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Use of Nanocomposite Materials (SNL-NCP) to Entrap and Immobilize Highly Volatile/Soluble Radionuclides

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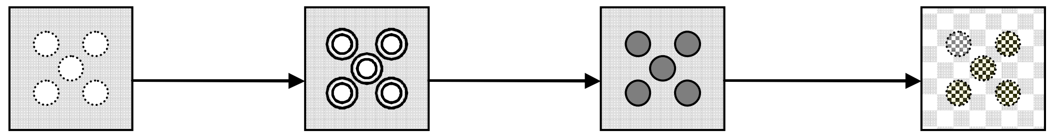
Technical Basis & Objective

- **Technical basis:** Under the support of LDRD, we have developed a suite of inorganic nanocomposite materials (SNL-NCP) that can effectively entrap various radionuclides, especially for I-129 and Tc-99 (Patents pending).
- **Objective:** We propose to further evaluate and optimize SNL-NCP materials for entrapping and immobilizing I-129 and radioactive noble gases (Kr and Xe).

Development of a New Generation Waste Forms through Nano-immobilization & Nano-encapsulation

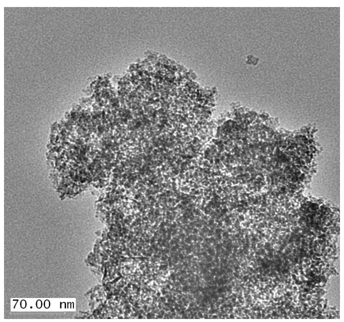


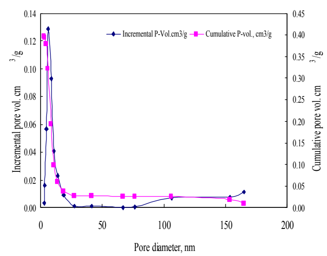
Nanoporous material

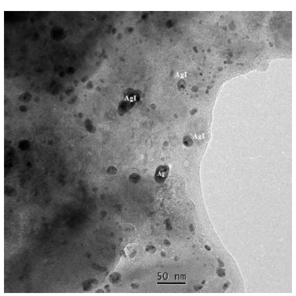
Functionalized pore surface

Sorption & immobilization

Fixation & encapsulation







Wang et al. (to be submitted); 1 patent pending; 1 patent in preparation

Why Nano-scale?

- Novel sorption capability: e.g. I-129, Kr & Xe
- Fast sorption kinetics
  - Getter materials
  - Membrane separation
- Easy to encapsulate
  - Durability: host minerals
  - Flexibility
- Mechanic strength
- Chemical durability

Nanopore Structures & Radionuclide Retention

Material	I sorption (ppm)	% of I lost during fixation	% of I lost during vitrification
Particles	98	~100%	~100%
Activated particles	8700	45	65
Nanoporous material	25000	~ 0	~ 0

- Sorption of I onto nanoporous silica is not detectable, surface chemistry is an important factor controlling RN sorption
- Nanoporous structures not only enhance I sorption but also help to retain I during the fixation and encapsulation.

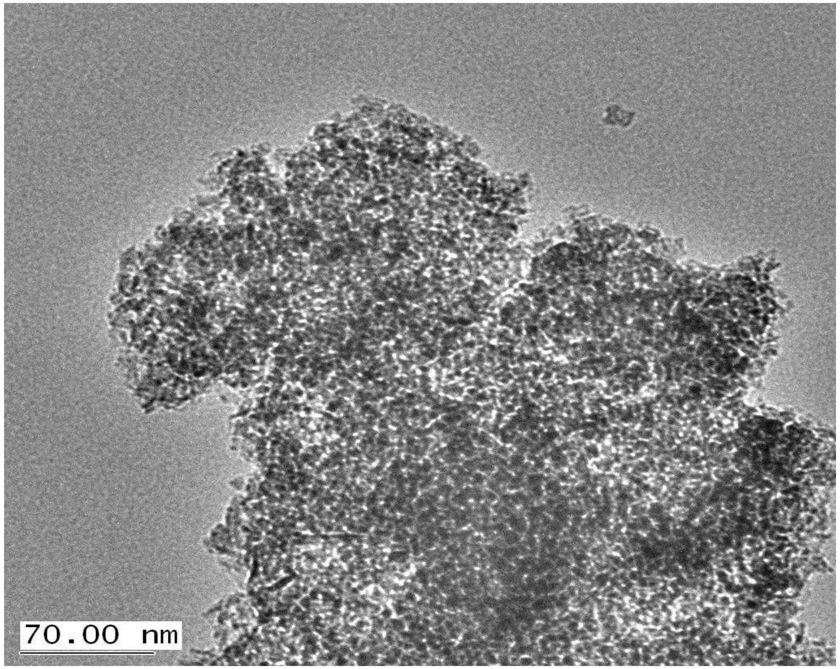
- No silver is needed for I retention.

