INL/CON-22-69521-Revision-1

Stable Element Doping of Sol-gel Toward Simulating Environmental Matrix in Surrogate Explosive Nuclear Debris

October 2022

Justin Teryl Cooper, Mathew S Snow, George Diehl

Changing the World's Energy Future

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Justin Cooper, P.h.D

NuFor Conference 2022

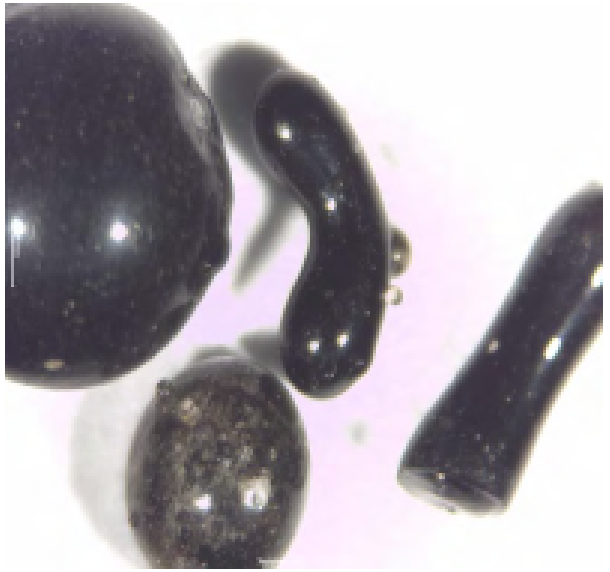
Idaho National Laboratory

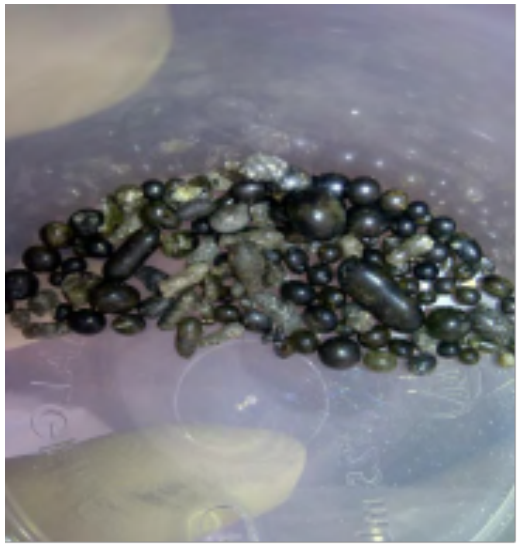
Surrogate Nuclear Explosive Debris (SNED)

- What is surrogate nuclear explosive debris?
  - Synthetic material that mimics some aspect of nuclear debris (physical, chemical, radiological etc).

- Need
  - Realistic surrogate nuclear debris needed for training nuclear forensic capabilities of first responders (in-field collections, laboratory analysis)

- Requirements
  - Size
  - Color
  - Elemental composition (matrix)
  - Radionuclide content
  - Dose





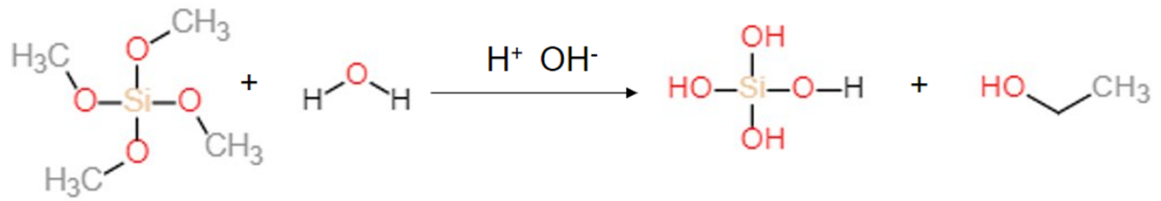
How is SNED produced?

