

# **Sandia Materials for the Capture, Storage or Purification of Cs, Sr and Fission Gases**

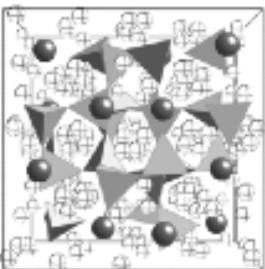
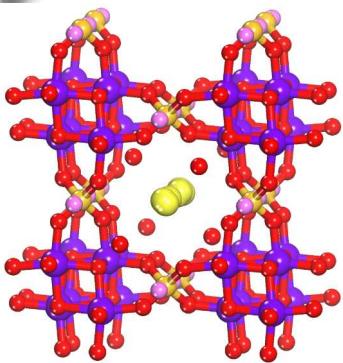
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Albuquerque, NM 87185

**Areva - SNL Visit  
December 19, 2013**

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

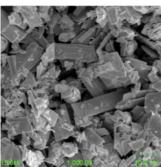
Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

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



*CST, Cs<sup>+</sup> removal from water to Pollucite Waste Form*

*Applied Geochem, 2011, 26, 57*



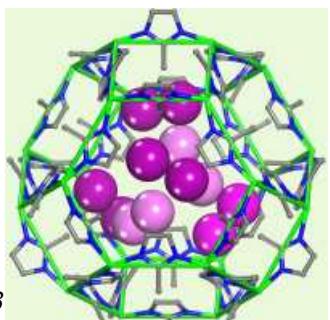
*In-situ Iodine removal from water*

*I<sub>2</sub>/MOF, Isolation to Waste Form*

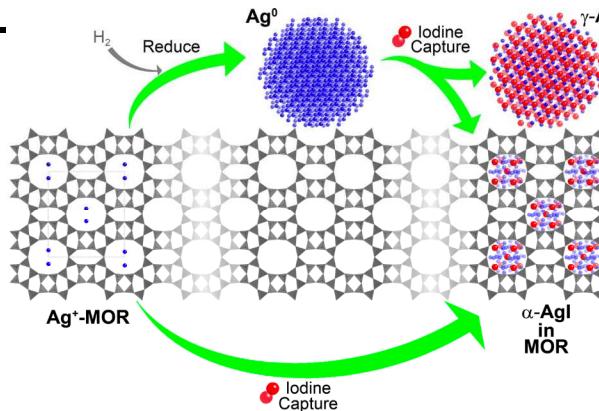
*JACS, 2011, 133(32), 12398*

*Ind. Eng. Chem. Res, 2012, 51(2), 614*

*Provisional Patent Oct 2013*

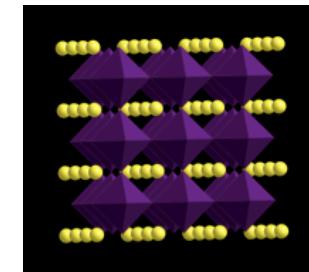
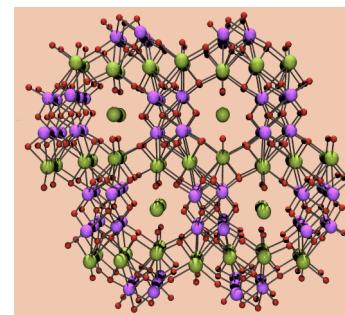


Areva 12/19/13

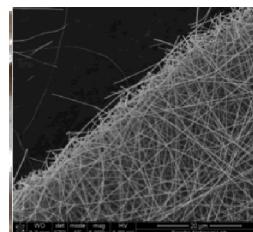


*Ag-MOR  
I<sub>2</sub>(g) capture & mechanisms*

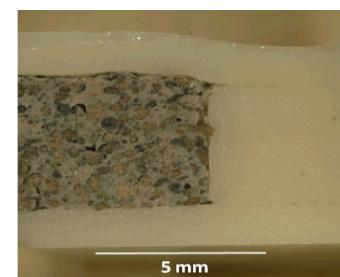
*JACS, 2010, 132(26), 8897  
J Phys Chem Letters, 2011, 2, 2742*



*Sr<sup>2+</sup> getter, 1-step to Perovskite waste form*  
*JACS, 2002, 124(3), 1704*



