Carbon Aerogels-Hot Catchers for Exotic Isotopes and/or Molecular Species

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Small Business
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Argonne NATIONAL LABORATORY

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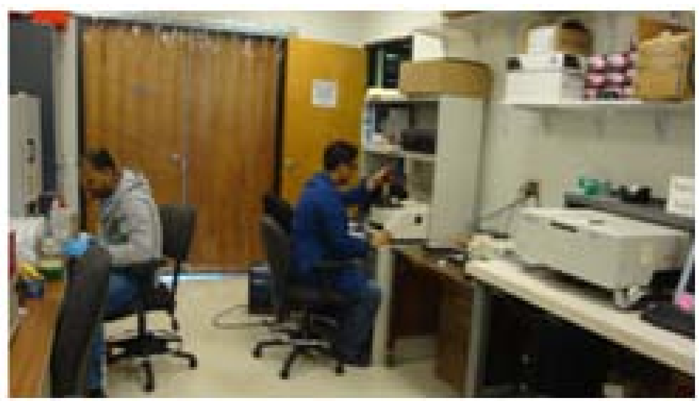
InnoSense LLC

Presentation Overview

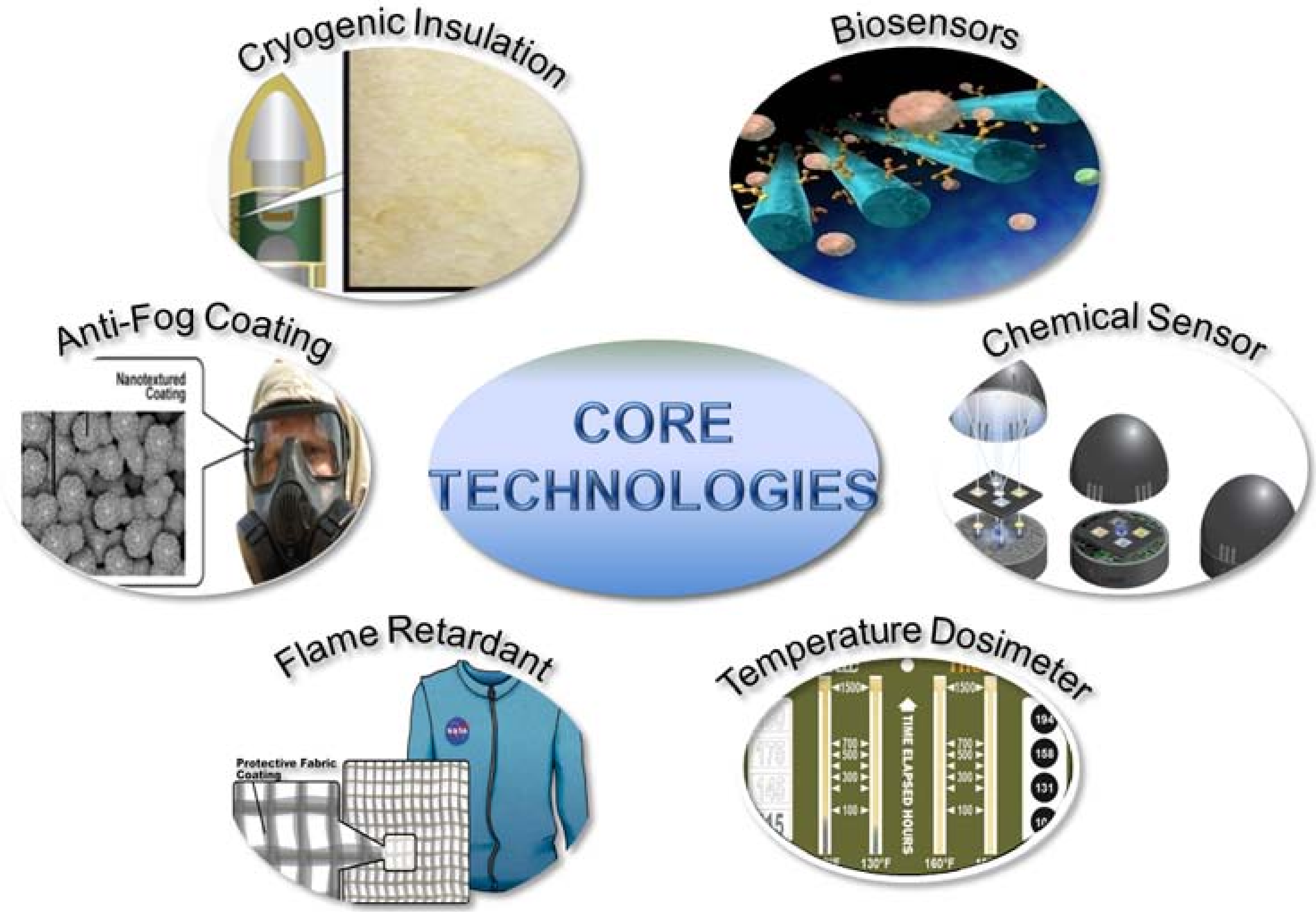
- About InnoSense LLC
- Phase II Project Goals
- Relevance to Nuclear Physics Programs
- Schedule/Deliverables
- Accomplishments to Date
- Summary/Future Plans
- Project Team
- Acknowledgments

