Radioxenon Emissions Abatement

May 25, 2021

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WOSMIP 2 Remote

May 26, 2021

PNNL is operated by Battelle for the U.S. Department of Energy

PNNL-SA-162587

Pacific Northwest
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Our History

1940s

Lab began in support of Manhattan Project

1965

Pacific Northwest Laboratory is established

1980s

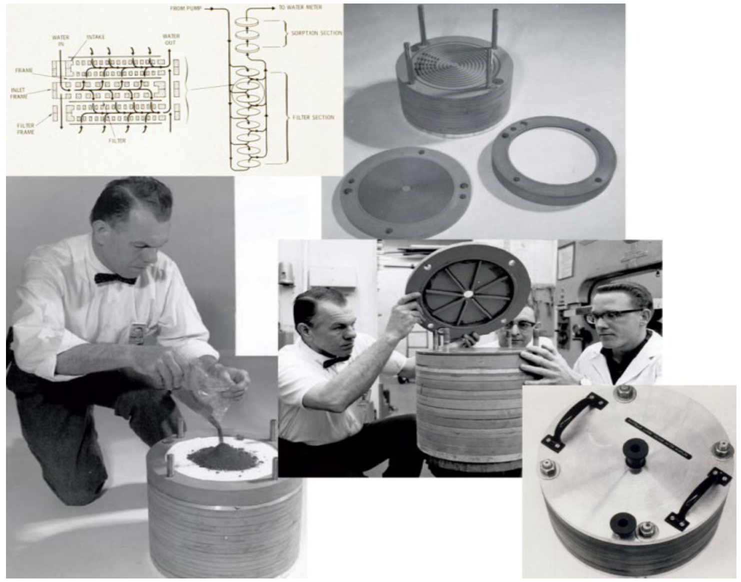
Cleanup effort begins following the shutdown of the last Hanford reactor

1990s

Trace detection expertise is leveraged to support nuclear nonproliferation and explosion monitoring

Today

PNNL continues to support the U.S. Government in emission reduction and trace detection of radionuclides in the environment



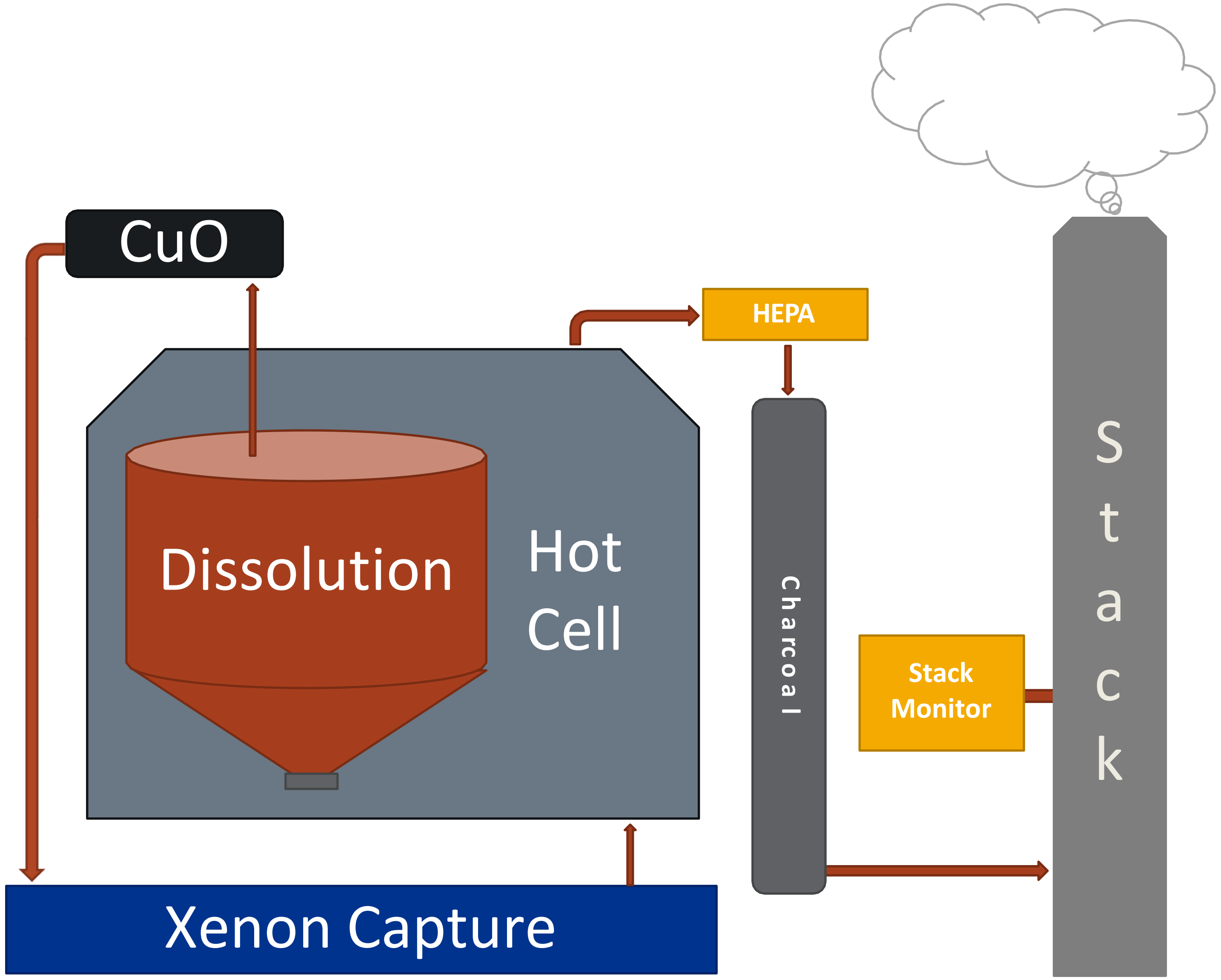
Xenon International – PNNL use of Cold Adsorbent to Collect Xenon from Air

