



Actinide Science for Post Detonation Nuclear Forensic Analyses

March 2023

Changing the World's Energy Future

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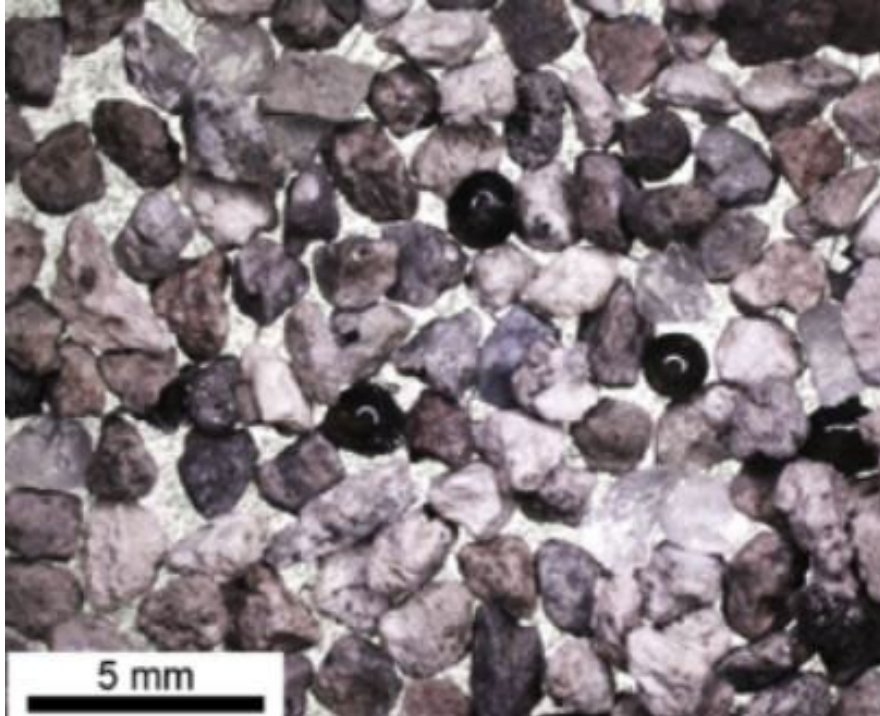
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March 20, 2023

TMS 2023, San Diego, Ca

Producing training simulants for emergency response/nuclear forensics teams



- Spherical glass matrix
- Known radioactive isotopic composition
 - altered from fission spectrum

Simulants – interlaboratory measurements and training and method

– Debris characteristics

- Short-lived fission products ($T_{1/2} < 48$ hours)
- Actinides
- Activation products
- Natural soil background
- Particle size
- Color/luster

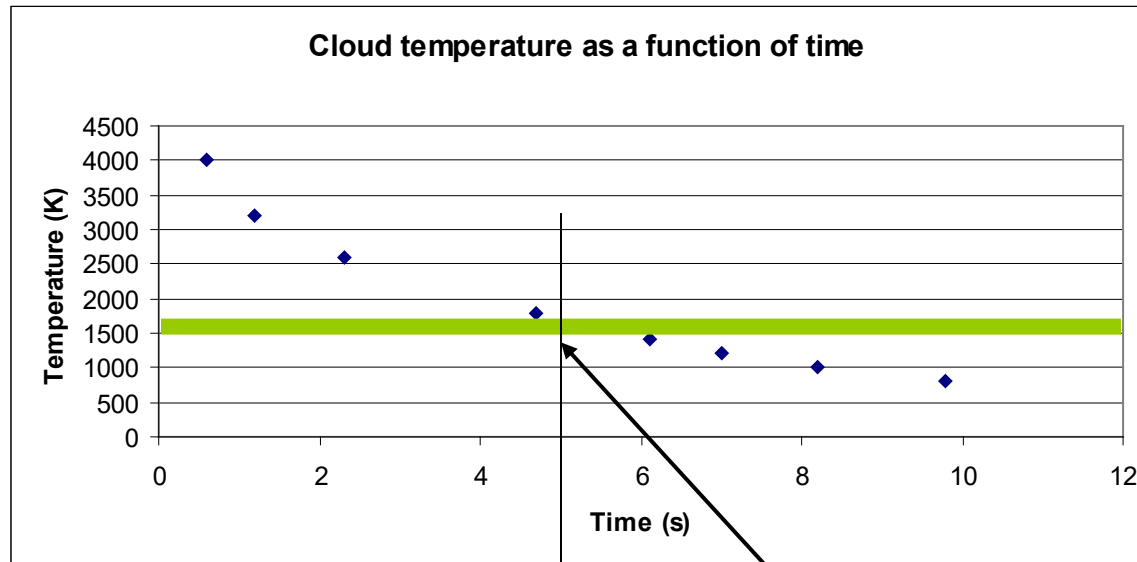
– Training restrictions – radiological hazards

- Sample measurement time 48hrs from end of fission.
- Limits on:
 - No detectable transuranics (alpha emitters: Pu, Am)
 - long-lived isotopes (Sr-90 and Cs-137)
- Sample size – grams
- Particle size range: 100 microns to millimeters

Cloud/Fallout Chemistry

~5s post-detonation debris condensation onset.

