INL/CON-20-60462-Revision-0

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Prepared for the
U.S. Department of Energy

Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517

Multi-Column Evaluations for Capturing and Separating Krypton and Xenon using Mordenite-based Sorbents

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Materials Research Society, Fall 2020 Meeting
November 30, 2020

U.S. DEPARTMENT OF ENERGY

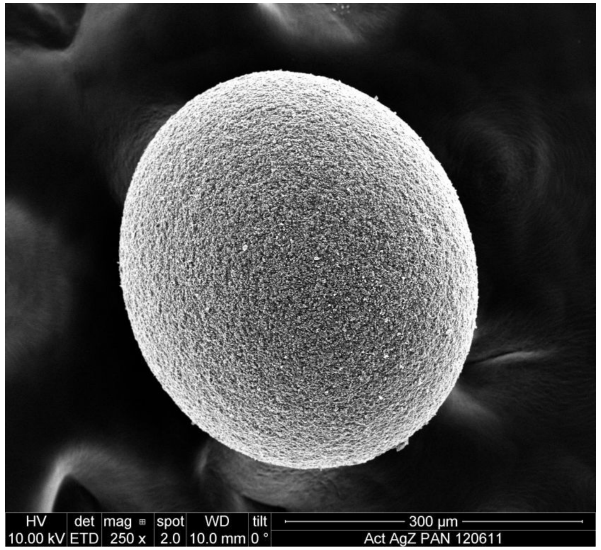
Material Recovery & Waste Form Development

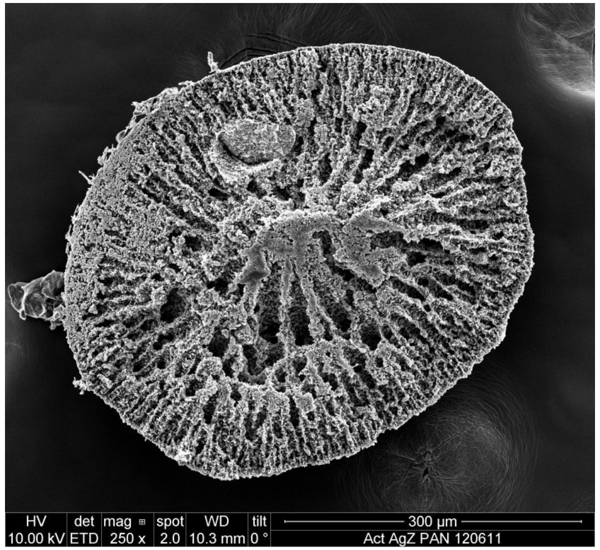
Introduction

- Off-gas compositions generated through the reprocessing of used nuclear fuel include volatile fission product isotopes
  - Kr-85, I-129, H-3, C-14, and various Xe isotopes
- Radioactive emission levels are addressed by the U.S. EPA in 40 CFR 190
  - Establishes annual dose limits resulting from nuclear fuel cycle activities in the commercial sector
  - To meet these dose limits, Kr-85 may need to be captured and immobilized to minimize its release into the environment
  - Due to the short half-lives of Xe isotopes, the Xe will be stable and have a high commercial value but will compete with Kr capture
- Solid-phase adsorption (physisorption)
  - Activated carbon is well-known as a highly selective material for capturing Kr and Xe at reduced temperatures.
  - Fire hazards associated with carbon-based sorbents have been reported.
  - Mordenite-based sorbents have shown adsorption capacities comparable to activated carbon.