Material Synthesis

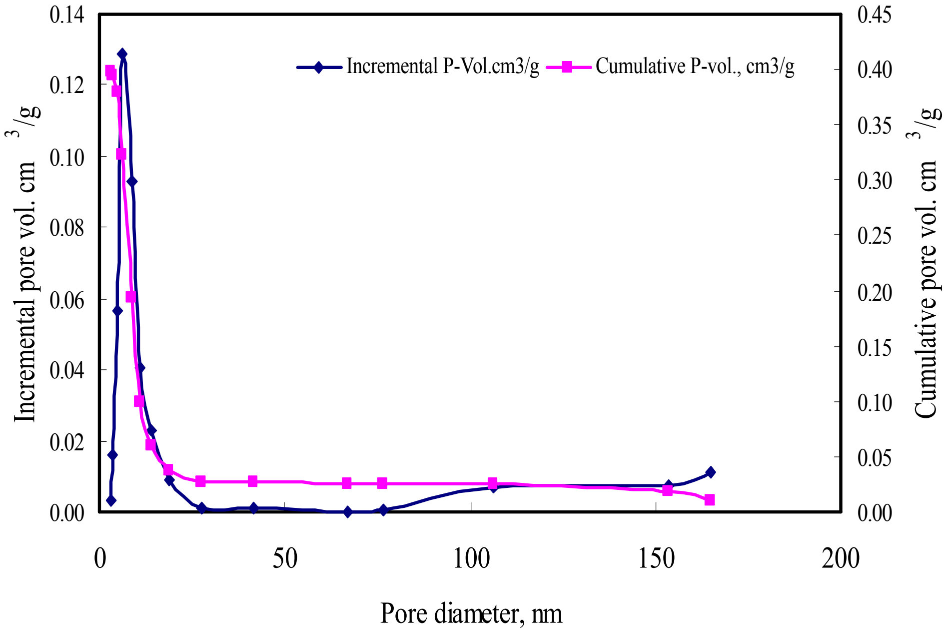
- General route for synthesizing nanoporous metal oxides
  - Based on a sol-gel method
  - Inorganic precursors & block copolymer (as a structural template)
  - Inexpensive, scalable for a large quantity production
- One-pot synthesis
  - Multiple metal oxides
  - Compositional & structural homogeneity ensured
- Formation of monolith
  - Preferred for material handling
- Total > 300 materials synthesized and tested

U.S. Patent No. 7,238,288; two pending patents

Material Synthesis (cont.)



TEM image of nanoporous double metal oxide synthesized using the one-pot route, showing worm-like pore structures.



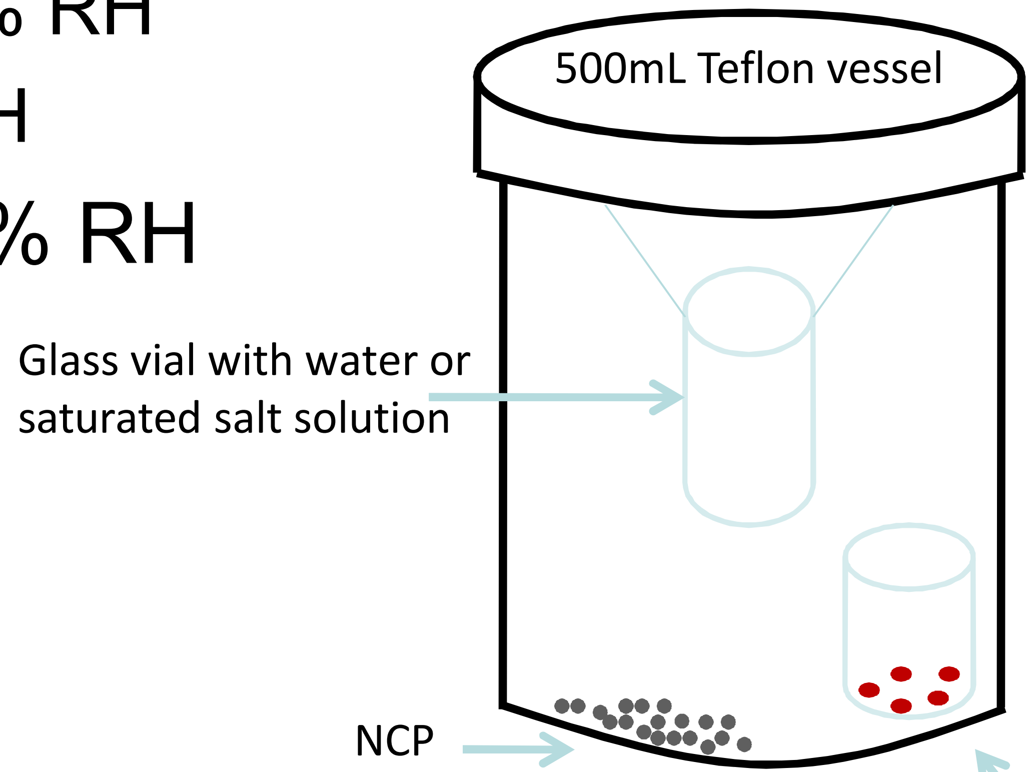
BET measurements of nanoporous double metal oxide

