Solid Catcher Materials Development at InnoSense for a Variety of Rare Isotopes

Nuclear Physics SBIR/STTR Exchange Meeting
August 6-7, 2014

Sponsor: Office of Nuclear Physics, DOE
Program Officer: Dr. Manouchehr Farkhondeh
Phase II Contract Number(s): DE-SC0004265; DE-SC0007572

Small Business
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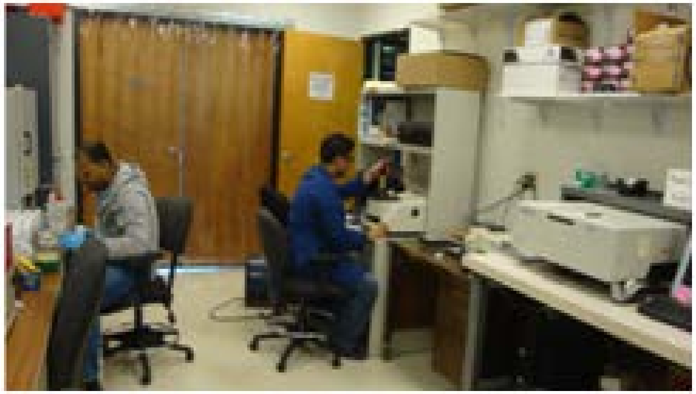
Presentation Overview

- About InnoSense LLC
- InnoSense LLC Team
- Overall Objectives
- Relevance to Nuclear Physics Programs
- Technical Objectives/Milestones
- Materials Under Investigation
  - Refractory Carbon Aerogels
  - Refractory Porous Oxides
- Simulations
- Off-line and In-Beam Testing
- Summary
- Acknowledgments

About InnoSense LLC

- Established in 2002 by private investment, housed in a recently expanded 9,000 square feet laboratory facility located in Torrance, California.
- Key laboratories include five “wet” chemical facilities equipped with fume hoods, a clean room, a spectroscopy facility, optics and testing laboratory, and two machine shops.
- 18 employees, including 4 PhD, 4 MS and 2 MBA degree holders.



Team at InnoSense LLC

Uma Sampathkumaran, PhD, Vice President, R&D

- Development of ORMOSILs and aerogel structures
- Processing refractory, magnetic and dielectric ceramics
- Materials characterization

David Hess, PhD, Senior Scientist, R&D

- Polymer chemistry
- Supercritical CO_2-asssited processing of nanostructures
- Materials development and characterization

Raymond Winter, MS, Senior Engineer

- Ceramics processing and sintering
- Statistical design of experiments
- Materials characterization

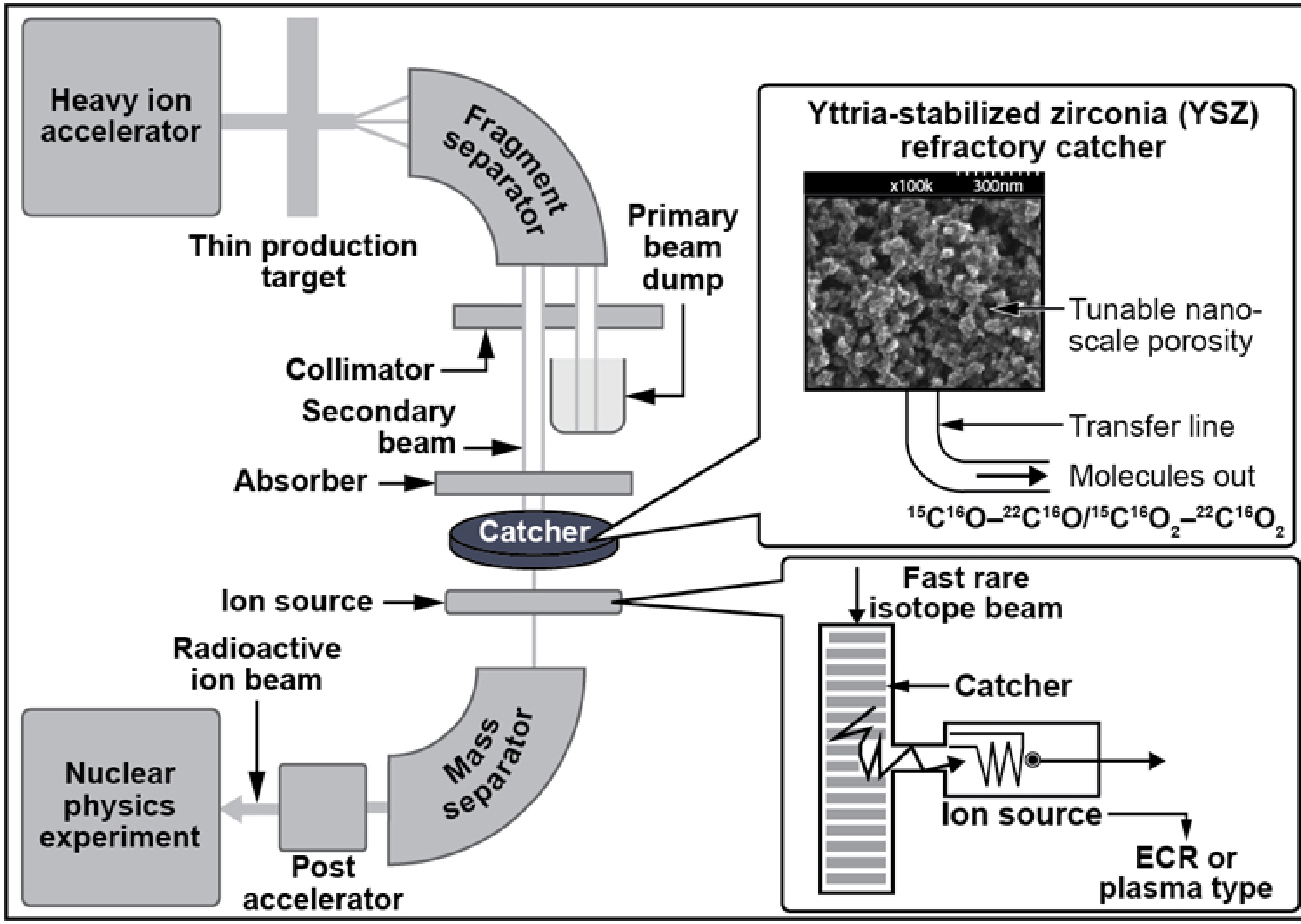
Mohammad Mushfiq, BSEE, Research Engineer

- Materials processing and characterization
- Supercritical drying of aerogels

John Abolencia, BS, Research Engineer

- Materials processing and characterization
- Tape casting

Refractory Hot Catchers for Rare Isotopes



Features

- Refractory YSZ of tunable porosity and geometry
- Stable in energetic radioactive beams
- Supplementary to the gas catcher

Advantages

- Higher stopping power (~100 times) over gas catchers
- Allows diffusion/effusion of short-lived isotopes as atoms or molecules
- Enable understanding of the formation of the cosmos

Collaborators

Argonne National Laboratory

National Superconducting Cyclotron Laboratory at Michigan State University

- *Design heater and ion source*
- *Beam-line studies*

Primary purpose of porous oxide monolith is to catch ^{9}C–^{22}C isotopes and convert them to $^{9}C^{16}O$– $^{22}C^{16}O$ expected to be released almost as a noble gas.

