

## Experimental Characterization of Pu Separation by PUREX Process on a Low-Burnup, Pseudo-Fast-Neutron Irradiated DUO<sub>2</sub> for Product Decontamination Factors and Nuclear Forensics

A PhD. Prelims Defense by: Paul Mendoza

Chair: Dr. Sunil Chirayath

Committee Members: Dr. Sean McDeavitt  
Dr. Craig Marianno  
Dr. Charles M. Folden III

Monday, February 27, 2017, 10:00 am  
AIEN 304



**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]

# Definitions

- ❖ Special Nuclear Material (SNM)
  - Plutonium,  $^{233}\text{U}$ , or  $^{235}\text{U}$
- ❖ 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

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

# National Context

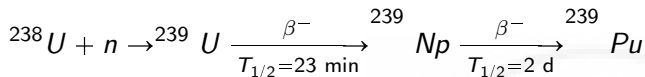
“The United States has developed a nuclear forensics capability that has been demonstrated in real-world incidents of **interdicted materials** and in exercises of actions required after a nuclear detonation. The committee, however, has concerns about the program and finds that without strong leadership,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

Major areas of concern include:

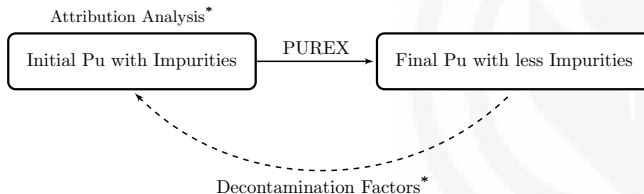
- ❖ Organization
- ❖ Sustainability
- ❖ **Workforce and Infrastructure**
- ❖ **Procedures and Tools (predetonation)**

# Forensic Context

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

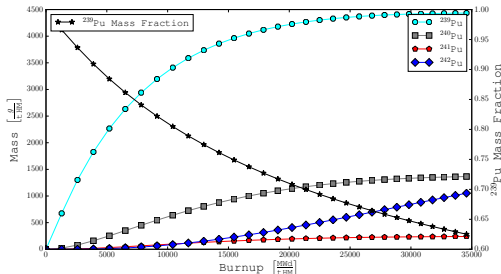


- ❖ Attribution for used or spent fuel has been previously studied [5, 6, 7]



# 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



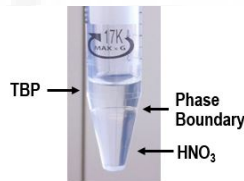
# Chemical Context

- ❖ 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)

$$D = \frac{C_o}{C_{aq}}$$

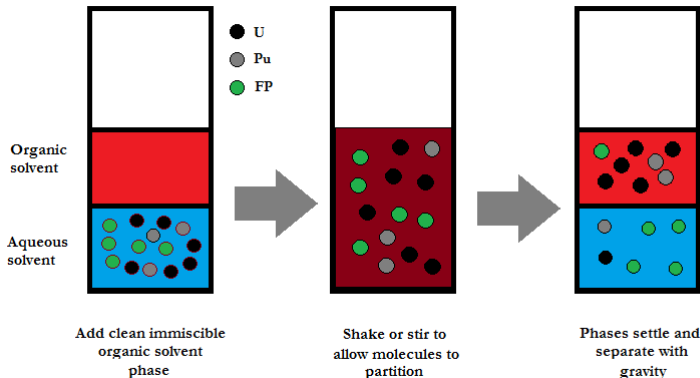
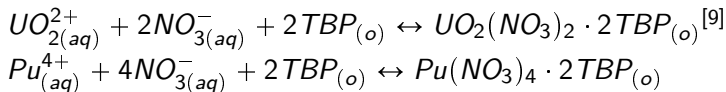
- Specific element to element
- Vary widely<sup>[8]</sup>
- The fraction of mass,  $f_o$  deposited in the organic phase, assuming a volume ratio between the aqueous and organic phases,  $V_R$ , is:

