INCORPORATION OF EICHROM TECHNOLOGY ANALYTICAL RESINS IN ORNL'S PU-238 PRODUCTION DEMONSTRATION

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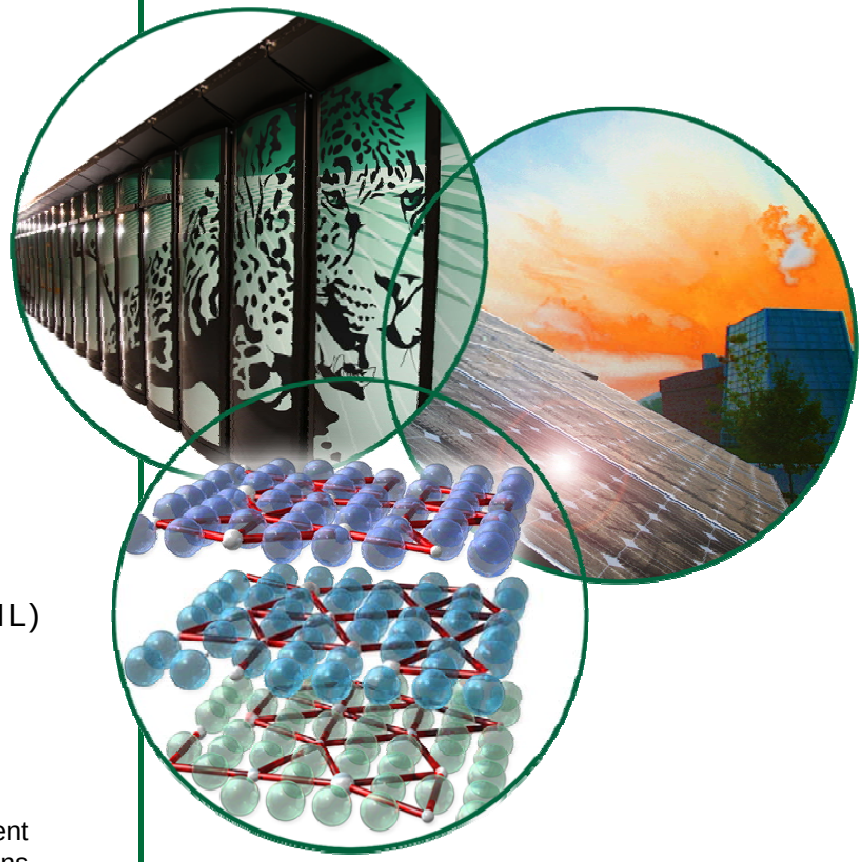

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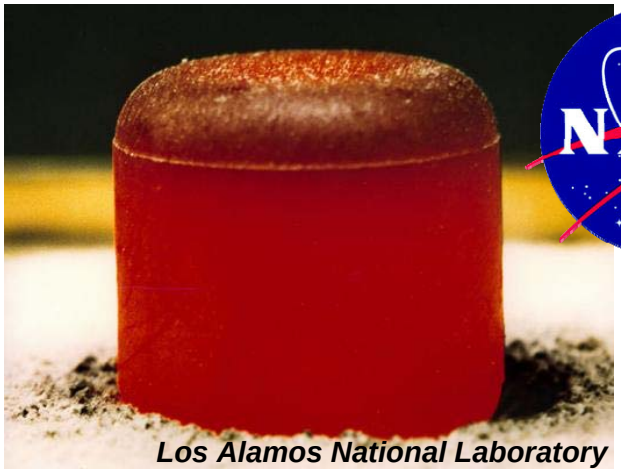
Background: Plutonium-238 is Essential to NASA Missions



Los Alamos National Laboratory

"RTGs provided by DOE have enabled American scientists to explore the solar system for many years….

