>
- ❖ Analysis of purified plutonium isotopics for reactor type<sup>[13]</sup>
- ❖ PUREX co-processing DF values for U and Pu<sup>[14]</sup>
- ❖ PUREX D-values and DF values under numerous circumstances<sup>[9, 15, 16, 17, 18, 8]</sup>
  - DF values for  $^{106}\text{Ru}$  and  $^{95}\text{Zr}$ <sup>[8]</sup>
  - Compilation of D-values for U, Th, and Pu<sup>[19]</sup>
  - D-values for rare earths, Pu, Th<sup>[17, 20, 21, 22, 23, 24, 25, 26, 27, 28]</sup>
  - Ga D-values<sup>[28]</sup>

# Procedure



# Analytical Procedure

❖  $12.9 \pm 0.1$  mg of  $\text{DUO}_2$  irradiated at HFIR

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

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

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

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

# Analytical Procedure - Burnup

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

$$\begin{aligned}
 BU &= \frac{\text{Power}[\text{MW}] \cdot \text{days}}{m[HM]} \\
 &= \left[ \frac{N^B}{N_0^{HM}} \right] \frac{N_A E_R}{\gamma_B} \cdot \frac{1}{M_0^{HM}}
 \end{aligned}$$



# Back-Extraction

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

# Back-Extraction

$(n, \gamma)^{148}_{60}\text{Nd} \quad \beta^{-1} \rightarrow ^{147}_{61}\text{Pm}$  $^{147}_{60}\text{Nd}$  11 days  0.025 eV: 380 Epi: 630	$(n, \gamma)^{149}_{60}\text{Nd} \quad \text{Stable} - 5.8\%$  $^{148}_{60}\text{Nd}$  0.025 eV: 2.2 Epi: 14	
	$\beta^{-1} \rightarrow ^{147}_{60}\text{Nd}$  $^{147}_{59}\text{Pr}$  13.4 min	$\beta^{-1} \rightarrow ^{148}_{60}\text{Nd}$  $^{148}_{59}\text{Pr}$  2.29 min

# 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  $\phi$ , single group x-sections
- Constant fissionable material,  $\sigma_a$  for  $^{148}\text{Nd} \approx 0$
- Cumulative yield instantly arrives at  $^{147}\text{Nd}$
- Utilizes a Taylor series expansion