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

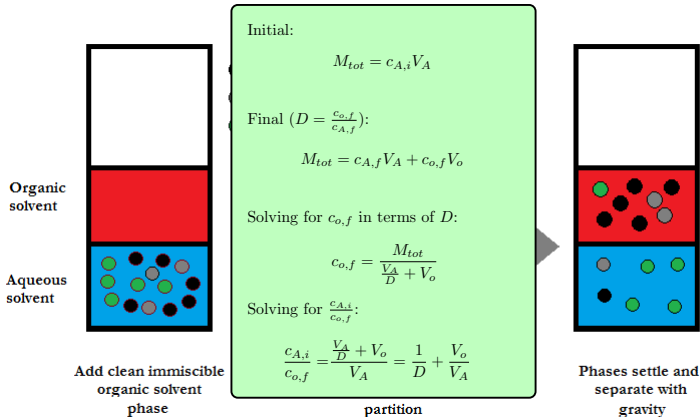
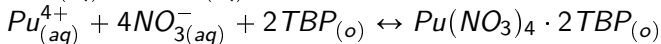
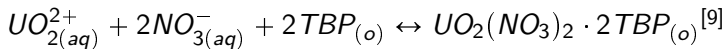




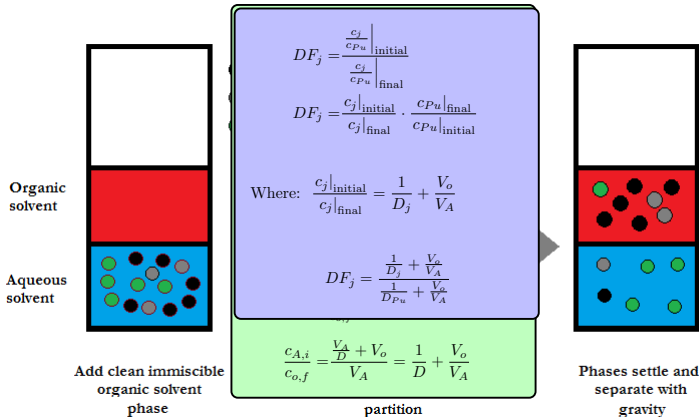
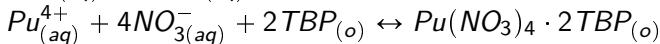
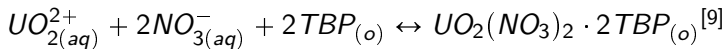
# Extraction



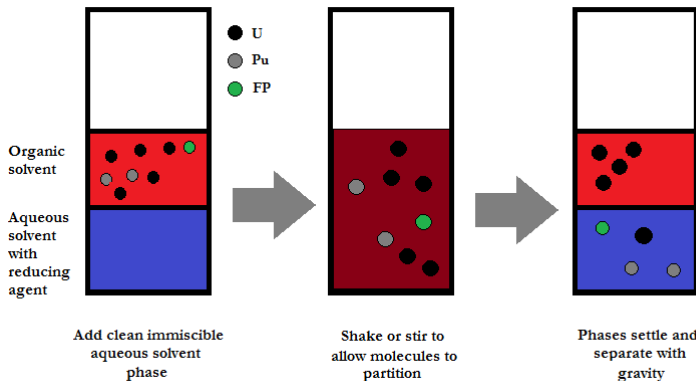
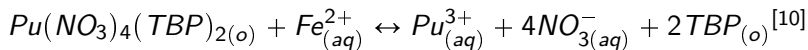
# Extraction



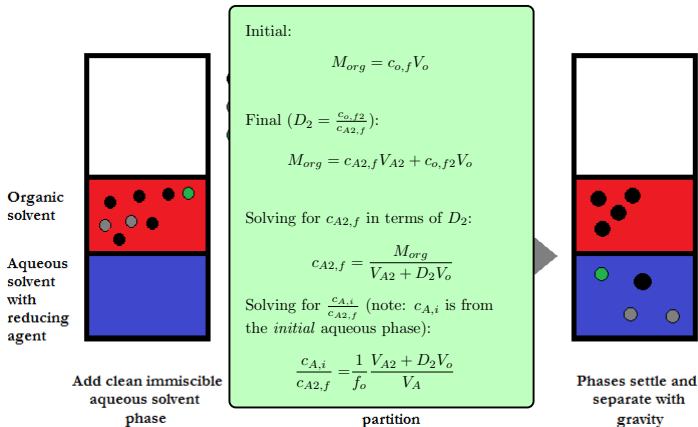
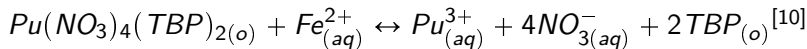
# Extraction