Sol-gel Particle Synthesis for Surrogate Aerodynamic Fallout

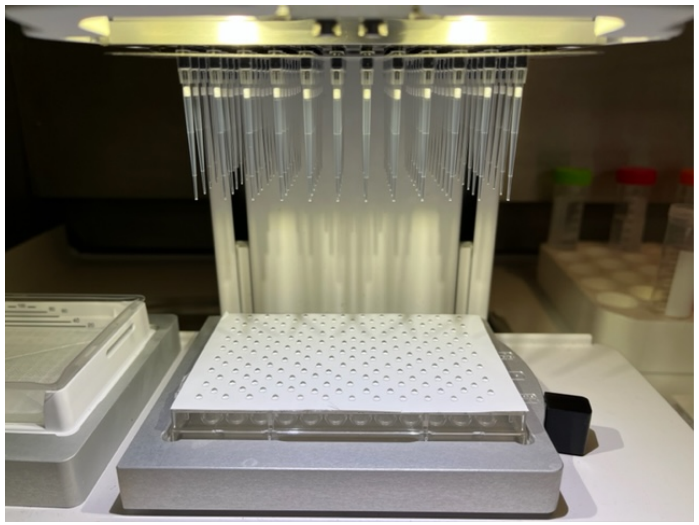
- Bottom-up Method for generating solid materials from small molecules (Tunability)
  - Monomeric Silicate precursor (TEOS)
  - Acid catalyzed hydrolysis and poly-condensation
  - High-throughput drop casting onto hydrophobic surface
- Tunable particle sizes
  - Near-field to mid-range field nuclear fallout (2 mm to 200 um particles)

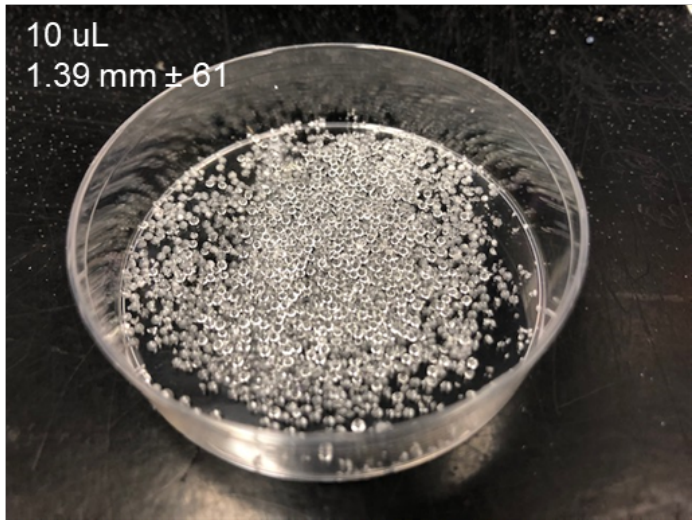
Sol-gel Synthesis Method

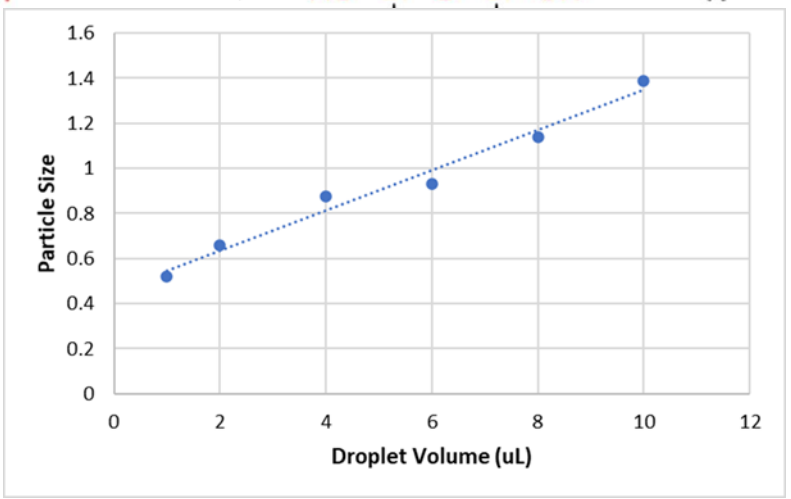
- Hydrolysis of Alkyl silicate precursor
  - Can be acid or base catalyzed



