SAND2014-16557PE

Sandia Materials for the Capture, Storage or Purification of Radiological Ions and Gases

Tina M. Nenoff
Nanoscale Sciences Department
Sandia National Laboratories
Albuquerque, NM 87185

August 2014
Sandia Japan Visit

Separations and waste forms research is currently funded under the DOE/NE-FCR&D Separations and Waste Form Campaign.

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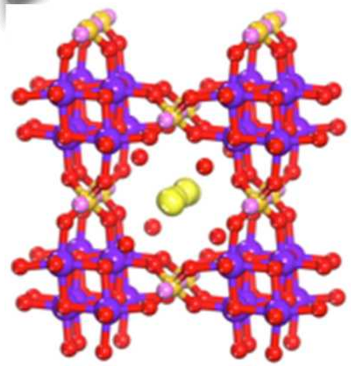
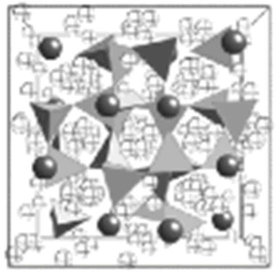
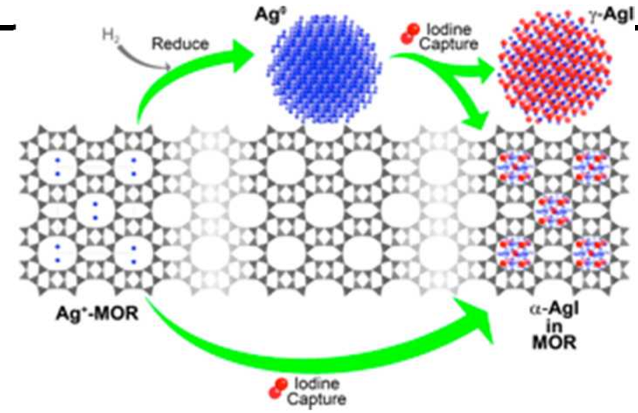
Nenoff, et.al., Portfolio of NE related Technologies: Novel Separations and Waste Forms



CST, Cs^+ removal from water to Pollucite Waste Form

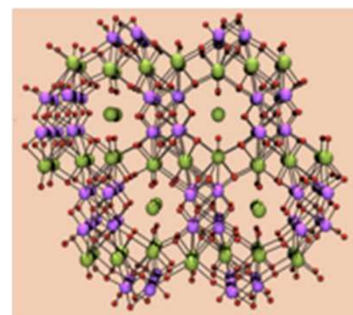


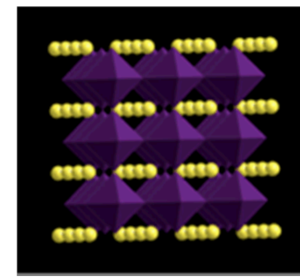
R&D100 1996
JACerS, **2009**, *92(9), 2144*
JACerS, **2011**, *94(9), 3053*
Solvent Extr. & Ion Exch, **2012**, *30*, 33



Ag-MOR
$I_2(g)$ capture & mechanisms

JACS, **2010**, *132*(26), 8897
J Phys Chem Letters, **2011**, 2, 2742

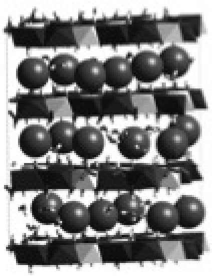


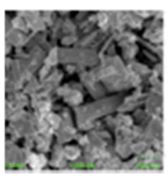


Sr^{2+} getter, 1-step to Perovksite waste form
JACS, **2002**,*124(3),* 1704

Fundamental Research to Applied to Commercial Products
Design the Separation Material To Develop the Waste Form