- Xenon International is a xenon monitoring system that makes use of **low temperature adsorption** to collect xenon from atmospheric gases.
- Xenon from a 100 slpm flow, is removed from air using approximately 20 W of cooling.
  - Cooling is provided by two RICOR Stirling coolers (500 watts of power).
- This performance is enabled using PNNL developed microchannel heat exchangers and a good thermal design with minimal losses.

Hayes, James C., Paul H. Humble, Mark E. Panisko, Warren W. Harper, and Timothy L. Stewart. "Xenon collection method and system." U.S. Patent 10,005,018, issued June 26, 2018.

Emissions Reduction – What can be done?

- Each facility is different
  - No single solution
- The level of emissions released from a facility is dependent on three principals:
  - Operational Sharpness
  - Chemistry
  - Abatement systems/engineered systems
- Stack Monitoring
  - Real time feedback



Abatement system. Diagram for description of facility abatement systems. This is a simplified diagram and actual systems would be engineered according to the chemistry and facility needs.

- Iodine
  - Charcoal Filters
  - Silver Zeolite Filters
  - Alkaline Scrubbing
  - Mercurex Process
  - Silver iodide formation
  - Containment/Delay Tanks
- Particulate
  - HEPA Filtration
- Xenon
  - Charcoal beds
  - Cooled charcoal beds
  - Containment/Delay Tanks

Xenon Abatement Objectives

- PNNL is modeling xenon abatement trap designs to develop new abatement technology.
  - Modeling ambient temperature abatement traps
  - Modeling cooled abatement traps
  - Developing cryocooled abatement traps

- Basic Assumptions
  - Charcoal used for xenon abatement
    - ✓ Nucon charcoal is approved for nuclear power plant xenon abatement
    - ✓ Other charcoal manufacturers adsorption properties better understood
  - Xenon holdup needs to be approx. 80 days for 1e5 reduction in xenon
- Overall goal is to demonstrate cost effective options for isotope producers to meet requirements and minimize xenon emissions

