Xenon Abatement Studies

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Pacific Northwest National Laboratory

2018 Mo-99 Topical Meeting – Knoxville, TN

Outline

- PNNL's policy for engaging on medical isotope production
- PNNL work on noble gas capture
  - Adsorption Measurements
  - Adsorption Material Considerations
- Modeling Adsorption System
  - Model Equations and Assumptions
- Current progress
  - Work performed under current project

PNNL Policy for Engaging on Medical Isotope Production

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- PNNL has been involved with the mitigation of the effects of medical isotope production on the non-proliferation environment
  - In particular, emissions from fission-based Mo-99 production are known to be the dominant source of airborne radioactive isotope background
  - Emissions of xenon isotopes are particularly problematic for the detection of nuclear explosions

PNNL Policy for Engaging on Medical Isotope Production

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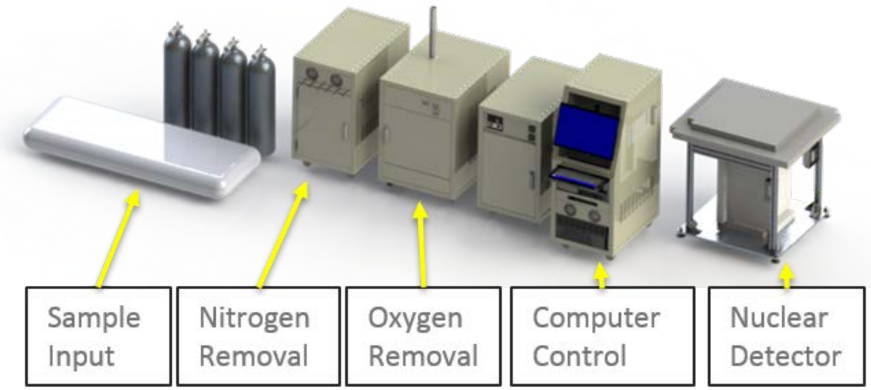
- To avoid conflicts of missions, we will continue to work with isotope producers with the following caveats. PNNL will:
  - Not permit the use of its facilities for the commercial production of Mo-99
  - Engage with domestic and international producers of medical isotopes to reduce the impact of production on nuclear explosion monitoring systems through the use of:
    - Emissions control systems
    - Stack monitoring
    - Other activities that may reduce emissions
  - Be open and transparent and provide all of our R&D on an equal basis to any producers interested in those results
    - Proprietary issues, will of course, be honored

Xenon Emissions Capture

- Investigation of emission capture technologies for SHINE's Mo-99 production facility
  - Task 1 - Evaluation of Adsorbent Materials (continuation of FY18 support)
    - Continued evaluation of new materials, such as silver mordenite and metal organic frameworks (MOFs) to make a compact design
    - Produce preliminary flow sheets
      - Designed to keep emissions below NRC requirements
  - Task 2 - Detailed Exploratory Testing to Verify Emissions Levels
    - Computational models and experiments may be used to test
      - Different adsorbents
      - Cooled abatement traps
      - Trapped gas management and long term storage
      - Trap regeneration
      - Trap design

PNNL Gas Processing Experience

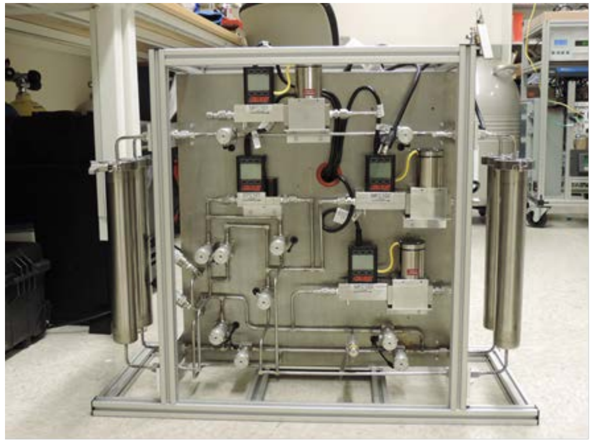
Radioxenon Sampler Analyzer (ARSA)



Argon 37 Field System

Xenon International

Microchannel Distillation



Dual Reflux Pressure Swing Adsorption

Adsorbent Investigations

- Isotherm data for activated carbons and other adsorbents being investigated are collected at several temperatures to allow calculation of the heat of adsorption
- Gases used for the initial round of data collection include xenon, nitrogen, CO_2 and H_2O
- Kinetic data is also collected
- Initial adsorbents include three coconut shell based activated carbons (Yakima, Alamo-Water, Nusorb GXK)
- Isotherm data for promising MOF’s will also be evaluated