Catchers Under Investigation

Refractory Catcher	Projectile	Expected Isotope
Tungsten-coated SiO_2 Aerogel	^{18}O (typical)	$^{8\text{-}11}Li$ $^{6,8}He$
Carbon Aerogel	^{16}O, ^{48}Ca, etc.	$^{12}C^{14}O$–$^{12}C^{24}O$ $^{12}C^{14}O_2$–$^{12}C^{24}O_2$
Yttria-Stabilized Zirconia /Other Porous Oxide Monoliths	^{12}C, ^{48}Ca, etc.	$^{9}C^{16}O$–$^{22}C^{16}O$ $^{9}C^{16}O_2$–$^{22}C^{16}O_2$

Background on ISOL Target Materials

Isotope Separation On-Line (ISOL) used to generate radionuclides

- Targets are used with high power beams
- Isotopes are produced by reactions of the beam with target material
- Target must be dense enough to stop energetic beam, yet porous enough to allow rapid diffusion of radionuclides to the accelerator source
- Must be thermally conductive to withstand beam power
- Targets are heated to > 2000 °C to increase diffusion rates of radioactive nuclides

Benefits When Used in Catcher Mode

- Catchers used to stop high energy radioactive isotopes created in a separate production target up stream
- In the catcher mode, thermal conductivity is less relevant since the beam power is deposited in the thermally separated production target irradiated with heavy ion beams
- No radiation damage when used in catcher mode since only secondary radioisotope beams impinge on it
- Selection of materials is open to new approaches that cannot work with ISOL targets, e.g. aerogels with low thermal conductivity
- The porous refractory materials will theoretically offer more stopping power and fast-release for the generation of intense rare isotopes
- The refractory nature potentially allows them to be used as:
  - Compact isotope catcher/ion source placed in the first focal plane of the fragment separator with the capability of selective harvesting isotopes for different applications

Technical Objectives and Milestones

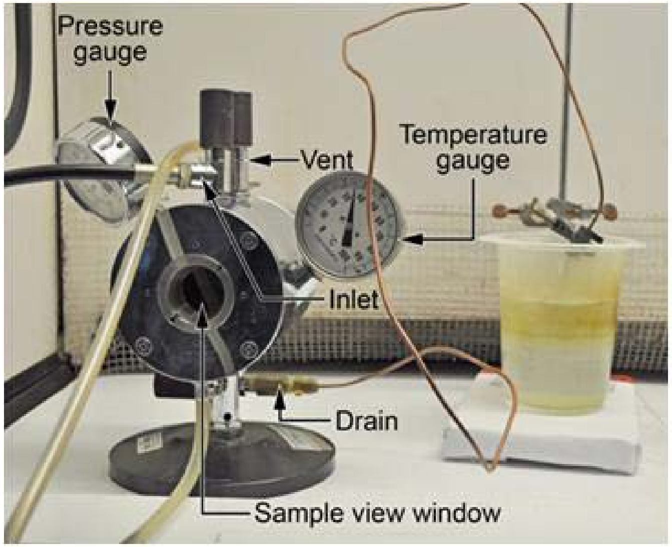
Objectives

1. Refine formulations and processing conditions to reproducibly fabricate porous, solid catchers (carbon aerogels, porous oxide monoliths).
2. Evaluate the physical properties of the porous monoliths after low temperature processing
3. Screen stability and open pore structure of refractory porous materials at isotope production temperatures (1000 to 2000 °C) for beamline studies.
4. Evaluate prescreened refractory porous materials off-line and on-line for suitability as reactive diffusion targets for molecular species of CO.

Milestones

- Fabricate refractory porous samples with moderate densities (1 g/cm^3) that retain their open nanoscale porosity upon heating.
- Refractory carbon aerogels and porous oxides maintain structural and dimensional stability after heating to temperatures ranging from 1000–1500 °C.
- Off-line release characteristics are studied using RGA and stable beams.
- On-line measurement of the release times of radioactive CO and CO_2 for the most promising samples completed.

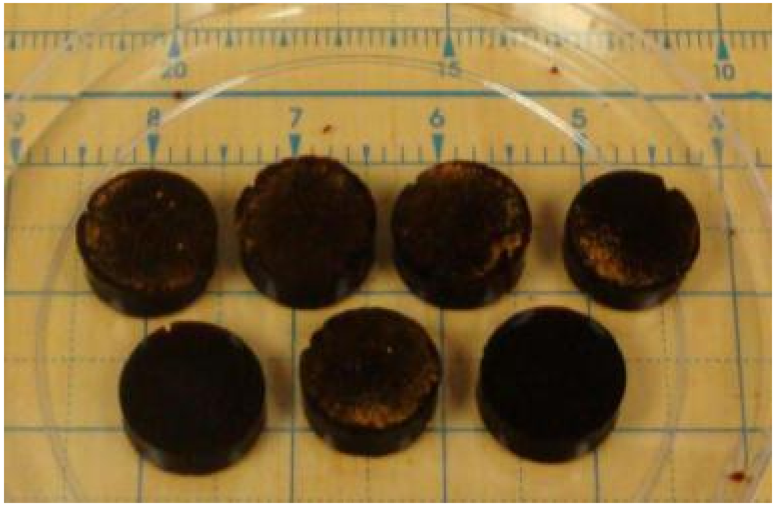
Supercritical CO_2 Drying of Polymer Aerogels



The $SCCO_2$ drier

Polymer aerogel in drier

- Supercritical CO_2 extraction of the solvent in pores enables aerogel fabrication without pore collapse
- Highly porous, low density, polymer aerogel monoliths with minimal reduction in dimensions.



Apparent Densities
~ 0.08–0.27 g/cm^3

Pyrolysis of Polymer Aerogel to Carbon Aerogel


Muffle furnace


Retort in furnace

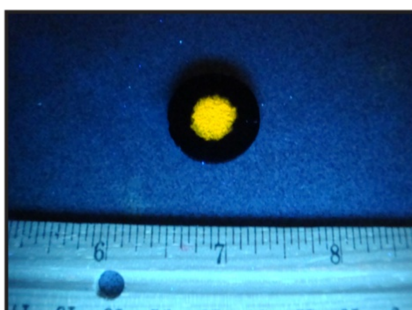
Before pyrolysis


$SCCO_2$ dried polymer aerogel

After pyrolysis


Carbon aerogel

Dye penetration viewed with UV light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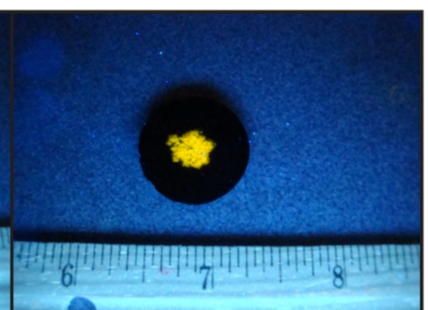
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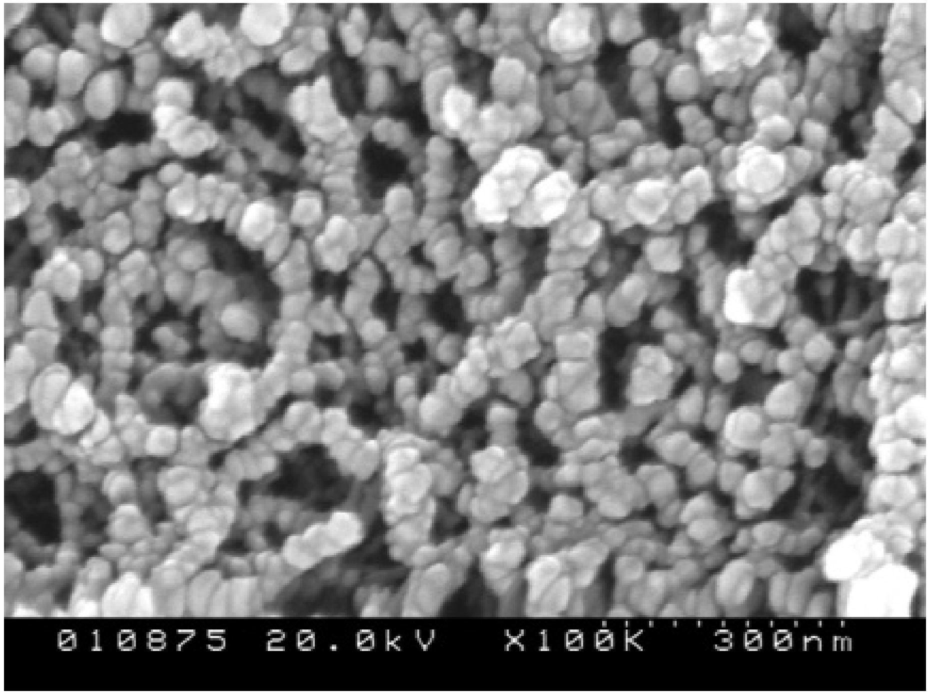
30% reduction in monolith dimension after pyrolysis

