Rapid Isolation of Neptunium from Solution and Soil

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Outline

- Background
- Rainwater Analogues
  - Preparation
  - Yields
- Soil Samples
  - Preparation
  - Yields
- Best Methods
- Further Steps

Value of Actinides

- Chernobyl
  - 2.6 PBq Pu
  - 1.5 ± 0.5% nuclear fuel inventory
- Fukushima
  - 2.7×10^{-4} PBq Pu
  - How much of its nuclear fuel inventory?
- Determine severity of nuclear accident using actinides

If using Pu…

- Isotopic ratios
  - $^{240}Pu/^{239}Pu$
  - $^{238}Pu/^{240+239}Pu$
- Sources
  - Pu-powered reactors (0.4-0.6, <0.1)
  - Global fallout (0.1-0.2, <0.1)
  - Weapons grade (<0.1, <0.1)
  - Naval reactors (<0.2, 0.1-0.45)
  - Civil reactor waste (0.2-0.6, 0.2-0.6)
- Will the release of plutonium significantly change the existing environmental ratio?

If using Pu…

- Alpha energies:
  - ^{238}Pu ($T_{1/2}$ = 87.74 yrs): 5.50 MeV
  - ^{239}Pu ($T_{1/2}$ = 2.4 x 10^4 yrs): 5.16 MeV
  - ^{240}Pu ($T_{1/2}$ = 6563 yrs): 5.17 MeV
- Mass spectrometry for $^{240}Pu/^{239}Pu$
- Consider sample preparation

If using ^{239}Np…

- Short-lived ($T_{1/2}$ = 2.4 d)
- Emits β^- and γ
  - E_γ = 106 keV (26.3%), 228 keV (11.1%), 278 keV (14.4%)
  - Rapid detection by gamma spectroscopy
- Must consider spectral interferences
  - ^{131}I
  - Radioactive Tellurium

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Motivation

- K. Shozugawa et al.
  - 106 keV, 278 keV, 121 keV (Pu X-ray)
- Attempt isolation using Eichrom® extraction chromatography resins.

Sample Prep: Radionuclides

- Neutron Activation at USGS TRIGA® Reactor

Sample	Quantity	Irradiation Position	Irradiation Time	Cooling Time
$CsNO_3$	50 mg Cs	Reflector Tube	2 h	48 h
$UO_2(NO_3)_2 \cdot 6H_2O$	20 mg depleted U	Reflector Tube	2 h	48 h
$Te(OH)_6 \cdot 2H_2O$	30 mg Te	Rotary Rack	12 h	5 d

- 1 mg/mL solutions

Nuclide	Energy (keV)	CPS in 100 µL	Activity (Bq/100µL)
^{123m}Te	159	9.5±3.7	93±37
^{239}Np	278	15.3±2.3	1244±187
^{134}Cs	605	6.0±1.4	132±30

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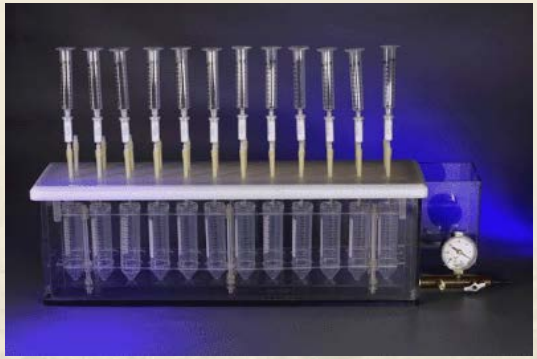
Rainwater Analogues

- Samples:
  - ~3 cps ^{239}Np
  - ~150 cps ^{134}Cs
  - ~100 cps ^{123m}Te

- Vacuum Box
- Resins
  - Actinide
  - RE
  - TRU
  - UTEVA



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Sample Prep: Rainwater Analogues