Isotopes fractionate in the atmosphere due to differing rates of condensation



Soil solidification
Temperature (1673K)

Volatile, intermediate and refractory elements

By Mass Chains (Gibson)

By Boiling Point:1673K (Izrael)

<u>Volatile</u>	<u>Intermediate</u>	<u>Refractory</u>
⁸⁹ Sr	¹⁰³ Ru	⁹⁵ Zr
⁹⁰ Sr	¹⁰⁶ Ru	⁹⁹ Mo
⁹¹ Y	¹³¹ I	¹⁴⁴ Ce
¹³² Te	¹³⁶ Cs	¹⁴⁷ Nd
¹³⁷ Cs	¹⁴¹ Ce	¹⁵⁶ Eu
¹⁴⁰ Ba		

<u>Volatile</u>	<u>Intermediate</u>	<u>Refractory</u>
Br	Te	Zr
Kr	Sb	Mo
Xe	Tc	Sr
I	Sn	Y
	Rh	Nb
	Ru	Ba
	Pb	La
		Nd
		Pr
		Pm
		Sm
		Ce

Izrael YA (2002) "Radioactive Fallout after Nuclear Explosions and Accidents"
 ISBN: 9780080438559, Elsevier Science Ltd

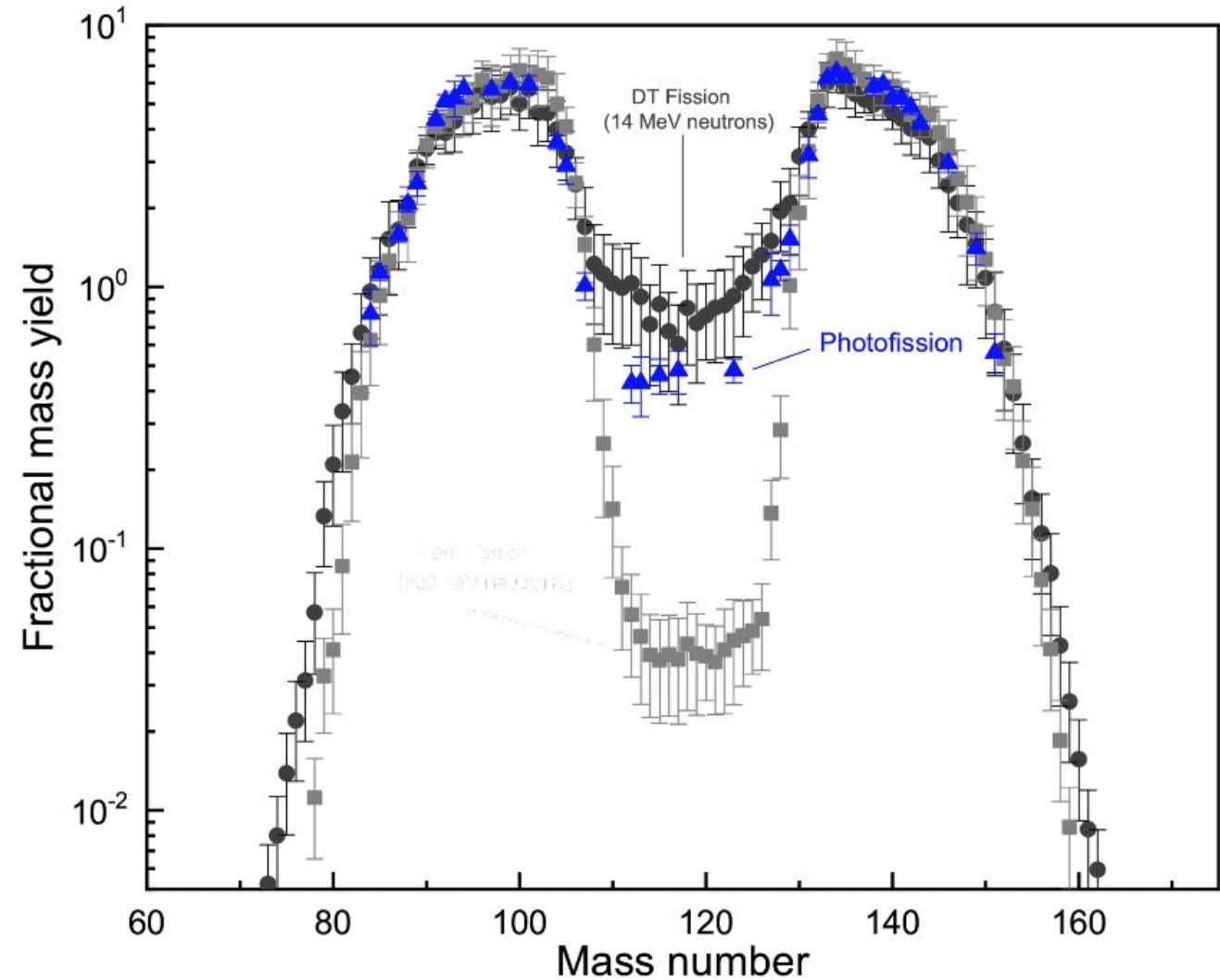
Gibson TA (1968) "Observed Fractionation in Ground Level Fallout
 from Three Nuclear Cratering Detonations,
 University of California, Lawrence Radiation Laboratory report: TID-4500 UC-35