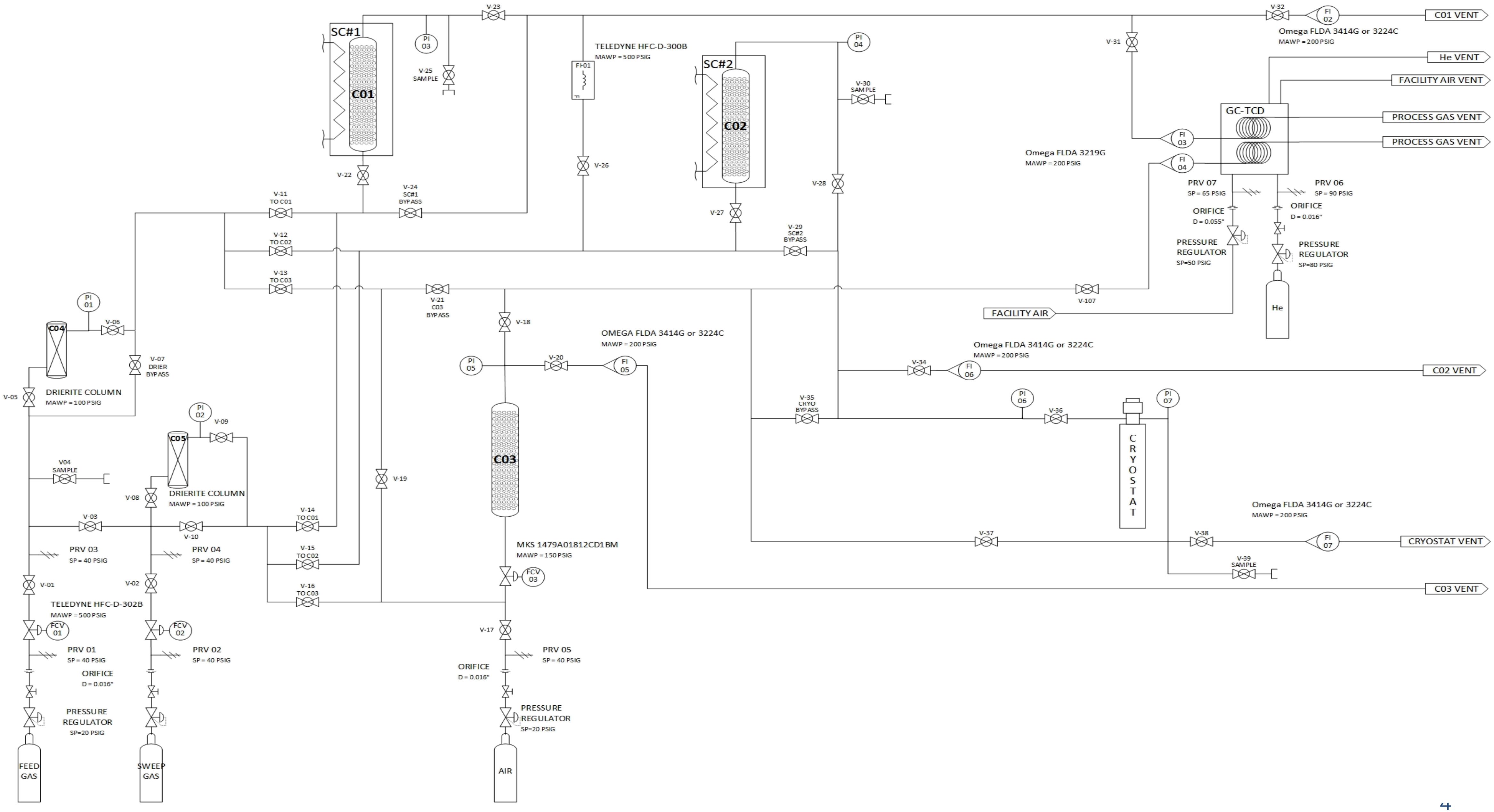
Sorbent Development

- Mordenites
  - Synthetic material commercially available in powder form (Na and H)
  - Limited availability of engineered forms that typically have low surface areas
  - Physisorption necessitates high surface area engineered forms
- The polyacrylonitrile (PAN) macroporous polymer was used to bind mordenite powders
  - Good thermal and radiolytic stability
  - 80-wt% mordenite in PAN mixtures
  - Ag and H forms prepared
  - Original surface areas maintained
  - Beads between 0.30 and 1.4 mm
  - U.S. Patent 8,686,083 B2