Hiden Gravimetric Adsorption Analyzer

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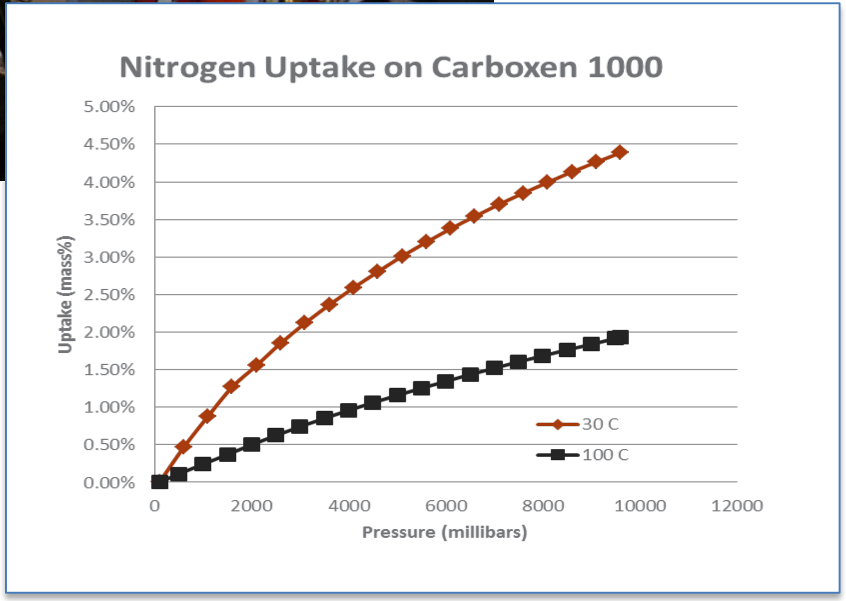
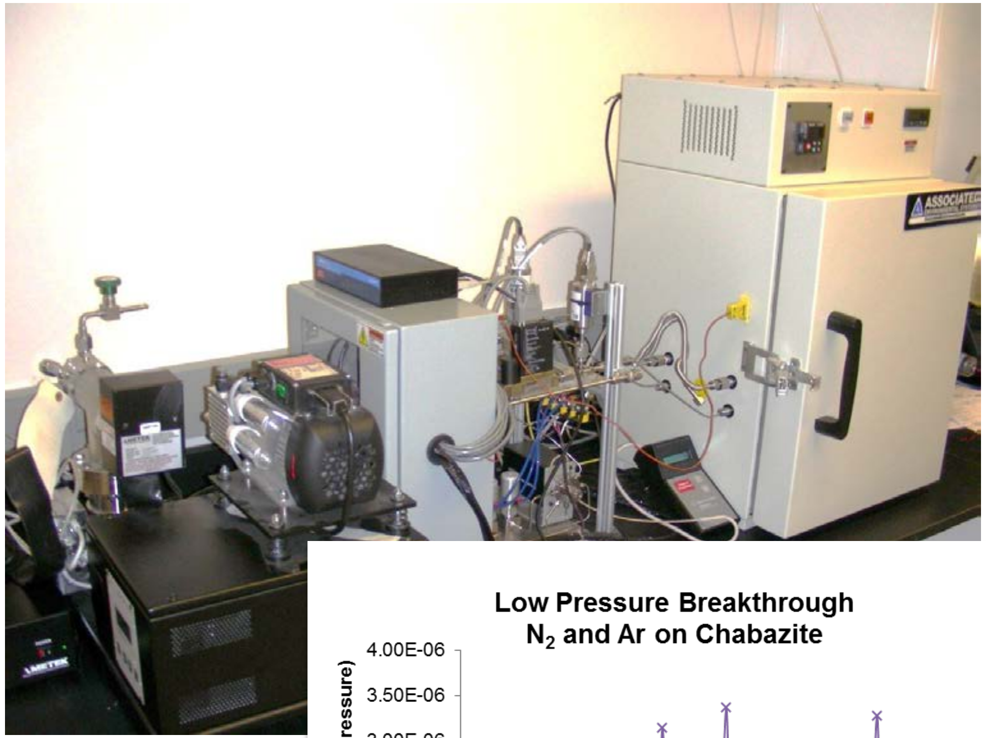
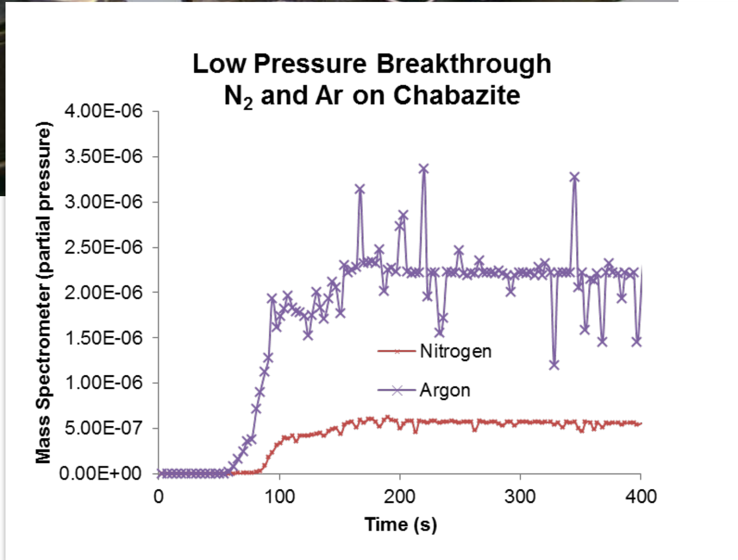
- This instrument can output the rate of uptake, as well as the final amount adsorbed at a given pressure and temperature.
- Able to take measurements at high temperatures as well as cryogenic temperatures.
- The rate of uptake is fit to a linear driving force model in the analysis software.
- All data collected is stored and can be used for future analysis.