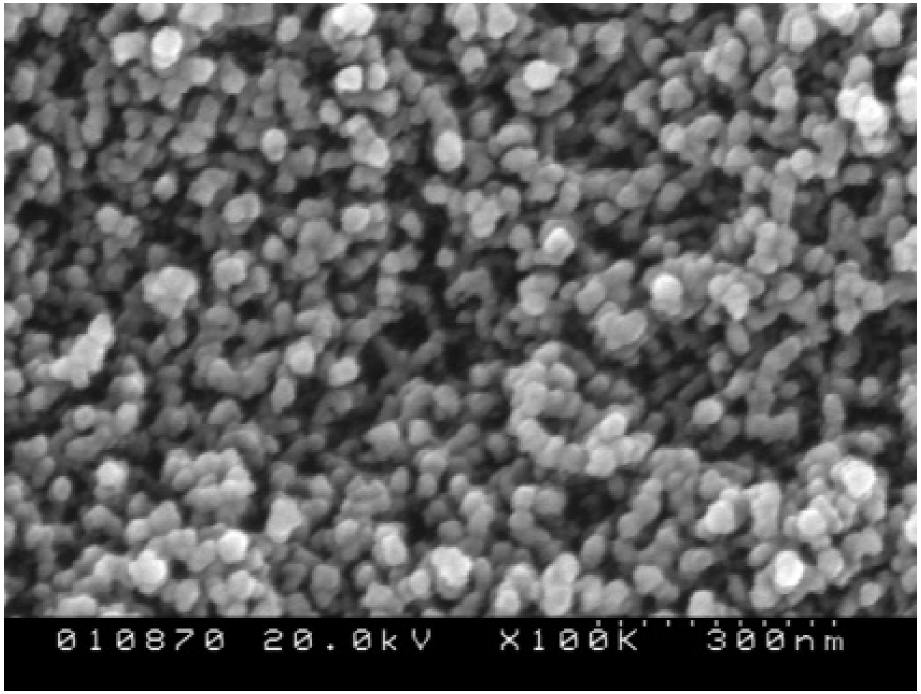
Apparent Density ~ 0.07–0.35 g/cm^3

SEM of Undoped Carbon Aerogels



Supercritical CO_2 dried polymer aerogels

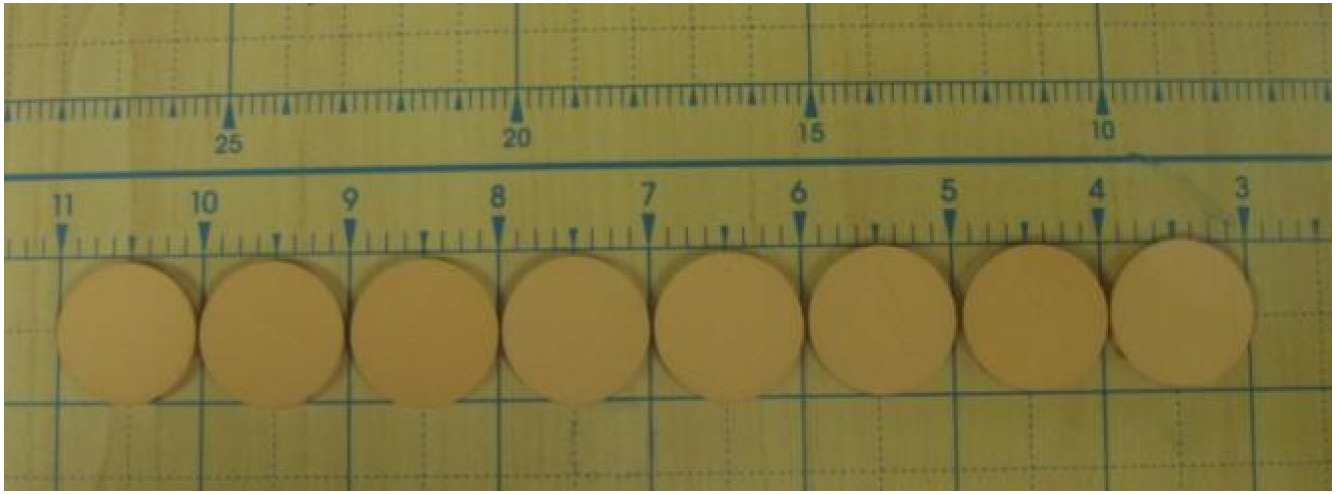
Pore size ~60–300 nm



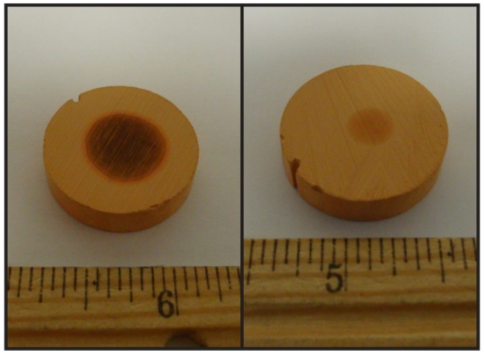
Polymer aerogel pyrolyzed to form carbon aerogel

Pore size ~60–180 nm

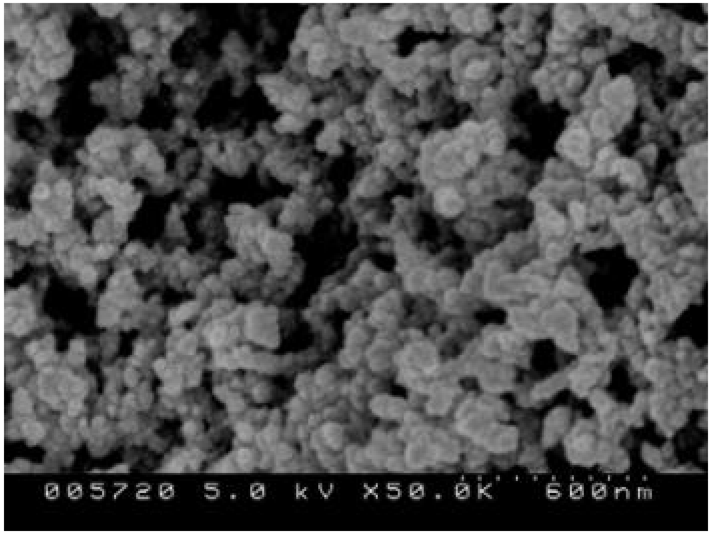
W-Doped Polymer Aerogels



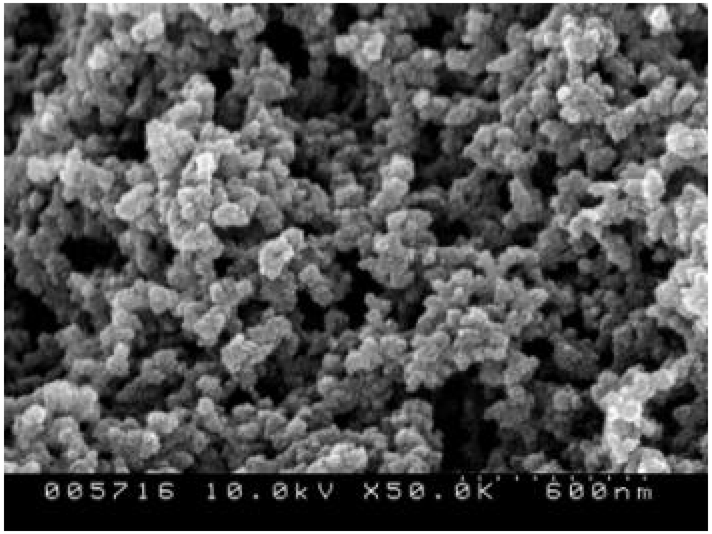
W-doped polymer aerogels after $SCCO_2$ drying