Actinide Isotopes

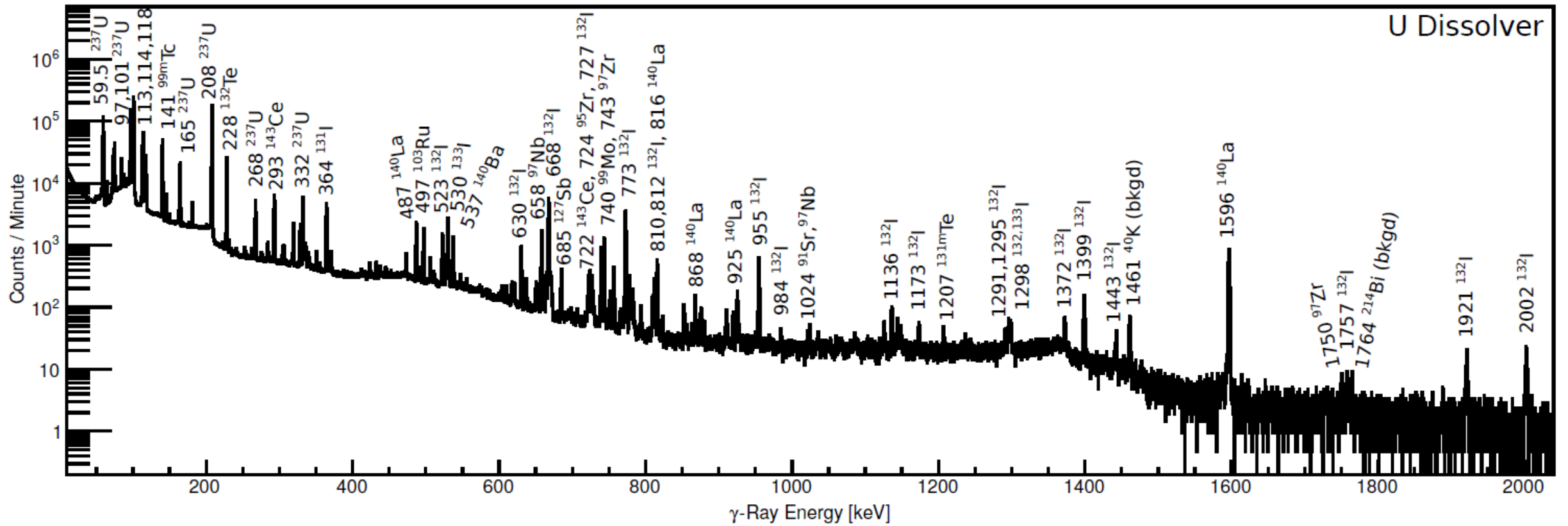
- Uranium
 - Fission products from fission of nuclear material – U, Pu
 - Spectrum – fast fission
- Matrix
 - Soil matrix – natural abundance
 - U-238 natural background: 8 ppm.
- Restrictions
 - High purity starting base
 - Limit to how much U-238 and U-235

Photofission U-238

- Fission product distribution similar to fast fission
- Generated with 30 MeV bremsstrahlung linear accelerator
- Rapid removal, shipment and handling for chemistry