- Condensation

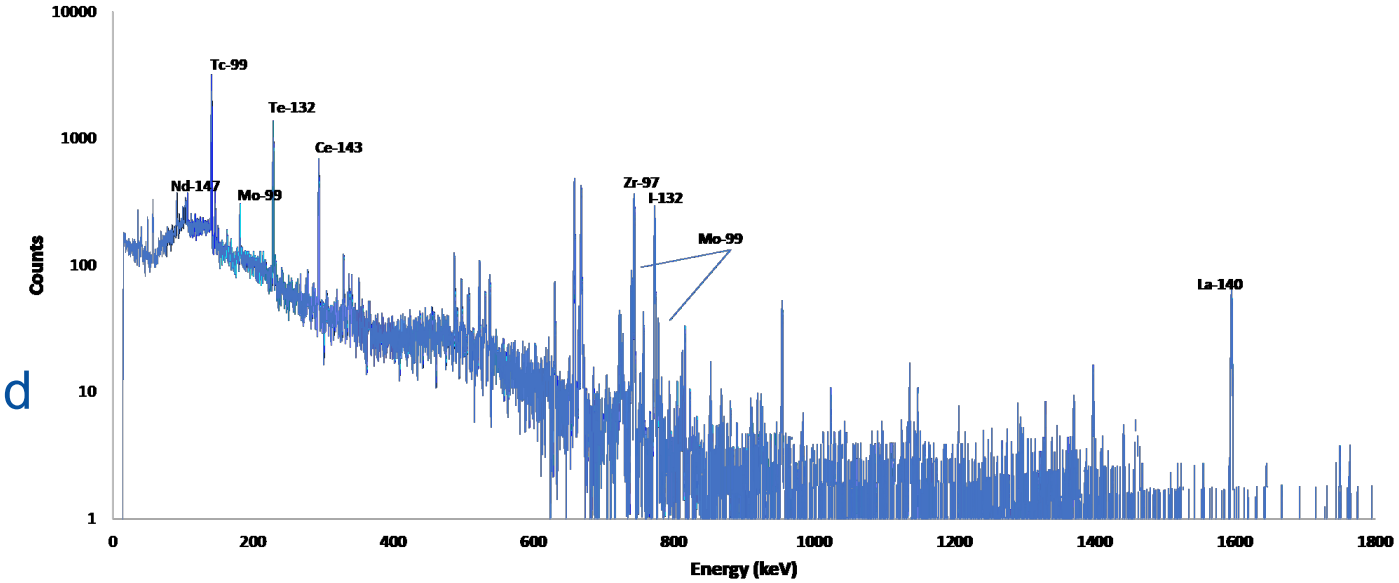






Sol-gel Particle Synthesis for Surrogate Aerodynamic Fallout

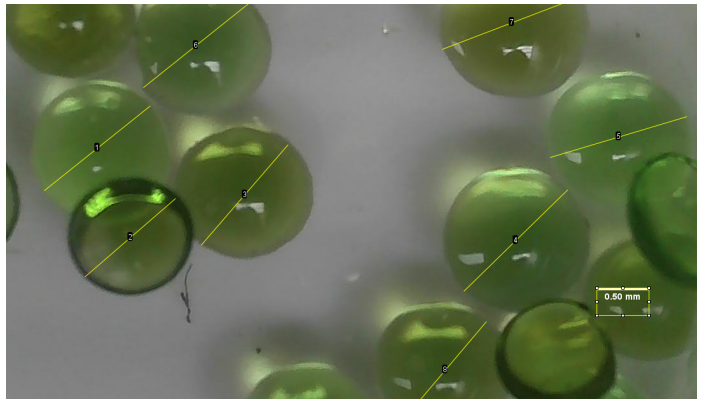
- Quantitative encapsulation of radionuclides
  - Included into acid fraction of sol-gel precursor solution.
  - Standard solution in → Standard solid out (within measurement error)
  - Tunable radionuclide ratios

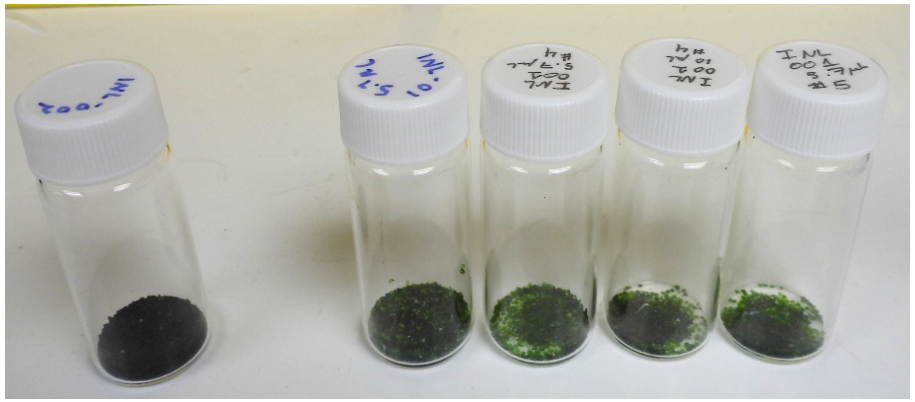


Example Gamma Spectrum of Sol-gel surrogate Debris

- Tunable Color
  - Black vitreous luster
  - Dark green
  - Blue, red, orange, etc.