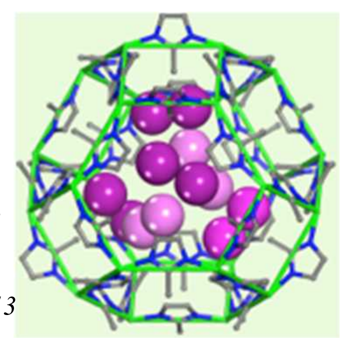
Applied Geochem, **2011**, *26*, 57

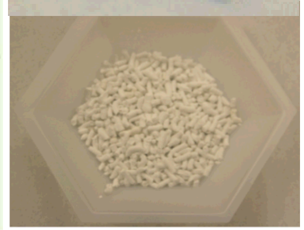


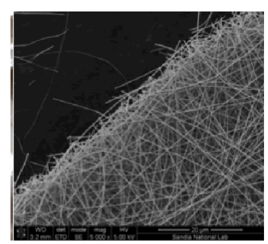


In-situ Iodine removal from water

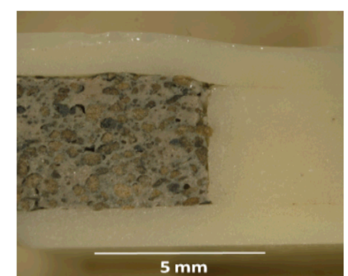
I_2/MOF, Isolation to Waste Form
JACS, **2011**,*133(32),*12398
Ind.Eng.Chem.Res, **2012**, *51*(2),614
Provisional Patent Oct 2013







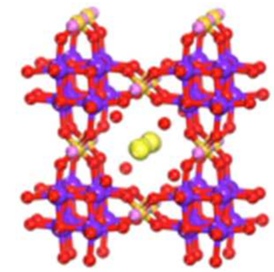
Nanoporous Nanofibers Volatile Gas Removal
US Patent Application, 2011



Universal Core-Shell Glass Waste Form Iodine & Getter
JACerS, **2011**, *94*(8), 2412
US Patent 8,262,950

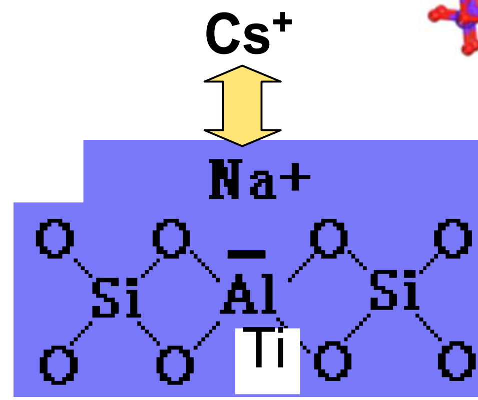
Removal of Rad-Cs^+ from Pooled Seawater (heat treat to Pollucite WF or add into SNL GCM)

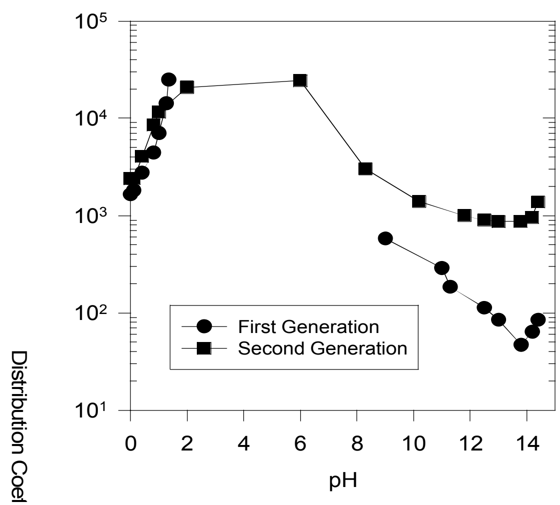
Crystalline Silicotitanates (CSTs)
With exceptional Cs^+ selectivity, and mechanical, thermal and radiological stability

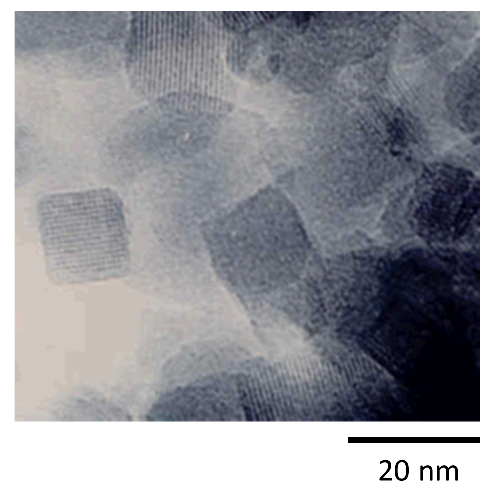


CST properties:

- Removes 1 part Cs per 100,000 parts Na
- Stable over entire pH range
- Stable in extreme environments
- *Commercially available as IONSIV™ IE-910 & IE-911*