SEM of $SCCO_2$ dried aerogel



SEM of carbon aerogel



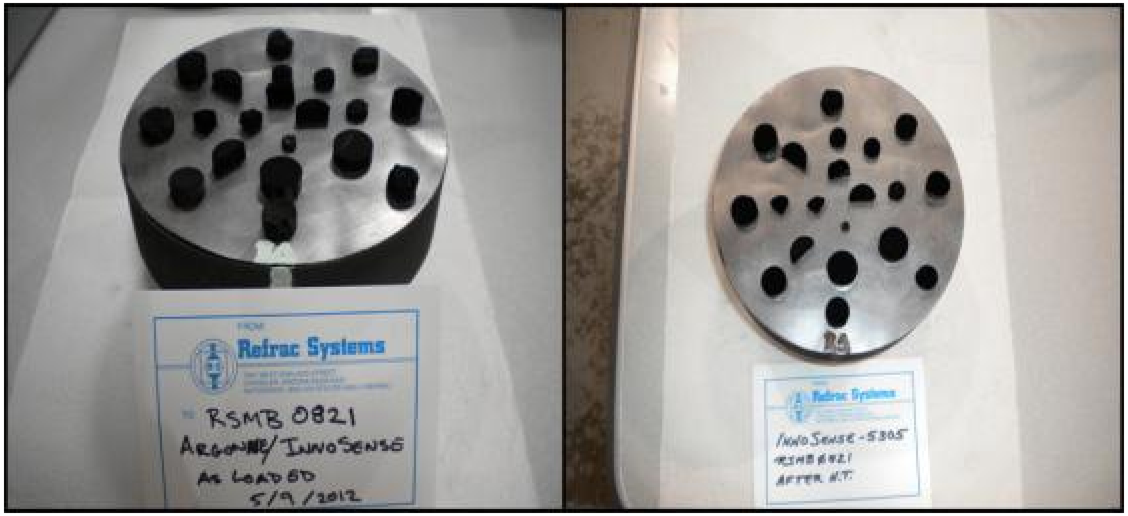
Pore size
~0.3–3 µm

Apparent Density
~0.36–0.40 g/cm^3

~0.39–0.41 g/cm^3

Samples Mechanically Stable After Vacuum Heat Treatment at 1500 °C

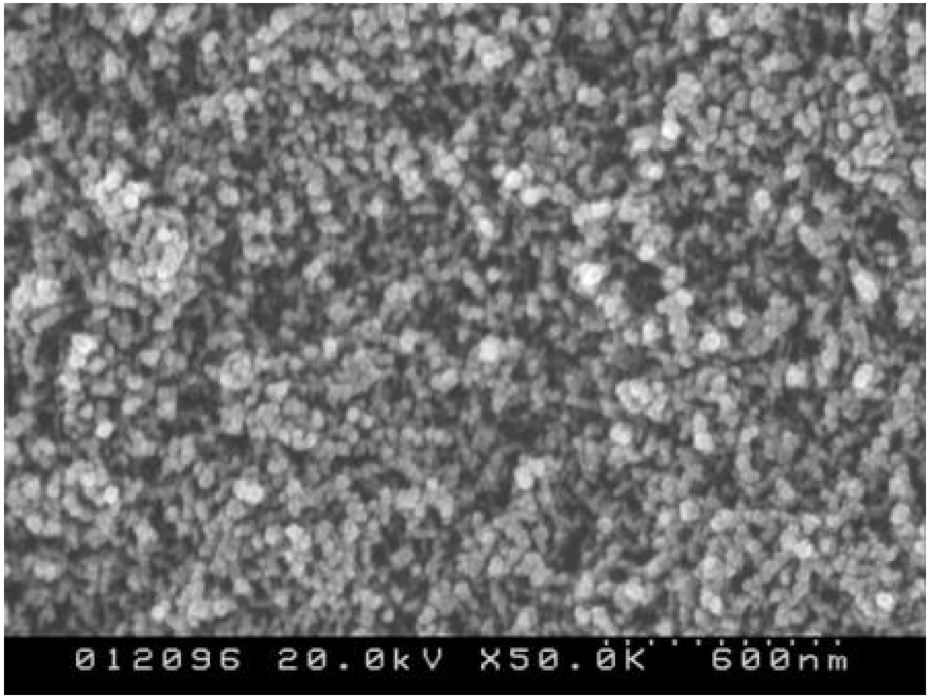
Vacuum induction furnace



C-aerogels before (left) and after (right) vacuum heating

- Samples vacuum heated at 1500 °C for 2 hour at Refrac, Chandler, AZ
- Minimum-to-no out gassing was observed indicative of fully pyrolyzed samples
- Monolithic samples remained mechanically intact

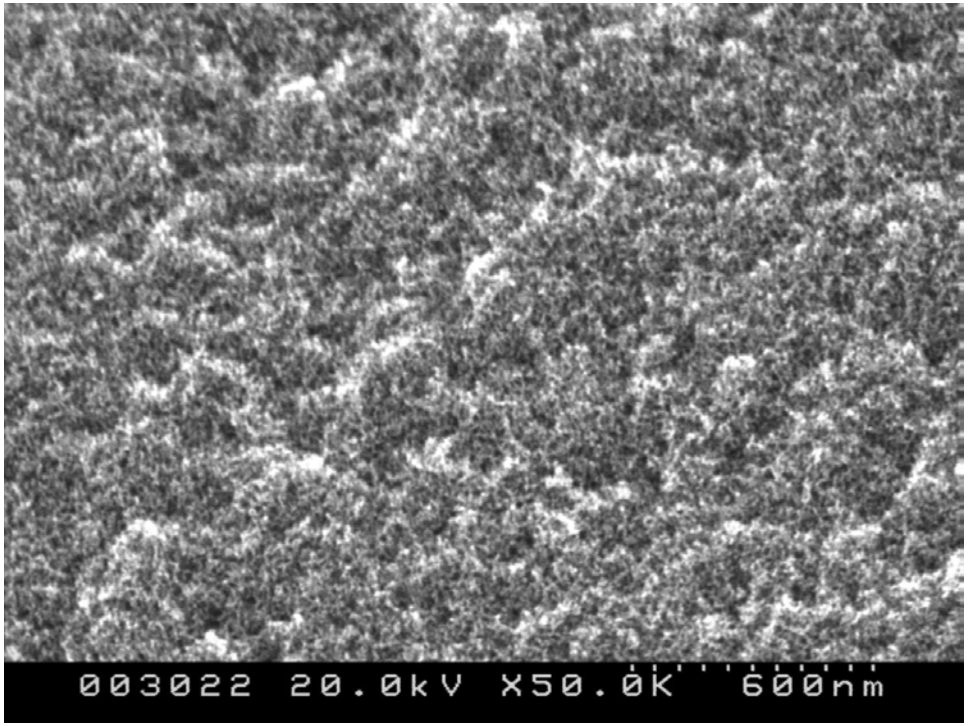
SEM Images of Undoped Carbon Aerogels After Vacuum Heat Treatment at 1500 °C