Using Stable Dopants

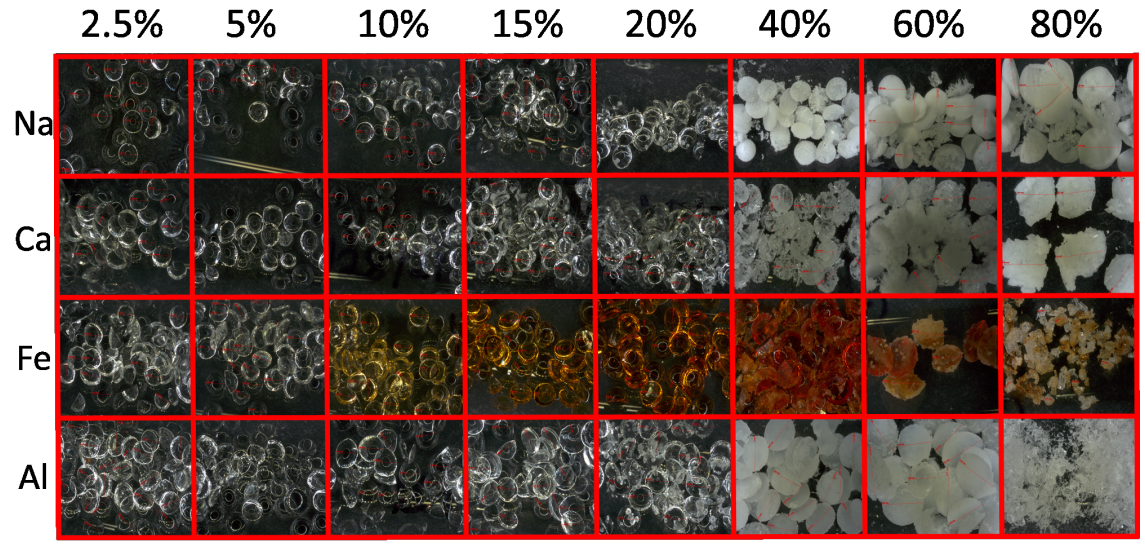




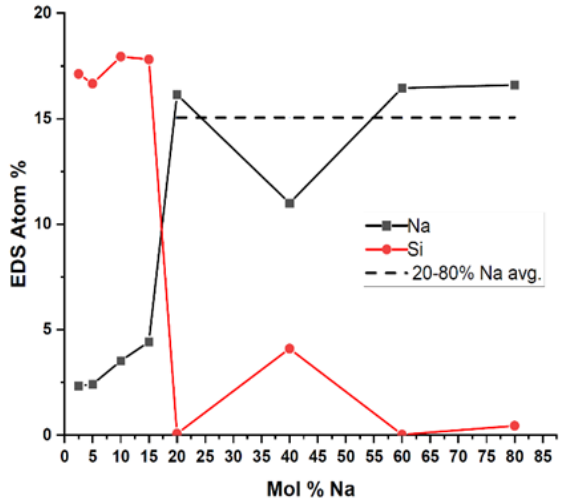
Example Green Sol-gel surrogate Debris

Stable Elemental Loading in Sol-gel

- Systematic loading of single elements
  - Al, Fe, Na, Ca nitrate salts
  - 0 to 80 mol % (relative to Si)
  - Characterization via
    - Optical microscopy
    - SEM
    - EDS