Modeling Adsorbent Bed Xenon Hold-up

- **Develop a PNNL multiphysics model** for xenon abatement adsorbent beds to:
  - Develop a universal adsorption bed model that can answer questions about xenon abatement trap designs and adsorbent effectiveness.
  - Incorporate **heat of decay** from high activity radioxenon gas emissions trapped on abatement beds.
  - Evaluate ambient temperature bed designs and cooled bed designs.
- **Adsorption Process Modeling**
  - Aspen Adsorption
  - Finite element simulation using COMSOL Multiphysics
- **Hiden Gravimetric Adsorption Analyzer**
  - Can output the rate of uptake, as well as the final amount adsorbed at a given pressure and temperature
- **Adsorption Breakthrough Instrument**
  - Can perform breakthrough experiments for gas mixtures (up to three gas species and a carrier).

PNNL Modeling Equations using COMSOL

$$\varepsilon \frac{\partial c_i}{\partial t} + \nabla \cdot \left(-D_i \nabla c_i - c_i \frac{\kappa}{\mu} \nabla \left(RT \sum_{j=1}^{m} c_j \right) \right) = R_i - \lambda_i c_i$$

Gas Phase Mass Balance

$$\frac{\partial c_{i,a}}{\partial t} = -R_i$$

Adsorbed phase mass balance for each species

$$R_i = k_i \left[c_{i,a} - \frac{\rho_b N_i b_i(T) c_i}{1 + \sum_j b_j(T) c_j} \right]$$

Rate of Mass transfer between gas and adsorbed phase

$$\rho_i(T,P) C_{p,i} \frac{\partial T}{\partial t} + \rho_i(T,P) C_{p,i} \vec{u} \cdot \nabla T + \nabla \cdot (-k_i \nabla T) = q_{\mathrm{decay,i}} + a h (T_b - T_i)$$

Heat Transfer Gas Phase

$$\rho_b C_{p,b} \frac{\partial T}{\partial t} + \nabla \cdot (-k_b \nabla T) = q_{\mathrm{decay,i}} - a h (T_b - T_i)$$