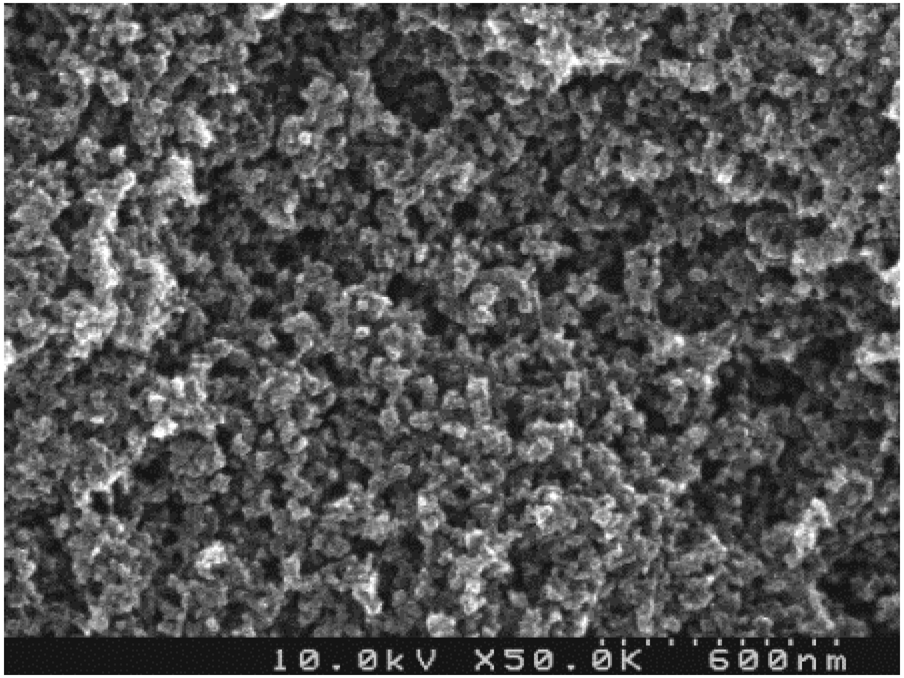
Apparent Density ~0.09–0.36 g/cm^3

Pore Size ~15–120 nm

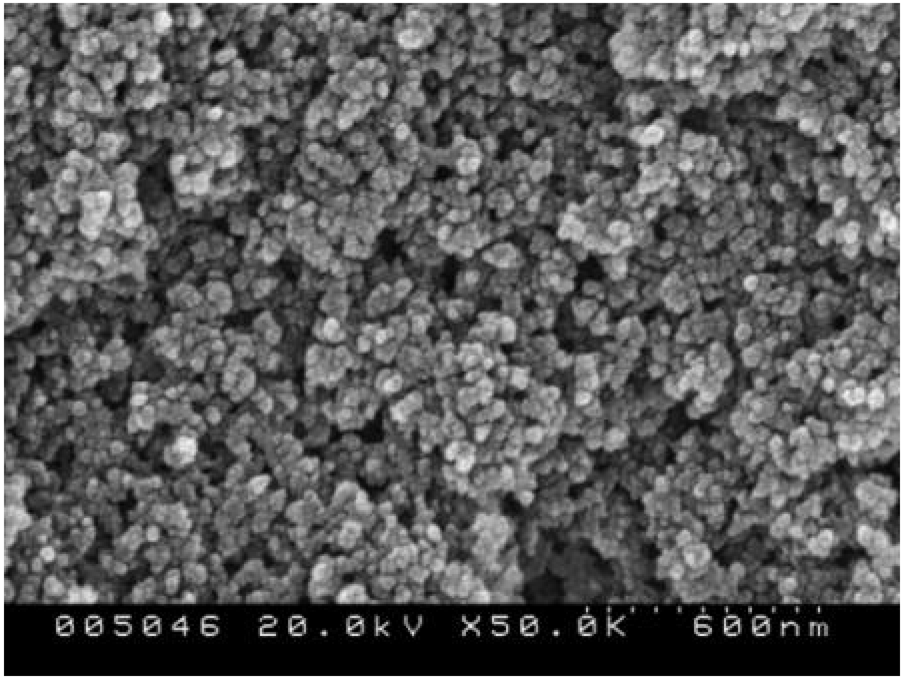
Different starting formulations to tune pore size and density