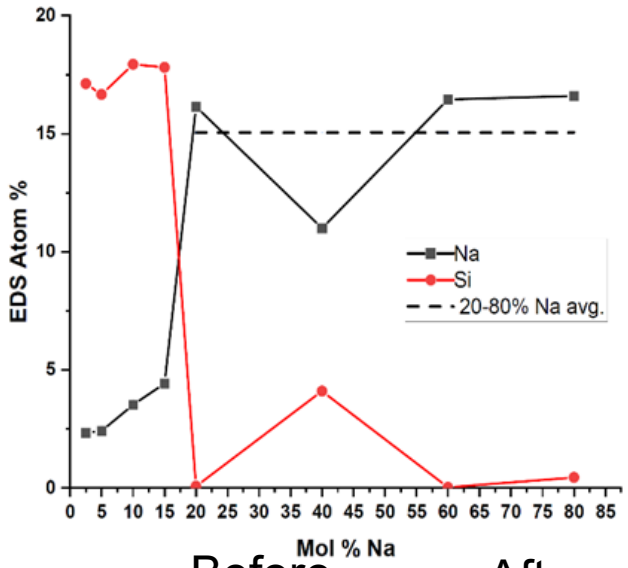


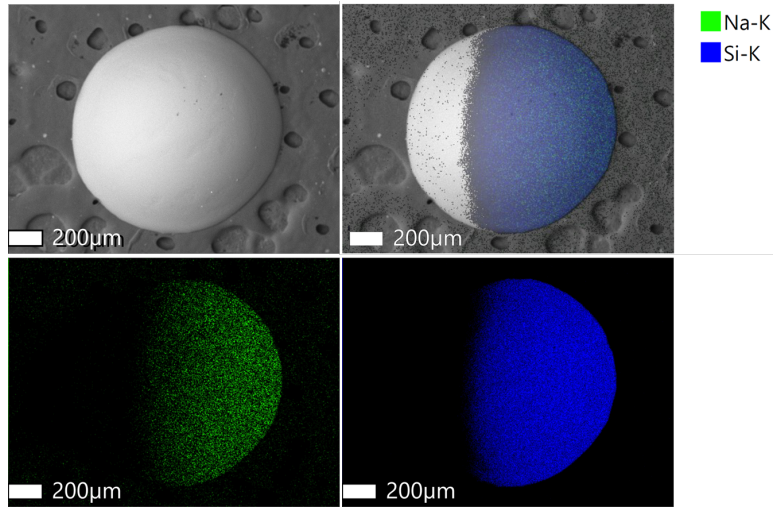
- Predictable linear incorporation at low loading levels (< 10 mol %)
- Predictable matrix inversion at high loading levels (>60 mol %)
- Most interesting at (10-20 mol %)