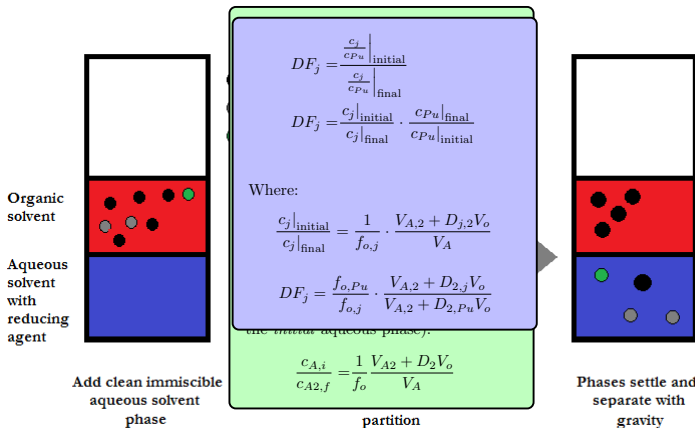
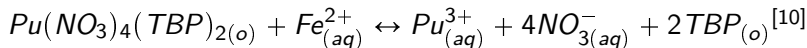
# Back-Extraction



# Back-Extraction

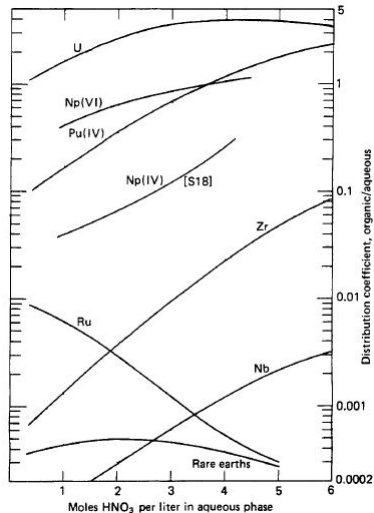


# Back-Extraction



# Chemical Context

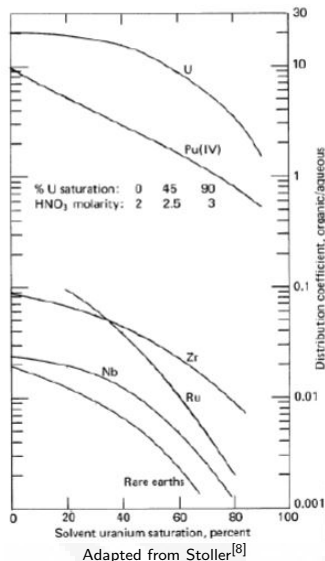
- ❖ Lack of literature on decontamination factors and distribution coefficients for useful forensic elements (Cs, Sb, Eu, Rb, Sr, Nd, Pm, and Sm)
- ❖ With a known process and D-values, DF values for individual elements can be determined



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

# Chemical Context

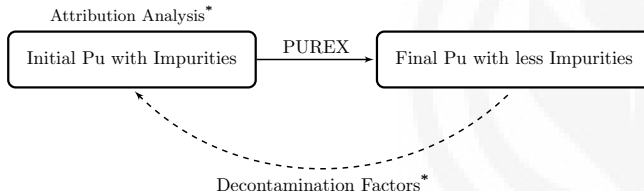
- ❖ Lack of literature on decontamination factors and distribution coefficients for useful forensic elements (Cs, Sb, Eu, Rb, Sr, Nd, Pm, and Sm)
- ❖ With a known process and D-values, DF values for individual elements can be determined





# 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 (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)
- Mathematically connect D-values to DFs
  - Derive equations with uncertainty propagation
  - Analyze connection with uncertainty

# Objectives II

## Determine attribution indicators:

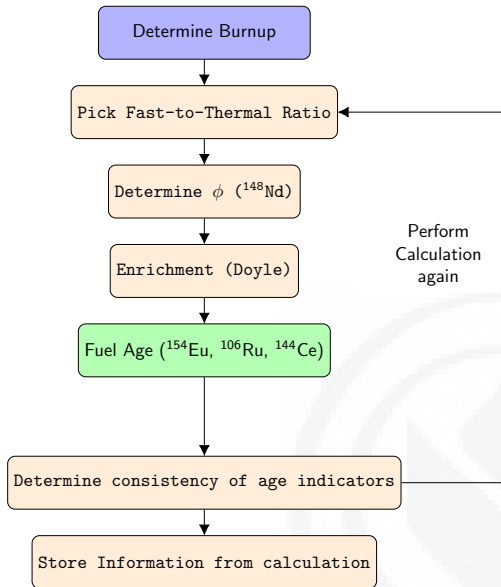
- Mathematically derive equations for above indicators with respect to one of the isotopes determined above
  - ✓ □ Burnup
  - ✓ □ Fluence Rate
  - ✓ □ Initial Enrichment
  - ✓ □ Decay Time
    - Fast-to-thermal ratios (requires iteration)
- Program a system to iteratively solve for these parameters 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<sup>[11]</sup>
- ❖ Determine burnup, enrichment, and decay time 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 <sup>106</sup>Ru and <sup>95</sup>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>
  - Gd D-values<sup>[28]</sup>