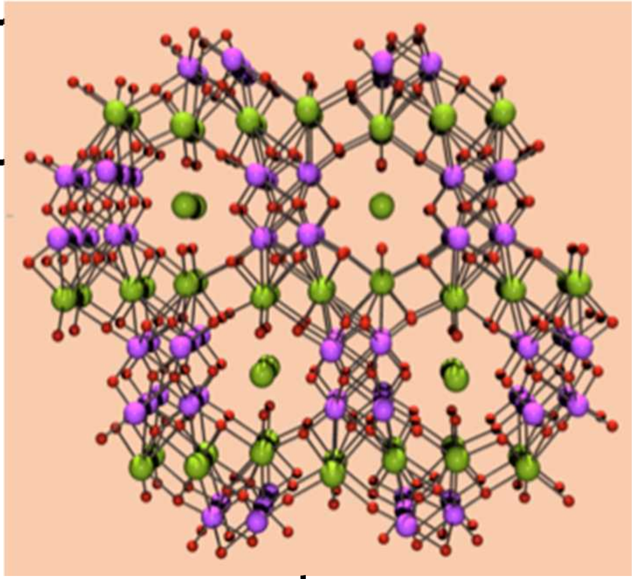




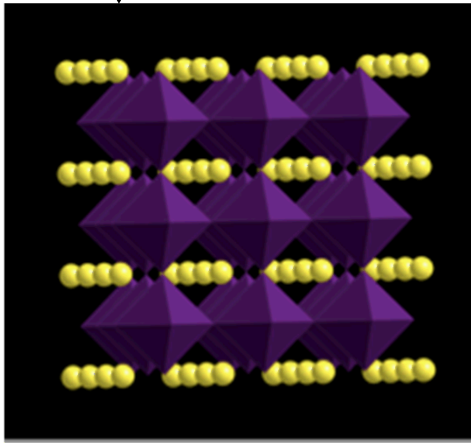
Sandia Octahedral Molecular Sieves (*SOMS*) Selectivity (heat treat to perovskite WF)

	metal ion	Ti-niobate phase Nb:Ti = 1:4	Zr-niobate phase Nb:Zr = 1:6
Radionuclides	Ba^{2+}	> 99,800 *	> 99,800 *
	Sr^{2+}	> 99,800 *	> 99,800 *
High concentration in natural systems	Ca^{2+}	2,300	2,657
	Mg^{2+}	226	458
Industrial Waste Metals	Pb^{2+}	66,467	22,022
	Cr^{3+}	> 99,800*	> 99,800 *
	Co^{2+}	> 99,800 *	> 99,800 *
	Ni^{2+}	> 99,800 *	> 99,800 *
	Zn^{2+}	> 99,800 *	> 99,800 *
	Cd^{2+}	> 99,800 *	> 99,800 *
	Cs^{+}	150	169
	K^{+}	95	153
	Li^{+}	8	35

Instead use CST: high selectivity



450°C heat treat
1 step to waste form
to perovskite



$K_d = [M]_{ie} / [M]_{sol}$

** 0.1 ppm detection limit*

K_d obtained from 50 ppm metal ion solutions (no competing ions)

Separations of non-burnable volatile fission products and lesser actinides

Gamma Irradiation Studies at Sandia's GIF Show structure stability and iodine retention

At Sandia, we have a number of new phases with high selectivity for various fission gases

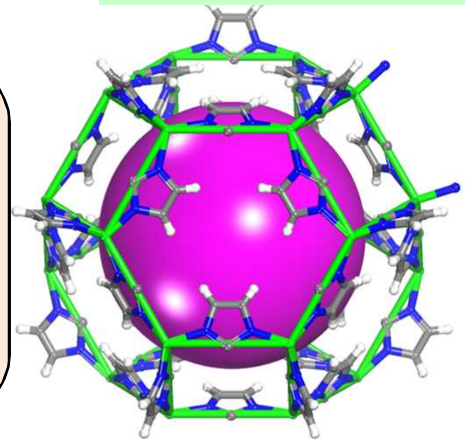
Use of Metal Organic Frameworks (MOFs) for Radiological Gas Sorption

Depending on Selectivity Needs:
Tunability of MOFs

Size (Pore) vs. Open Metal Center
Hydrophobic vs Hydrophilic MOFs
For I_2 Sorption in complex stream (eg., H_2O)