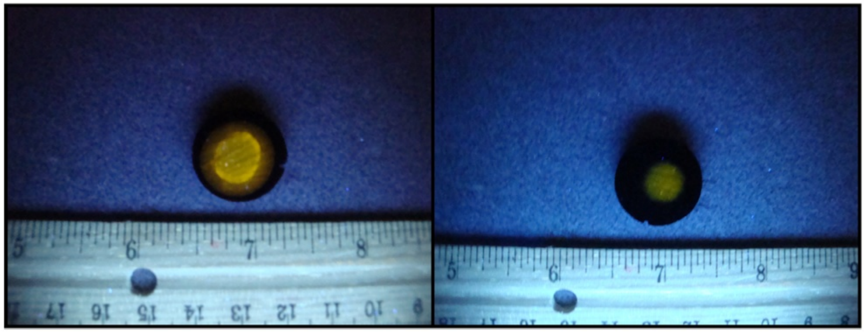


SEM Images of W-Doped Carbon Aerogels After Vacuum Heat Treatment at 1500 °C



- Retains open porosity
- Low doping level increases density significantly





Top Bottom

Apparent Density ~ 0.41–0.51 g/cm^3

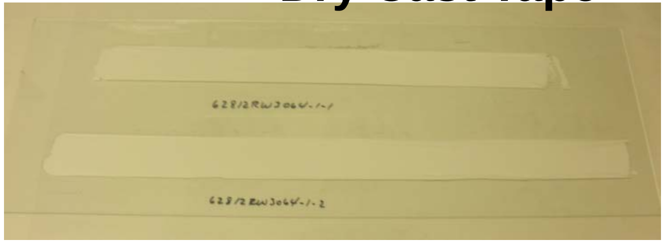
Pore size ~60 nm–1.2 µm

Tape Casting Process to Fabricate Porous Oxides

Slip in Ball Mill

Wet Cast Tape

Dry Cast Tape



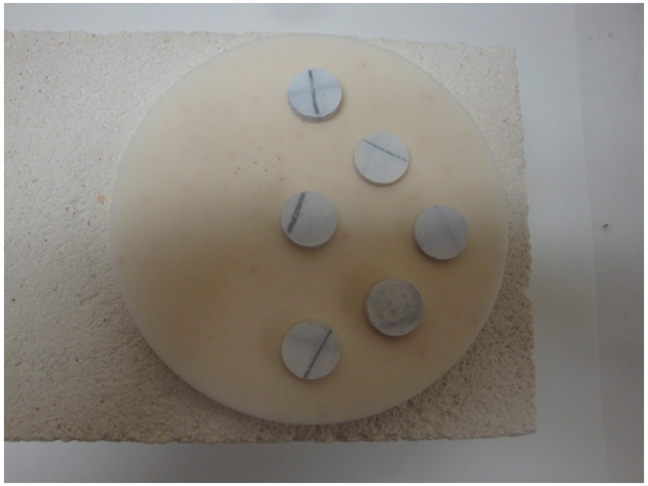
½" Die Punch

Pressed Green Disk

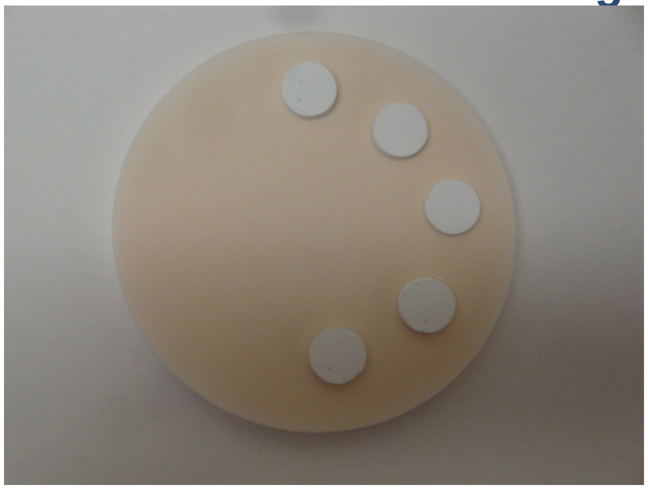
Carver Hydraulic Press

Tape Cast Porous Oxide Monoliths

Diced Stacked Hot Pressed Tape



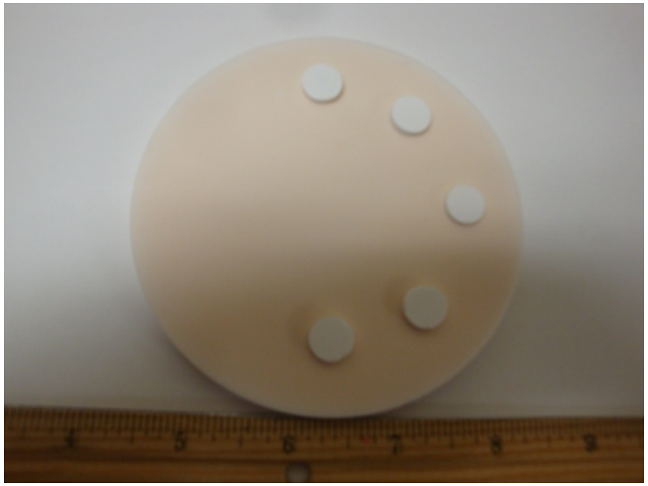
Disks after 800 °C Firing



Apparent Density ~ 2.7–4.5 g/cm^3

Open porosity ~15–49%

Disks After 1500 °C Firing

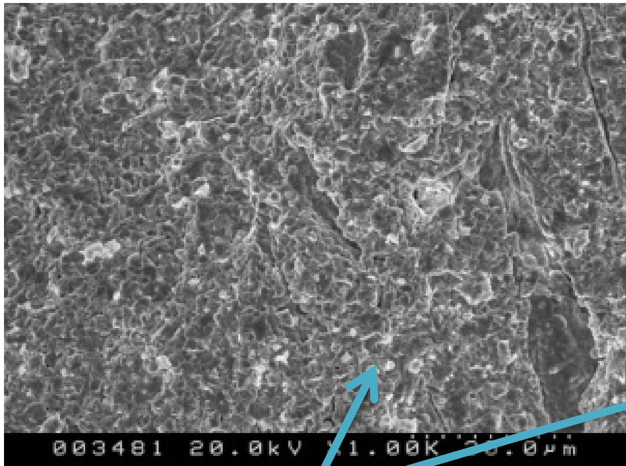


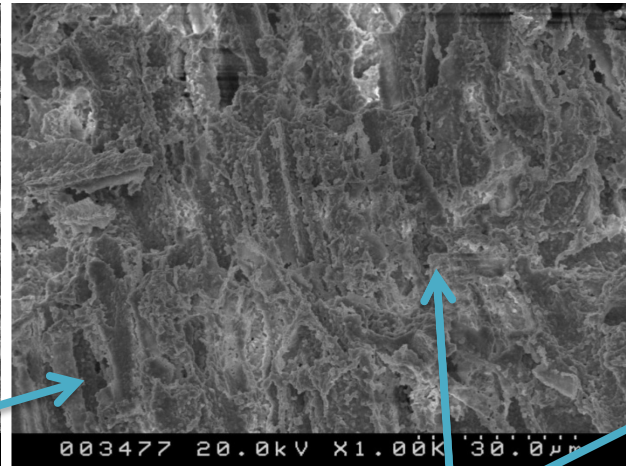
Dye Penetrant Test

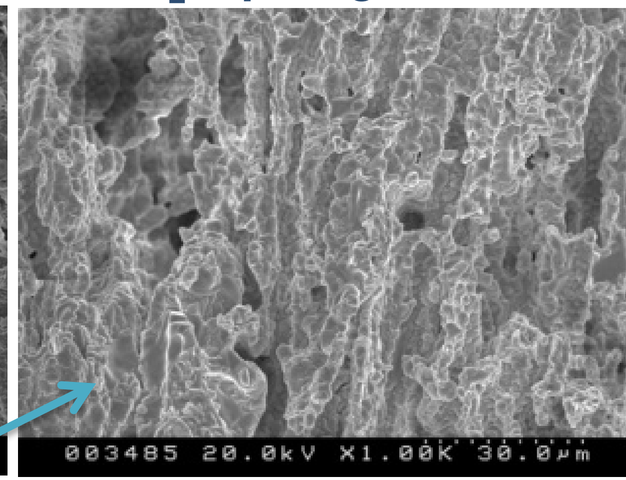
Top Bottom

Grain Growth Inhibition Demonstrated

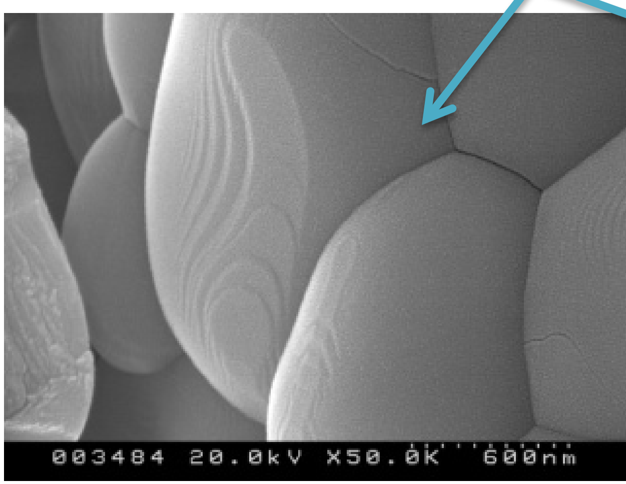
HfO_2 after 24 h@1500 °C	HfO_2 + acicular porogen	HfO_2 + porogen + GGI

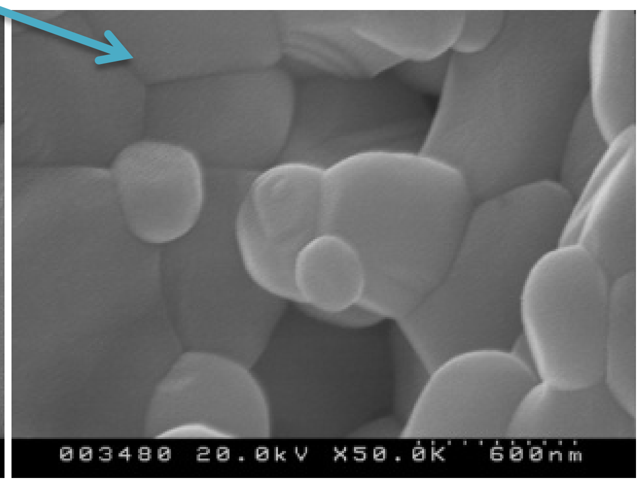


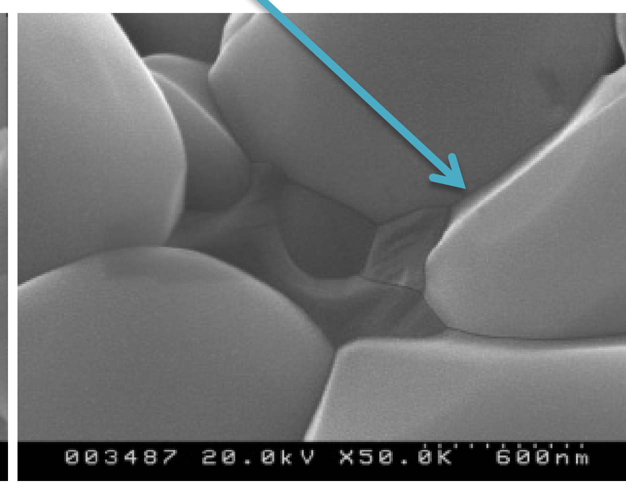




Imaged at x1k and x50k | **Needle like pores** | **Grain boundary phase**



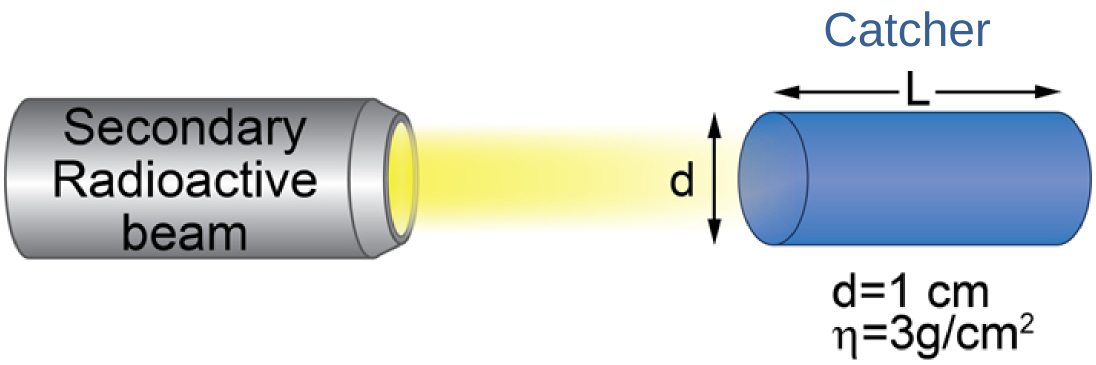




Summary of Material Properties

Material	Processing	Density (g/cm^3)	Pores Sizes	Through Porosity (Y/N)
Undoped polymer aerogel	$SCCO_2$ dried	0.08–0.27	60–300 nm	Yes
Undoped carbon aerogel	800 °C/4 h in N_2	0.07–0.35	60–180 nm	Yes
Undoped carbon aerogel	1500 °C/2 h	0.09–0.36	15–120 nm	Yes
W-doped polymer aerogel	$SCCO_2$ dried	0.36–0.40	0.3–3 µm	Yes
W-doped carbon aerogel	800 °C/4 h in N_2	0.39–0.41	0.3–3 µm	Yes
W-doped carbon aerogel	1500 °C/2 h	0.41–0.51	0.1–1.2 µm	Yes
Hafnia	1500 °C/7 day	4.34	1–30 µm	Yes (15%)
Hafnia + acicular porogen	1500 °C/7 day	2.76	7W x 30L	Yes (49%)
Hafnia + acicular porogen+ grain growth inhibitor	1500 °C/7 day	4.79	5–7W x 30–40L	Yes (49%)