Heat Transfer Solid Phase

$$q_{\mathrm{decay,i}} = \lambda_i c_{\mathrm{tot,i}} Q'_{\mathrm{i}}$$

Radioactive Decay

$$S = \frac{6(1-\varepsilon)}{d_{\mathrm{p}}}$$

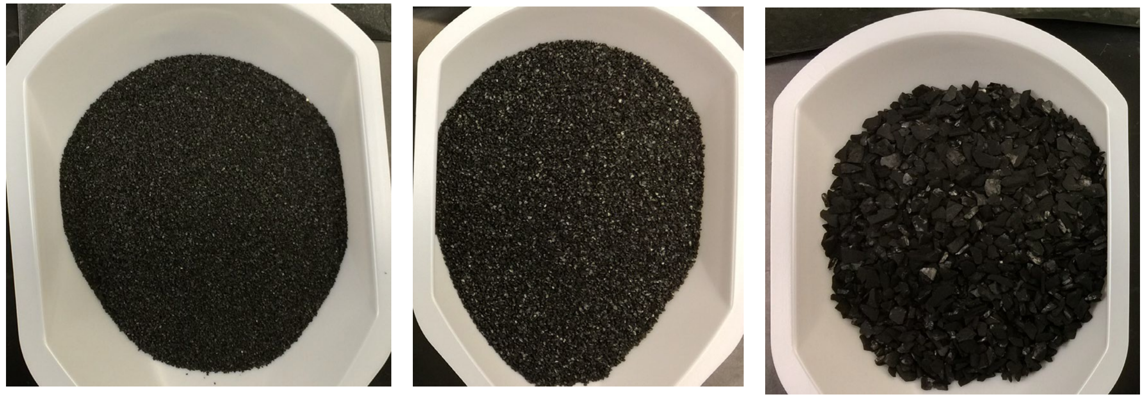
Adsorbent Surface Area

$$\dot{E}_i(t) = c_{i,\mathrm{out}}(t) \dot{V} N_A \lambda_i$$

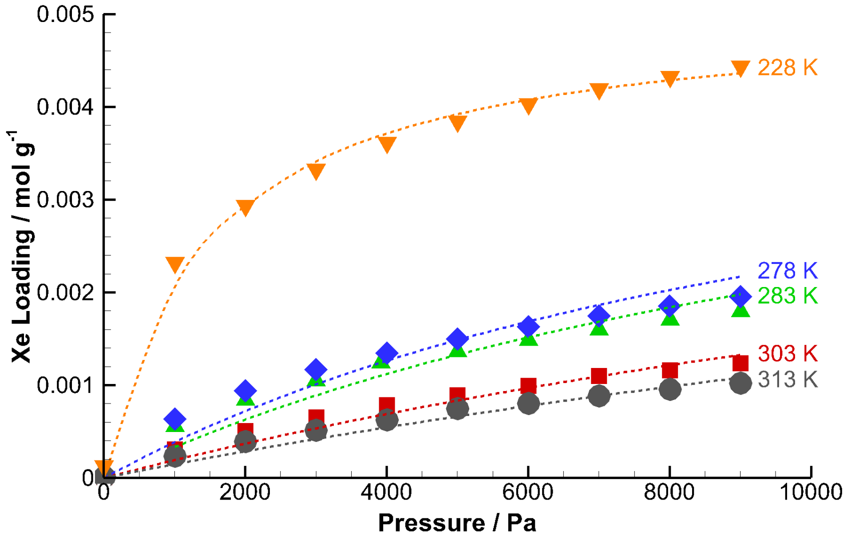
Instantaneous emission rate at end of column

Carbon Materials

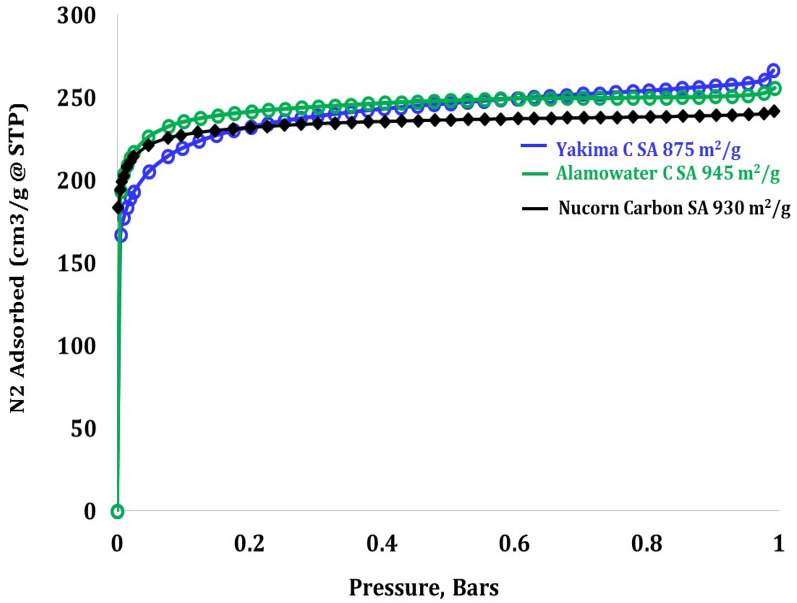
- Three commercially available activated carbon materials were identified and tested for the various gases of interest at multiple temperatures
- The NUCON GXK carbon is marketed as a noble gas delay carbon for the nuclear industry.
- All three carbon materials were studied for noble gas adsorption at various temperatures including water