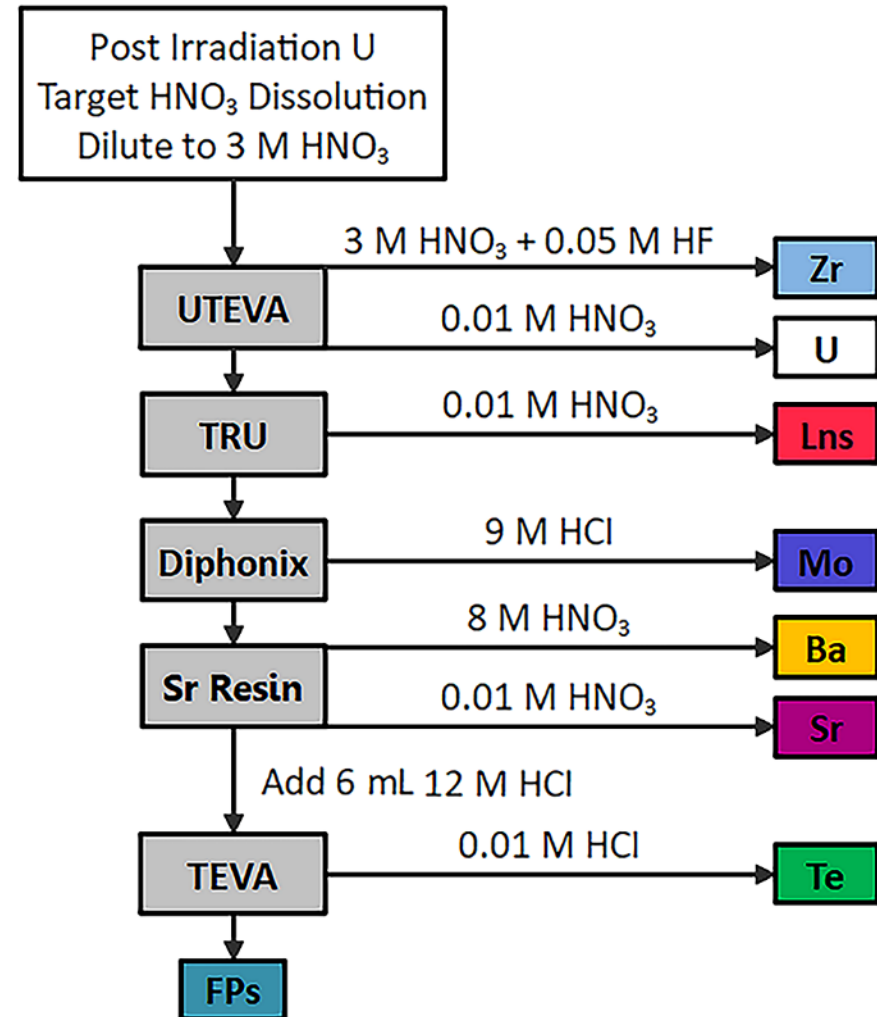
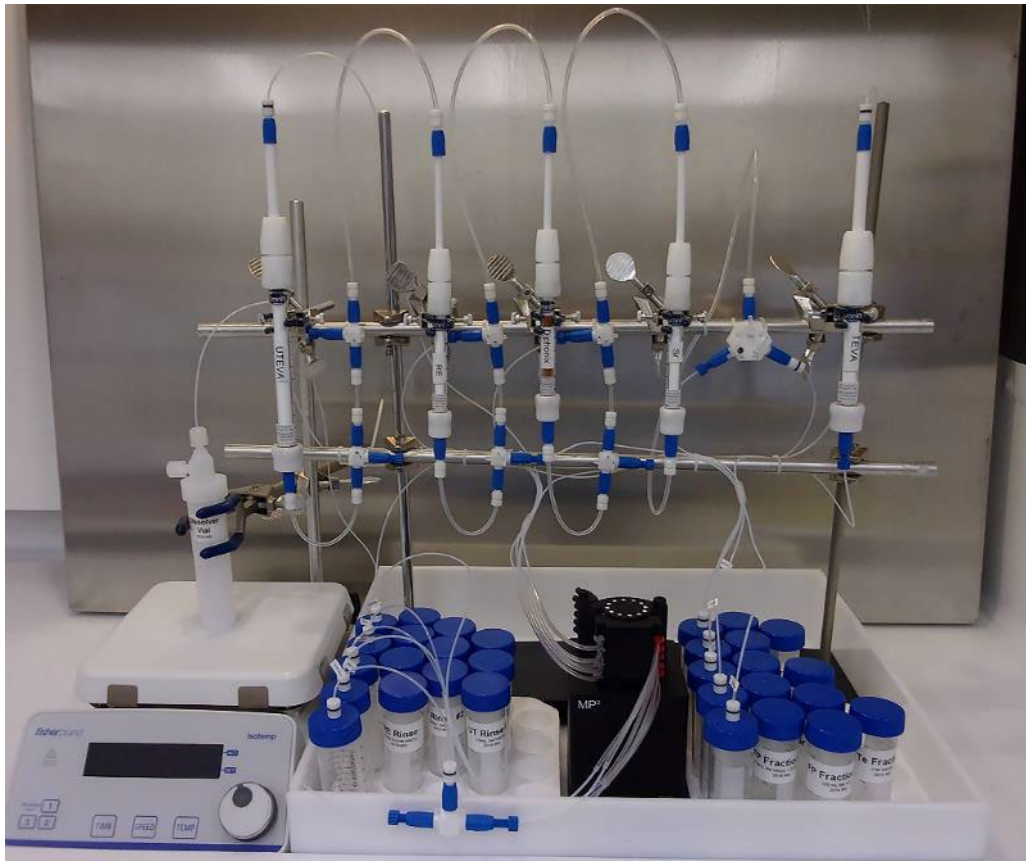


Gamma spectrum of dissolved, irradiated U-238 foil.



Rapid Chemistry

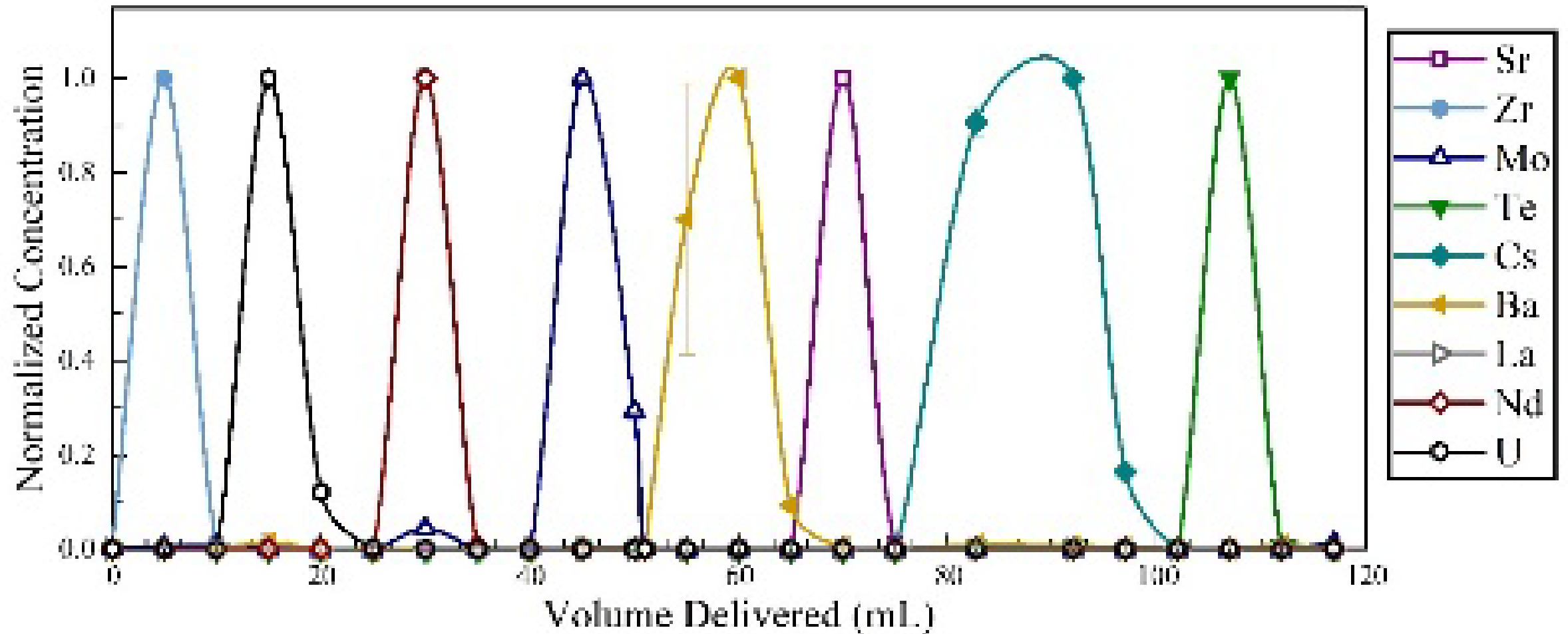
Five columns in series (load) and parallel (elution)