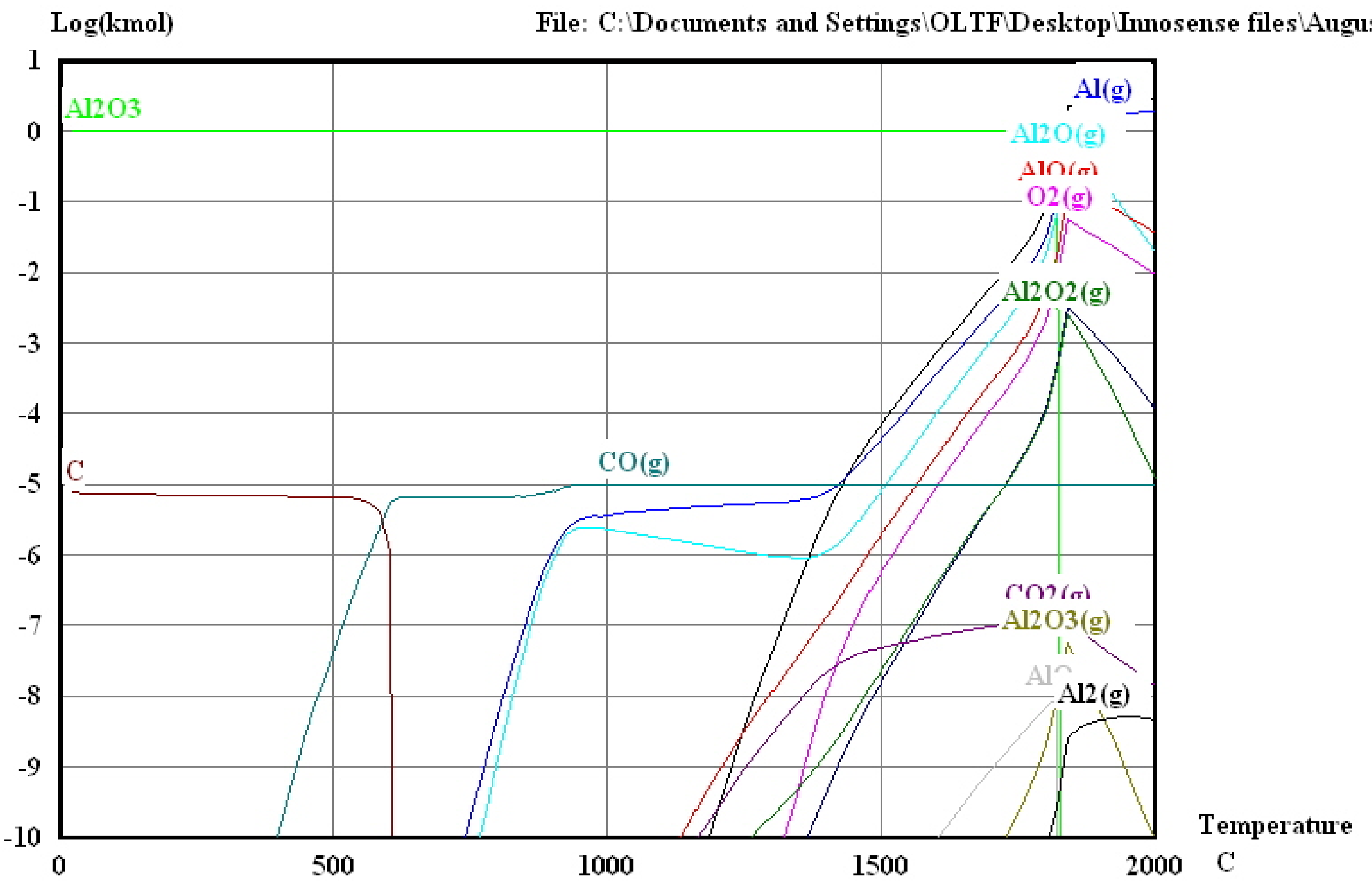
Catcher Thickness Considerations



- Desired area density (η) for efficient isotope capture is ~ 3 g/cm^2 or more
- Area density can be related to the volumetric apparent density (ρ) measured by:
  - $\eta = \rho L$
- This value are used to screen catcher disks after the 1500 °C vacuum heat treatment

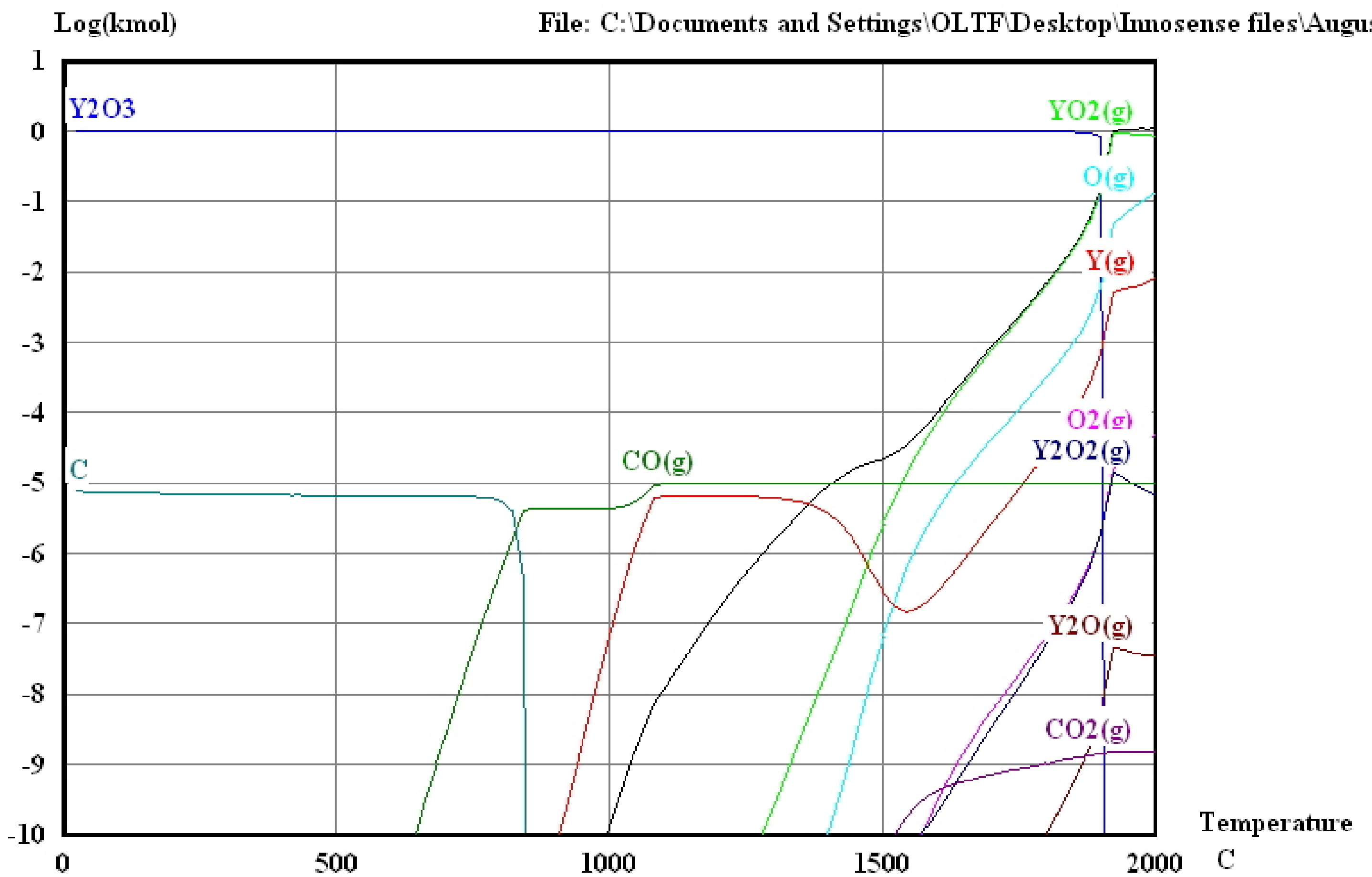
- W-doped carbon aerogels are @50% of targeted density
- Tape cast porous oxides are ~3-4x the targeted density with 50% open porosity

Simulation of CO Release from Alumina



- **HSC Chemistry Software for simulations**
- **CO release can take place at 700 °C.**