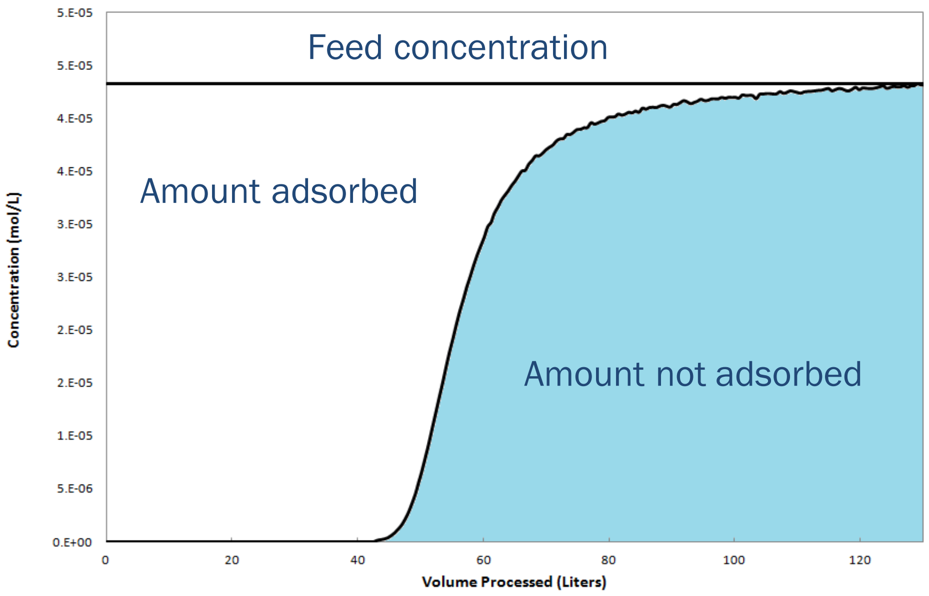


Adsorption Testbed



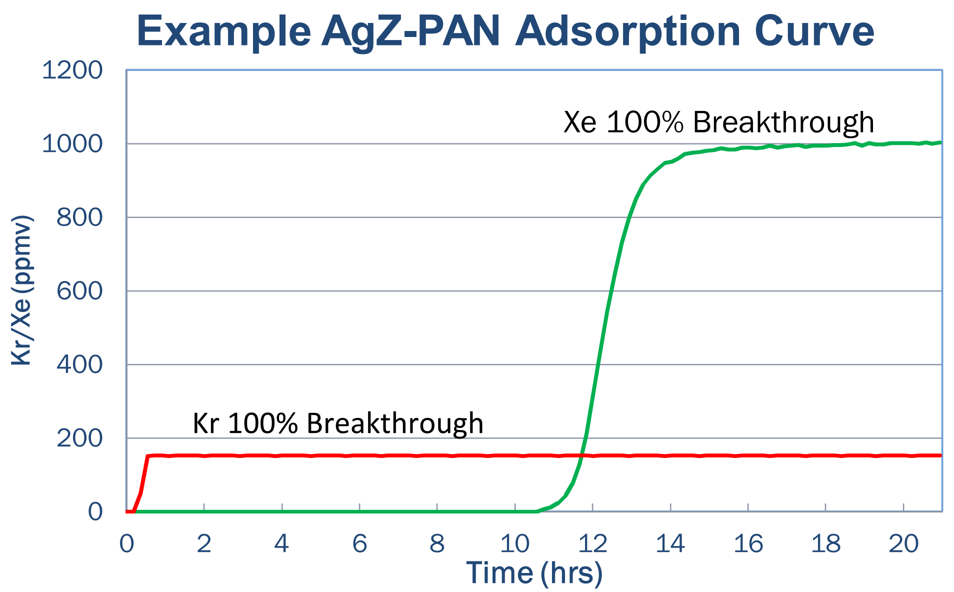
Xe and Kr Adsorption Testing

- Xe and Kr adsorption capacities
  - Various feed-gas compositions
  - Multiple sorbent temperatures
  - Breakthrough curves generated from GC data
  - Jandel Scientific's TableCurve utilized for determining the area under the curve
  - AgZ-PAN and HZ-PAN evaluated
- Xe and Kr adsorption results
  - AgZ-PAN
    - Highly selective for Xe at ambient temp.
    - Little to no Kr adsorbed
    - No thermal degradation observed
  - HZ-PAN
    - Adsorbs Kr at -82 °C
    - No thermal degradation observed
    - Kr is easily desorbed by raising temperature