Iodine Sorption Tests: Materials Tested

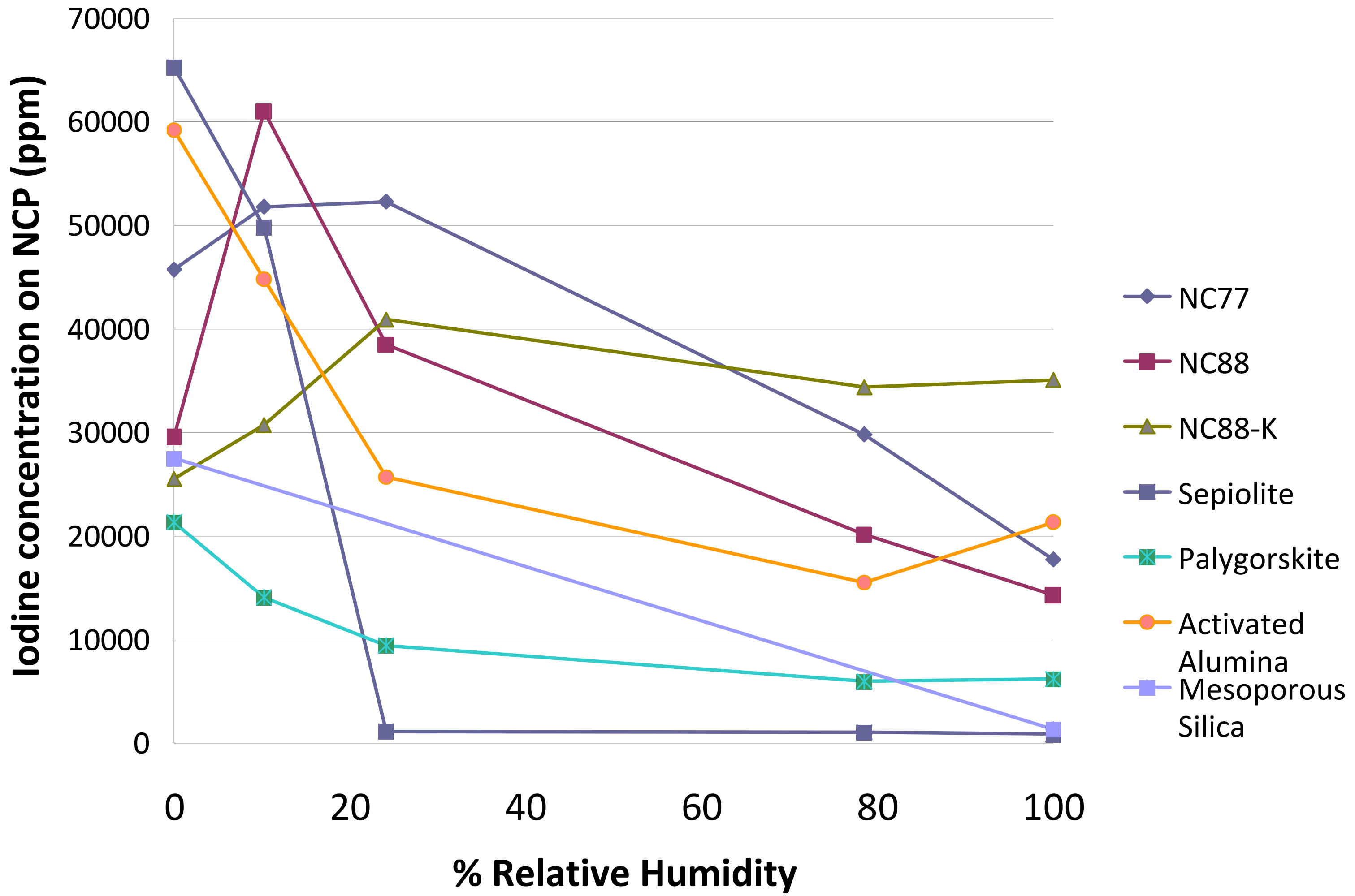
- SNL-NCP materials
  - NC-77
  - NC-88: Two preparations
  - Mesoporous Silica
- Other materials
  - Sepiolite
  - Palygorskite
  - Activated Alumina

Relative Humidity Testing

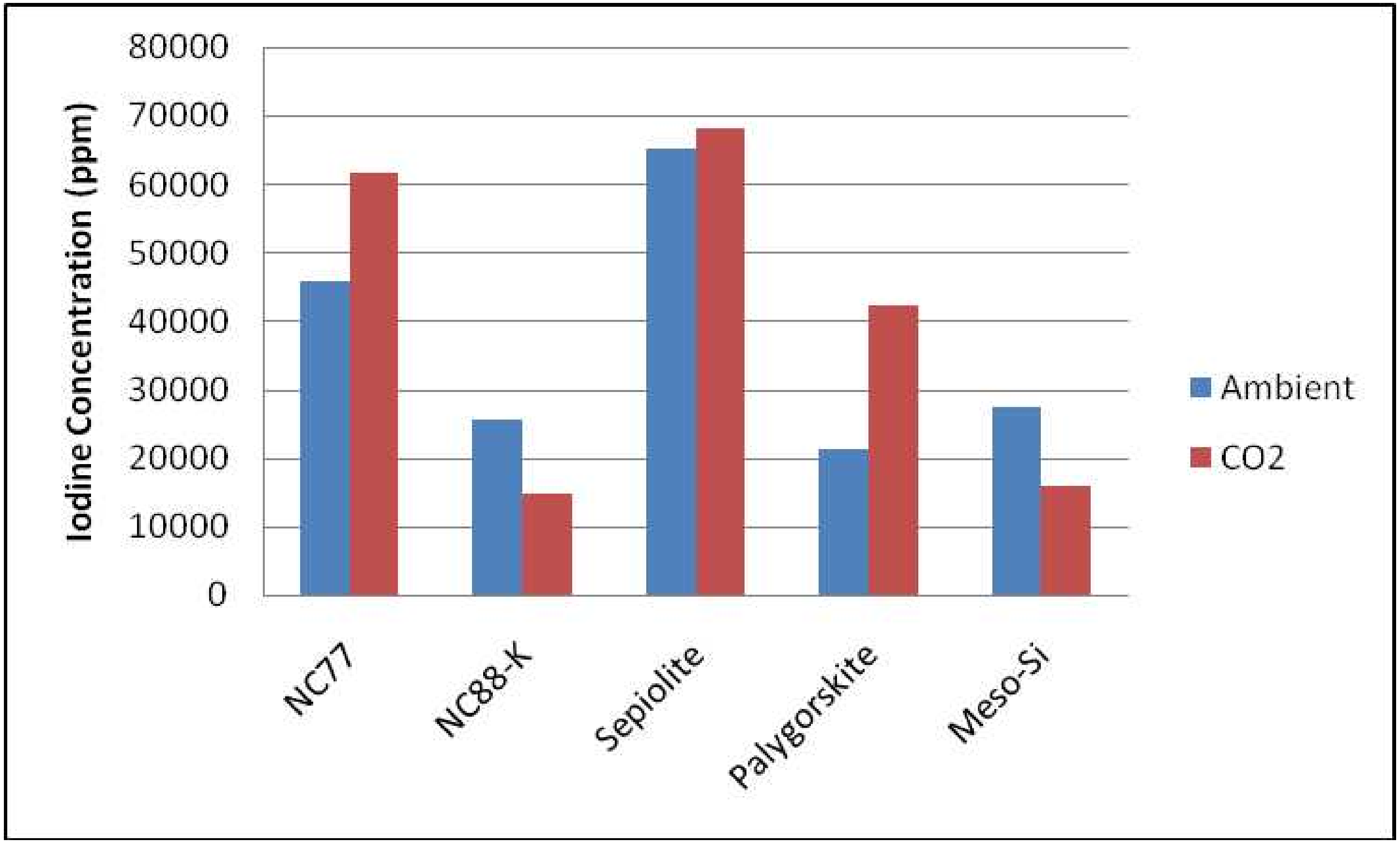
- Saturated Salt Solutions
  - LiCl = 10.23% RH
  - $MgCl_2$ = 24.12% RH
  - KCl = 78.5% RH
- DI Water = 100% RH