# Analytical Procedure - Initial Enrichment and Fuel Age

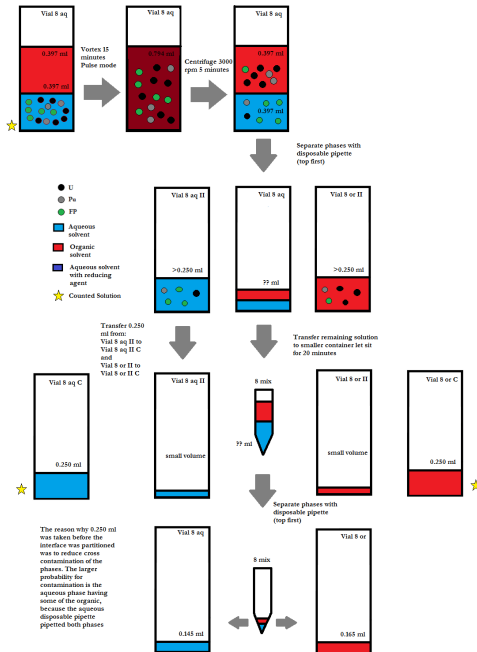
## Initial Enrichment

- ❖ 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  $^{238}\text{U}$

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

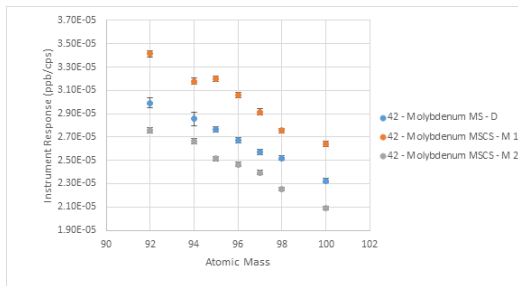
## Fuel Age

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



# Experimental Procedure - Mass Spectrometry

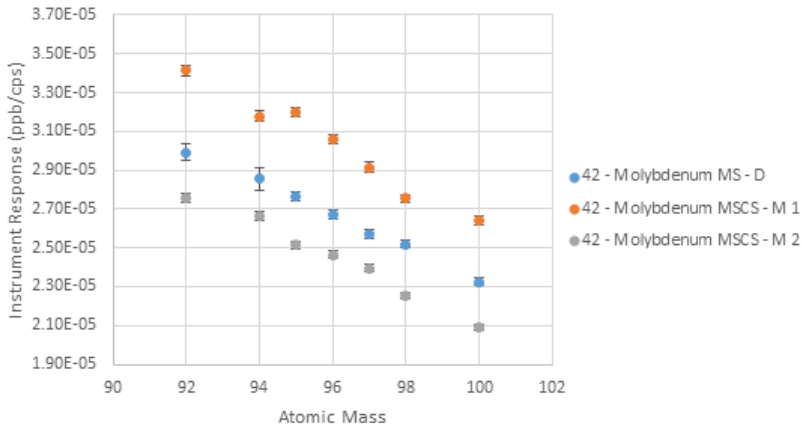
- ❖ NexION 300X quadrupole
- ❖ 7 different concoction standards (including Pu and U standards)
- ❖ Two point calibration curves
- ❖ Extrapolation for radioactive species



# Experimental Procedure - Mass Spectrometry

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

# Mass Spec





# Experimental Procedure - Gamma Spectrometry



# Experimental Procedure - Alpha Spectrometry



## Current and Expected Results

# Irradiation

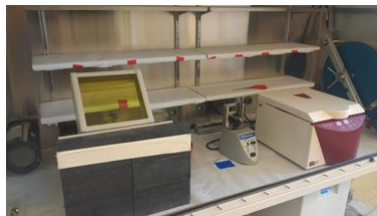
- ❖  $12.9 \pm 0.1$  mg of  $\text{DUO}_2$  was irradiated
  - High Flux Isotope Reactor at Oak Ridge National Laboratory
- ❖ Burnup was  $4.43 \pm 0.31$  GWd/tHM<sup>[29]</sup> from  $^{137}\text{Cs}$
- ❖  $0.196 \pm$  mg of total Pu was produced as measured by ICP-MS



## 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: 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

## 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	$(92.7 \pm 6.0)\%$	$(0.45 \pm 0.03)\%$



# Mass Spectrometry Results

## Decontamination Factors

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

# Gamma Spectroscopy Results



# Initial vs Final solutions



## Subsection 5

### Future Work

# Objectives I

## Characterize a 4 extraction 3 back-extraction PUREX process

- Collect D-values for each step
  - ✓ □  $^{144}\text{Ce}, ^{155}\text{Eu}^*, ^{154}\text{Eu}^*, ^{125}\text{Sb}, ^{106}\text{Ru}, ^{134}\text{Cs}, ^{137}\text{Cs}$  (Measured Gamma in triplicate)<sup>[5]</sup>
  - ✓ □ Convert Gamma Spectrum to D-values
  - ✓ □  $^{85}\text{Rb}^*, ^{90}\text{Sr}^*, ^{97,98,100}\text{Mo}, ^{101,102,104}\text{Ru}, ^{110}\text{Pd}, ^{112}\text{Cd}, ^{133}\text{Cs}^*, ^{140,142}\text{Ce}, ^{143}\text{Nd}^*, ^{147}\text{Pm}^*, ^{151}\text{Sm}^*, ^{154}\text{Eu}^*, \text{U}^*, \text{Pu}^*$  (Mass Spec)
  - Convert all mass spec data to D-values
- Collect DF-values for the process
  - ✓ □ Prepare alpha samples for each step (triplicate)
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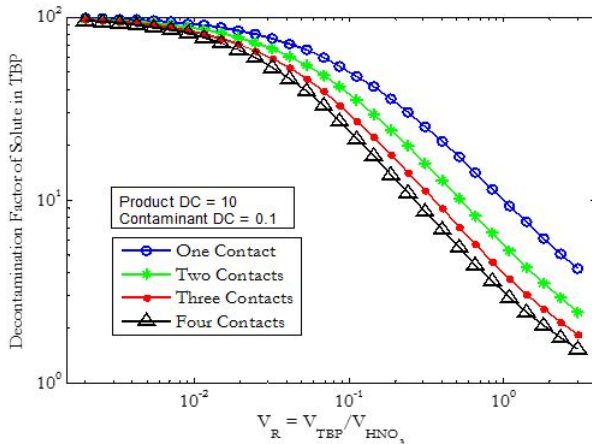
## Objectives II

### Determine attribution indicators:

- Mathematically derive equations for above indicators with respect to one of the isotopes determined above
  - ✓  
□ Burnup
  - ✓  
□ Fluence Rate
  - ✓  
□ Initial Enrichment
  - ✓  
□ Fuel Age
    - 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
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□ 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?

## Previous Experiment Results



Decontamination Factors for multi-contact extraction.



## References I

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