*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*

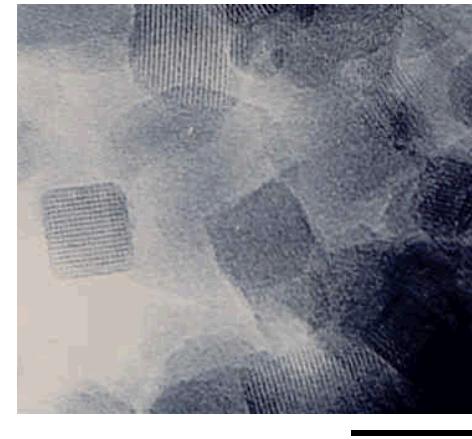
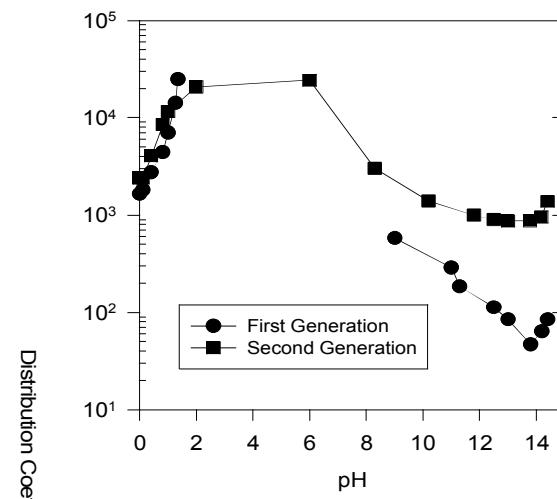
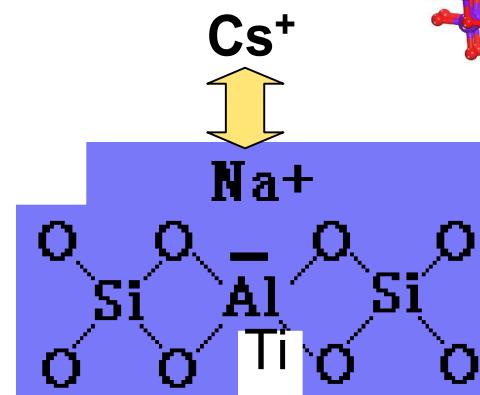
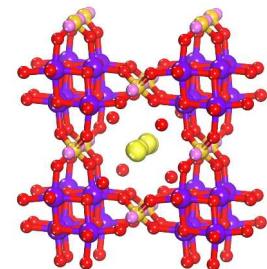
# Removal of Rad-Cs<sup>+</sup> from Pooled Seawater (heat treat to Pollucite WF or add into SNL GCM)

## Crystalline Silicotitanates (CSTs)

With exceptional Cs<sup>+</sup> 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*



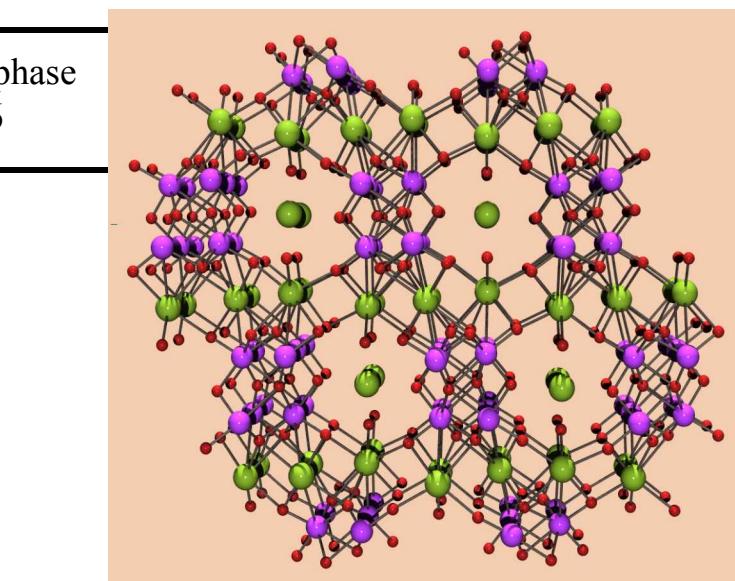
# Sandia Octahedral Molecular Sieves (SOMS) Selectivity (heat treat to perovskite WF)



Radionuclides  
High concentration  
in natural systems

Industrial  
Waste  
Metals

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



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

\* 0.1 ppm detection limit

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

# Long Term Storage of I<sub>2</sub> Capture Materials: Waste Forms

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

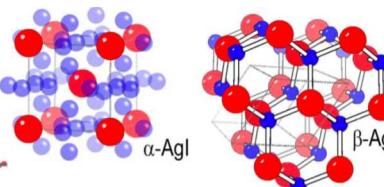
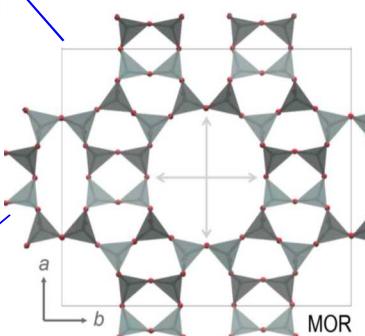
JACerS, 2011, 94(8), 2412

All These waste forms have been made with the SNL Low Temperature Sintering Oxide Glass