I_2 Sorption: Effect of Humidity



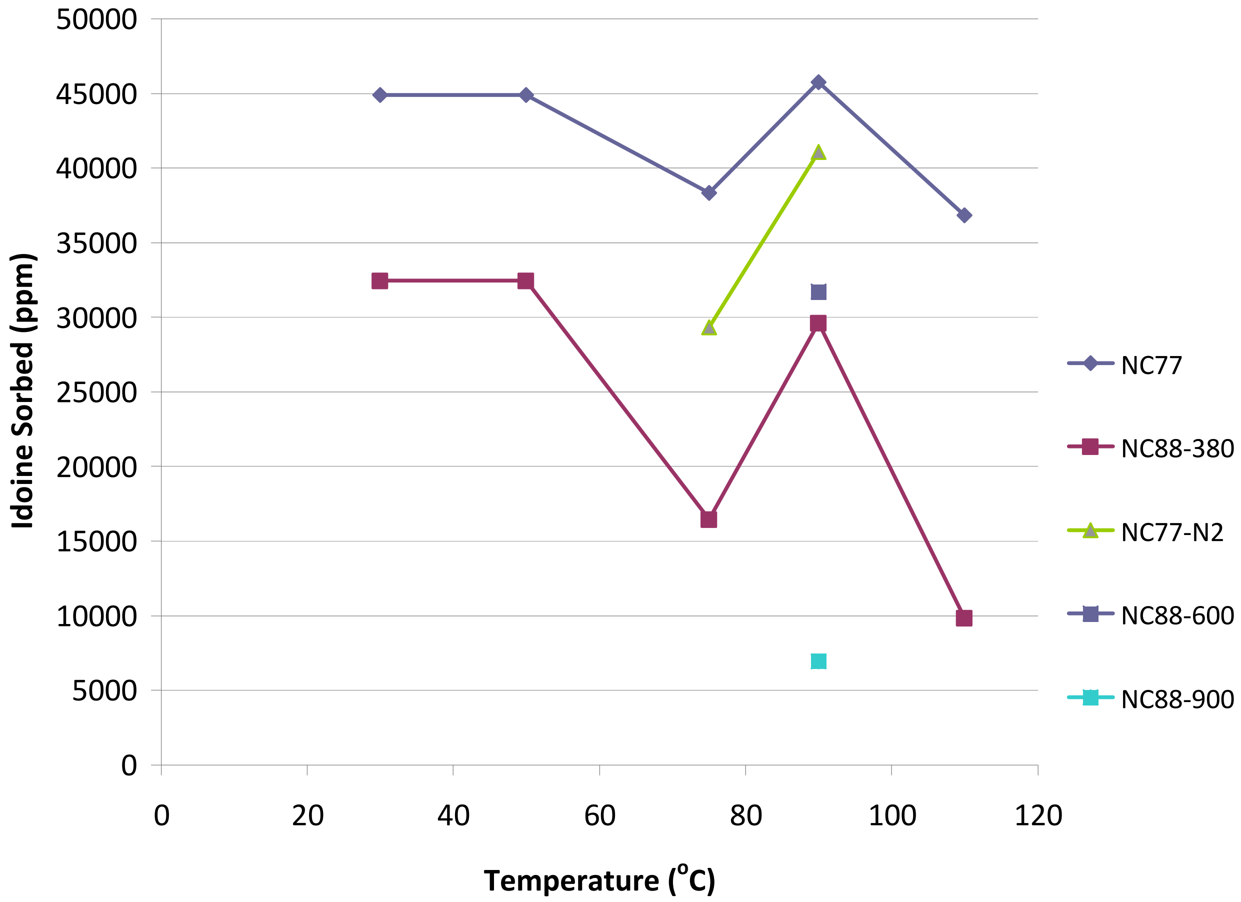
I_2 Sorption: Effect of CO_2



I_2 Sorption: Effect of N_2O

	Ambient	CO_2	N2O
NC77	45728	61814.44	32987.32
NC88-380	25536	14907.62	15773.96
NC88-600			34808.18
Sepiolite	65270	68101.24	
Palygorskit	21343	42162.79	
Meso-Si	27496.25368	15774.66	

I_2 Sorption: Temperature Effect



Kr & Xe Sorption Tests

- TGA analysis
- Two stages:
  - Degassing

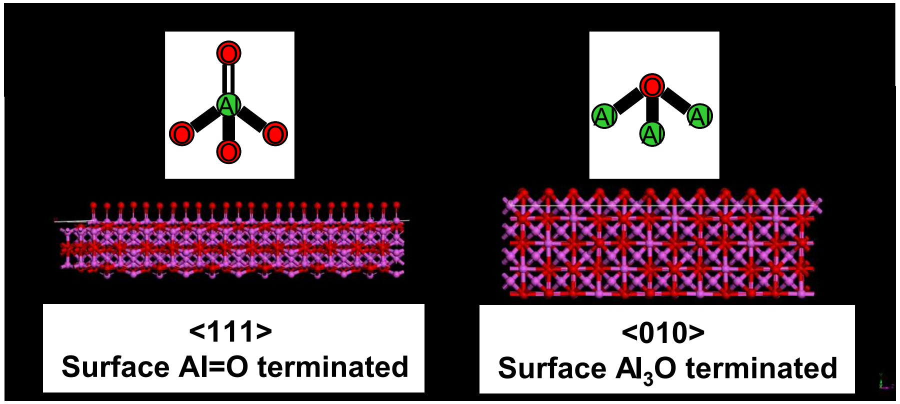
Ar sorption @ room temperature & 0.9 atm