Apollo missions…, the **Viking missions** to Mars, the **Pioneer, Voyager, Ulysses, Galileo and Cassini** missions….

all used this safe, efficient and long-lasting power source." **www.energy.gov**


www.NASA.gov

Mars Science Laboratory Curiosity:
8 general-purpose heat source modules, 4.8 kg of PuO_2


http://solarsystem.nasa.gov/multimedia/gallery/PIA17036.jpg

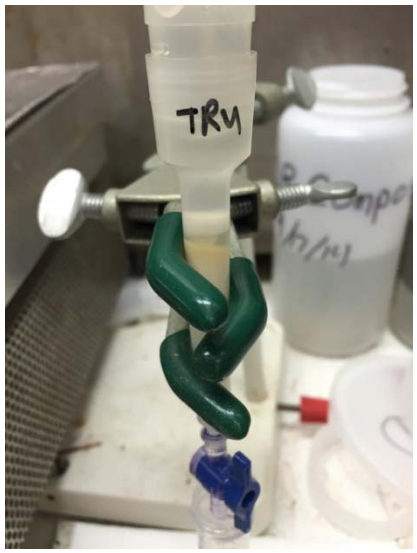
Launched in 1977, Voyager 1 is currently 1.91×10^{10} km from Earth

Surrogate solution for “cold” testing of pilot-scale chemical processing

- Np solutions produced from project stock of NpO2
- Legacy PuBe neutron sources currently being deinventoried in Building 7920 hotcell bank
- ~60 grams of Pu-238 in the REDC inventory from this process and is available for use to test pilot scale chemical processing for Np and Pu separation and recoveries
- Analytical testing required to ensure Be is reduced to acceptable levels before Pu-238 material is approved for this use
- Project required detection limits are low enough to require reduction of the Pu-238 alpha activity before ICPMS

Beryllium Impurity Analysis Bench Tests

Mixed valence Pu sample reduced to Pu(III) (left) followed by oxidation to Pu(IV) (right) for optimum retention



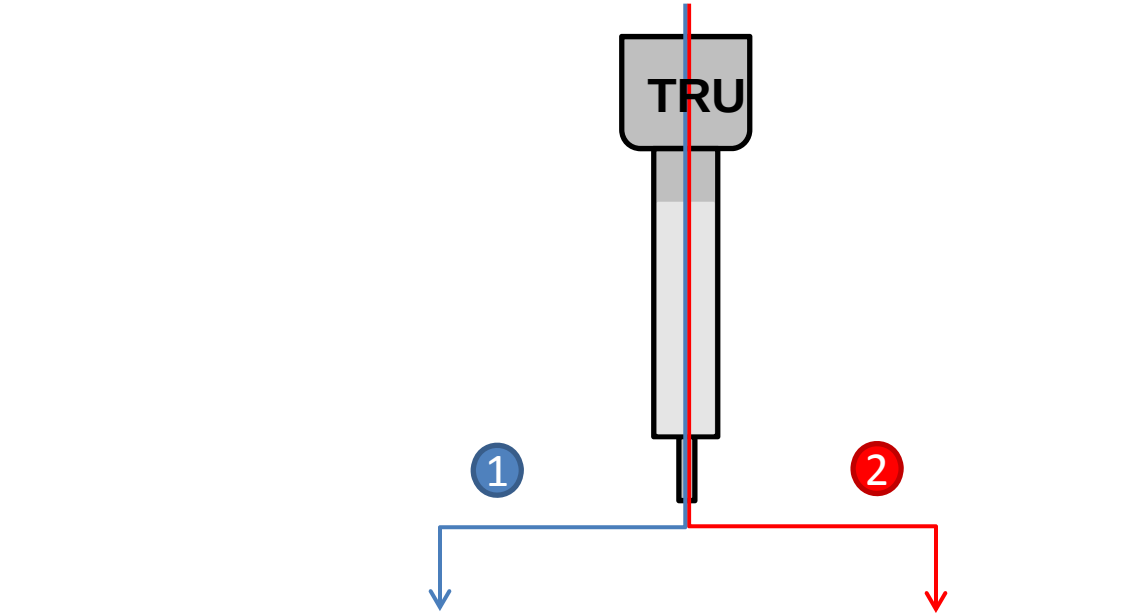
Method development using:

- SNM-334 (99.86% Pu-242) 1 mg/mL
- 10 µg/mL beryllium standard
- Eichrom TRU® resins