Adsorption Breakthrough Instrument

- This instrument can perform breakthrough experiments for gas mixtures (up to three gas species and a carrier).
- Chromatography experiments can also be performed.
- Equilibrium data can be extracted from the breakthrough time.
- Kinetic data can be extracted from the slope of the breakthrough curve, or the shape of chromatographic peaks.
- Extraction of kinetic data requires fitting the experimental data to a model (Aspen Adsorption).





Activated Carbon Data

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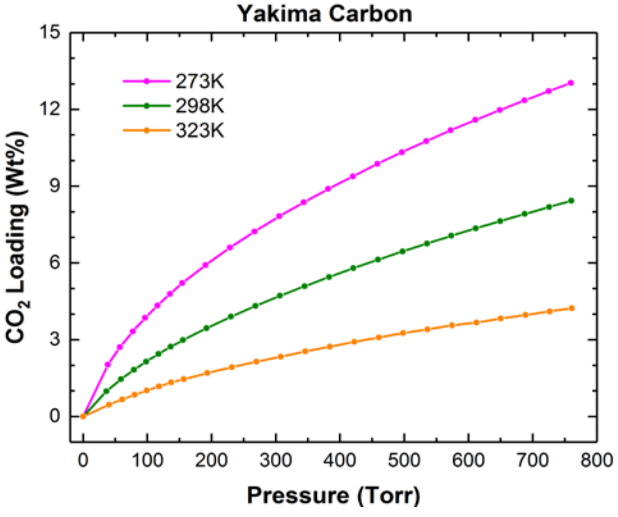
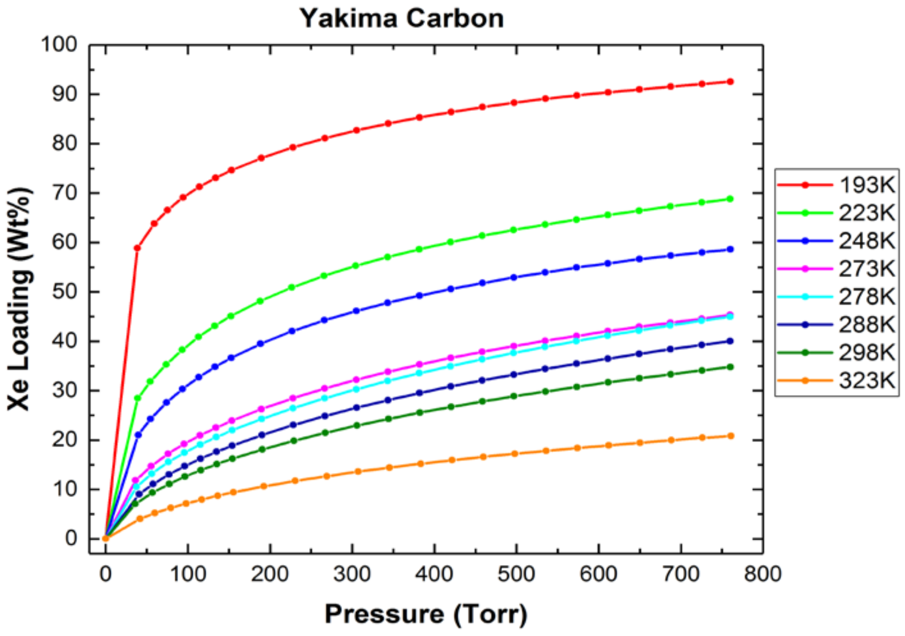
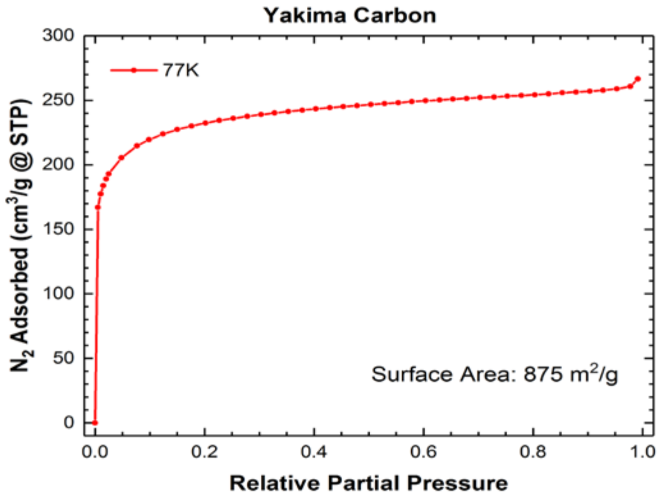
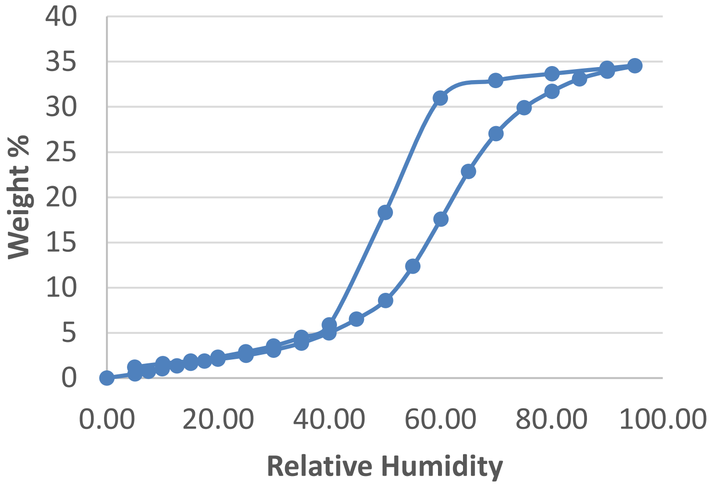
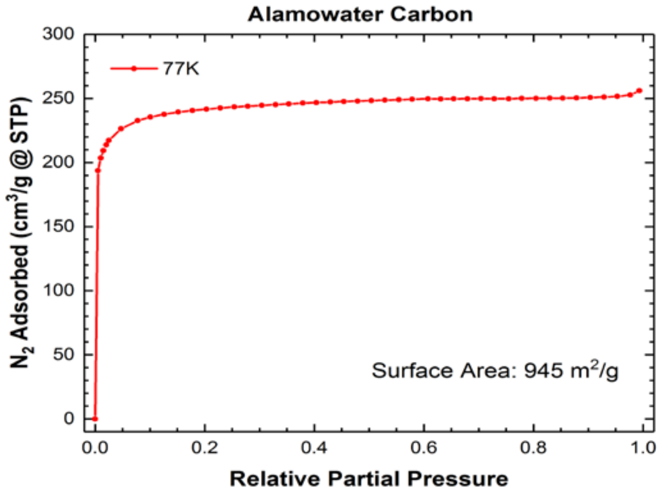