Resin	Ascorbic Acid 1.5 M (mL)	5 mg/mL Fe(II) (mL)	1 mg/mL U (Np) (µL)	Loading Solution (20 mL)	1 mg/mL Te (mL)	1 mg/mL Cs (mL)
Actinide	1.25	0.1	20	0.1 M HCl	1	2.5
RE	1.25	0.1	20	8 M HNO_3	1	2.5
TRU	1.25	0.1	20	3 M HNO_3	1	2.5
UTEVA	1.25	0.1	20	10 M HCl	1	2.5

- Samples run in quadruplicate (trials I – IV)
- Day 4
  - Use 500 mL for III and IV
  - Test different eluents
    - UTEVA[HNO_3]
    - UTEVA[H_2O]
    - RE[H_2O]
    - TRU[H_2O]

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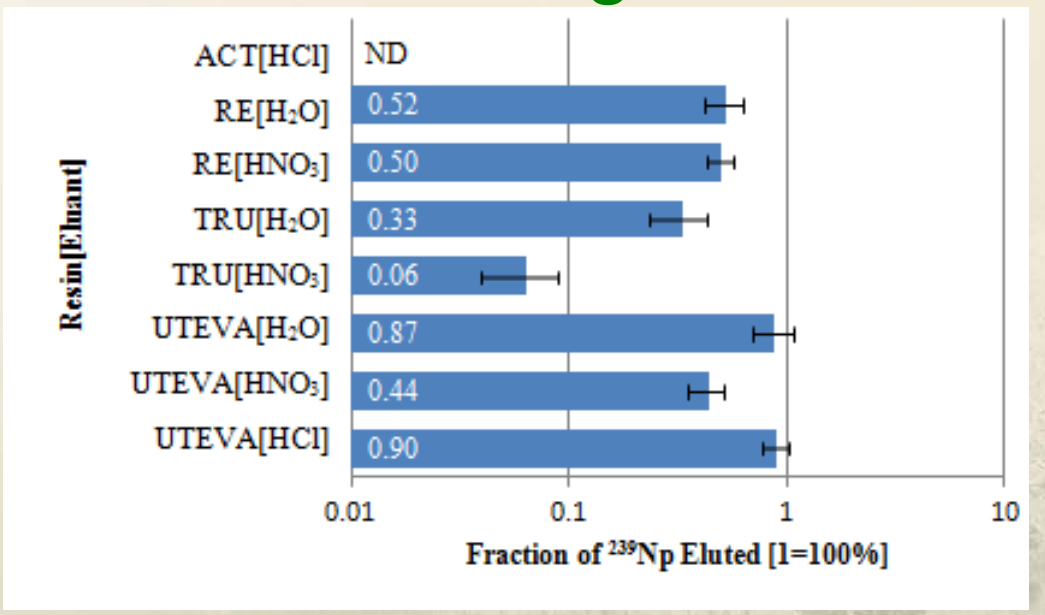
Fractional Yield:

- From Rainwater Analogues:



- ^{131}I retained on the columns! (~100 cps)