About InnoSense LLC

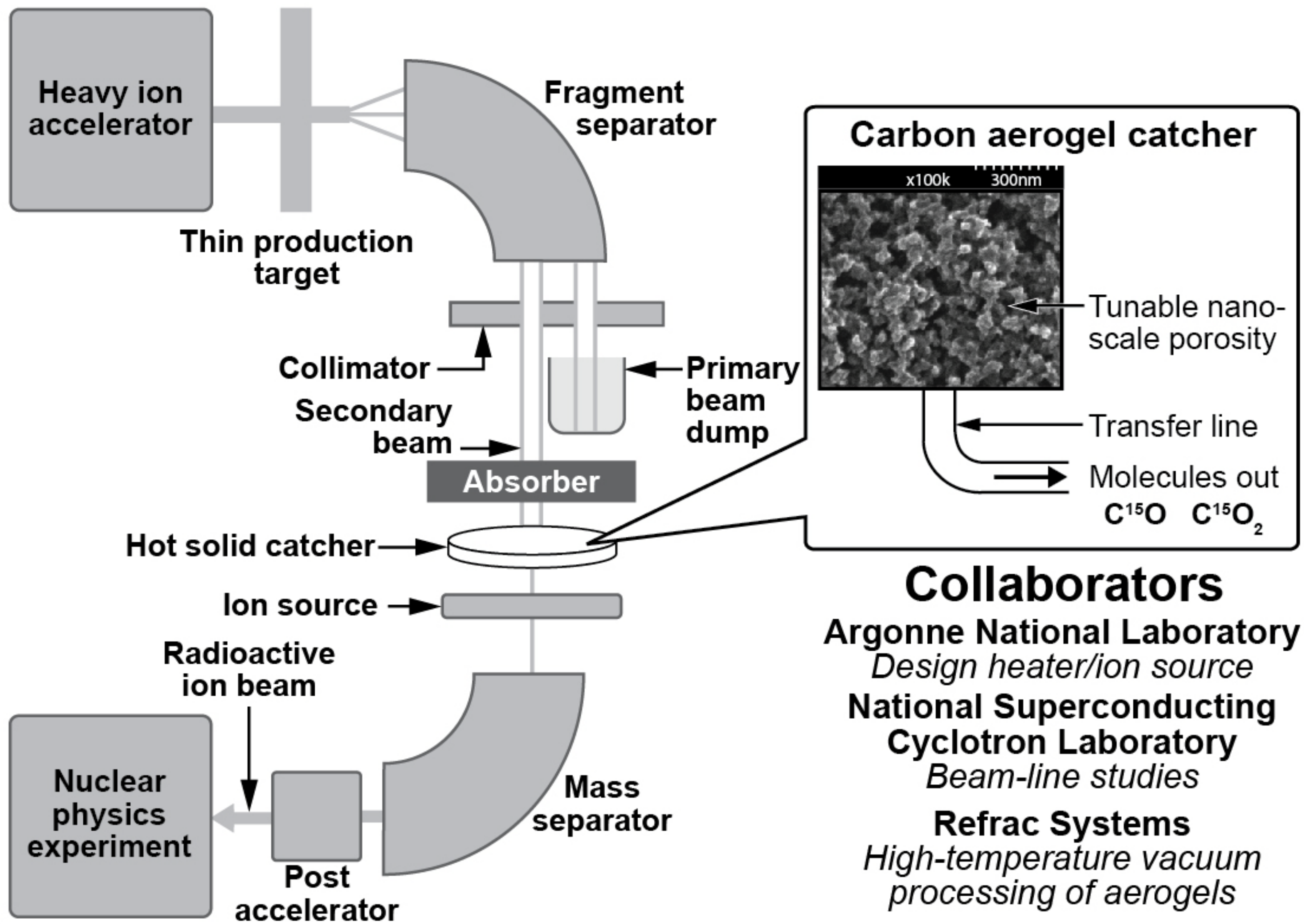
- Established in 2002 by private investment, housed in a recently expanded 9,000 square feet laboratory facility located in Torrance, California.
- Key laboratories include five “wet” chemical facilities equipped with fume hoods, a clean room, a spectroscopy facility, optics and testing laboratory, and two machine shops.
- 20 employees, including 5 PhD, 2 MBA, and 5 MS degree holders.



InnoSense LLC – Core Technologies



Refractory Hot Catchers for Rare Isotopes



Collaborators

Argonne National Laboratory
Design heater/ion source

National Superconducting Cyclotron Laboratory
Beam-line studies

Refrac Systems
High-temperature vacuum processing of aerogels

Features and benefits

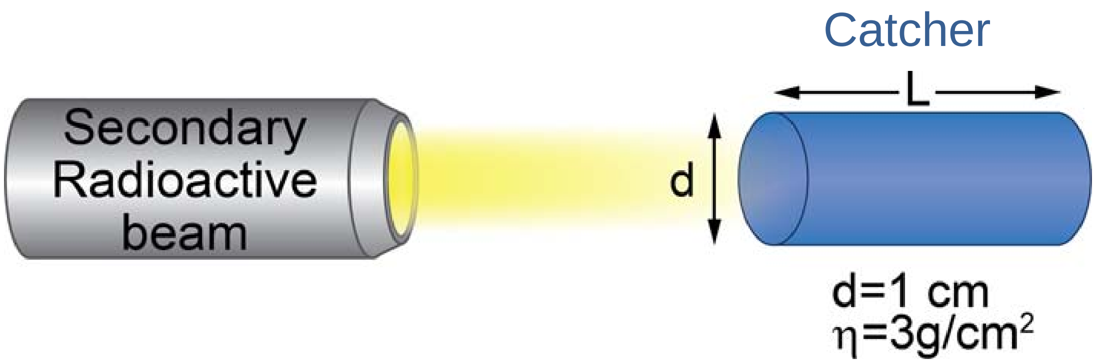
- Monolithic carbon aerogel with tunable porosity
- Stable in energetic radioactive beams
- Supplementary to the gas catcher
- Stopping power ~100 times greater than gas catchers
- Allows diffusion/ effusion of short-lived isotopes as atoms or molecules

