Lawrence Livermore National Laboratory

Porous Materials for Laser Target Applications

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Summary / Outline

Summary:

We are developing unique materials synthesis capabilities focused on producing porous materials with a broad range of compositions and properties for laser target applications

Outline:

Motivation

Aerogels

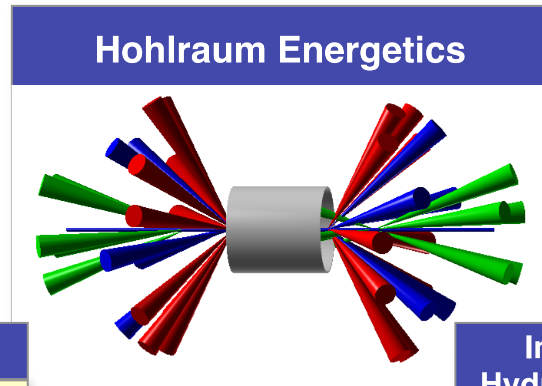
Experimental Platforms Fielded

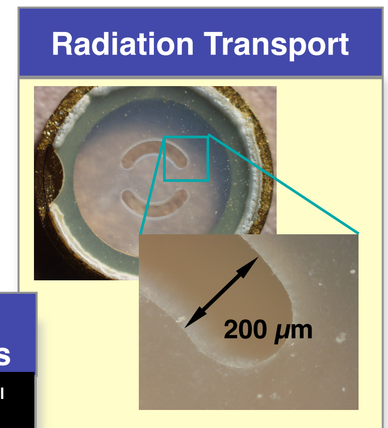
Current Development Example

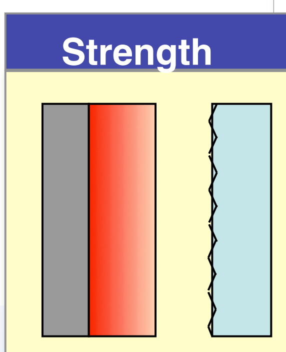
High Energy Density Physics (HEDP) experiments require materials that meet a broad range of specifications

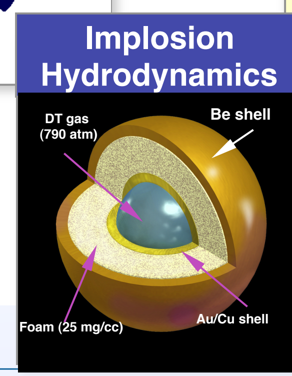
- **Many designs require porous materials with challenging attributes:**
  - ***Machined or cast*** **high- and low-Z materials (inorganic and organic)**
  - **Range of densities, density gradients**
  - **Single component, composites, or doped materials**

- **To meet these challenges, we endeavor to develop reliable methods for preparation of materials with controlled:**
  - **Composition**
  - **Cell sizes**
  - **Uniformity/homogeneity**
  - **Dopant levels**
  - ***Fashioned into a target***