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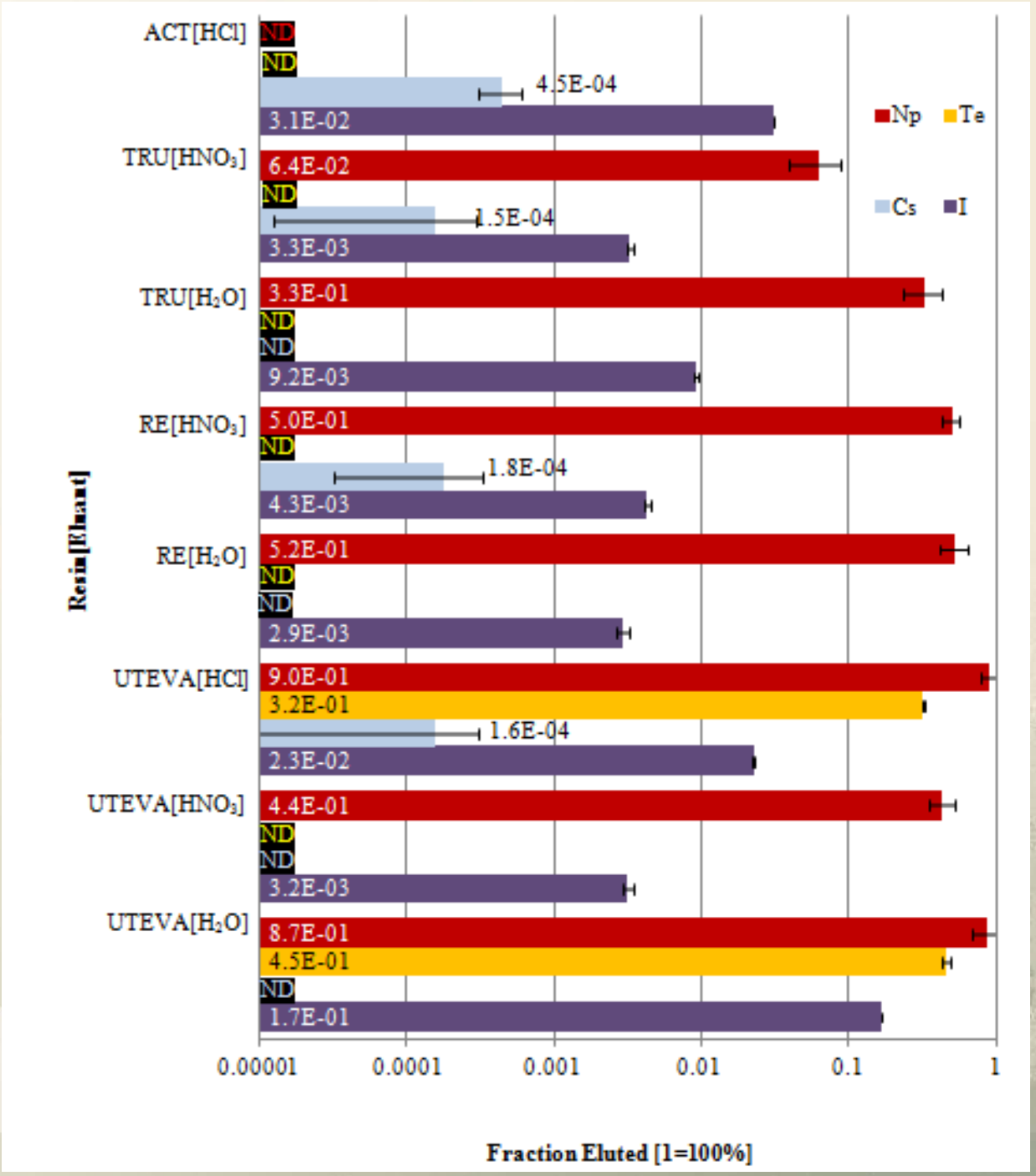
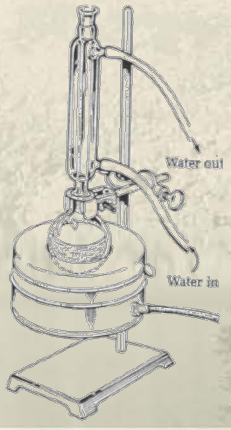
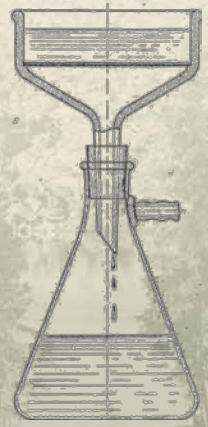