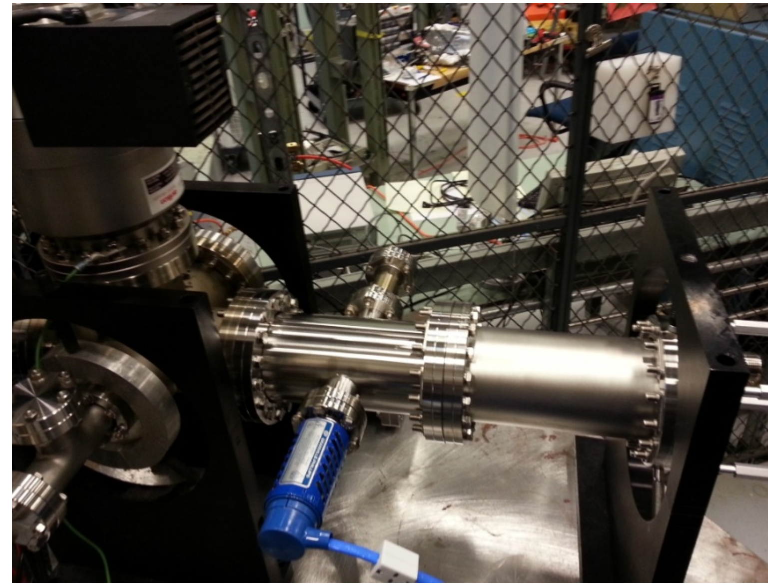
Simulation of CO Release from Yttria

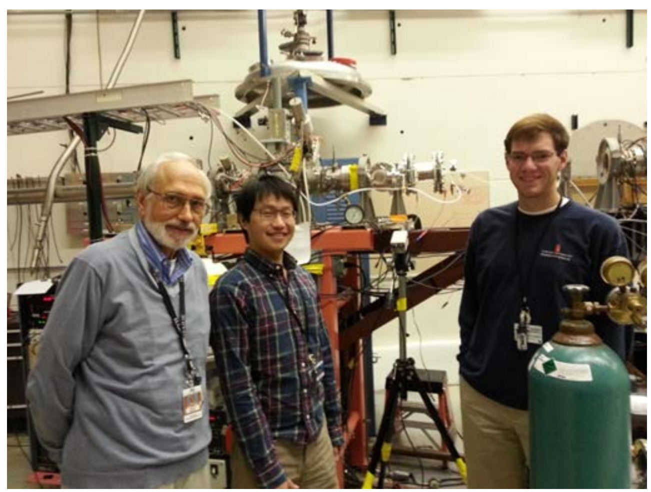


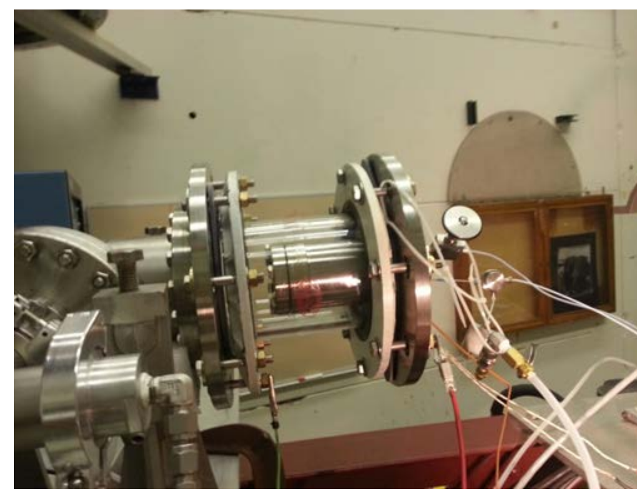
CO release takes place at ~800 °C.

New RGA Method for Release Efficiency Under Development at Argonne

Extrel
Core Mass Spectrometers

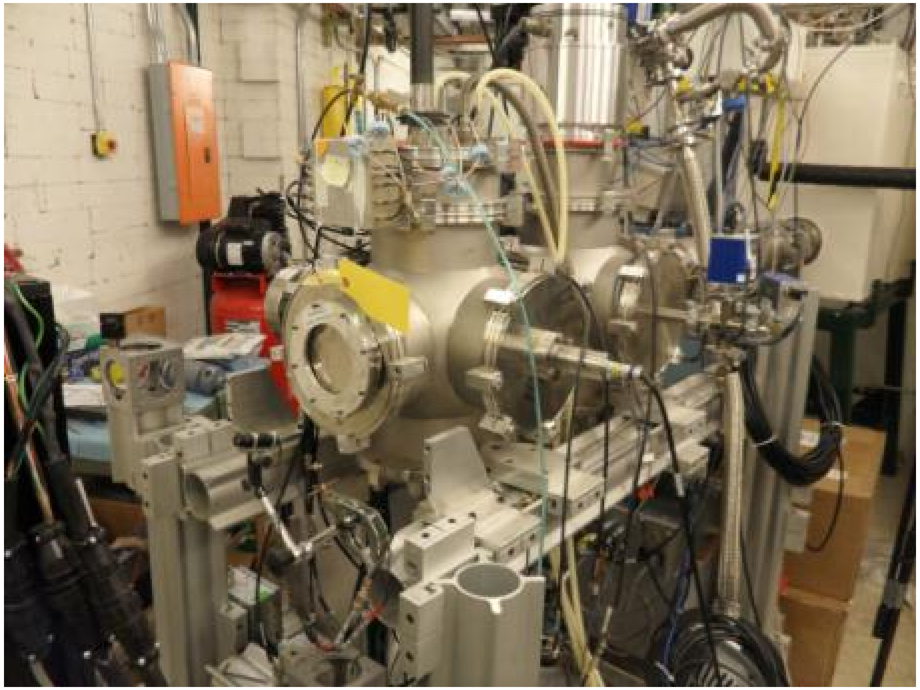




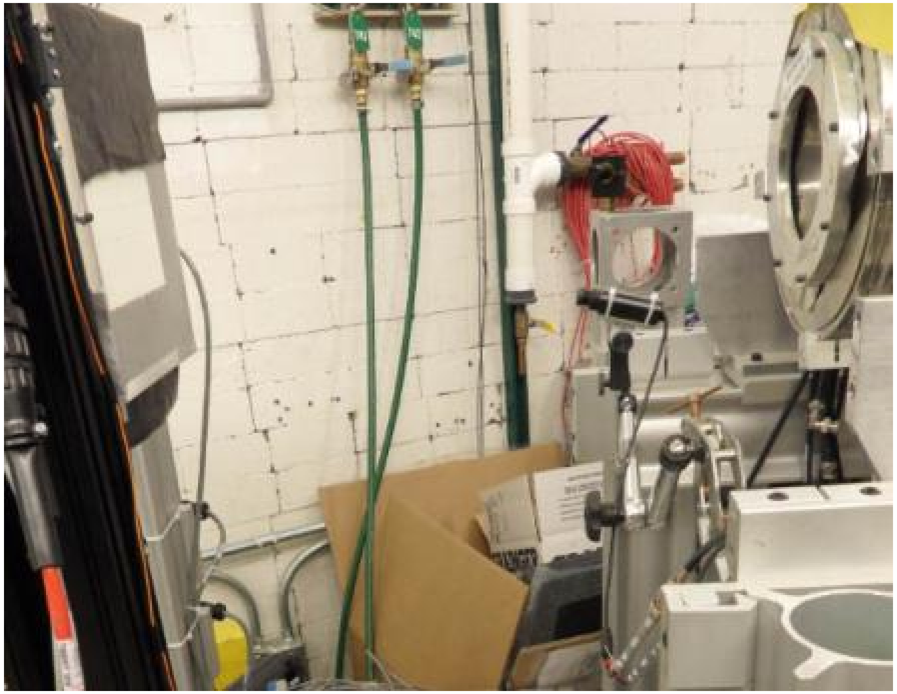


Irradiation Beamline at NSCL

The beam exits through the thin window shown in this view. Beam intensity and profile detectors are in the beam line before the thin window.



The thin window is at the right side of this view. At the left is a scintillating screen to view the beam spot. The space between these is available for the stopper/heater/ion source and decay detector for release time measurements.



Summary

- Density of W-doped carbon aerogels ~0.42–0.51 g/cm^3 @1500 °C
- Density of undoped carbon aerogels ~0.22–0.36 g/cm^3 @1500 °C
- Carbon aerogel disks intact when heated to 1500 °C
- Carbon aerogels retain through porosity confirmed by dye test
  - 15–120 nm pores (undoped); 0.1–1.2 µm pores (W-doped)
- Density of tape cast oxide monoliths ~2.76 g/cm^3 (1500 °C/7 days)
  - 50% Hg intrusion porosity
  - Needle like pores ~30 nm wide –54 µm long
- Grain growth inhibition is demonstrated for Hafnia
- New RGA method for release characteristics of stable isotopes is being developed, trace ^{13}C implanted in Al_2O_3
- New heater design completed for in-beam studies
- In-beam tests pending for all catcher materials at FRIB

Acknowledgments

DOE and the Office of Nuclear Physics to support these efforts through the following grants DE-SC0004265 and DE-SC0007572

Program Officer – Dr. Manouchehr Farkhondeh

Dr. Georg Bollen for technical discussions and sustained interest to evaluate the catcher materials at FRIB

Dr. Dan Stracener for the HSC Chemistry simulations of release studies