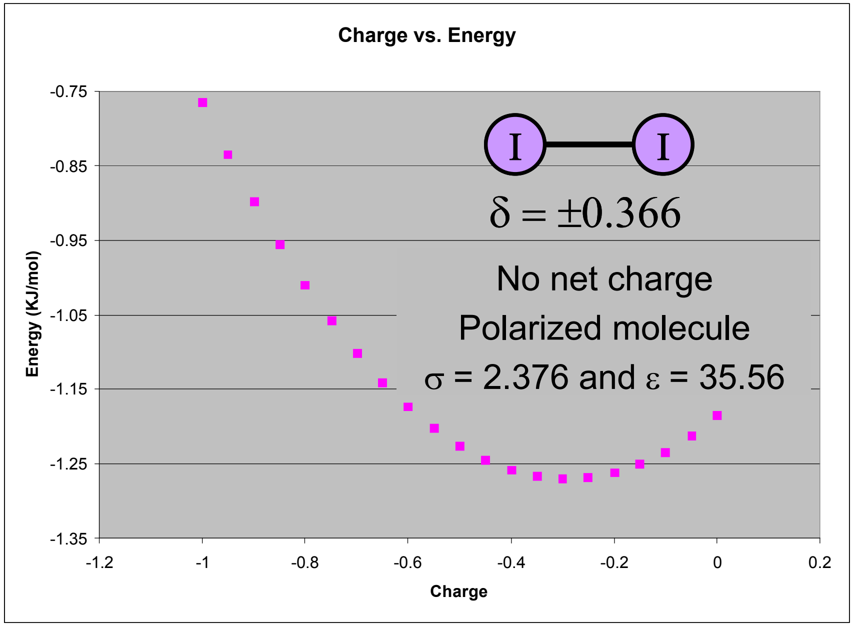
Adsorbents	Weight percentage of noble gas adsorbed (wt.%)
Zeolite-M1	0.07
Zeolite-X1	0.08
NC 77 (SNL Nanocomposite)	0.33
NC 95 (unground) (SNL Nanocomposite)	0.23
NC 95 (ground) (SNL Nanocomposite)	0.31
S51HF (Nanostructured carbon)	0.68
Hydromagnesite	0.02
Carbon-D (Nanostructured carbon)	0.72
SNL-CBD-1 (SNL Composite)	0.42±0.06
SNL-CBD-2 (SNL Composite)	0.37±0.01
S159-2-B (unground)	0.20
Microporous-P	0.26
Microporous-S	0.36
Zeolite-N	0.27
Zeolite-NA	0.12

Kr sorption @ room temperature & 0.9 atm

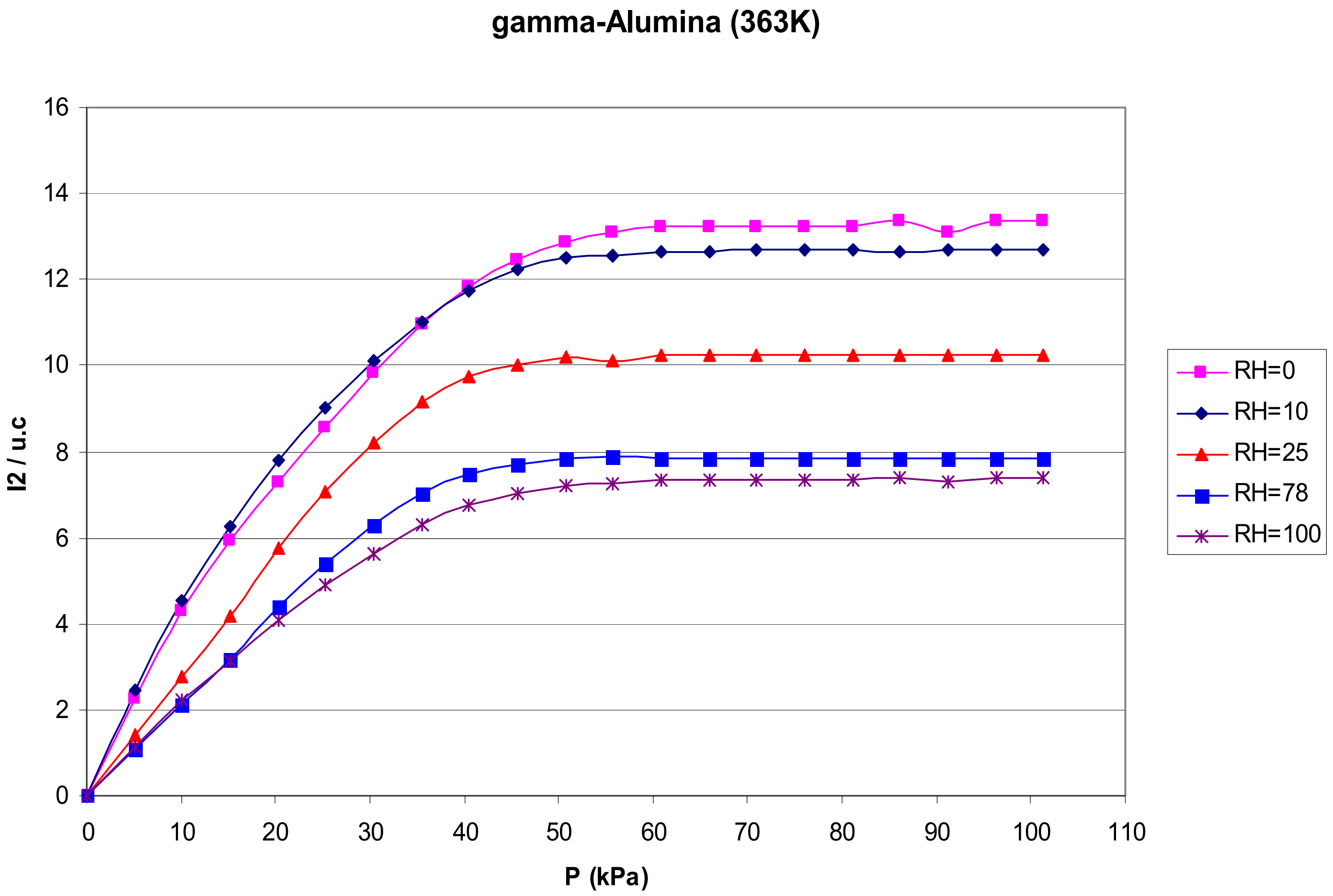
Adsorbents	Weight percentage of noble gas adsorbed (wt.%)
Zeolite-M	0.36±0.08
Zeolite-X	0.64
NC 77	0.24
Hydromagnesite	0.05
Carbon-D	3.60
SNL-CBD-1	3.01
SNL-CBD-2	2.51
Microporous-S	0.71
SNL-CBD-3^{A}	1.93
SNL-CBD-3B^{A}	1.97
SNL-CBD-3C	2.56
SNL-CBD-4	1.63
SNL-CBD-5	2.69
Zeolite-N	1.74
Microporous-P	1.03
S159-2-B (Surface-grafted NC)	0.30
S185-4 (Surface-grafted NC)	0.30
S185-5 (Surface-grafted NC)	0.42