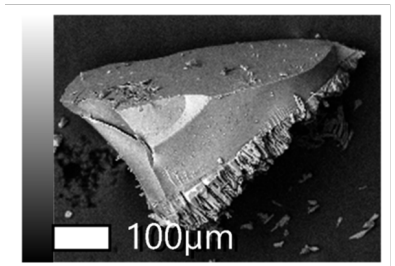


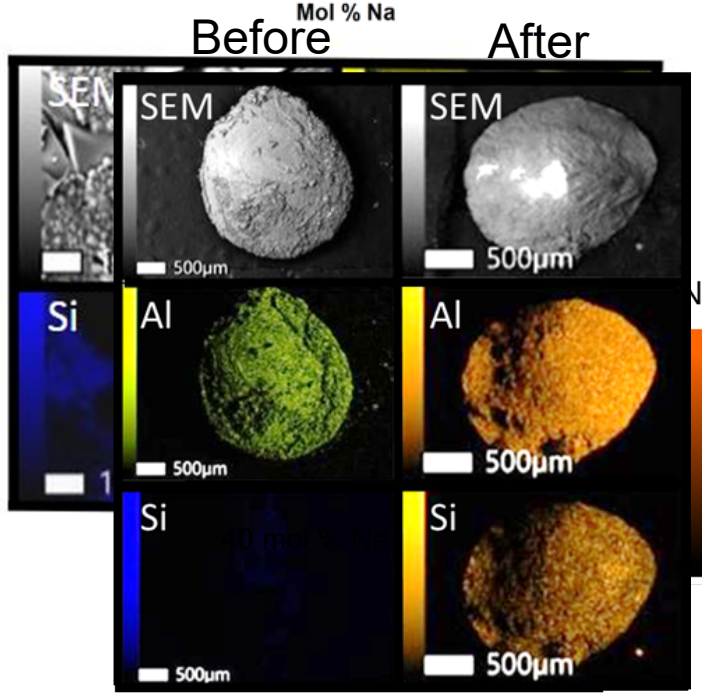
Stable Elemental Loading in Sol-gel

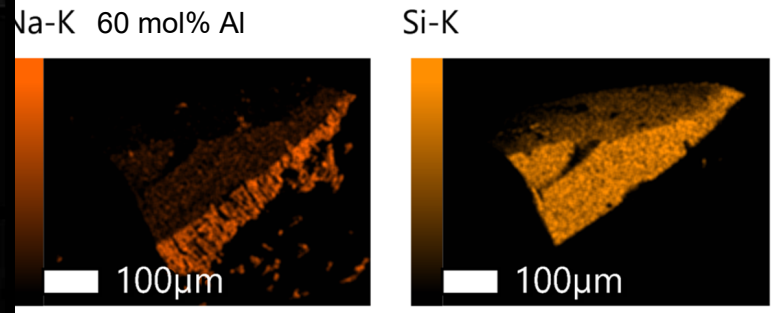
- Prior to 15 mol % loading level
  - Smooth topographical structure
  - Homogenous elemental distribution
  - Co-localization w Si
- Above 15 mol % loading level
  - Rough topo. surface features form
  - Enriched in dopant element
  - Obscuring or not co-localized w Si.
  - Dopant Capacity reached at ~15-20%
- Thermal Treatment at 500 deg C
  - Re-homogonizes elemental distribution









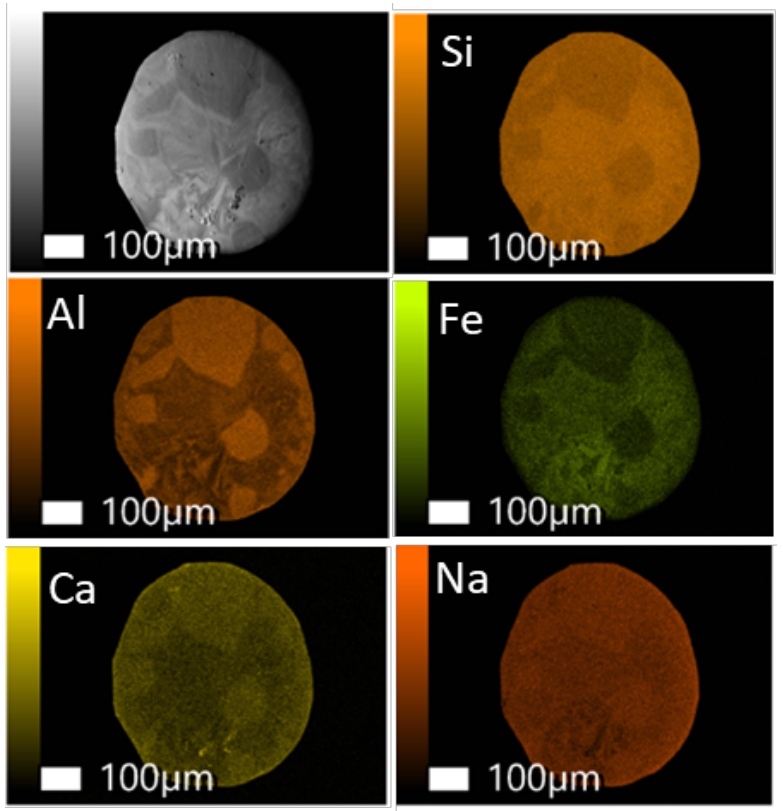


40 mol % Na

Soil Matrix and NTS Debris

- Soil Matrix
  - Al, Fe, Na, Ca doped into approximate wt. % to mimic average earth crustal abundance
  - Particles thermally treated at 500 deg C.