## Subsection 1

### Mathematical methodology



# 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**

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



# Analytical Procedure - Burnup

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

# Analytical Procedure - Burnup

$(n, \gamma)^{148}_{60}\text{Nd} \quad \beta^{-1} \rightarrow ^{147}_{61}\text{Pm}$  $^{147}_{60}\text{Nd}$ 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 yeild instantly arrives at  $^{147}\text{Nd}$
- 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  $^{238}\text{U}$

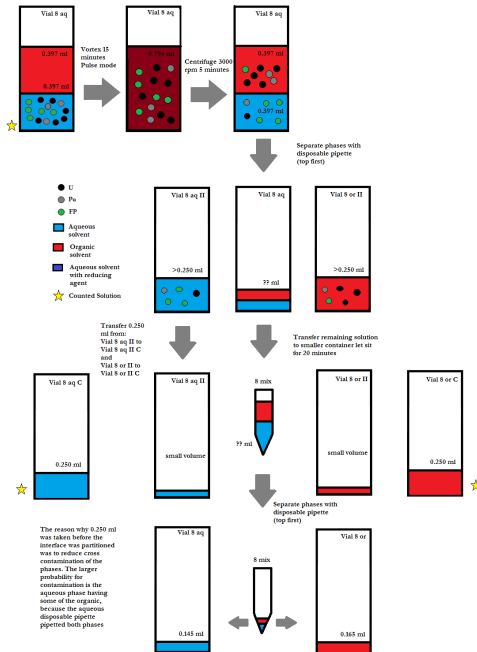
$$\epsilon_0 = \frac{N^{U238}(T)}{N_0^U} \left[ \frac{N^{U235}(T)}{N^{U238}(T)} + \frac{N^{U236}(T)}{N^{U238}(T)} \right] + \frac{M_0^U}{N_A E_R} BU(T) - G^{238} - G^{239} - G^{240} - G^{241}$$

## Decay Time

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

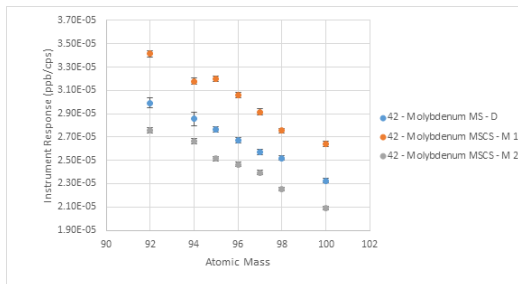
## Subsection 2

### Experimental Procedure



# Experimental Procedure - Mass Spectrometry

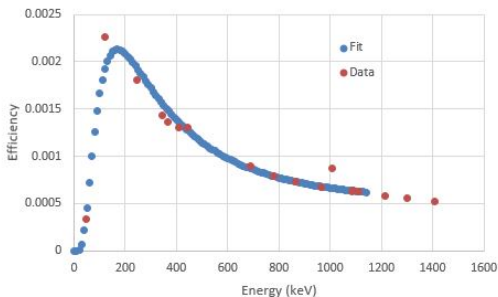
- ❖ NexION 300X quadrupole
- ❖ 7 different concoction standards (including Pu and U standards)
- ❖ Two point calibration curves
- ❖ Extrapolation for radioactive species



# Experimental Procedure - Gamma Spectrometry

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

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

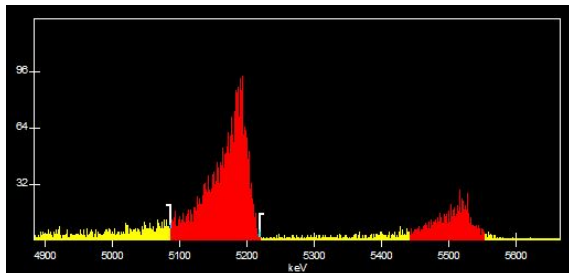




# Experimental Procedure - Alpha Spectrometry

- ❖ Passivated implanted planar silicon (PIPS) detector
- ❖ 4-peak Pu, Am, Gd, and Cm source from Eckert and Ziegler
- ❖ Samples prepared by evaporating 10-20  $\mu$ l on aluminum surface