Primary purpose of carbon aerogel is to catch ^{15}O isotopes and convert them to $C^{15}O$ expected to be released almost as a noble gas.

Catchers Under Investigation

Refractory Catcher	Projectile	Expected Isotope
W-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 porous monolith	^{12}C, ^{48}Ca, etc.	$^{9}C^{16}O$–$^{22}C^{16}O$ $^{9}C^{16}O_2$–$^{22}C^{16}O_2$

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 at ISL by:
  - $\eta = \rho L$
- This value will be used to screen catcher disks after the 1500 °C vacuum heat treatment
- Presently W-doped carbon aerogels are at 50% and undoped carbon aerogels are at 25% of targeted volumetric density
- YSZ catchers are at targeted density of ~3.1 g/cm^3 and ~ 48% porosity

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 for isotopes for different applications

Technical Objectives and Milestones

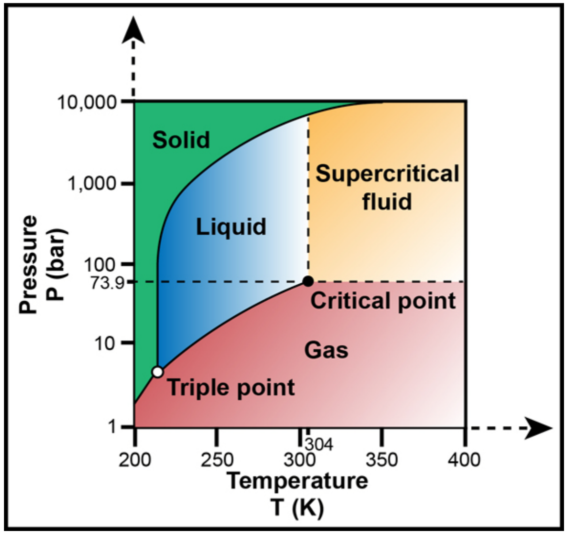
Objectives

1. Refine formulations and processing conditions to reproducibly fabricate polymer and carbon aerogels.
2. Evaluate the physical properties of the polymer aerogels after supercritical CO_2 drying and carbon aerogels after heating to 800 °C.
3. Screen refractory carbon aerogel materials for high temperature stability from 1000 to 2000 °C and open pore structures for beamline studies.
4. Evaluate prescreened refractory carbon aerogels on-line for suitability as reactive diffusion targets for molecular species of ^{15}O.

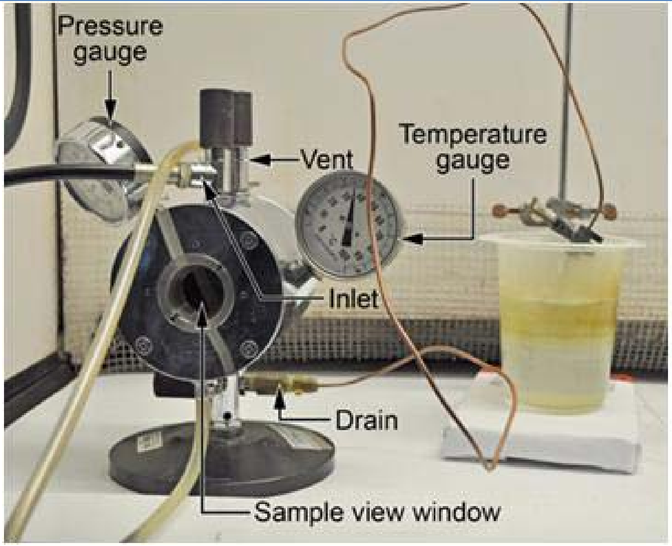
Milestones

- Fabricate carbon aerogel samples with moderate densities 1 g/cm^3 that retain their open nanoscale porosity upon heating.
- Fabricated carbon aerogel samples maintain structural and dimensional stability after heating to temperatures ranging from 1000–1500 °C.
- On-line measurement of the release times of radioactive $C^{15}O$ and $C^{15}O_2$ for the most promising samples. Test carbon catchers with a thickness of ~3 g/cm^2 are required for in-line beam tests.