Element	Target Wt. %[1]	Measured Wt. %	Mol % to SiO_2	Crustal Wt. %[17]
Fe	4.47	7.39	4.94	5.00
Na	5.24	6.18	10.03	2.83
O	-	58.72		46.6

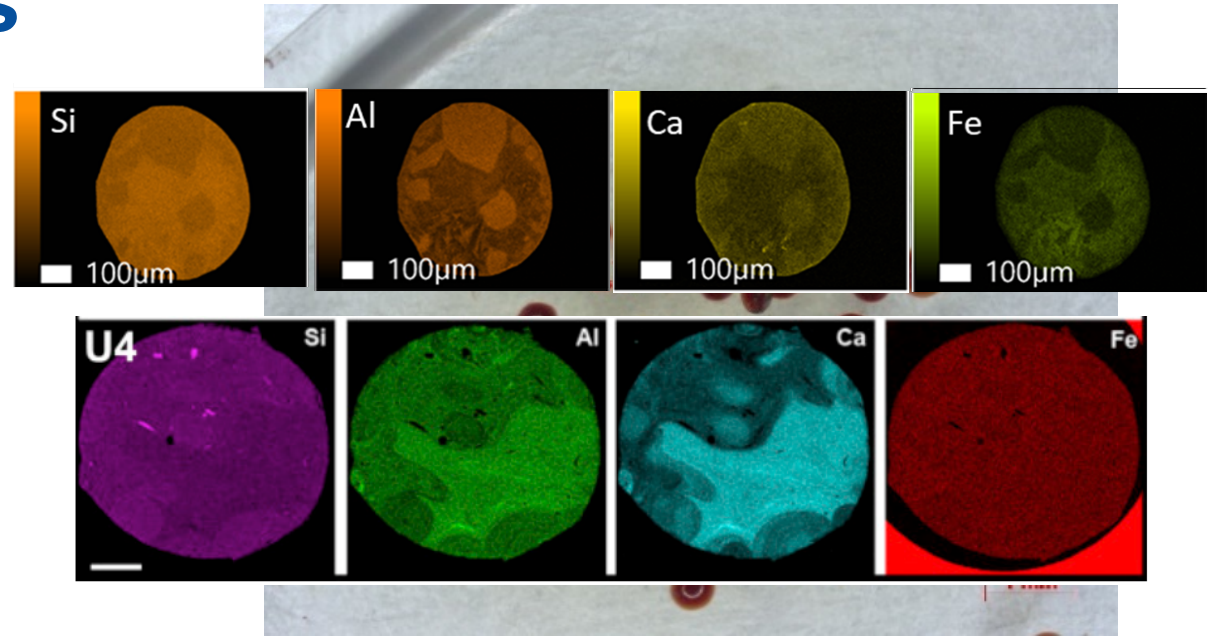


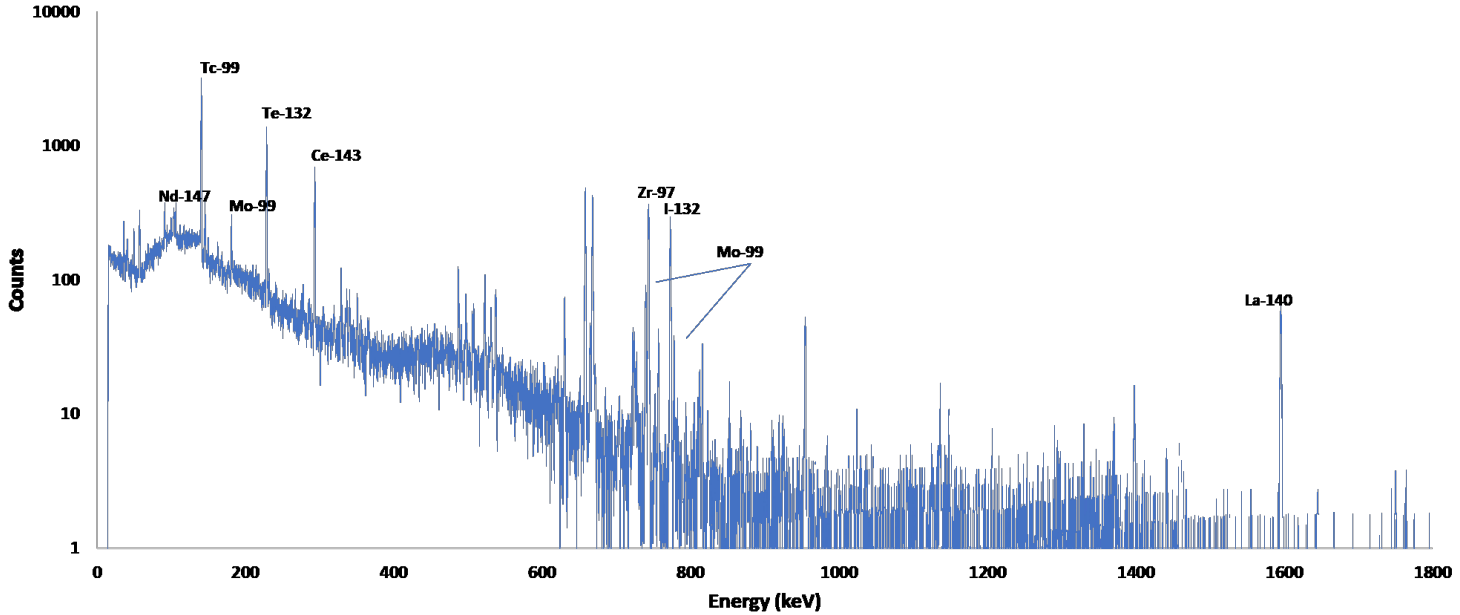
SEM and EDS of soil matrix sol-gel particles.