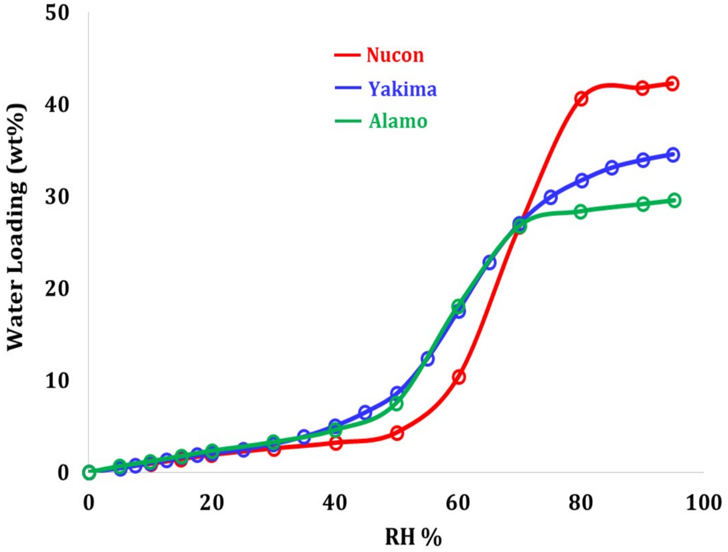
Yakima *Alamowater* *Nucon*



Nucon GDX isotherm data and fits



The BET surface area



Water adsorption isotherms

Abatement Trap Cooling

- PNNL has modeled 3 different cooling methods that provide different temperatures:
  - Liquid cooled jacket
  - Cold head at inlet of trap
  - Liquid nitrogen cooled jacket
- Hold time is dependent on;
  - Charcoal type
  - Other considerations
    - ✓ Humidity
    - ✓ Temperature
    - ✓ Time of use
- Temperature (rule of thumb):
  - Every 20 °C will drop bed size by half
  - Half flow rate cuts bed size by half

Ideal Calculations (4ft column) -120ºC

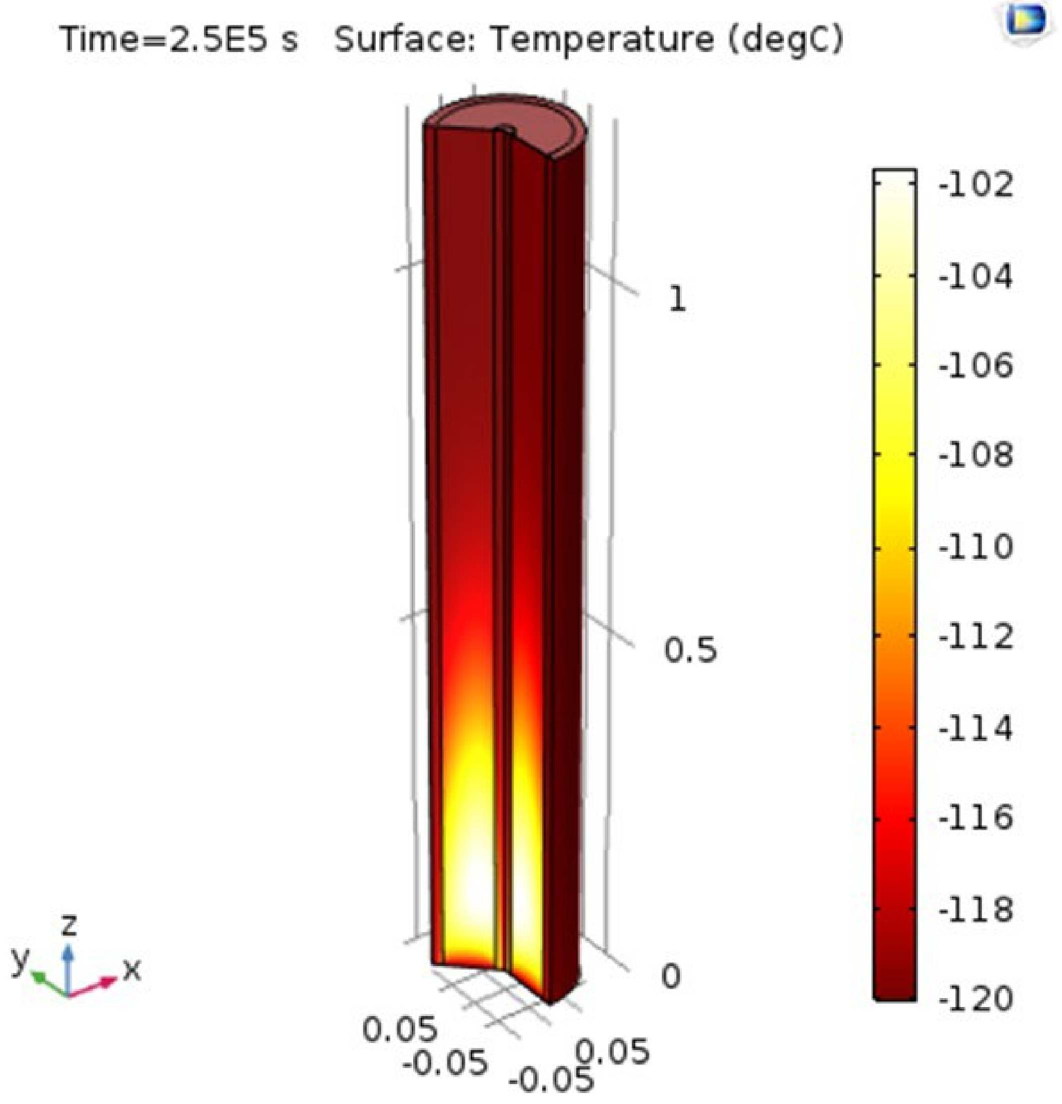
- 0.034 to 0.22 psi bed pressure drop at 100 LPM (depends on selected material's particle size)