Water loading on Coconut Shell Activated Carbon (Yakima)





Adsorbent Performance based on competitive isotherms (nitrogen and xenon)

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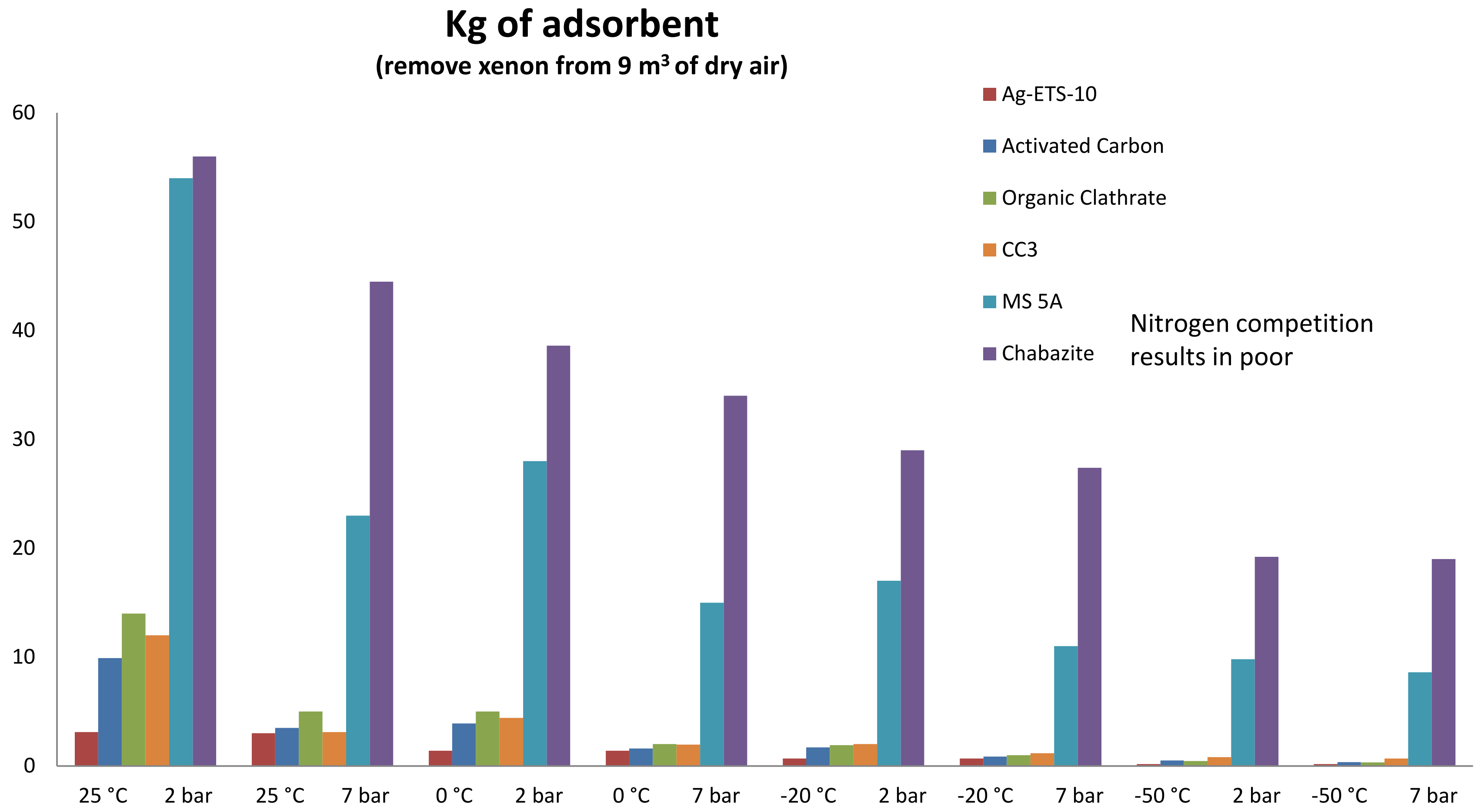
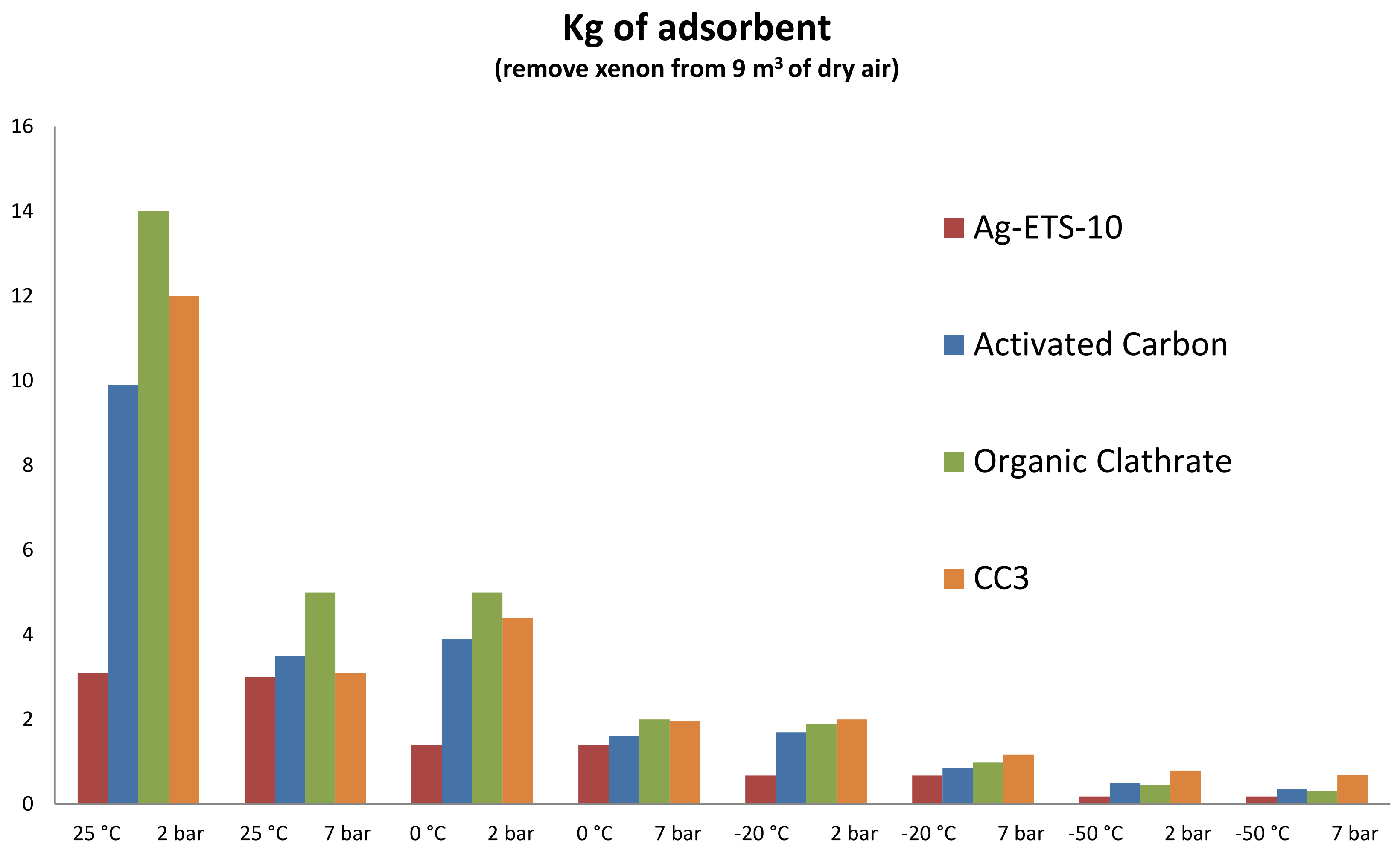