Supercritical CO_2 Drying of Polymer Aerogels



Phase diagram for CO_2



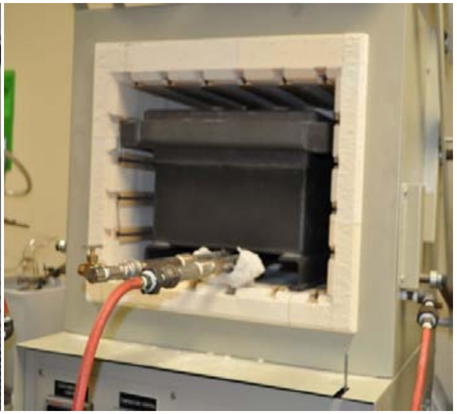
The $SCCO_2$ drier

Polymer aerogel inside drier

$SCCO_2$ drying leads to highly porous, low density, carbon monoliths

Pyrolysis of Polymer Aerogel to Carbon Aerogel

Muffle furnace



Retort in furnace

Before pyrolysis

$SCCO_2$ dried polymer aerogel

After pyrolysis

Carbon aerogel

- Heat treatment converted the polymer aerogel to carbon aerogel
- A carbon “getter” and nitrogen gas feed stream were used to reduce the likelihood of complete sample degradation.

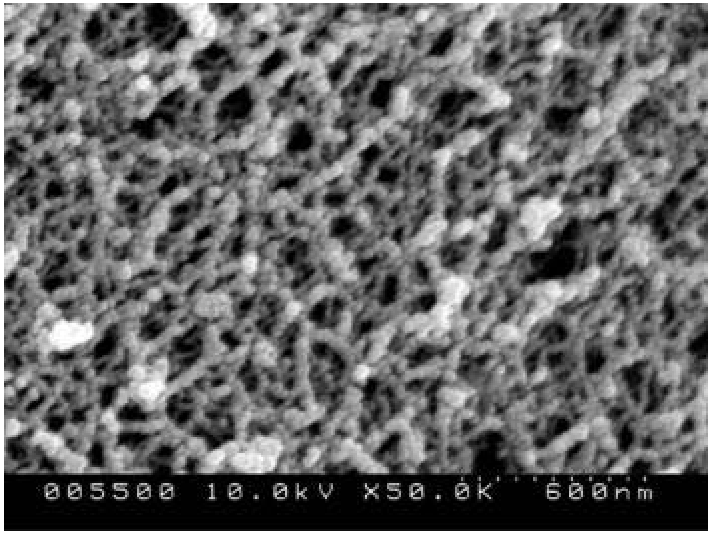
Samples Mechanically Stable After Vacuum Heat Treatment at 1500 °C

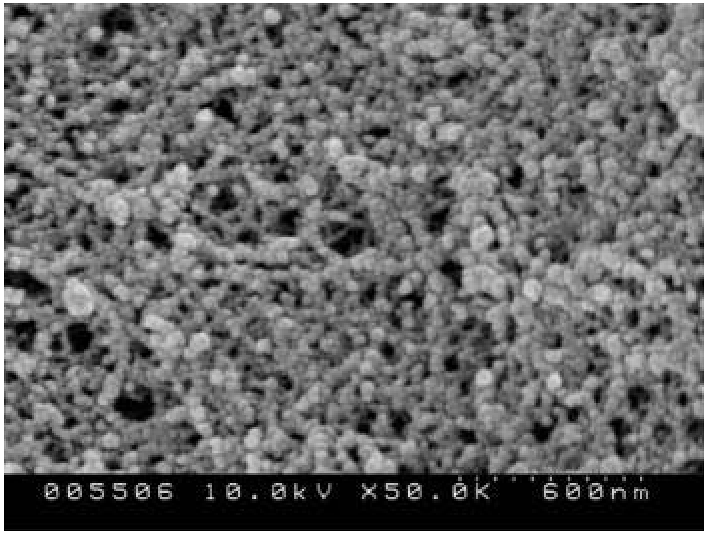
Vacuum induction furnace

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

- Samples vacuum heated at 1500C 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 of Undoped Polymer Aerogels