Sample ID	Measured counts for Be in sample	Recovery of Be Spike (%)	Decontamination Gross Alpha Activity (%)
COLUMN_1	9737.528	100.4	99.98
COLUMN _2	9944.635	102.6	>99.99
COLUMN _3	9755.345	100.6	99.97
COLUMN _4	9786.355	100.9	>99.99
COLUMN _5	9908.461	102.2	>99.99

Plutonium From Trace Impurities

Plutonium (IV) loaded

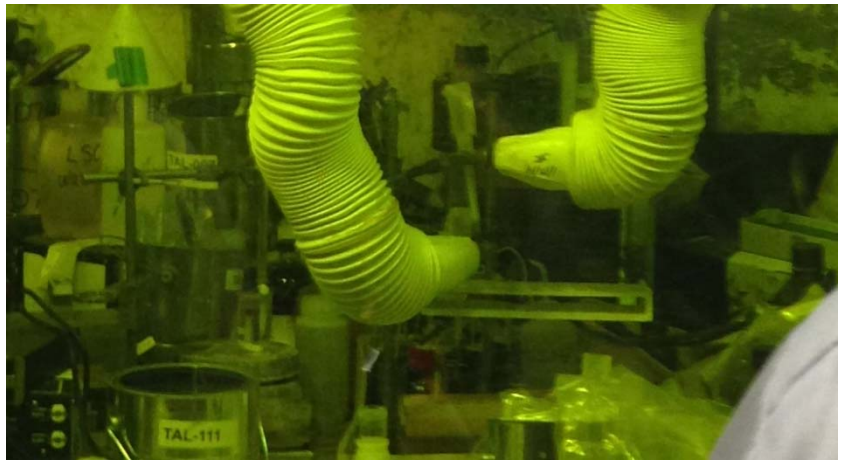


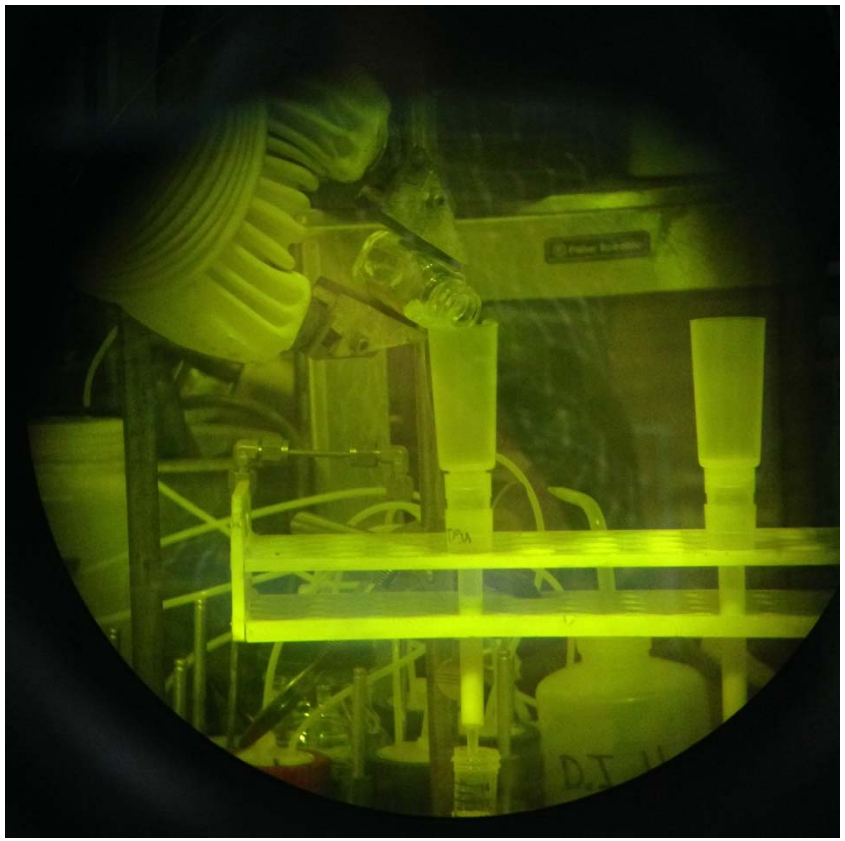
Impurities in the
+1 and +2 Oxidation state (+T-metals in +3)
removed in 8N HNO_3 + 0.01N $NaNO_2$