ACT[HCl]
TRU[HNO3]
TRU[H2O]
RE[HNO3]
RE[H2O]
UTEVA[HCl]
UTEVA[HNO3]
UTEVA[H2O]
Resin [Eluant]
ND
4.5E-04
3.1E-02
6.4E-02
1.5E-04
3.3E-03
3.3E-01
9.2E-03
5.0E-01
1.8E-04
4.3E-03
5.2E-01
2.9E-03
9.0E-01
3.2E-01
1.6E-04
2.3E-02
4.4E-01
3.2E-03
8.7E-01
4.5E-01
1.7E-01
Np
Te
Cs
I
0.00001
0.0001
0.001
0.01
0.1
1
Fraction Eluted [1=100%]

Sample Prep: Soil

- Dried overnight
- Spiked with 20 µL U (Np) Solution
- Refluxed (8 M HNO_3) and Filtered
- Added Ascorbic Acid and Fe(II)
- Eluted with Acetone

Sample	Mass (g)	Resin
SoI	5.86	RE
SoII	5.80	RE
SoIII	8.74	UTEVA
SoIV	6.66	UTEVA





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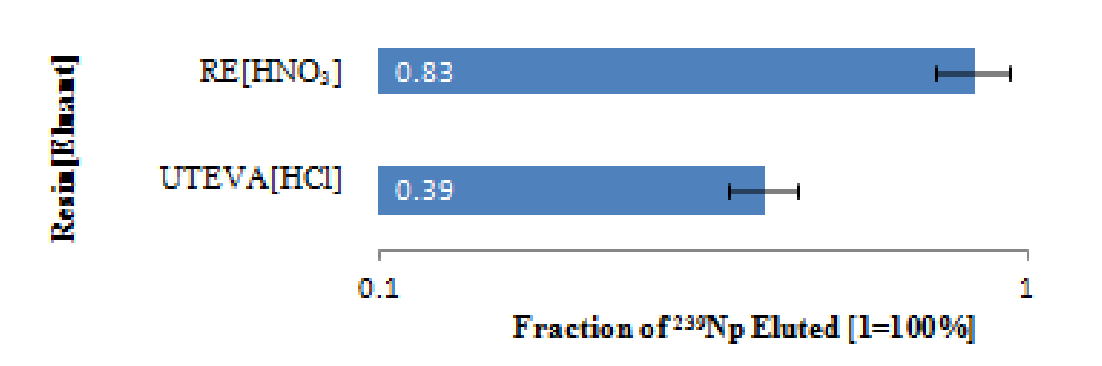
Fractional Yield:

- From Soil Samples:



Sample	Mass (g)	Resin
SoI	5.86	RE
SoII	5.80	RE
SoIII	8.74	UTEVA
SoIV	6.66	UTEVA

  - More soil mass used for UTEVA; less yield

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Best Methods:

- Aqueous Samples:
  - UTEVA[HNO_3]
  - RE[H_2O]
  - TRU[H_2O]
- Soil Samples:
  - Acetone may be required
  - Be wary of iodine

Further Steps:

- Assess TEVA Resin for activation/fission product retention
- Why is acetone required for soil samples?
  - Potentially no effect on Te and Cs retention
  - Must test using neptunium with volatile radionuclides
  - Dependent on soil type?

References

- Steinhauser, Georg; Brandl, Alexander; Johnson, Thomas, "Comparison of the Chernobyl and Fukushima nuclear accidents: A review of the environmental impacts," *Science of the Total Environment,* vol. 470-471, pp. 800-817, 2014.
- V. A. Kashparov, S. M. Lundin, S. I. Zvarych, V. I. Yoshchenko, S. E. Levchuk, Y. V. Khomutinin, I. M. Maloshtan and V. P. Protsak, "Territory contamination with the radionuclides representing the fuel component of Chernobyl fallout," *Science of The Total Environment,* vol. 317, pp. 105-119, 2003.
- S. Cagno, K. Hellemans, O. C. Lind, L. Skipperud, K. Janssens and B. Salbu, "LA-ICP-MS for Pu source identification at Mayak PA, the Urals, Russia," *Environmental Science: Processes & Impacts,* vol. 16, pp. 306-312, 2014.

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