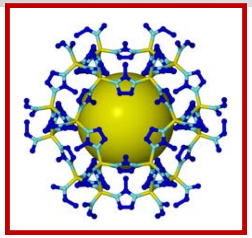
Nitrogen competition results in poor

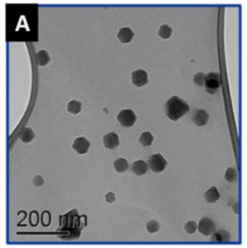
Adsorbent Performance based on Competitive Isotherms Modeling



Metal Organic Frameworks

- MOFs are constructed from precursor chemicals that in most cases are readily available
- Mild synthesis conditions provide options for in-house production as desired
- MOFs can be made in many different forms as needed:
  - Bulk powders
  - Nanoparticles
  - Fibers
  - Gels

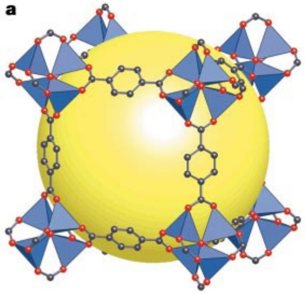




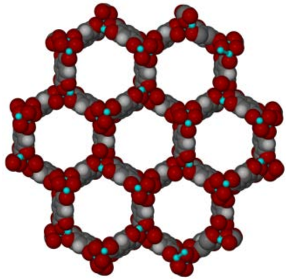
Nanoparticles

MOF Gel

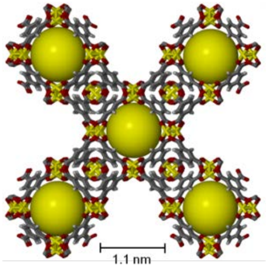
MOF Fibers



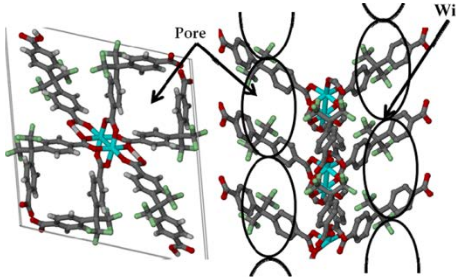
MOF-5 (Zn)