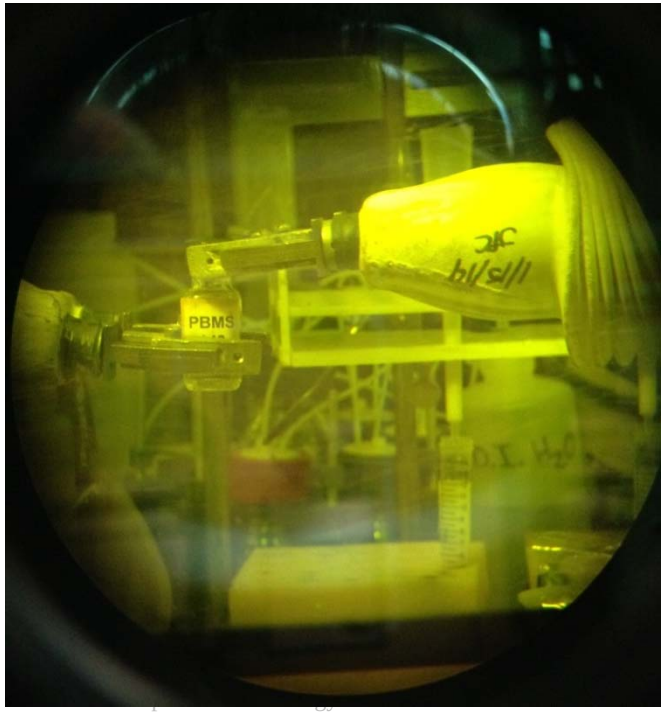
Plutonium (III) stripped
in 1N $NH_2OH.HNO_3$
and 50mM HNO_3

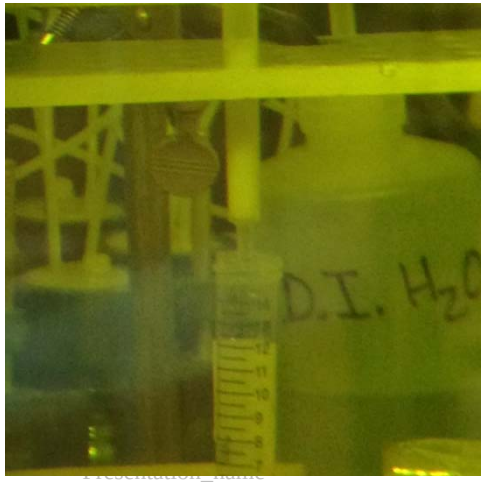
Quantitative separation of plutonium

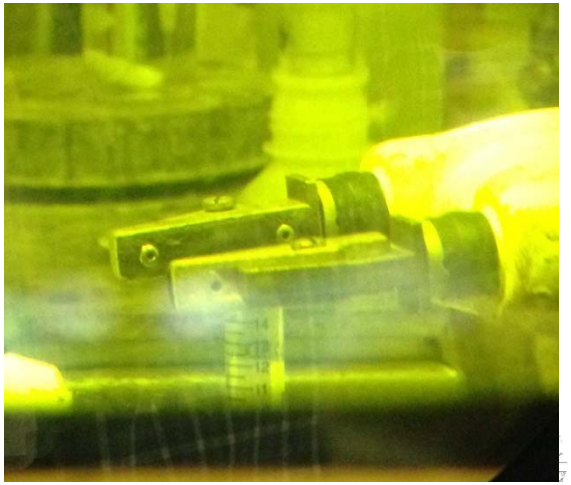
Hot-cell Resin Separation of Pu-238 for Beryllium and Impurities











Hot-cell Resin Separation of Pu-238 for Beryllium

- **Pu-238 stock solutions from the PuBe sources was purified using an anion resin**
- **Pu-238 material was analyzed for trace Be post-column using Eichrom TRU® resins**