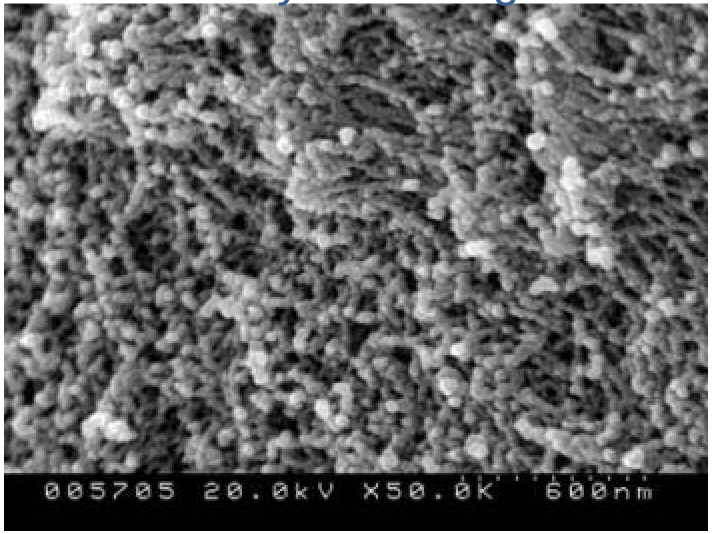


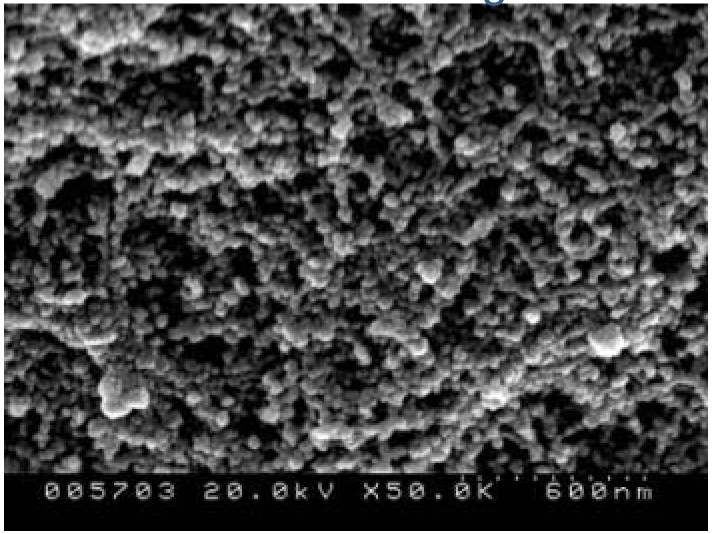


Supercritical CO_2 dried polymer aerogels

Polymer aerogel

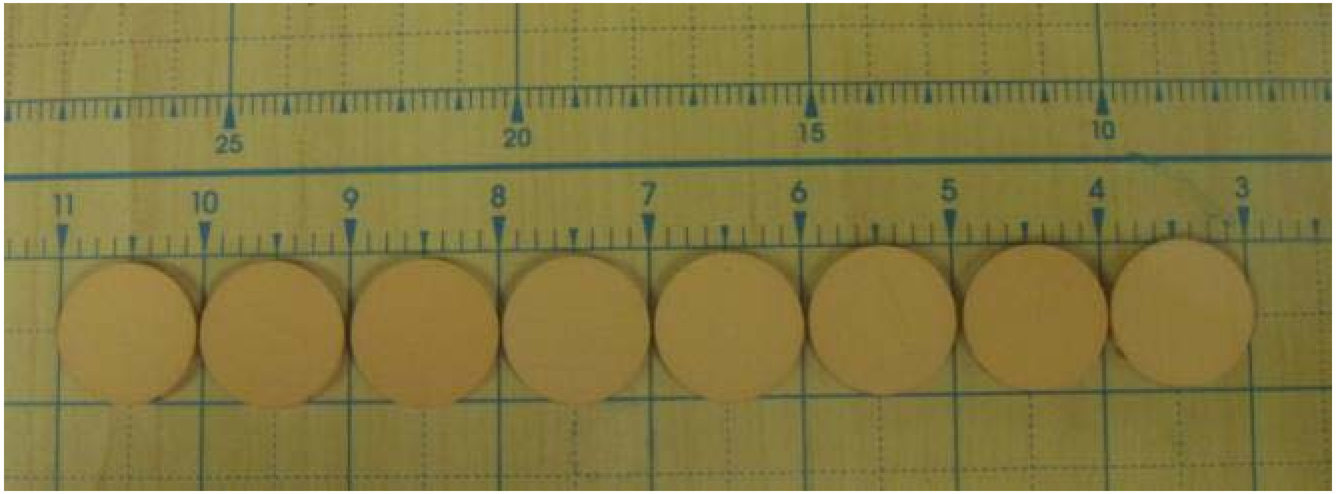
Carbon aerogel





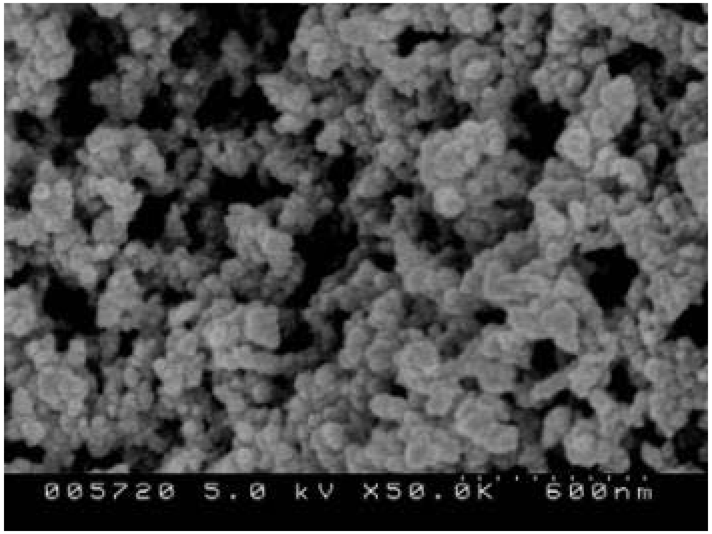
$SCCO_2$ dried polymer aerogel (left) pyrolyzed to carbon aerogel (right)