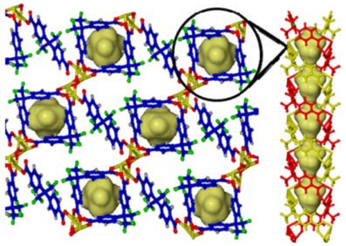
NiDOBDC



CuBTC

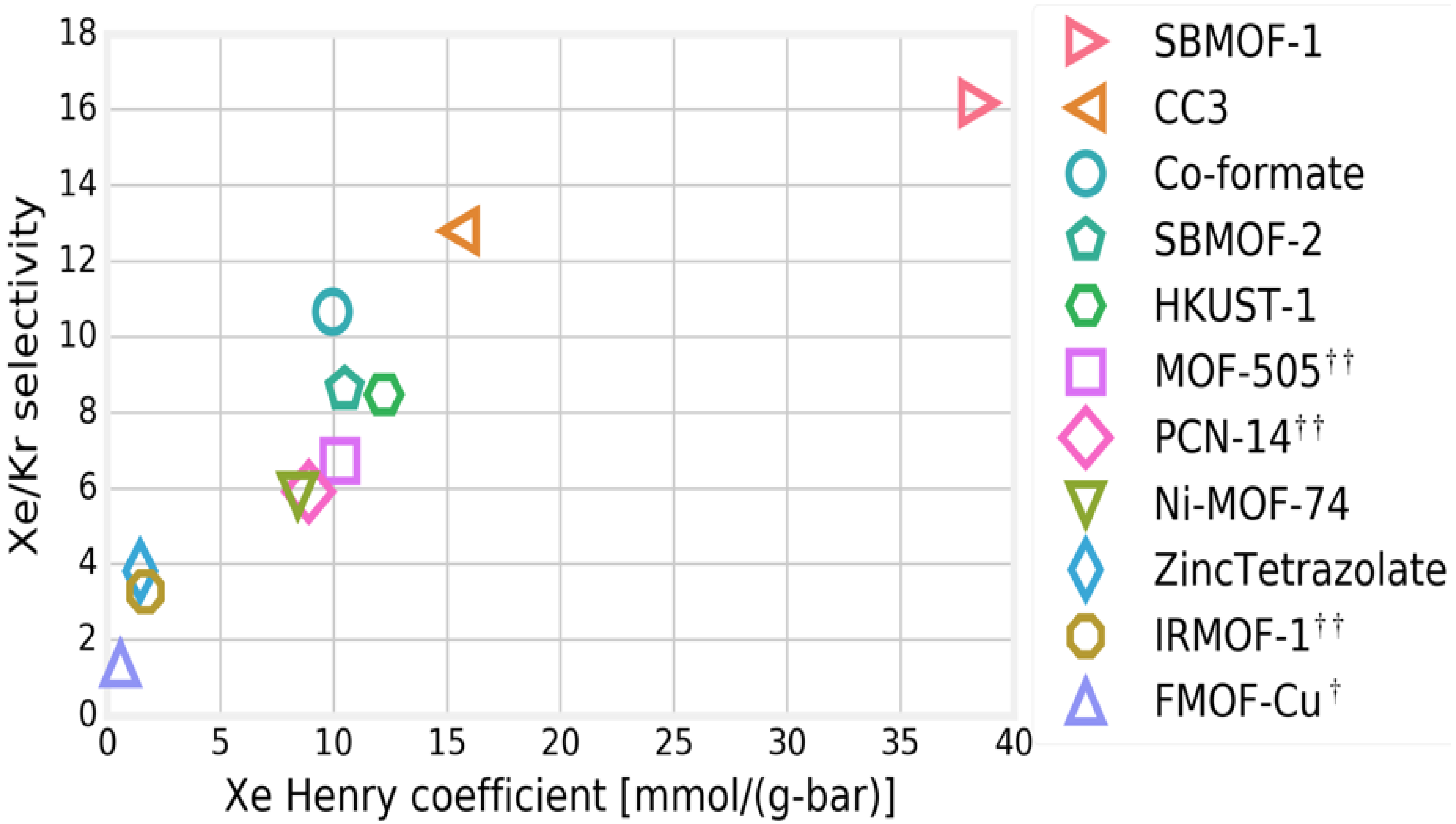


FMOFCu



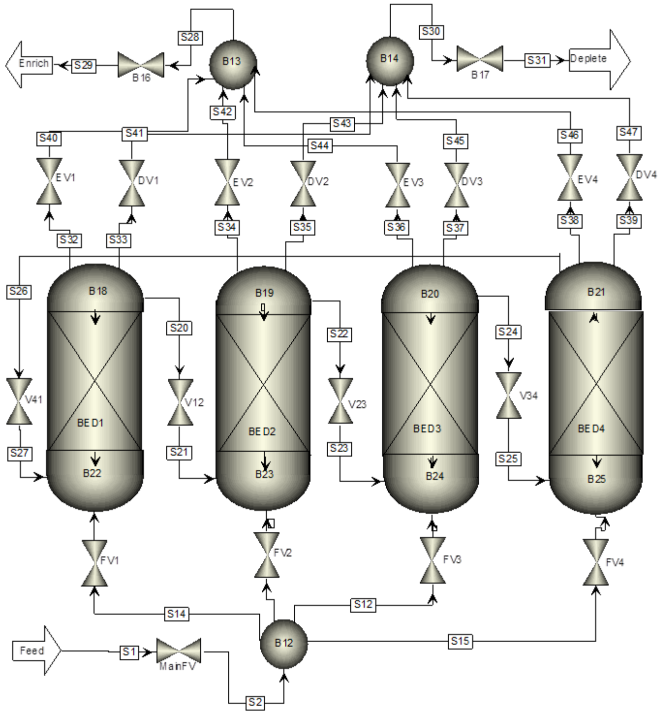
FMOFZn

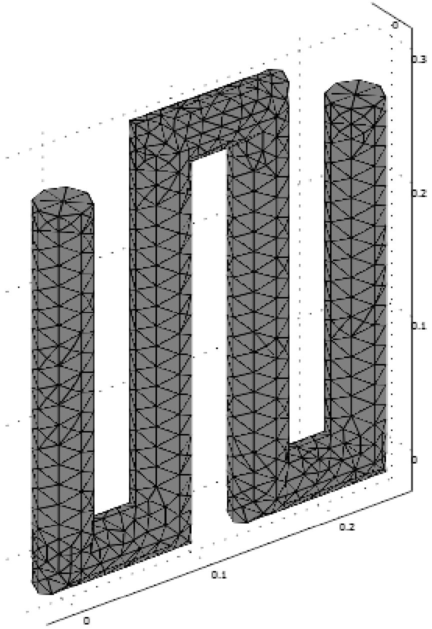
SBMOF-1 vs other MOFs

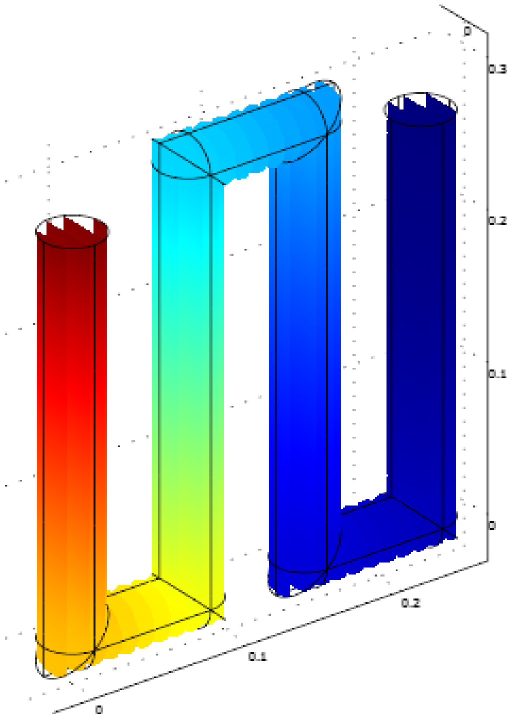


Adsorption Process Simulation

- Aspen Adsorption
  - Industry standard for modeling gas adsorption processes
- COMSOL Multiphysics
  - Finite element software, allows direct implementation and modification of model equations
  - Allows the modeling of complex bed geometries







Finite Element Modeling Equations

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Gas phase mass balance for each species

$$\underbrace{\varepsilon \frac{\partial c_i}{\partial t}}_{\text{Time derivative gas phase concentration}} + \nabla \cdot \left(-\underbrace{D_{igas}}_{\text{Dispersion}} \nabla \cdot c_i - \underbrace{c_i \frac{\kappa}{\mu} \nabla \cdot \left(R \cdot T \cdot \sum_{j=1}^{n} c_j\right)}_{\text{Convection (gas velocity)}}\right) = \underbrace{R_i}_{\text{Mass Transfer Rate}}$$

Adsorbed phase mass balance for each species

$$\underbrace{\frac{\partial c_{iad}}{\partial t}}_{\text{Time derivative adsorbed phase concentration}} + \nabla \cdot \underbrace{\left(-D_{isurf} \nabla \cdot c_{iad}\right)}_{\text{Dispersion (surface diffusion)}} = -\underbrace{R_i}_{\text{Mass Transfer Rate}}$$

Rate of mass transfer between gas and adsorbed phase

$$\underbrace{R_i}_{\text{Mass Transfer Rate}} = \underbrace{k_i}_{\text{Rate Constant}} \cdot \left[c_{iad} - \underbrace{\frac{\rho_{bed} R \cdot T \cdot N_{io} b_i(T) c_i}{1 + R \cdot T \cdot \sum_j b_j(T) c_j}}_{\text{Competitive Langmuir Adsorption Isotherm}} \right]$$