	PBMS-141B		PBMS-142B		PBMS-143B		10ppm Spike - recovery
	Result (ug/L)	+/-	Result (ug/L)	+/-	Result (ug/L)	+/-	
9Be	**1706**	170.6	**2940**	294	**10.68**	2.67	96.3%
24Mg	1667	166.7	1483.6	148.36	581.4	58.14	103.6%
52Cr	540.2	54.02	37960	3796	301.6	30.16	90.3%
59Co	31.3	3.13	4962	496.2	2.58	0.516	96.4%
60Ni	676.1	67.61	204600	20460	81.74	8.174	93.8%
66Zn	2411	482.2	3412	682.4	597.4	119.48	99.4%

Hot-cell Resin Separation of Pu-238

Activity reduction (Becquerel)
5×10^{9} - 1×10^{10} → $5X10^{3}$-$1X10^{4}$*

Determined via Gross Alpha

*Remaining Activity due Rn-224 and daughters potentially from the Pu-236 decay chain
(Pu(IV)-236→U(VI)-232→Th(IV)-228→**Ra(II)-224(3.6d)→Rn-220(55s)→Po-216(0.14s)**)

Plutonium-238 reduction
~99.9999%¥

Determined via Gross Alpha + Alpha spectrometry

¥Observable plutonium reduction greater than for Pu-242 due to the much higher specific activity of Pu-238

Hot-cell Resin Separation of Pu-238

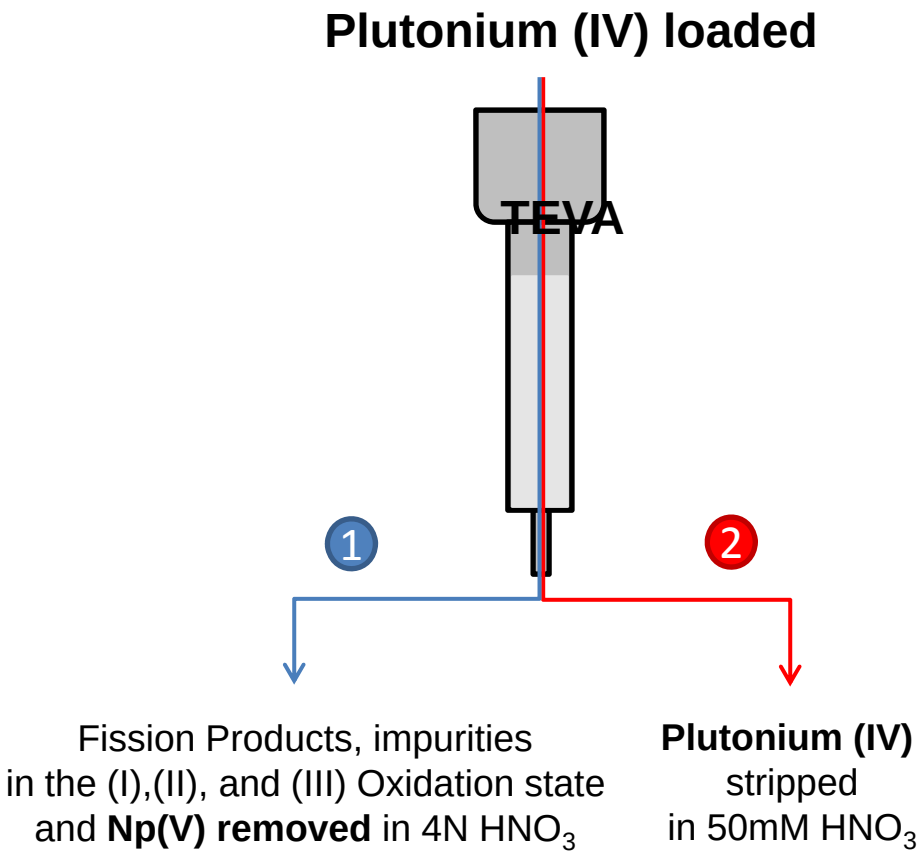
- **Analytical tests determined that a second cleanup of the stock solution was required**
- **Pu-238 batches were reanalyzed for Be post-column using Eichrom TRU® resins**