Snow et al. Anal. Chem. 2021, 93, 8, 3770-3777.

Snow, M.; Ward, J. U.S. Patent, US-2020-0109348, 2020.

Efficient separation of target elements



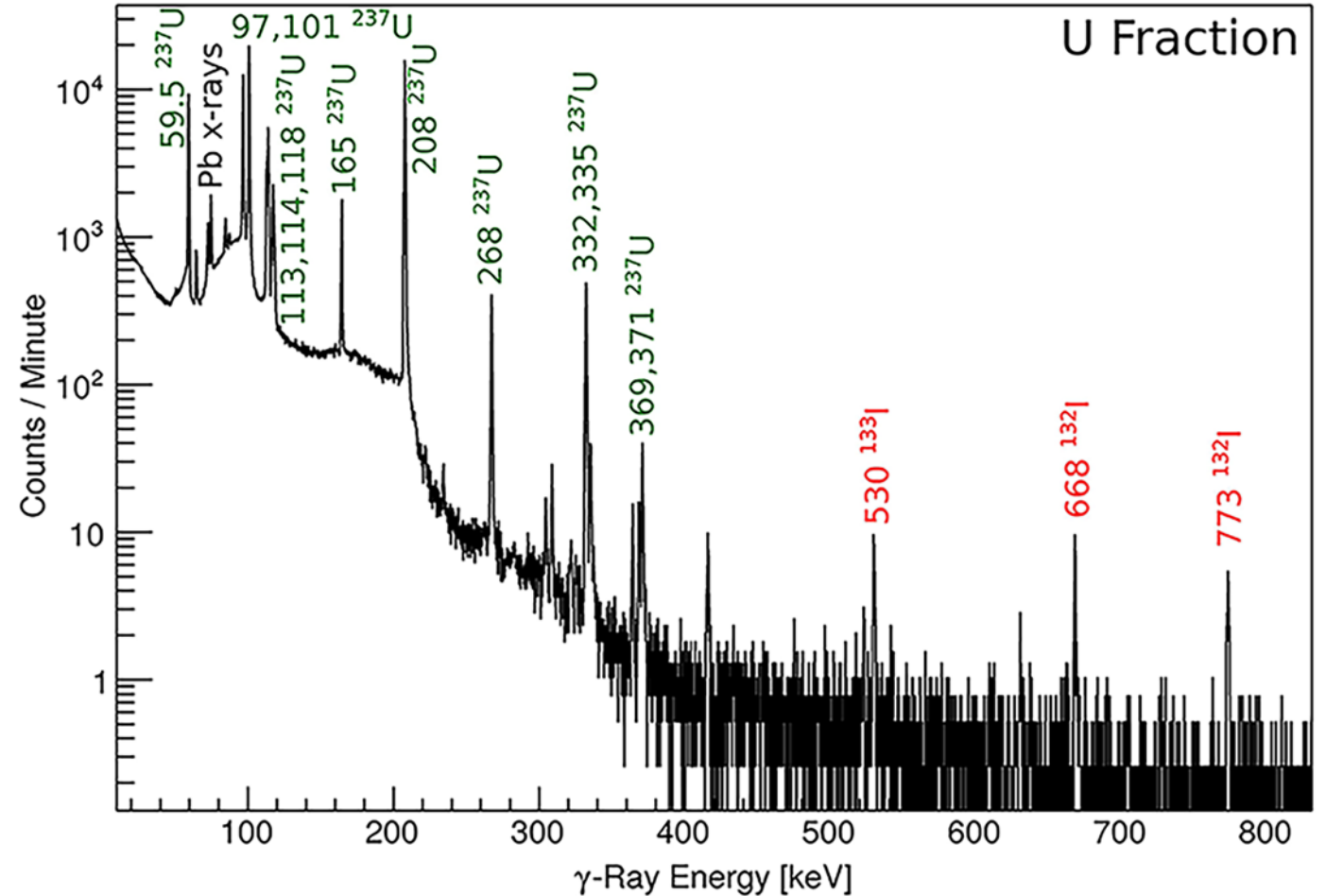
Snow et al. Anal. Chem. 2021, 93, 8, 3770-3777.