- -120ºC Alamowater
  - 2X abatement (37 days hold up time), 100 LPM (may need two or more columns)
  - 5X abatement (87 days hold up time), 40.4 LPM (goal)
  - 20X abatement (352 days hold up time), 10 LPM

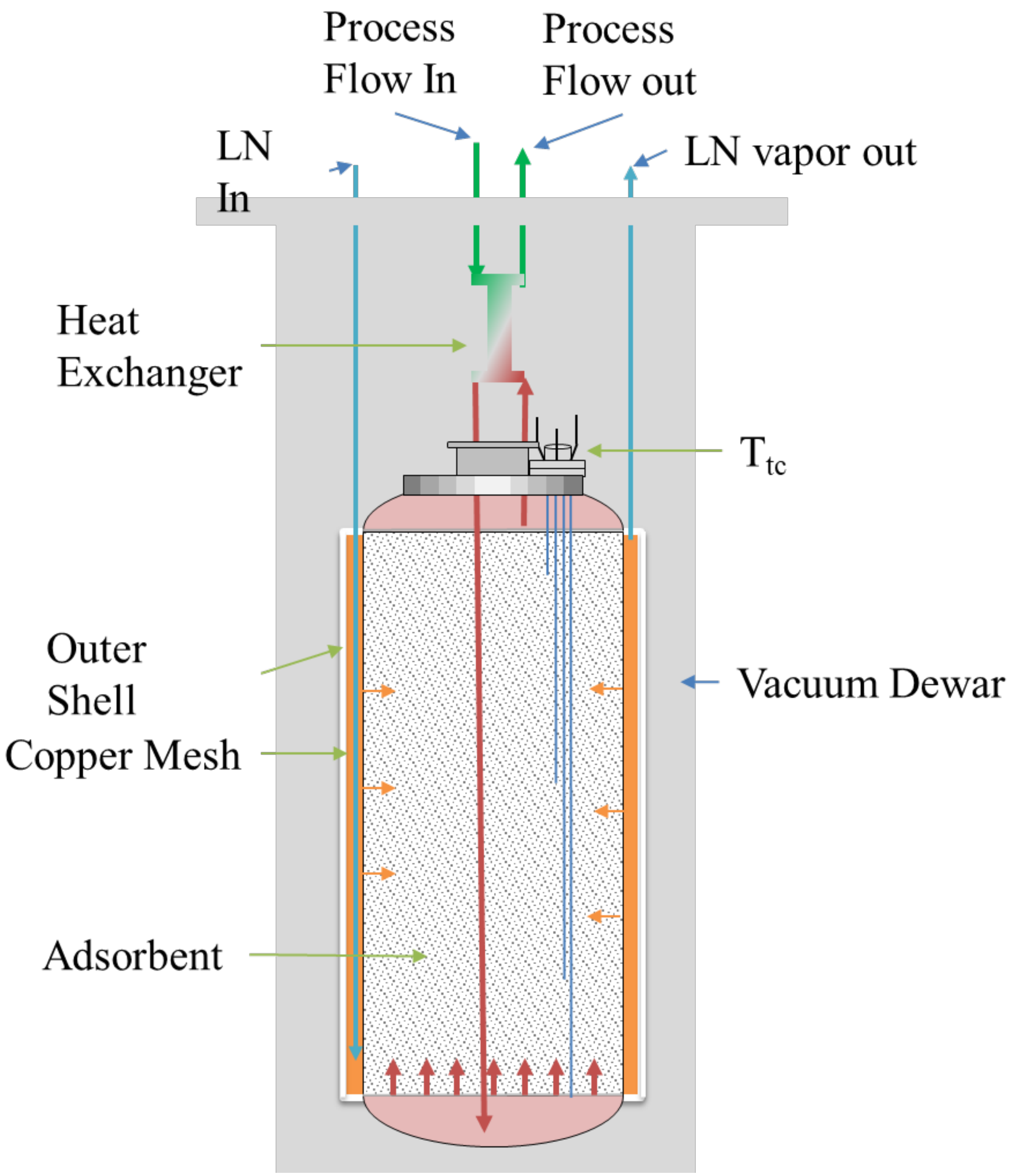
- -120ºC Nucon GDX (Nuclear industry approved at ambient temperature, no good data at cryogenic temperatures)
  - Less than 1X abatement (2.7 days hold up time), 100 LPM
  - Approaching 2X abatement (27.6 days hold up time), 10 LPM
  - 5X abatement (87 days hold up time), 3.16 LPM (goal)