JACS, **2011**, *133* (32), 12398

Basolite Z1200, ZIF-8
Constricted Pore Opening (≈ 3.4Å)
1100 – 1600 m^2/g
Pore Volume = 0.636 cc/g
stable in Air & H_2O
High Selectivity for I_2, slow kinetics

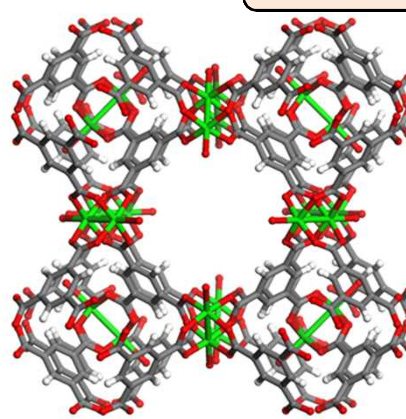


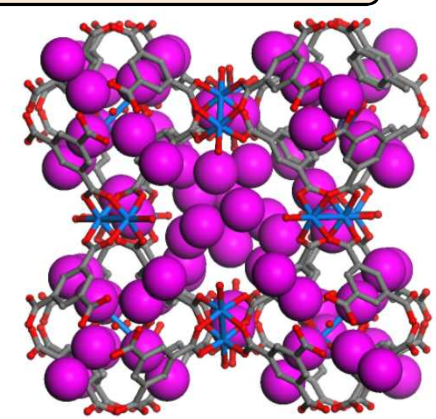
I_2@ZIF-8 ~ 125 wt.% I_2

I_2@HKUST-1 ~ 175 wt.% I_2

Chem. Mater., **2013**, *25* (13), 2591

Basolite C300, Cu-BTC, HKUST-1
Open Channels, ≈ 1nm in 3D
1500-2100 m^2/g
Exposed Metal Sites of Framework
I_2 selectivity in Humid Stream