Model System for Iodine Sorption





Molecular Dynamic Modeling: Iodine Sorption



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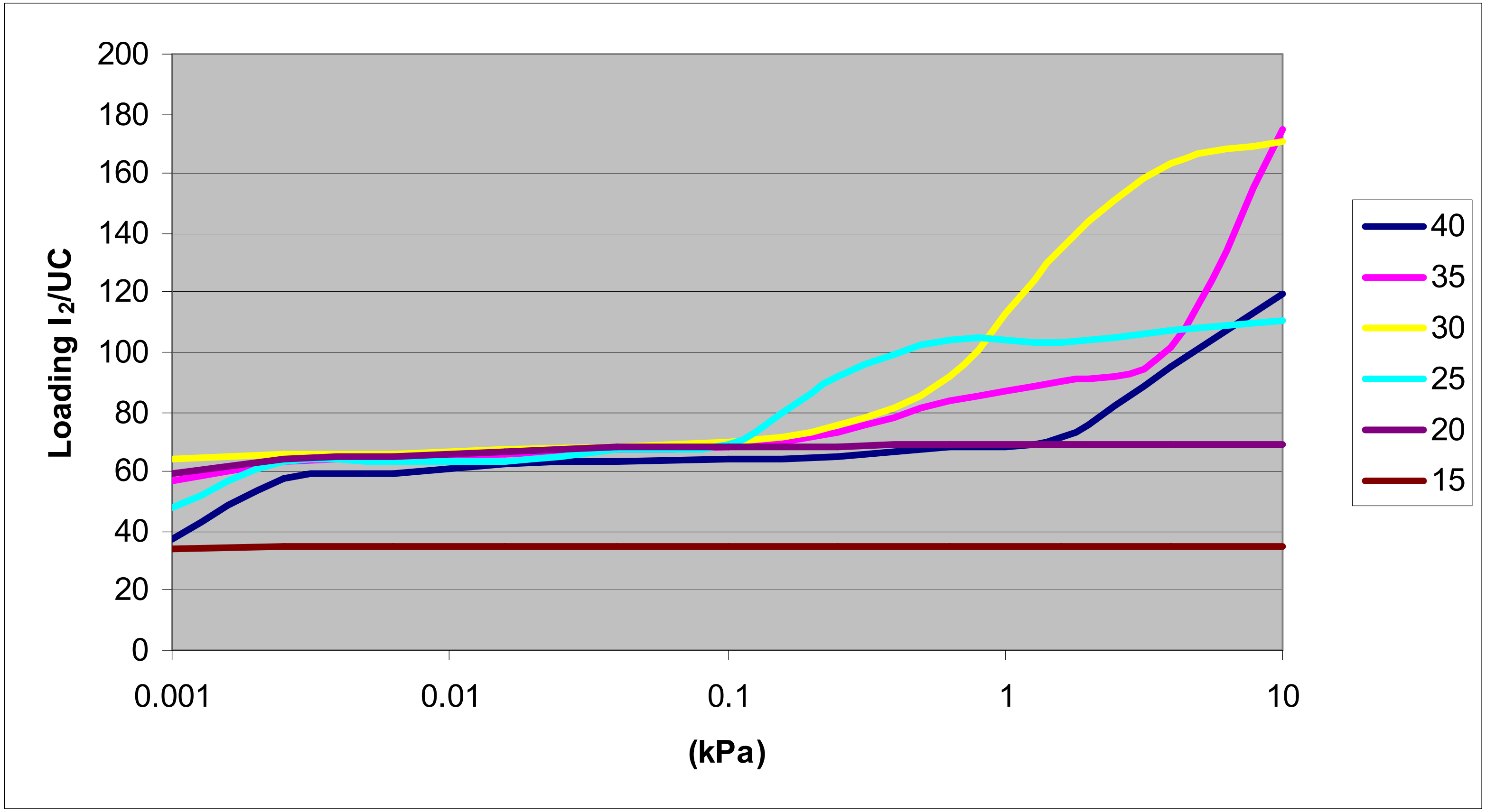
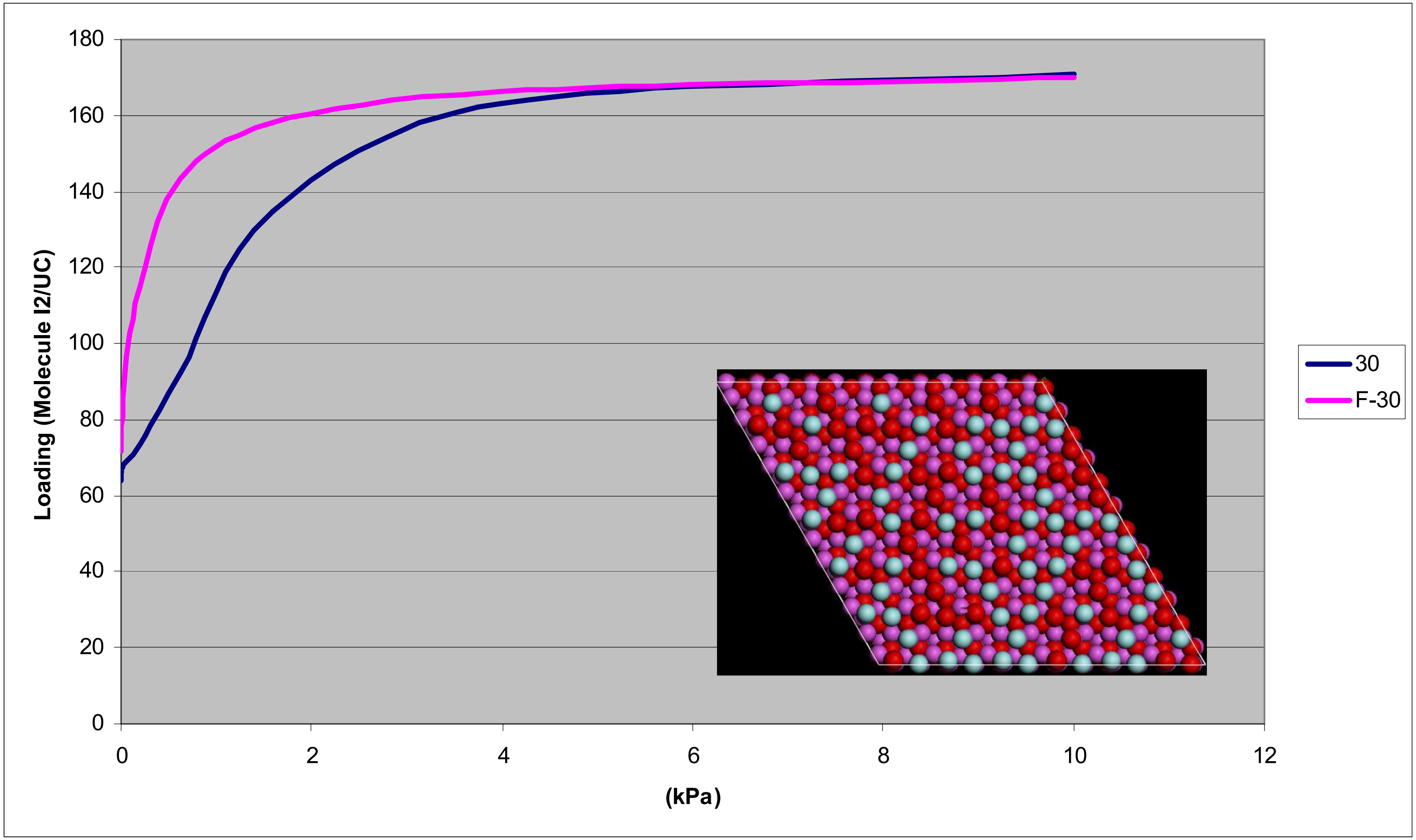
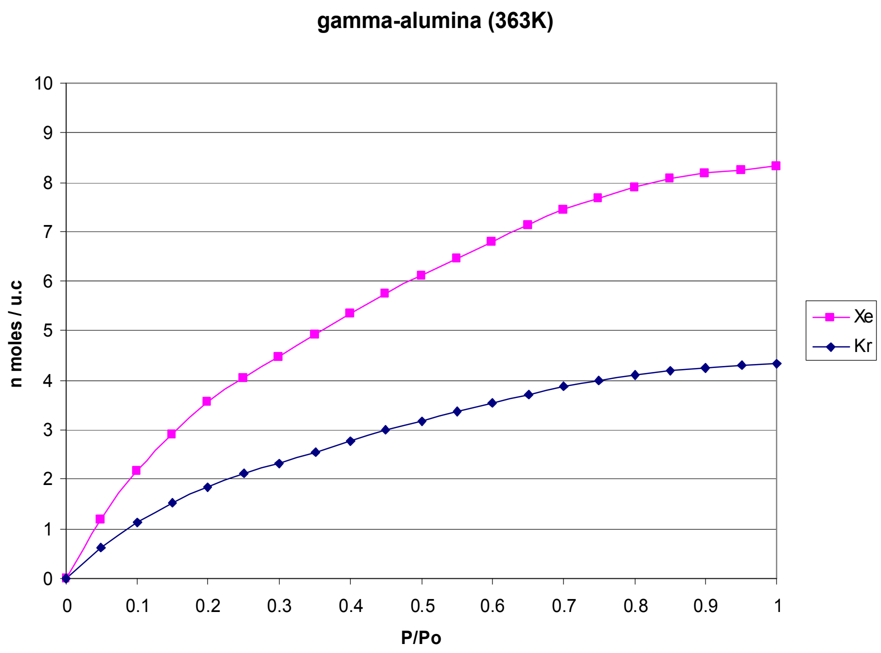
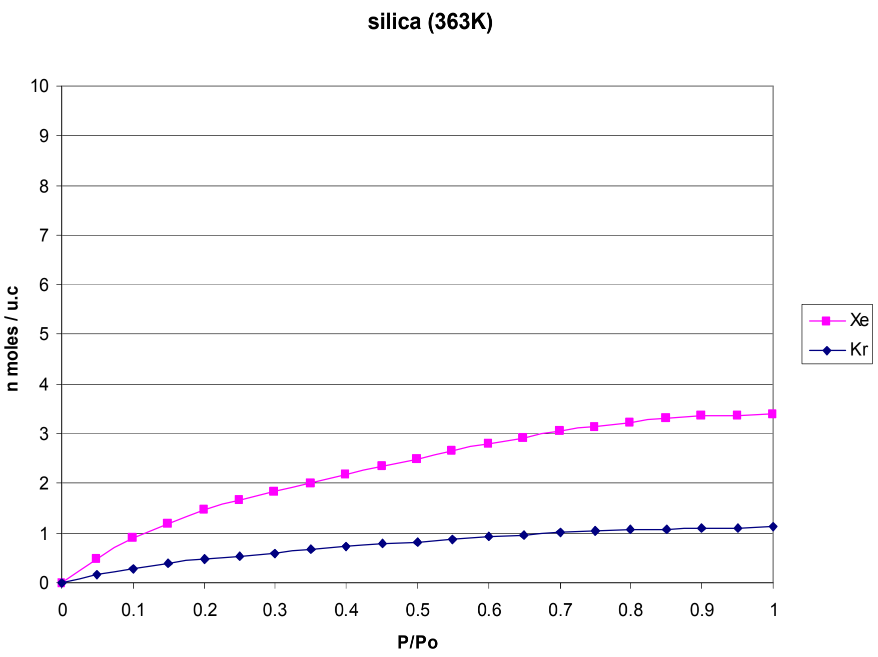