COMSOL Model Simulations (-120˚C) and Estimated Temperatures

- Alamowater carbon
  - 40 LPM of abatement gas flow
  - Expected 87-day delay time (Alamowater carbon simulated with no radioxenon heating)
- Including heating and dispersion
  - 0.155 LPM of liquid N_2 flow
  - Avg T = -115.12 ˚C
  - Max T = -101.87 ˚C
  - Actual simulated delay time <u>72.68 days</u>

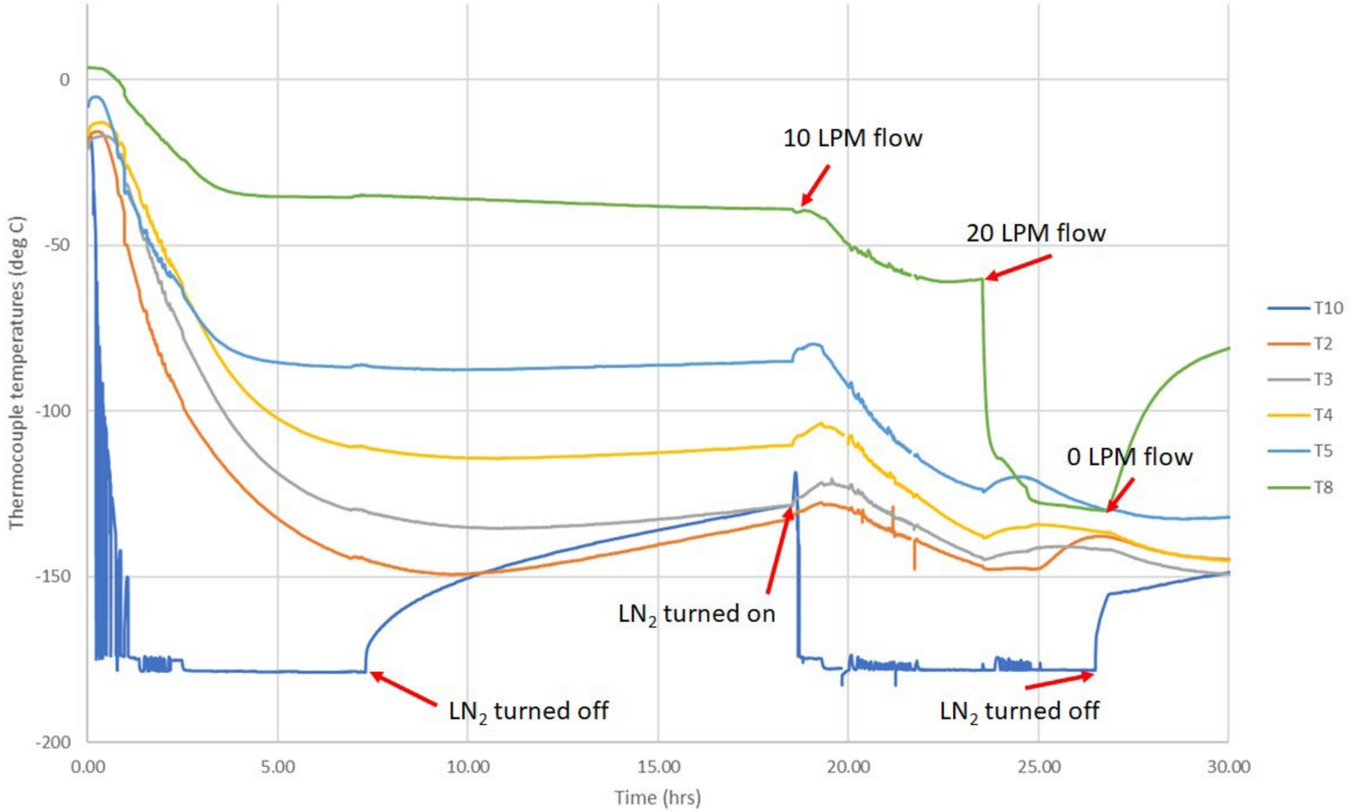


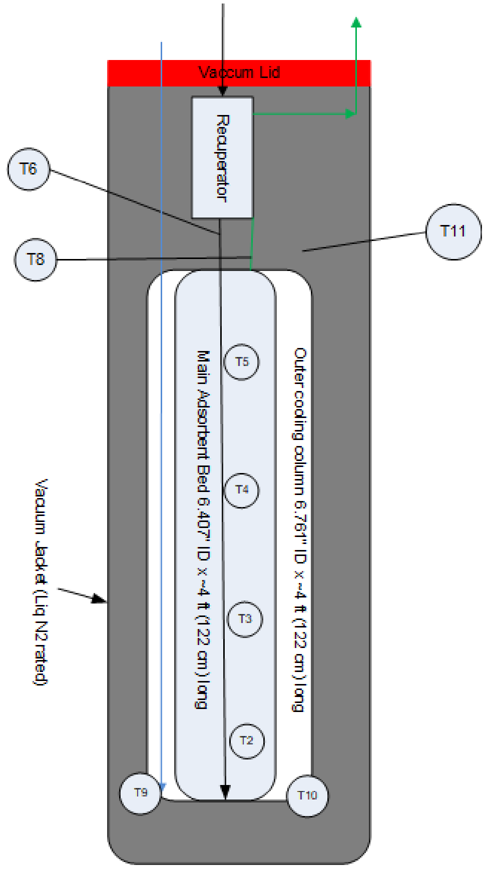
Prototype Abatement Trap Design



Prototype Radioxenon Abatement Trap

- Dimensions: 6-inch x 4-feet (15.3 cm x 122 cm)
  - Trap designed for 3000 Ci per day of radioactive Xe with up to 40 LPM process gas
  - 87 days of delay time of Xe is expected (5 orders of magnitude reduction of radioactive Xe)
  - Data below is preliminary trap temperatures without radioactive Xe in the process gas





Conclusion

- PNNL has generated adsorption models to estimate Xe emissions from both ambient temperature and cooled abatement traps that can be used for medical isotope production xenon abetment.
- PNNL has modeled and are developing a prototype cooled xenon abatement trap that is being tested in FY21 and FY22.
- PNNL's design and modeling expertise support the medical isotope production community in demonstrating cost effective means for minimizing Xe emissions and facilitating trace detection.

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Thank you