W-Doped Polymer Aerogels

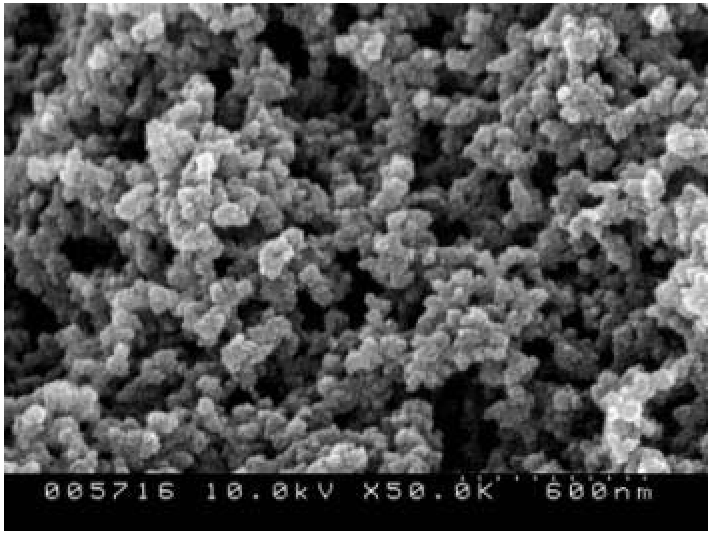


Porous monoliths are fabricated

SCCO$_2$ dried polymer aerogel

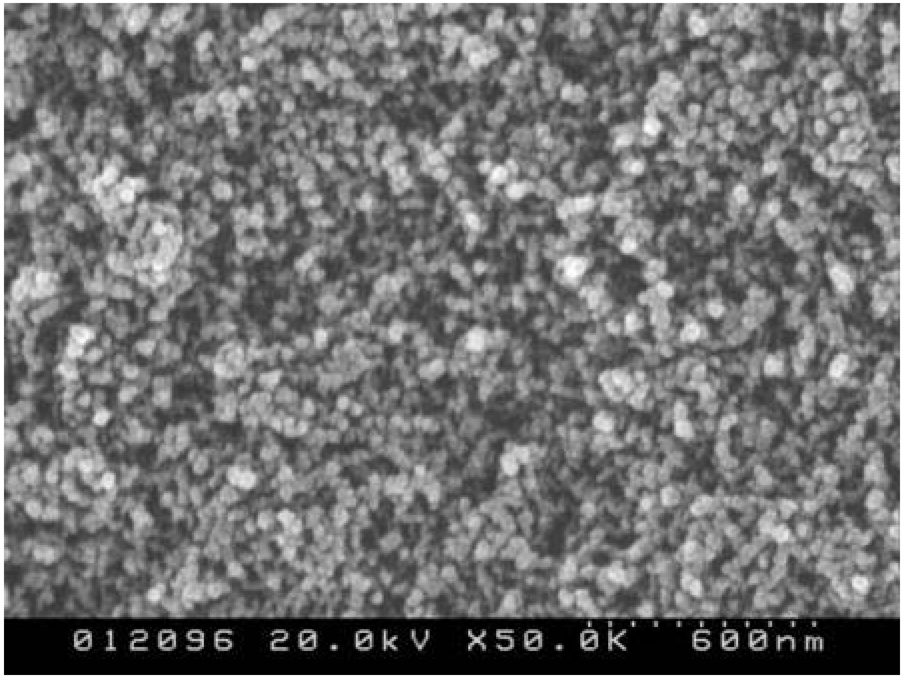


Carbon aerogel after pyrolysis

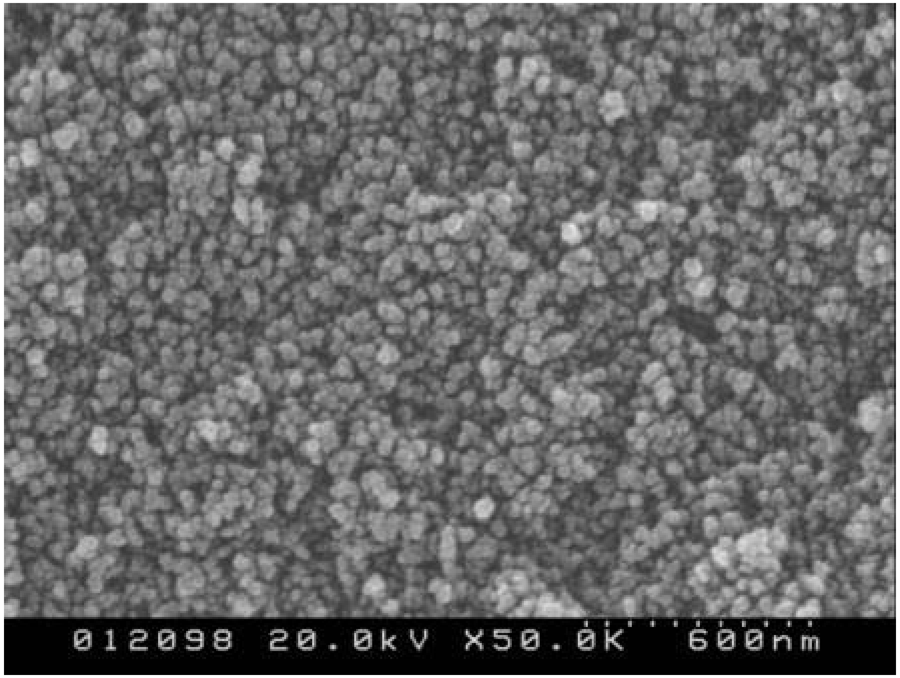


High degree of porosity

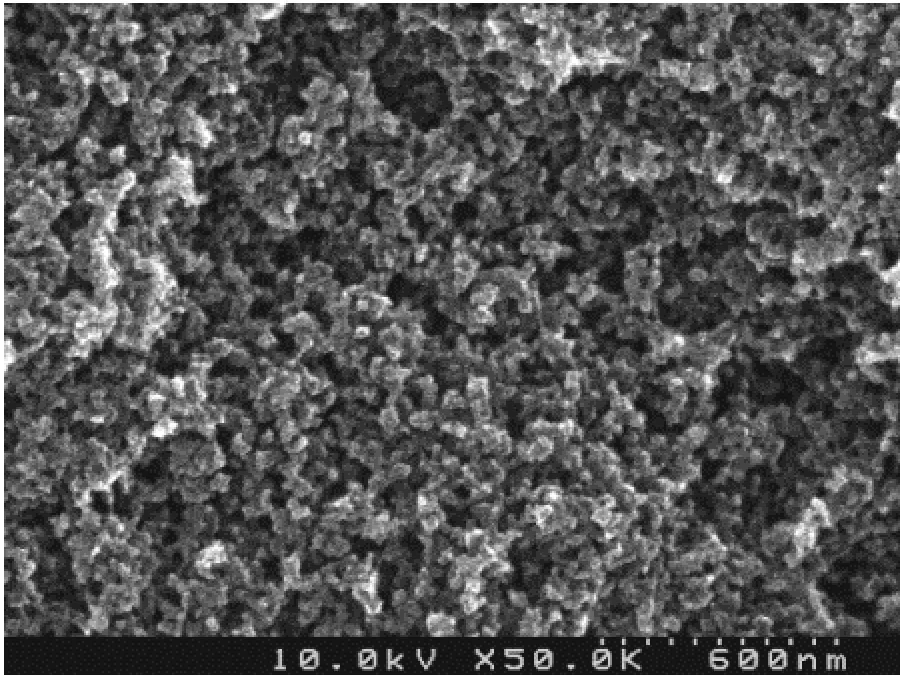
SEM Images of Undoped Carbon Aerogels After Vacuum Heat Treatment at 1500 °C



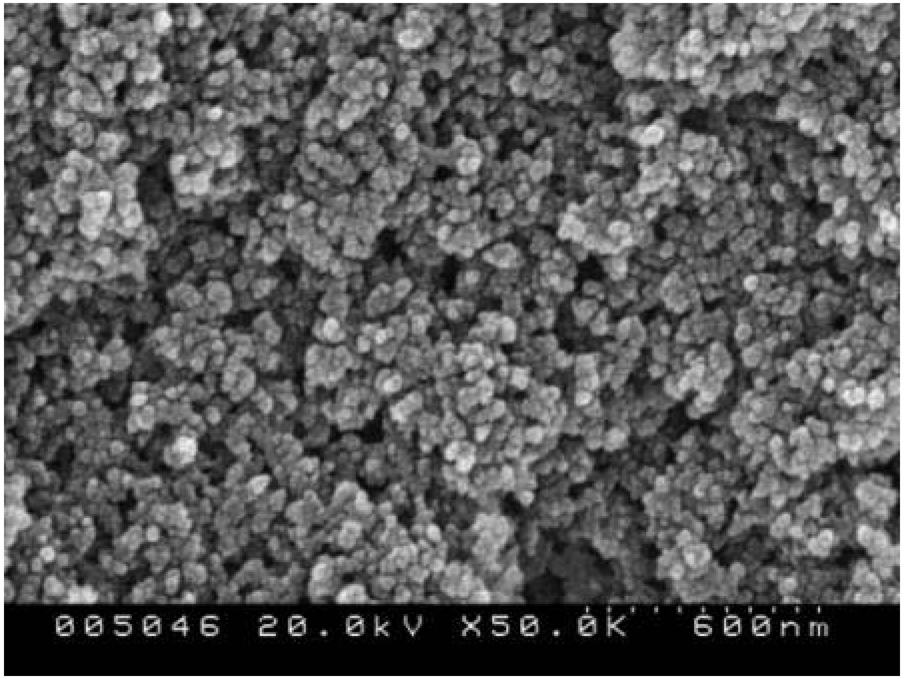
Different starting formulations to tune pore size and density



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



- **Retains open porosity**
- **Low doping increases density significantly**



Density of Polymer and Carbon Aerogels at Different Stages of Processing

Undoped polymer aerogels

- After $SCCO_2$ drying (polymer)
  - 0.08 to 0.27 g/cm^3
- After pyrolysis (carbon)
  - 0.07 to 0.35 g/cm^3
- After vacuum heating at 1500 °C
  - 0.09 to 0.5 g/cm^3

W-doped polymer aerogels

- After $SCCO_2$ drying (polymer)
  - 0.36 to 0.4 g/cm^3
- After pyrolysis (carbon)
  - 0.40 g/cm^3
- After vacuum heating at 1500 °C
  - 0.51 to 0.76 g/cm^3

Heater Design for In-Beam Studies

- Container/heater for the carbon aerogel material
- An ionizer to extract beams of radioactive isotopes of oxygen extracted from the catcher in the form of CO molecules.
- The expectation is that the oxygen isotopes will be released with delay times of ~100 milliseconds or less so that the efficiencies for very short-lived oxygen isotopes will be comparable to those of helium gas catchers without the intensity or stopping thickness limits of that technology.
- Preliminary design of the container/heater and CO ionizer complete. The design is modular, consisting mostly of commercially available hardware with very few custom made components.
- Molecular ionizer (in place of cold finger) is being designed for the release time measurements based on the expectation that the release times will be shorter than the delay times associated with diffusion of the CO molecules from the container/heater to the cold finger.
- Initial use of an RGA with stable beams is being considered for accurate release curve determination

Heater Design for In-Beam Studies (Cont.,)