$^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Am}$



## Current and Expected Results

# Irradiation

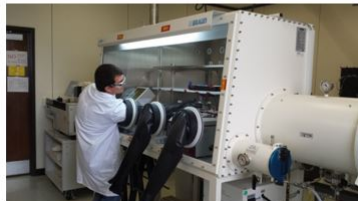
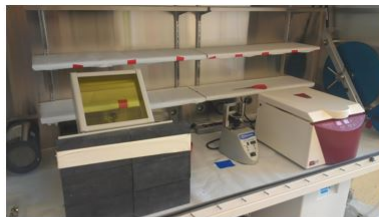
- ❖  $12.9 \pm 0.1$  mg of  $\text{DUO}_2$  was irradiated
  - High Flux Isotope Reactor at Oak Ridge National Laboratory
- ❖ Burnup was  $4.43 \pm 0.31$  GWd/tHM<sup>[29]</sup> from  $^{137}\text{Cs}$
- ❖  $0.196 \pm$  mg of total Pu was produced as measured by ICP-MS



## Dissolution of the spent fuel pellet



# Glovebox



# Experiments

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

- ❖ 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

## Recoveries of U and Pu

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

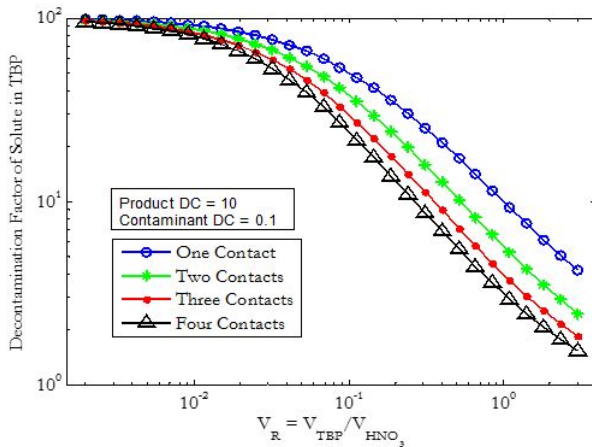
# Mass Spectrometry Results

## 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



## Previous Experiment Results



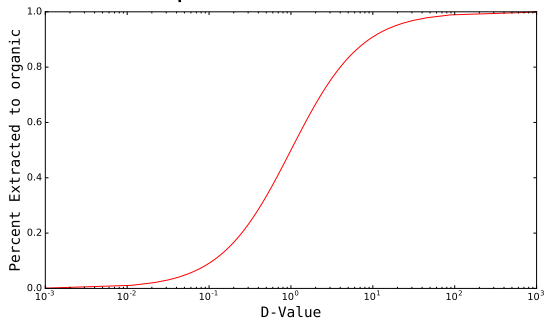
Decontamination Factors for multi-contact extraction.

## Gamma Spectroscopy Results: 4M HNO<sub>3</sub>, 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

### Equal Volume Extraction



## Initial vs Final solutions (process)

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

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

## Subsection 4

### Future Work

# Objectives I

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

- Collect D-values for each step
  - ✓  $^{144}\text{Ce}, ^{155}\text{Eu}^*, ^{154}\text{Eu}^*, ^{125}\text{Sb}, ^{106}\text{Ru}, ^{134}\text{Cs}, ^{137}\text{Cs}$  (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 (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)
- 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?

## References I

- [1] Dwight D Eisenhower. Atoms for peace speech. *Voices of Democracy*, 1953.
- [2] *Nuclear Forensics: A Capability at Risk (Abbreviated Version)*. The National Academies Press, Washington, DC, 2010.
- [3] AAAS/APS. Nuclear forensics: Role, state of the art and program needs. Report, AAAS/APS, 2008.
- [4] Kenton J Moody, Ian D Hutcheon, and Patrick M Grant. *Nuclear forensic analysis*. CRC Press, 2014.
- [5] 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 and Global Security*, 23(1):48–67, 2015.
- [6] Mark Robert Scott. *Nuclear forensics: attributing the source of spent fuel used in an RDD event*. PhD thesis, Texas A and M University, 2005.
- [7] 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.



## References II

- [8] Sidney M Stoller, Walter Henry Zinn, Stuart MacLain, and Atomic Energy Commission USA. *Reactor handbook. 2. Fuel reprocessing*. Interscience Publ., 1961.
- [9] M Benedict, H Levi, and T Pigford. Nuclear chemical engineering. *Nucl. Sci. Eng.:(United States)*, 82(4), 1982.
- [10] 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.
- [11] William S Charlton, Bryan L Fearey, Charles W Nakhleh, Theodore A Parish, Robert T Perry, Jane Poths, John R Quagliano, William D Stanbro, and William B Wilson. Operator declaration verification technique for spent fuel at reprocessing facilities. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 168(1):98–108, 2000.
- [12] Mark Robert Scott. *Nuclear forensics: attributing the source of spent fuel used in an RDD event*. Thesis, 2005.
- [13] 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.