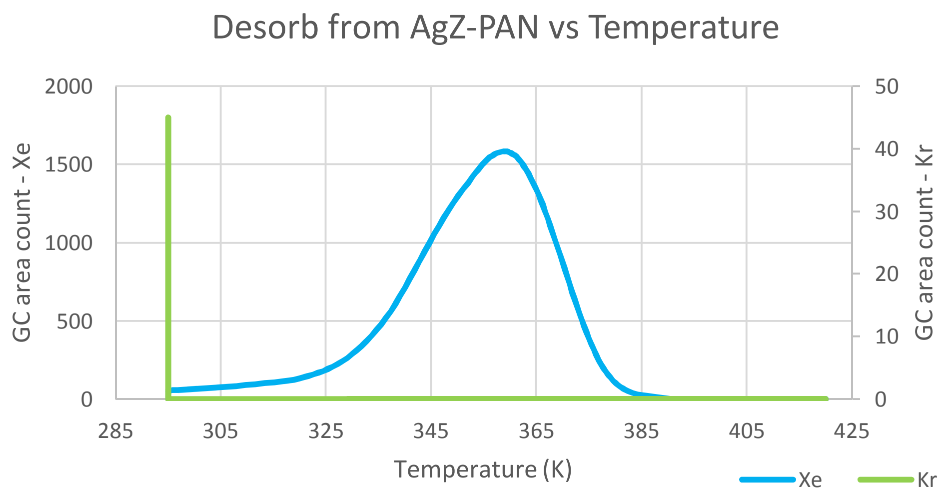
Xe and Kr Separation

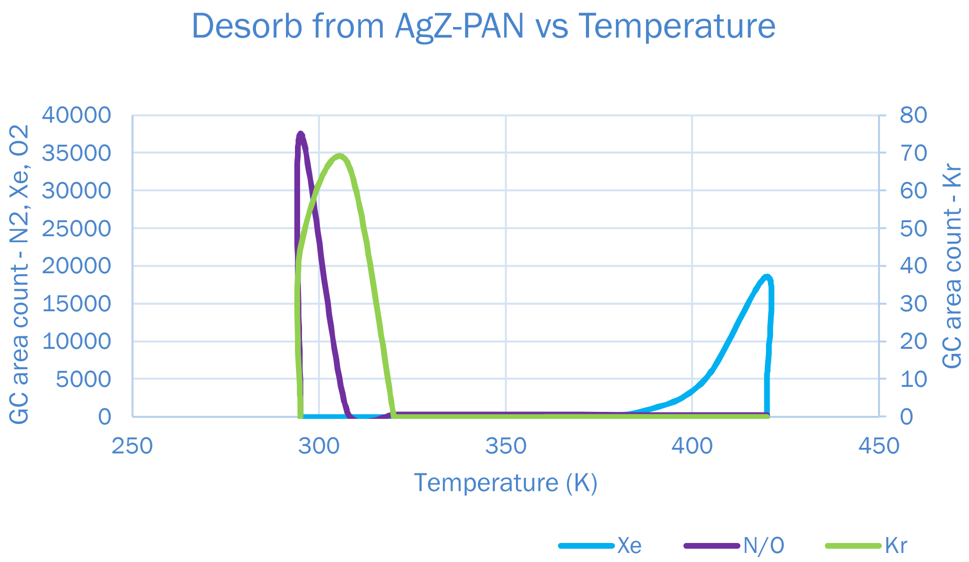
- AgZ-PAN column
  - Feed gas: 1000 ppmv Xe, 150 ppmv Kr in air
  - 44.3 grams AgZ-PAN
  - Flowrate: 50 sccm
  - Temperature: 22°C
- HZ-PAN column
  - Feed gas: effluent from AgZ-PAN column
  - 29.8 grams HZ-PAN
  - Flowrate: 50 sccm
  - Temperature: -82°C