Sample	result (ng/ml)	analysis dilution	Prep dilution	[Be] (ug/L)*	SPIKE recovery
Process Blank	0.0011	10	10	0.11	
Process Blank #2	0.0005	10	10	0.05	
PBMS-141B	0.0025	100	20	5	
PBMS-142B	0.0015	100	20	3	
PBMS-143B	0.2021	100	20	404.2	
10ppm LCS					94%

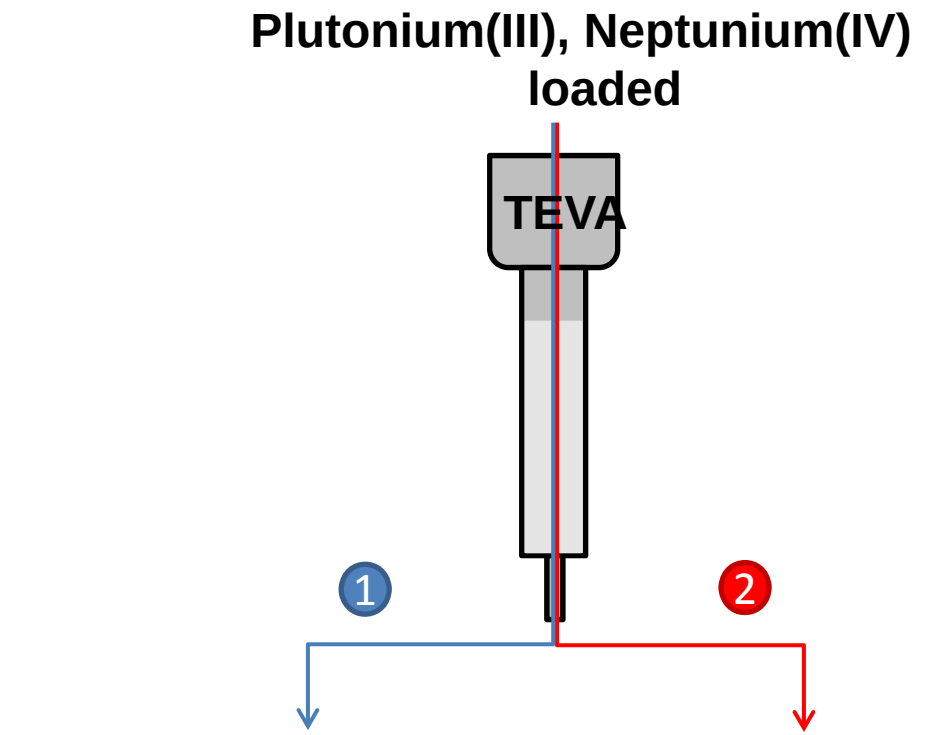
*PQL 5ug/L

This separation procedure will be used for qualification of the final Pu-238 product for trace impurities during production phase of the project

Plutonium from Fission Products, Impurities, and Neptunium



Neptunium From Fission Products, Impurities, and Plutonium



~99.9% reduction in plutonium

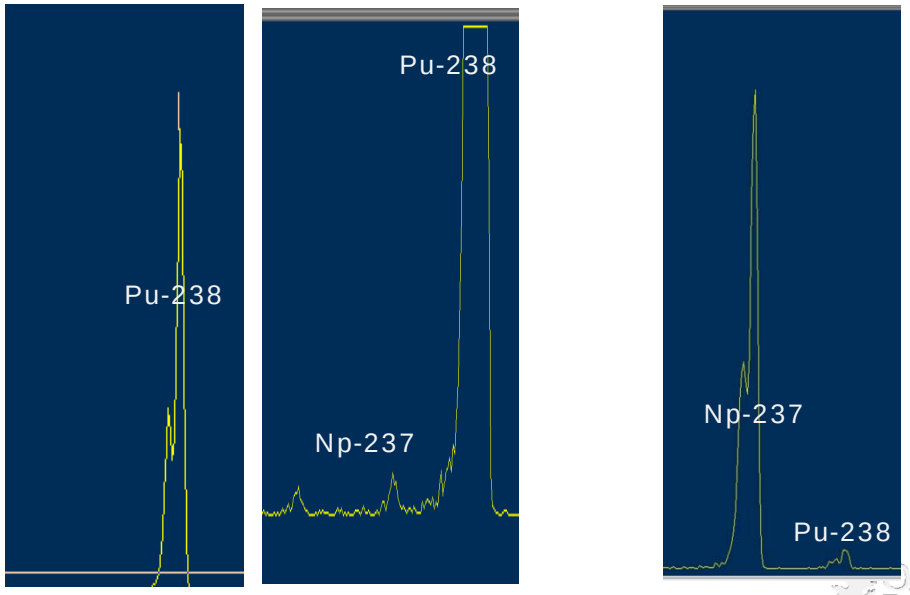
Neptunium From Fission Products, Impurities, and Plutonium