Uranium removal

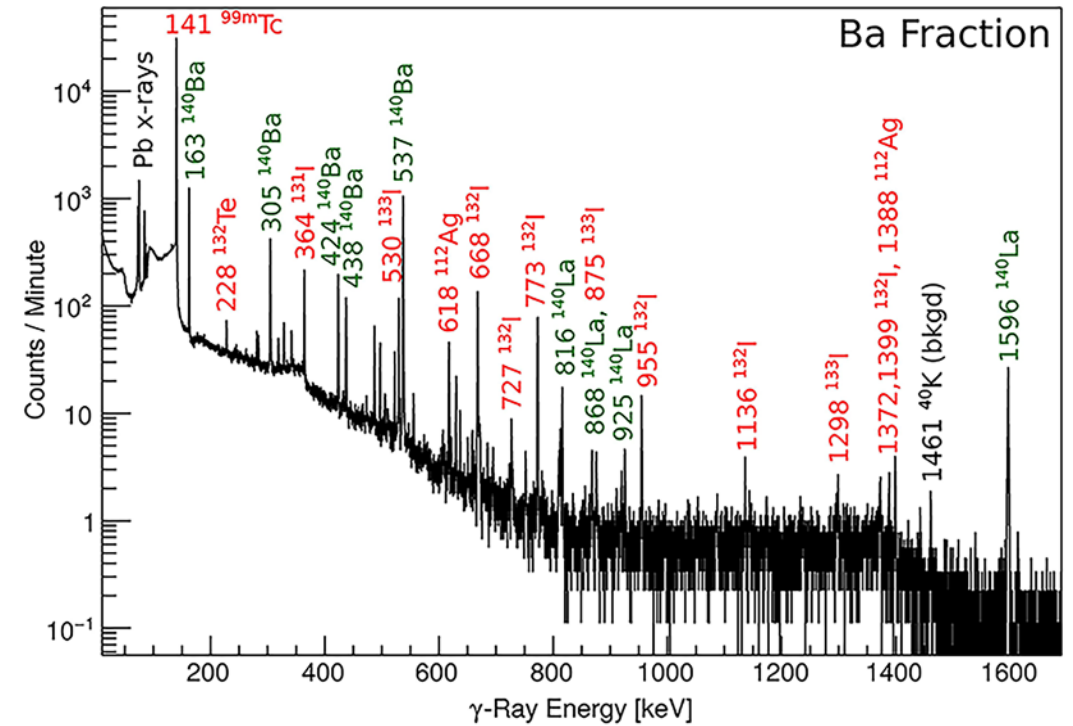
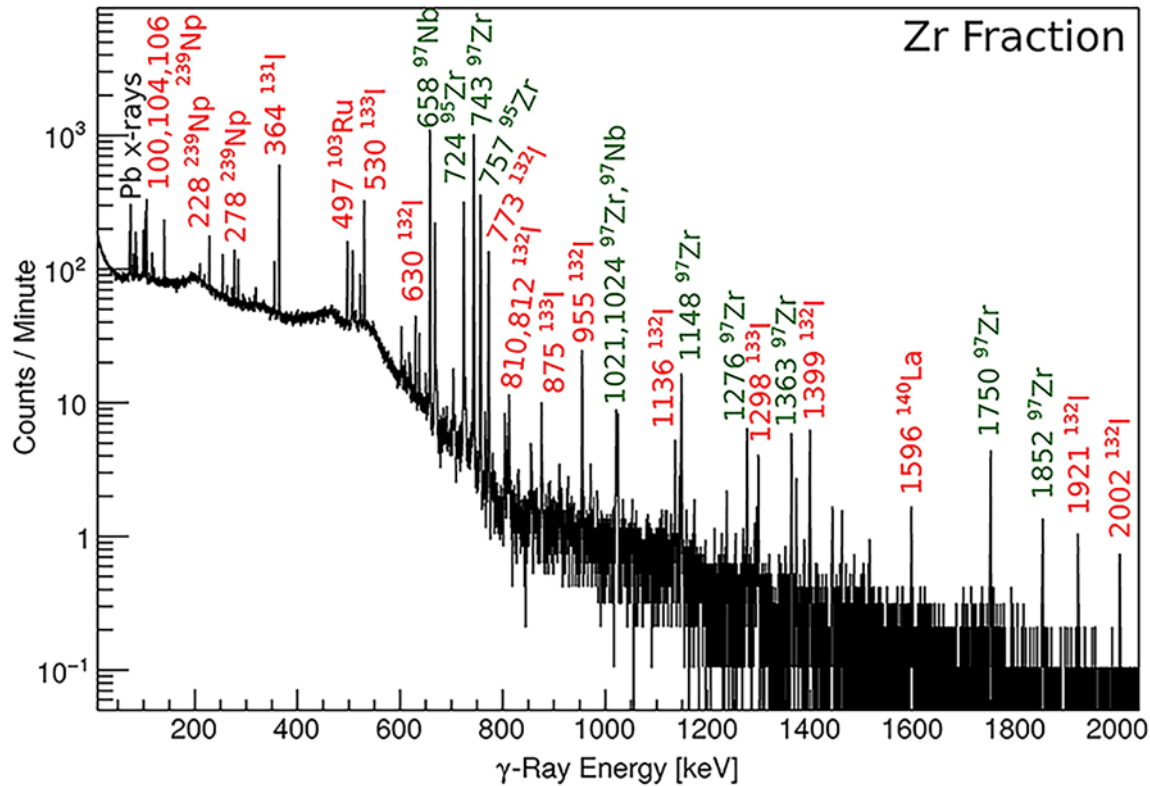
Photo-irradiation of a gram of U-238 produces millicuries of U-237

U-237 has a half-life of approximately 6 days

U-237 is a tracer for uranium removal
>99.999% (< 10 ppm)