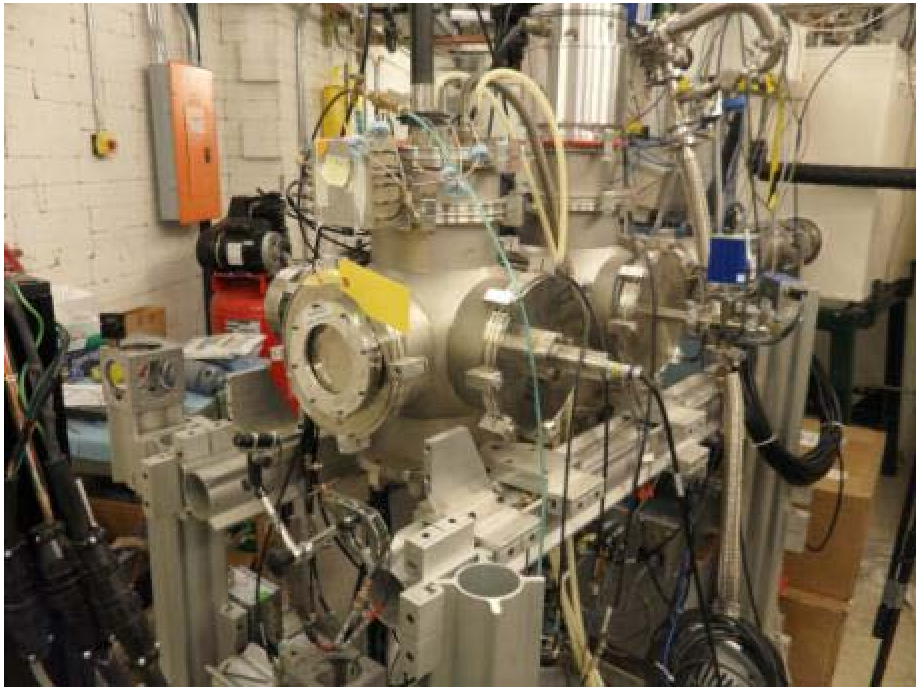
- The design is also compatible with carrying out the tests at the NSCL radioactive beam facility on the beamline known as The Single Event Effects (SEE) Test Facility.
- This beamline is a dedicated in-air irradiation station with complete diagnostic equipment and controls, located in the S2 experimental vault.
- Tentative choice of ^{15}O as the isotope of choice for these measurements has been made.
  - This is based on the high yields (>10^9/s) of this isotope at the NSCL combined with its convenient half-life of 122 s.
  - This half-life is long compared with the expected release time of the CO molecules, while being short enough to decay fast enough for detection of the isotope via direct counting of the relatively high energy positrons emitted.
  - The design is compatible with direct detection of the positrons with high efficiency via a plastic scintillator and photomultiplier tube in the air outside a thin vacuum window.

Irradiation Beamline at NSCL

- NSCL/FRIB has hired a staff member to develop isotope harvesting capabilities (J.J. Das from ORNL began August, 2012) and he is available to assist with these catcher projects as part of his new job.
- An existing irradiation station beam line is available and well equipped to locate our new catcher/heater/ion source apparatus.
- Beam profile detectors are in place just before the secondary radioactive beam exits through a vacuum window.
- The new apparatus will be placed in an independent vacuum enclosure just downstream of the beam line vacuum window (see photos on the next slide)

Irradiation Beamline at NSCL (cont.)

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.

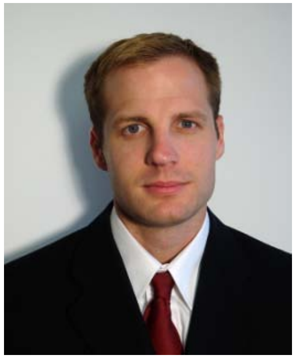
Summary / Future work

- W-doped carbon aerogels currently at ~ 0.5–0.76 g/cm^3 after 1500 °C
  - Use supercritical CO_2 extraction to process aerogels for open surface pores to improve density of monoliths
  - Vary W-loading to increase final density at 1500 °C to ~ 1 g/cm^3
- Undoped carbon aerogels currently at 0.2–0.4 g/cm^3 after 1500 °C
  - Refining process parameters to increase density of samples for use in beam line experiments.
- Mechanical stability of aerogel disks demonstrated at 1500 °C
- Heater design completed
- Beam line tests pending for all catcher materials

Key Project Personnel at InnoSense LLC

Uma Sampathkumaran, PhD, Vice President, R&D

- Development of ORMOSILs and aerogel structures
- Nanocomposites for optochemical sensing & protective coatings
- Self-assembled monolayers
- Materials characterization



David Hess, PhD, Deputy Director, 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

Commercialization Team

Kisholoy Goswami, PhD, President and Chief Technology Officer

- Eleven relevant U.S. patents
- First commercial fiber-optic sensor
- Optical sensors
- Raising private capital

Lexi Donne, MS, Technology Transition

- Technology transition
- Client interface
- Marketing
- Four U.S. Patents

Paul Levin, BSEE, MBA, Director of Engineering, R&D

- Circuit board and overall systems engineering and design.
- Analog fiber optic transmission systems.
- Design management, and program management.

Acknowledgments

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Program Officer – Dr. Manouchehr Farkhondeh

Dr. Jeff Elam and Dr. Anil Mane for W-ALD on the silica aerogels

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