1. Shapley P (2010) The Earth's Crust. http://butane.chem.uiuc.edu/pshapley/Environmental/L26/1.html. Accessed February 22 2022.

Soil Matrix and NTS Debris

- NTS Matrix w Fission Products
  - Quantitative fission product encapsulation
  - Encapsulated ~ 1E^{13} fissions/g sol-gel
  - Does rate ~ 350 mrem/h/g





Element	Target NTS Wt. %[1]	Measured Wt. %	Bias %
Al	7.28	9.0 ± 0.2	24
Fe	2.06	2.0 ± 0.2	3.7
Ca	1.34	1.4 ± 0.1	2.4
Mg	0.328	0.33 ± 0.04	1.1

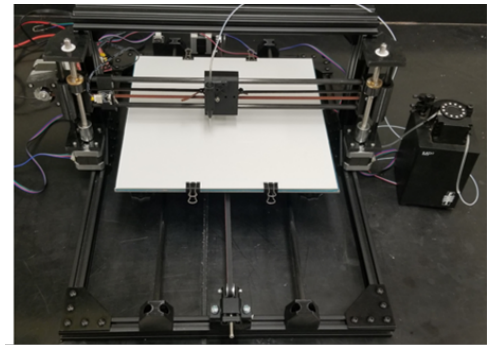
1. Weisz et al. *Geochim. Cosmochim. Acta* **2017,** *201*, 410-426.

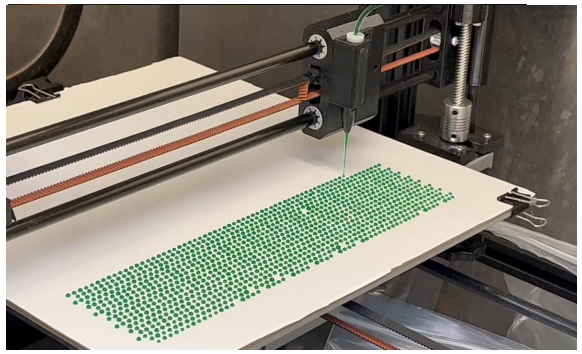
Summary

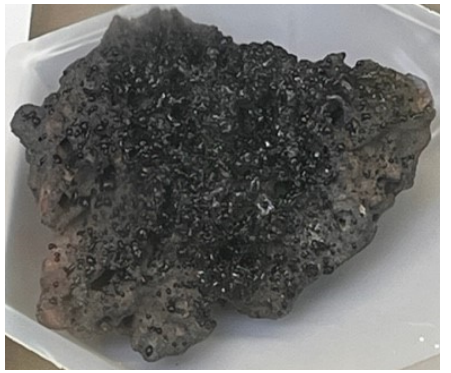
- Sol-gel Particle Synthesis for surrogate aerodynamic fallout
  - Tunable size, color, elemental composition, radionuclide content, dose rate
- Characterized the loading behavior and capacity of stable dopants into sol-gel particles
- Produced sol-gel particles with correct size, color, elemental composition, radionuclide content, dose rate as evaluated NTS fallout
- Sending sol-gel SNED to LLNL, LANL, AWE etc…

Future Directions

- Surrogate Far-field Particles
  - Alternate matrices e.g. urban etc.
  - Super hydrophobic surfaces
    - <100 um
    - Spherical
  - Advanced Manufacturing Techniques
    - Higher throughput
    - Other types of debris
  - “Ballistic splatter” type debris







Acknowledgments

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