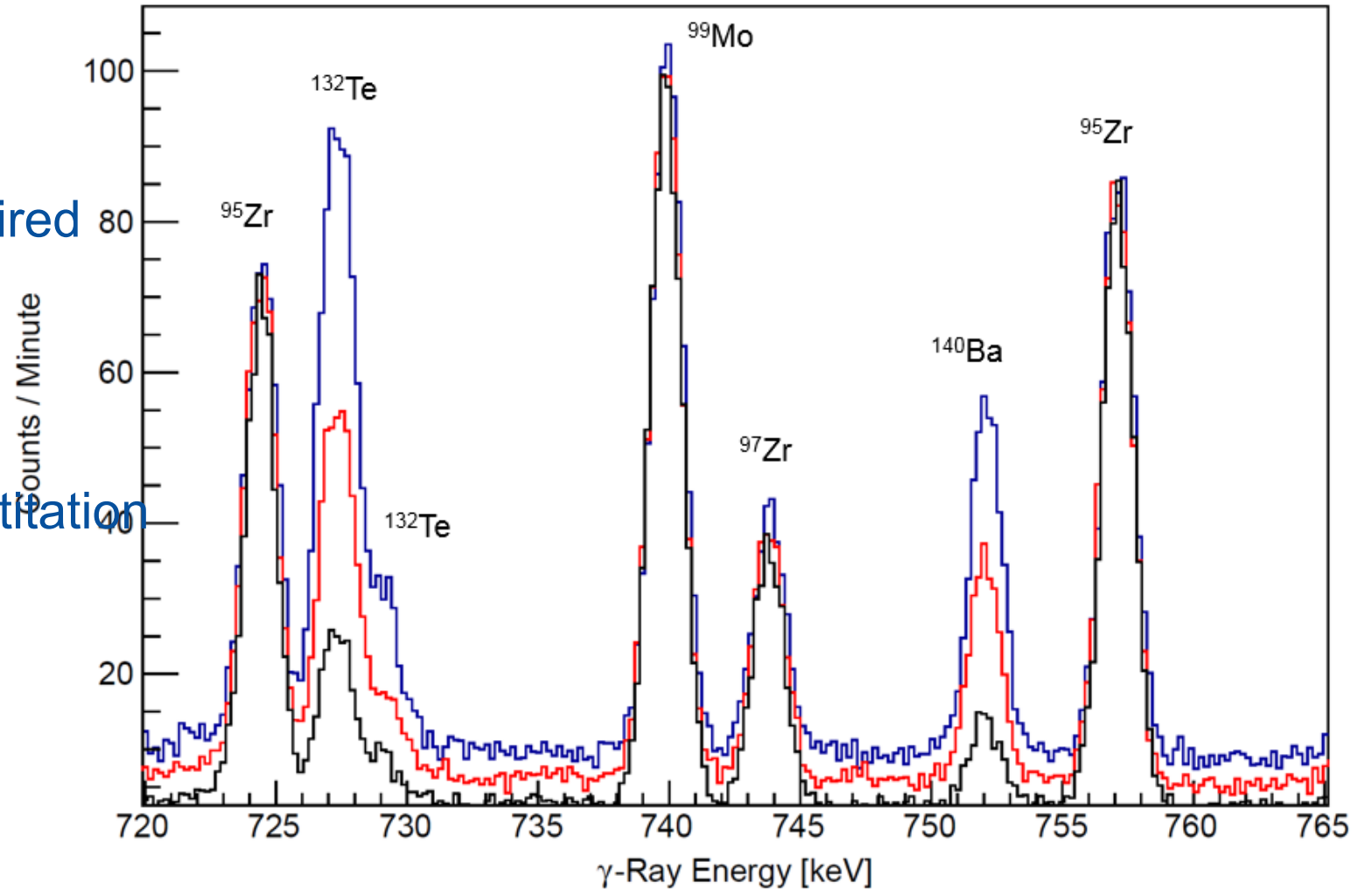


Historic fractionation monitors: Zirconium and Barium



Pre-encapsulation solution preparation

- Separation
- Re-mix isotopes into desired proportions.
- Quantitation
- Encapsulation
- Post-encapsulation quantitation



Glass encapsulation using sol-gel synthesis

- Solution based polymerization of tetra-ethyl ortho silicate
- Extremely pure silica matrix
- Acid catalyzed rapid polymerization