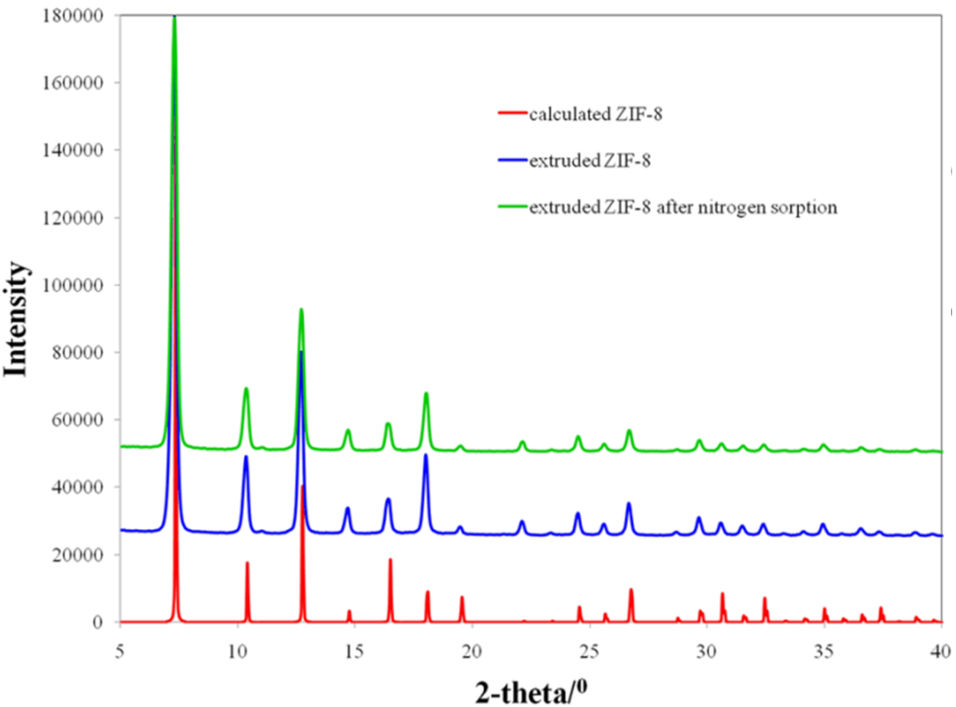


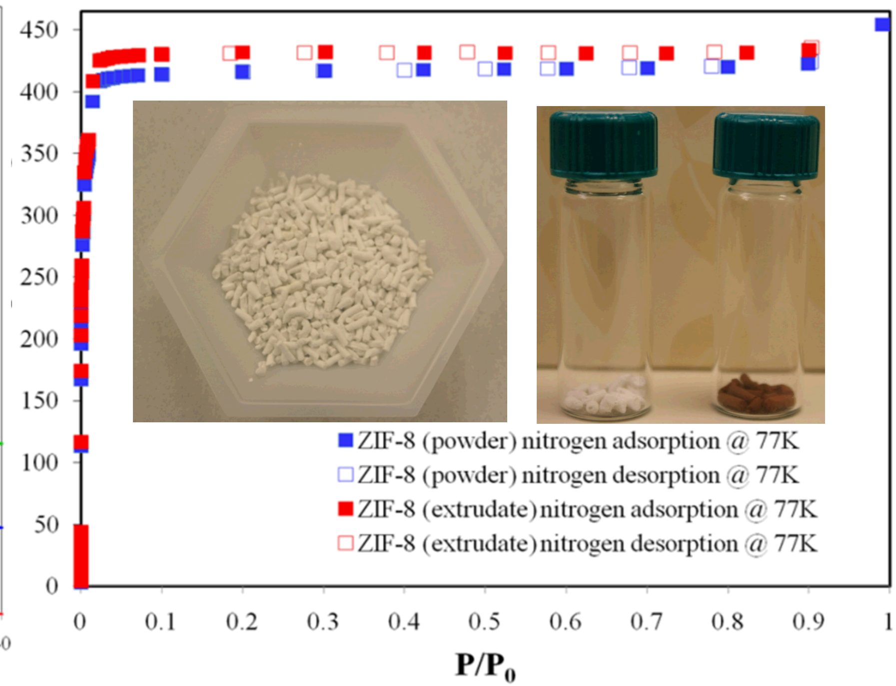


MOF Pelletizing / Characterization for Industrial Applications

Example utilizing ZIF-8

US Patent submitted, 2014





Regularly sized pellets
Maintained surface area of MOF
1850-1900 m^2/g
3.5 grams prepared

US Patent # 8,262,950; **2012**

Long Term Storage of Fission Gas Capture Materials: Waste Forms

All These waste forms have been made with the SNL Low Temperature Sintering Oxide Glass
No HIP-ping needed: Sintering 550°C
Accepting of all types of rad-loaded getters:
zeolites (AgI-MOR), Metals (AgI), MOFs (I_2-MOF), and Cs-CSTs
Durability studies: equal to better performance than basalt glass

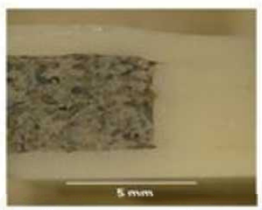
Homogenous Glass GCM: for
AgI or AgI-MOR off-gas capture and storage

50 wt% AgI/50 wt% Glass
500°C for 3 hr

50 wt% AgI/50 wt% Glass,
500°C for 3 hr

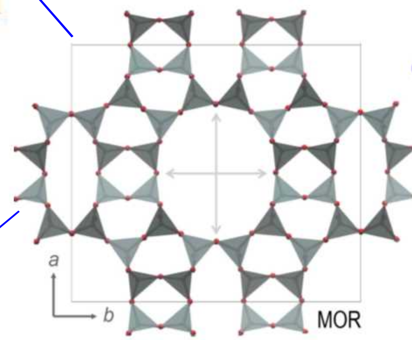
Core-Shell GCM Glass Waste Forms

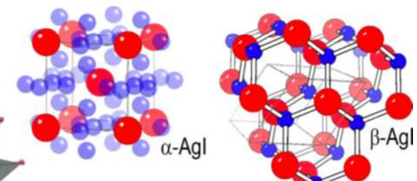
Glass shell, AgI/glass core, 75/25

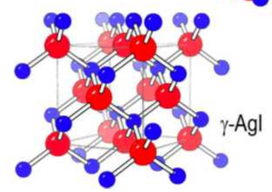


Glass shell,
AgI-MOR/Ag/Glass core 80/20/5

JACerS, **2011**, *94*(8), 2412

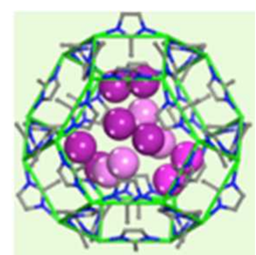




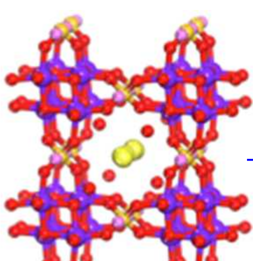


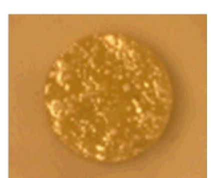
AgI bp 556° C

I_2/MOF, Isolation to Waste Form
JACS, **2011**,*133(32)*,12398
Ind. Eng. Chem. Res (Invited Article)
2012, *51*(2), *614*