Using an irradiated Np-237 pellet dissolution

Measurement		Pre-column	Post-column
G-ALPHA		3.2E+10	1.1E+07
4.80 MeV (Np-237)		**0.1**	**99.6**
5.15 MeV		0.1	N/A
5.50 MeV (Pu-238)		**99.8**	**0.4**
5.80 MeV		TRACE	N/A

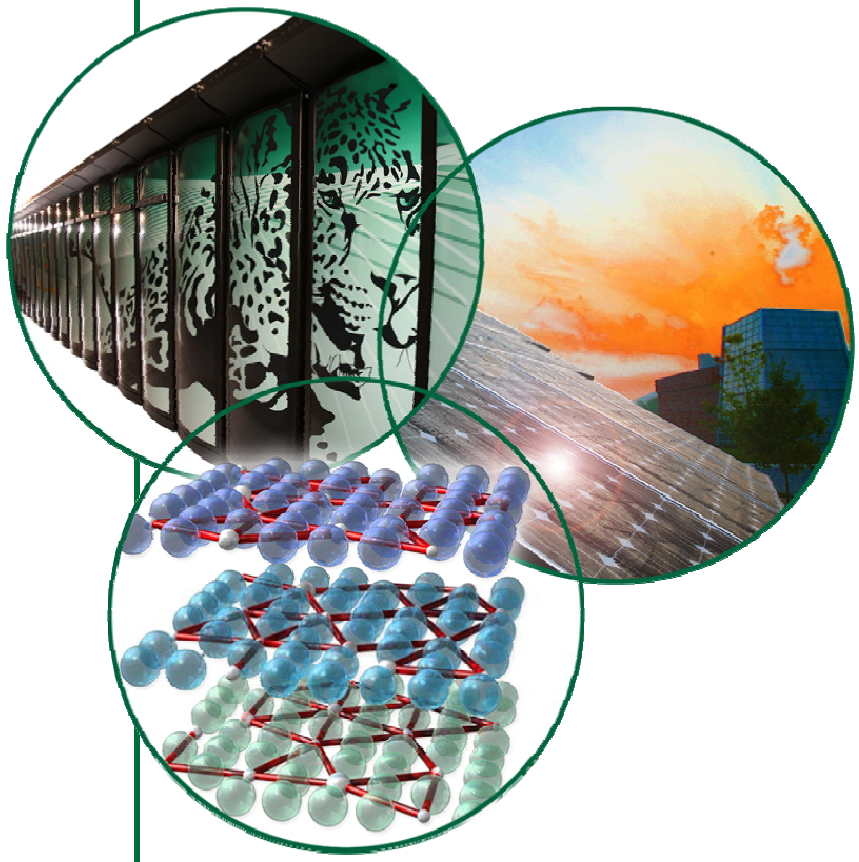
- **99.97% reduction in activity**
- **>99.9998% reduction in Pu-238**
- **Quality Assurance standards >95% Neptunium recovery**

The majority of the dose from these systems stems from the fission products present in the I, II, and III oxidation states which are also removed



Acknowledgments

All members of the Nuclear Analytical Chemistry and Isotopics Laboratories (NACIL) group at ORNL



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