## References III

- [14] Okan H Zabunoglu and Levent Ozdemir. Purex co-processing of spent lwr fuels: flow sheet. *Annals of Nuclear Energy*, 32(2):151–162, 2005.
- [15] W. B. Lanham and A. T. Gresky. Purex process laboratory development. *Oak Ridge National Laboratory*, USAEC Report ORNL-717, 1950.
- [16] A. J. Arker. Terminal report on purex program in kapl separations pilot plant. *Knolls Atomic Power Laboratory*, 1954.
- [17] D. O. Darby and J. M. Chandler. Terminal report for the ornl pilot plant investigation of the purex process. *Oak Ridge National Laboratory*, USAEC Report ORNL -1519, 1954.
- [18] E.R. Irish and Reas W.H. The purex process-a solvent extraction reprocessing method for irradiated uranium. 1957.
- [19] Thomas H. Siddall, W. E. Prout, Sidney G. Parker, U. S. Atomic Energy Commission, Laboratory Savannah River, E. I. du Pont de Nemours, and Company. *Equilibrium distribution data for purex and similar extraction processes*. DP ; 53. E.I. du Pont de Nemours and Co., Explosives Dept., Atomic Energy Division, Technical Division, Savannah River Laboratory, 1957.

## References IV

- [20] K Alcock, FC Bedford, WH Hardwick, and HAC McKay. Tri-n-butyl phosphate as an extracting solvent for inorganic nitrates-i: Zirconium nitrate. *Journal of Inorganic and Nuclear Chemistry*, 4(2):100–105, 1957.
- [21] K Alcock, FC Bedford, WH Hardwick, and HAC McKay. Tri-n-butyl phosphate as an extracting solvent for inorganic nitrates-i: Zirconium nitrate. *Journal of Inorganic and Nuclear Chemistry*, 4(2):100–105, 1957.
- [22] K Alcock, GF Best, E Hesford, and HAC McKay. Tri-n-butyl phosphate as an extracting solvent for inorganic nitrates-v: Further results for the tetra- and hexavalent actinide nitrates. *Journal of Inorganic and Nuclear Chemistry*, 6(4):328–333, 1958.
- [23] GF Best, E Hesford, and HAC McKay. Tri-n-butyl phosphate as an extracting agent for inorganic nitrates-vii: The trivalent actinide nitrates. *Journal of Inorganic and Nuclear Chemistry*, 12(1):136–140, 1959.
- [24] GF Best, HAC McKay, and PR Woodgate. Tri-n-butyl phosphate as an extracting solvent for inorganic nitrates—iii the plutonium nitrates. *Journal of Inorganic and Nuclear Chemistry*, 4(5):315–320, 1957.

## References V

- [25] E Hesford, EE Jackson, and HAC McKay. Tri-n-butyl phosphate as an extracting agent for inorganic nitrates—vi further results for the rare earth nitrates. *Journal of Inorganic and Nuclear Chemistry*, 9(3-4):279–289, 1959.
- [26] E Hesford, HAC McKay, and D Scargill. Tri-n-butyl phosphate as an extracting solvent for inorganic nitrates—iv thorium nitrate. *Journal of Inorganic and Nuclear Chemistry*, 4(5):321–325, 1957.
- [27] D Scargill, K Alcock, JM Fletcher, E Hesford, and HAC McKay. Tri-n-butyl phosphate as an extracting solvent for inorganic nitrates-ii yttrium and the lower lanthanide nitrates. *Journal of Inorganic and Nuclear Chemistry*, 4(5):304–314, 1957.
- [28] ED Collins, DO Campbell, and LK Felker. Measurement of achievable plutonium decontamination from gallium by means of purex solvent extraction. *ORNL/TM-1999/312*, Oak Ridge National Laboratory, 2000.
- [29] Mathew Wayne Swinney. *Experimental and Computational Assessment of Trace Nuclide Ratios in Weapons Grade Plutonium for Nuclear Forensics Analysis*. PhD thesis, 2015.
- [30] Kenneth D Kok. *Nuclear engineering handbook*, volume 60. CRC Press, 2009.