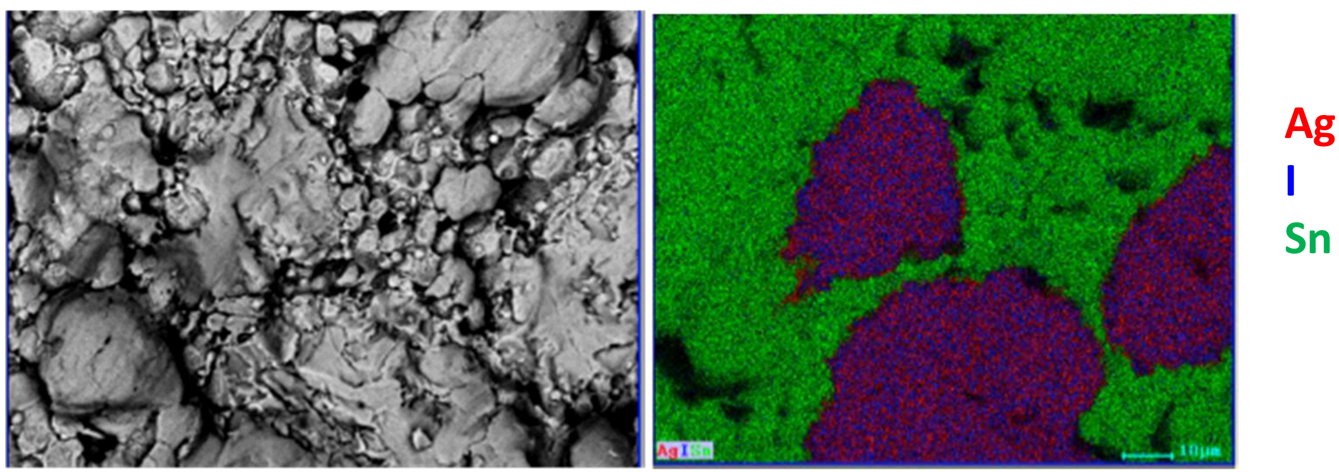
Cs-CST in Low Temp Glass Waste Form, No Cs Loss in Sintering





Metal Matrix Waste Form

- Highly attractive to encapsulate iodine-loaded zeolites and MOFs due to low temperature processability.
- This methodology prevents the use of expensive Ag for both the getter material and the waste form.
- Potential to incorporate a very high capacity of iodine into a final waste form.
- Waste form durability testing procedures need to be established.



SEM-EDS image of Sn with 25% AgI waste form

Nenoff, et. al., Patents Awarded and Pending Related to Nuclear Fuel and Legacy Waste

U.S. PATENTS
Awarded/Filed

- Cesium Silicotitanates for Ion Exchange and Waste Storage, 6,482,380, November 19, 2002.
- Niobate-based octahedral molecular sieves, 6,596,254, July 22, 2003.
- Niobate-based octahedral molecular sieves, 7,122,164, October 17, 2006.
- Low Sintering Temperature Glass Waste Form for Sequestering Radioactive Iodine, 8,262,950, September 11, 2012
- Mixed-Layered Bismuth-Oxygen-Iodine Materials for Capture and Waste Disposal of Radioactive Iodine, 8,383,021, February 2013

Applications

- An Inexpensive Method for bulk synthesis and Commercial Scale up of SOMS: Sandia Octahedral Molecular Sieves (2006)
- Pelletized Molecular Sieves and Method of Making Pelletized Molecular Sieves (Nonprovisional Patent Application, SD11971), 11/07/12.
- Metal Matrix Waste Forms for Fission Products. (Aug 2013)

Interested in learning more about Tech Transfer Possibilities in
(1) getter materials, (2) waste forms and (3) engineered/ pelletized getter materials