AgZ-PAN Xe Desorption

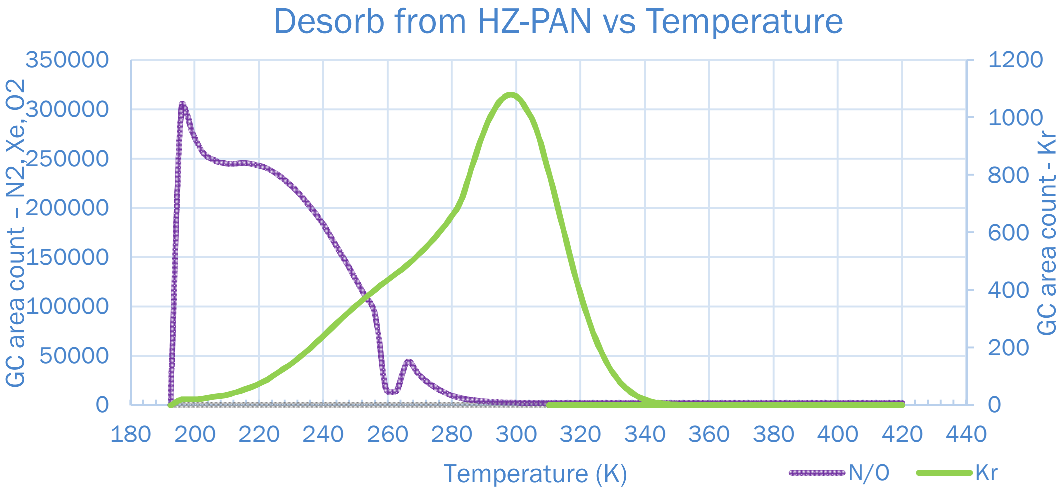
- AgZ-PAN column
  - Xe/Kr separation
  - Ambient temperature adsorption
  - Adsorbed phase contains Xe, Kr, and air
  - Desorption requires purge gas (air, N_2, Ar, or He)
  - Slowly ramp to temperature over 12 hours
  - Step temperature and soak 6 hours
  - 99–100% Xe separation

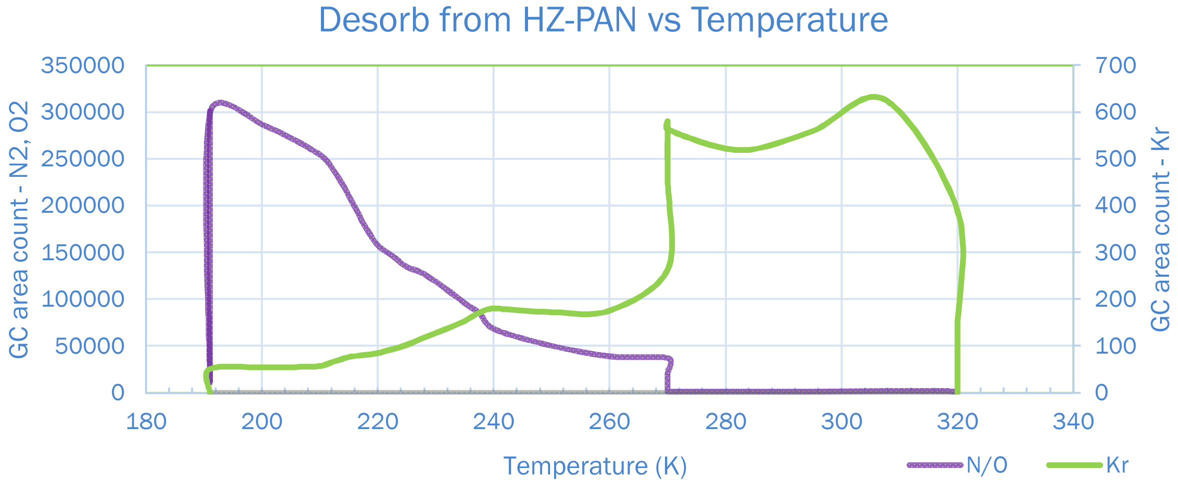




HZ-PAN Kr Desorption

- HZ-PAN column
  - Kr capture
  - -82 °C adsorption
  - Adsorbed phase contains Kr and air
  - Desorption requires purge gas (air, N_2, Ar, or He)
  - Slowly ramp to temperature over 12 hours
  - Step temperature and soak 9 hours
  - ~80% Kr separation
  - 3.5–5 times the Kr concentration





Summary

- Major accomplishments
  - Developed novel engineered-form sorbents
    - AgZ-PAN and HZ-PAN
    - Patent awarded for the process (U.S. Patent 8,686,083 B2)
  - Designed custom adsorption system
  - Evaluated the solid-phase adsorption of Xe/Kr
  - Published two journal articles
    - Journal of Nuclear Science and Technology, 51:4, 476-481, DOI: 10.1080/00223131.2014.877404
    - Journal of Nuclear Science and Technology, DOI:10.1080/00223131.2015.1126205
  - Demonstrated the separation of Xe from Kr in a dual-column system
  - Produced a pure Kr stream from a Kr/Xe/Air mixture
  - Evaluated the desorption of Kr and Xe

Acknowledgements

- Funding
  - U.S. Department of Energy's Office of Nuclear Energy
  - Material Recovery & Waste Form Development Campaign