Molecular Dynamic Modeling: Iodine Sorption

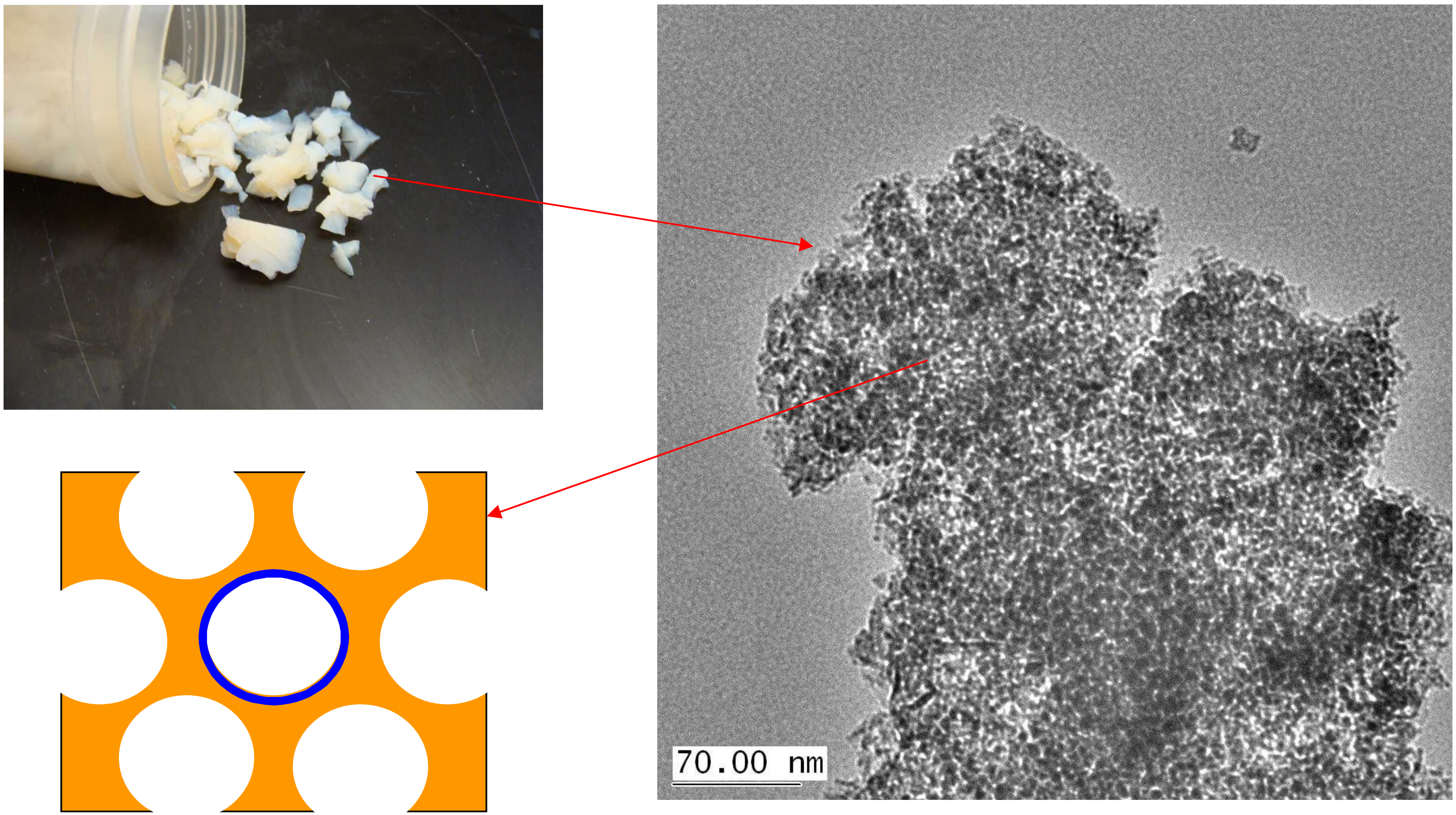


Molecular Dynamic Modeling: Kr & Xe Sorption





Functionalized Nanoporous Materials



Ongoing & Future Work

- Synthesis & optimization of NCP materials
  - Enhancement of Kr & Xe sorption
  - Two methods for nanopore surface functionalization under development.
- More tests for Gaseous Radionuclide sorption
  - Sorption kinetics
  - Collaboration with PNNL
- Mechanistic understanding of material performance
  - MD modeling