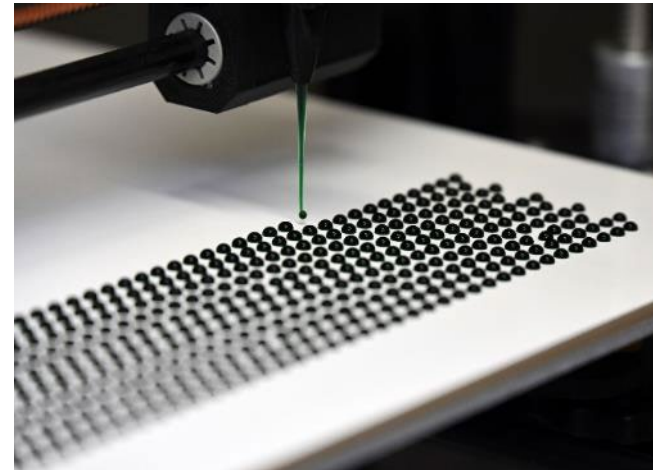
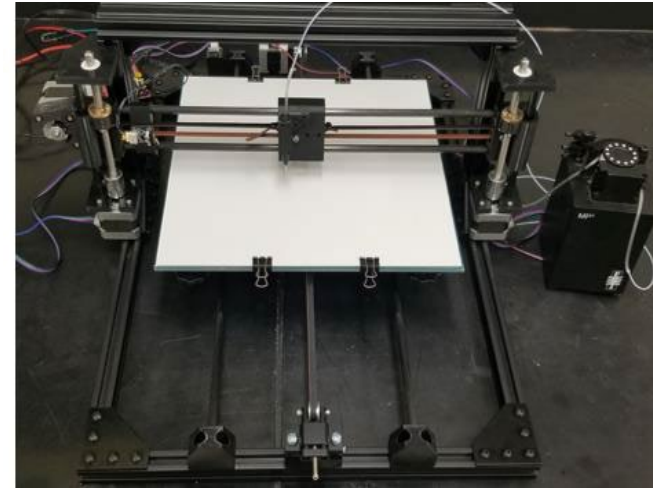
- Produces
 - Spherical glass beads
 - Retention is quantitative for most elements
 - Tailored isotope mixes
 - Control of particle size
 - Control of isotopic composition

Carney, K. P.; Finck, M. R.; Mcgrath, C. A.; Martin, L. R.; Lewis, R. R. The development of radioactive glass surrogates for fallout debris J. Radioanal. Nucl. Chem. 2014, 299, 363–372.

Glass bead product

- Produce gram quantities – under 48 hours irradiation to encapsulation with desired:
 - Well characterized uniform, durable test glasses
 - Desired matrix, particle size range/distribution
 - Desired fission product and actinide composition

Solid Debris Simulant Advanced Manufacturing



Glass annealing - element retention/loss

- Annealing affects glass porosity, density fragility
- Desired density ~2.4
- Loss of some volatile elements (I and Ru)

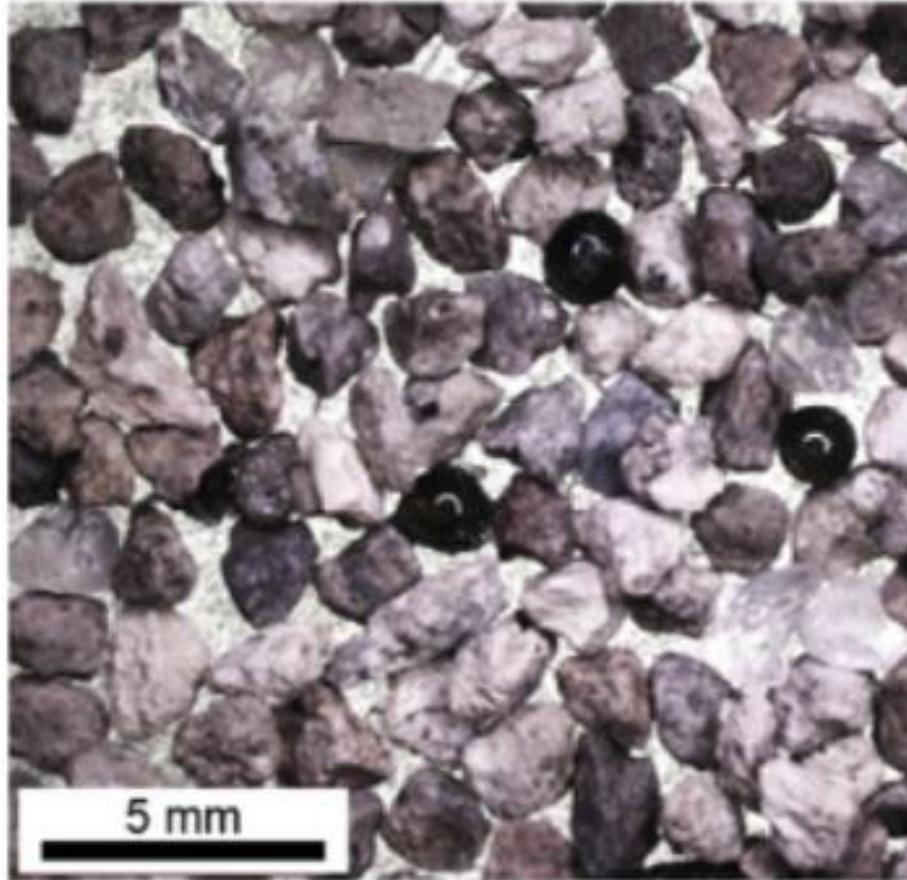
	Te	Ce	Nd	Ba	Mo	I	Zr	Rh	Ru	Sb
100°C	103 ± 2%	102 ± 2%	105 ± 4%	101 ± 4%	103 ± 2%	105 ± 22%	103 ± 2%	110 ± 16%	109 ± 4%	104 ± 4%
300 °C	104 ± 2%	105 ± 2%	106 ± 4%	105 ± 3%	106 ± 2%	0%	104 ± 3%	91 ± 14%	92 ± 2%	105 ± 4%
500 °C	98 ± 2%	101 ± 2%	99 ± 4%	99 ± 4%	105 ± 4%	0%	101 ± 4%	95 ± 18%	75 ± 4%	99 ± 6%
600 °C	102 ± 2%	101 ± 2%	103 ± 4%	101 ± 2%	101 ± 2%	0%	103 ± 2%	99 ± 14%	12 ± 2%	99 ± 4%



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Summary



Historic NTS Debris



INL Fallout Simulants