Radioactive decay is implemented as a first order reaction in the mass balance equation

Equations Continued

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Competitive Langmuir isotherm

$$c_{ad}^{*} = \left[\frac{\rho_{bed} R \cdot T \cdot N_{io} b_i(T) c_i}{1 + R \cdot T \cdot \sum_j b_j(T) c_j} \right]$$

The Langmuir isotherm is one possible model for equilibrium uptake, both software packages allow the user to implement other isotherm models.

Convection conduction equation in adsorption bed

$$\rho_{bed} Cp_{bed} \frac{\partial T}{\partial t} + \nabla \cdot (-k_{bed} \nabla T) = -Rate_{ad} (H_{ad}) - \rho_{bed} Cp_{bed} \vec{u}_{bed} \nabla T$$

Heat from radioactive decay is implemented in the convection conduction equation as a heat source tied to the first order reaction in the mass balance equation

Current Progress

- Isotherm data is being collected for several high surface area activated carbons, including data for competitive gas species.
  - This data will be used to compare potential adsorbents for xenon abatement, including the impact of competitive adsorption and rate of uptake.
- A finite element model that incorporates radioactive decay has been developed using literature isotherm data for coconut shell activated carbon.
  - The model currently does not include competitive adsorption from H_2O. This can be implemented.
  - The model will be updated when isotherm measurements are completed.
  - The 3-D finite element simulations will allow investigation of thermal effects in realistic geometries.
- PNNL will be open and transparent and provide all of our R&D on an equal basis to any producers interested in those results