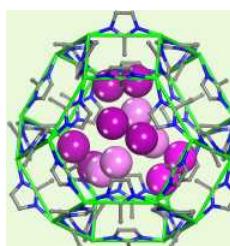
No HIP-ping needed: Sintering 550°C

Accepting of all types of rad-loaded getters:

zeolites (AgI-MOR), Metals (AgI), MOFs (I<sub>2</sub>-MOF), and Cs-CSTs  
Durability studies: equal to better performance than basalt glass



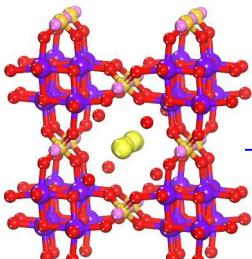
AgI bp 556°C



I<sub>2</sub>/MOF, Isolation  
to Waste Form

JACS, 2011, 133(32), 12398

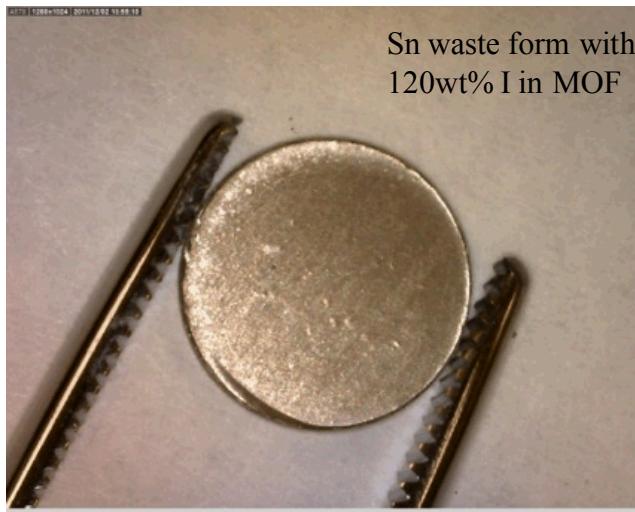
Ind. Eng. Chem. Res (Invited Article)  
2012, 51(2), 614



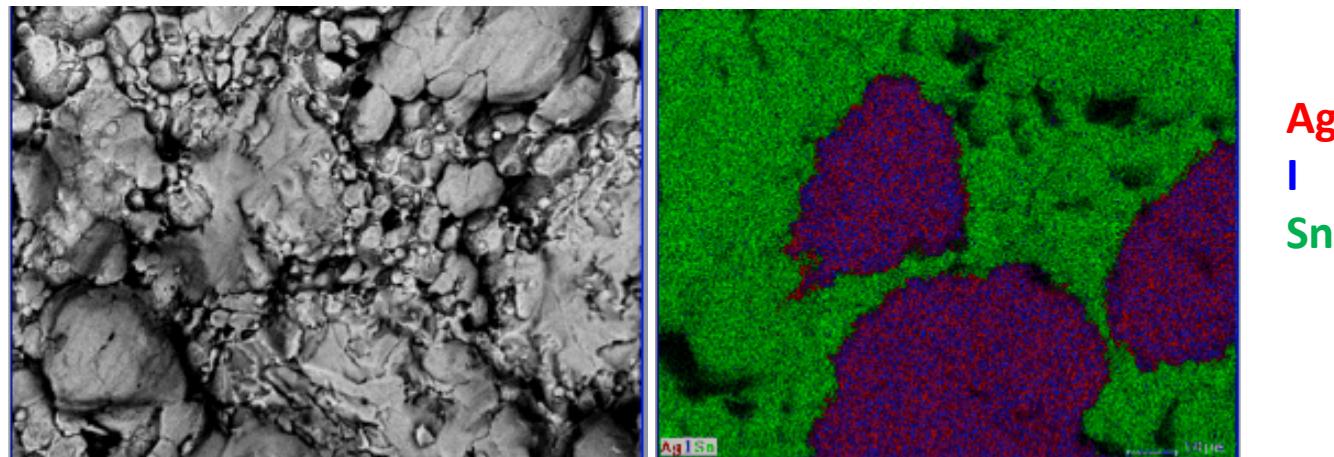
Cs-CST in Low Temp Glass  
Waste Form, No Cs Loss in Sintering



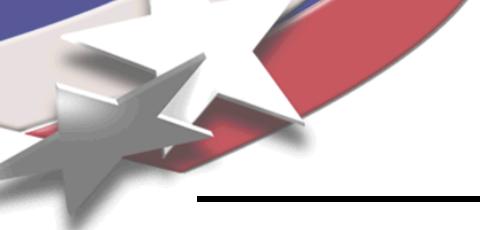
# Metal Matrix Waste Form



- 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

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## 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)

Working with Dan Jenkins and Brooke Garcia for Tech Transfer Possibilities in (1) waste forms and (2) engineered/ pelletized getter materials, along with others