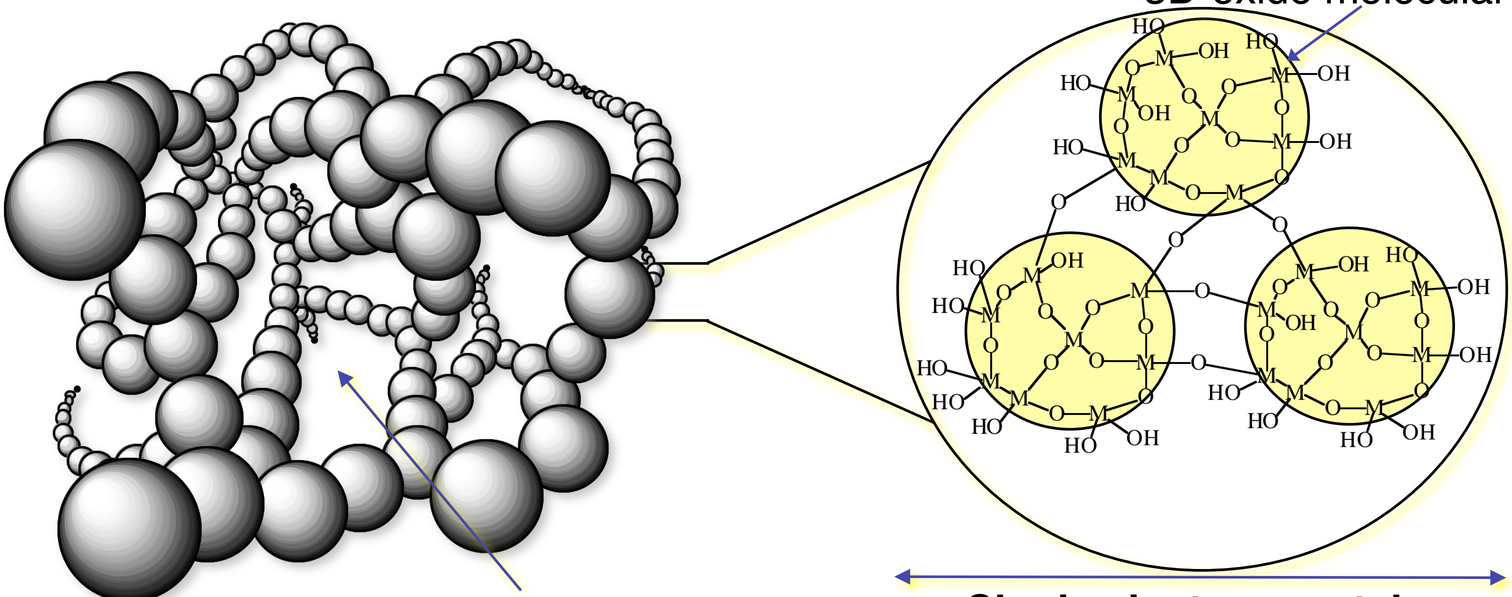






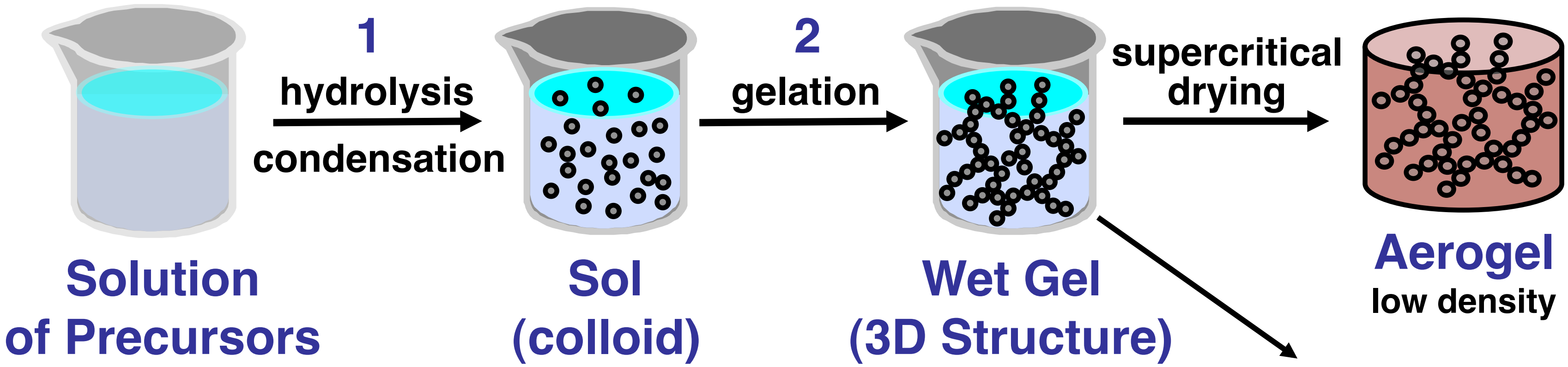
Aerogels possess a number of structural features that are desirable for laser target designs

- **Microstructure of aerogels consists of an interconnected 3D network of nanometer-sized particles:**
  - Continuous porosities (0.40-0.99+)
  - High surface areas (400 to 1000 m^2/g)
  - Low mass densities (1.8 to 0.001 g/cm^3)
  - Ultrafine cell/pore sizes (≤ 0.1 micron)

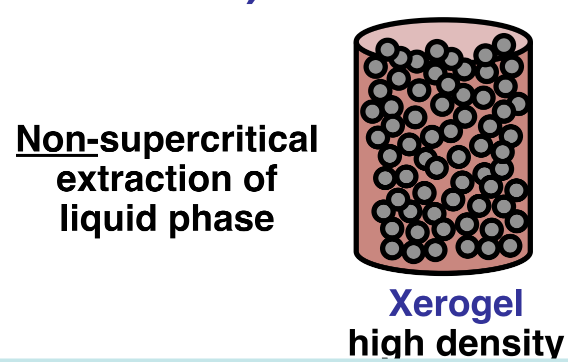


Aerogels are prepared using the sol-gel process

- Sol-gel chemistry involves two basic steps:

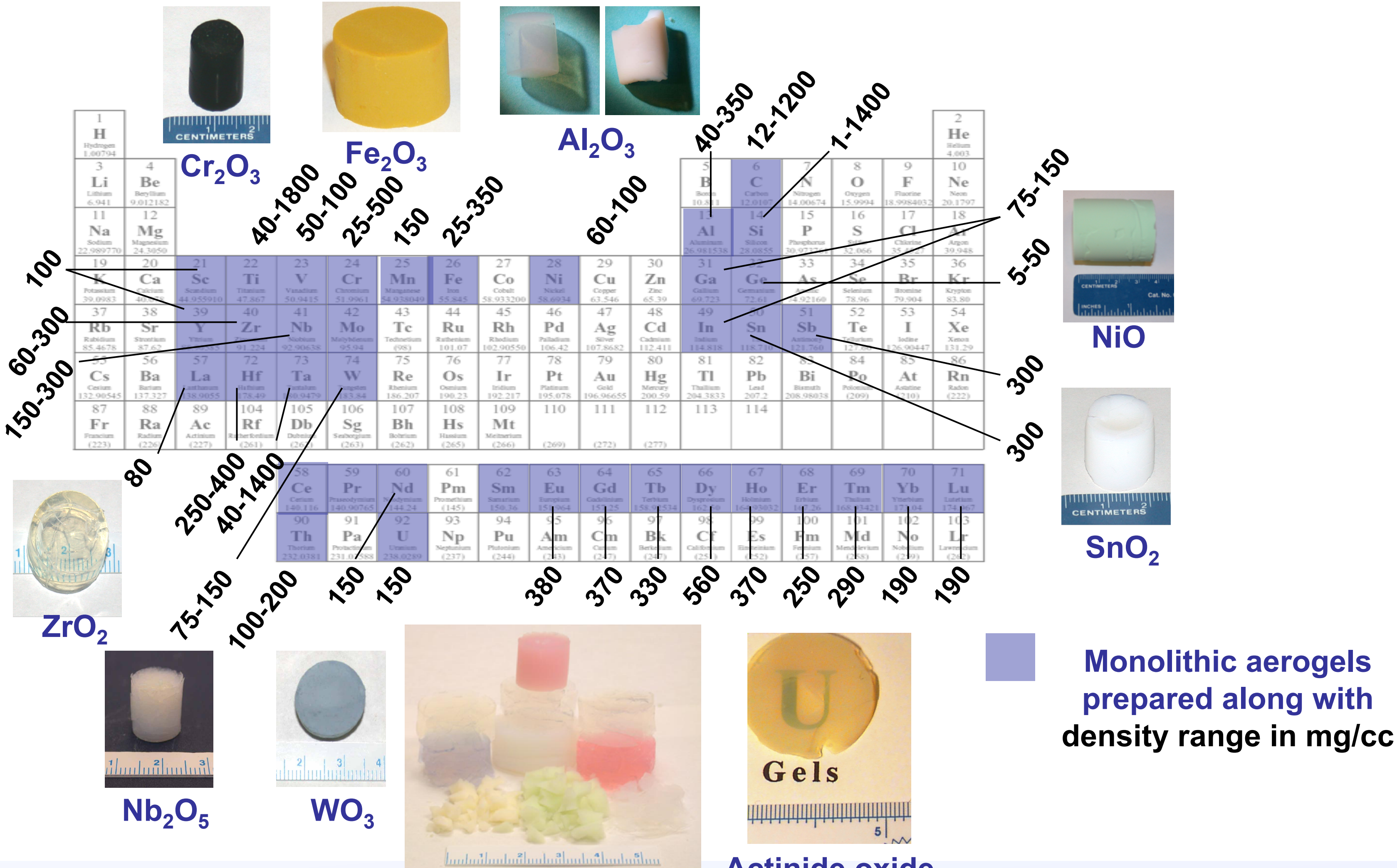


- Drying of wet gel is equally important in determining final material properties



Length scale (pore size) is a consequence of the process, not composition, however, chemistry defines structural details

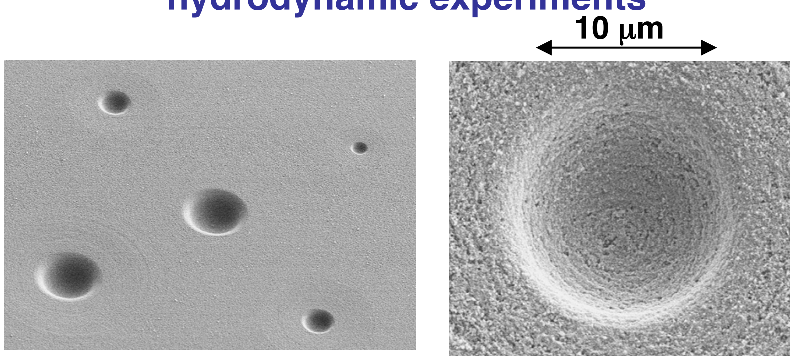
We have defined the synthesis protocols to synthesize a broad range of *monolithic* aerogel compositions



Example 1: In some designs, machineable inorganic aerogel materials are required

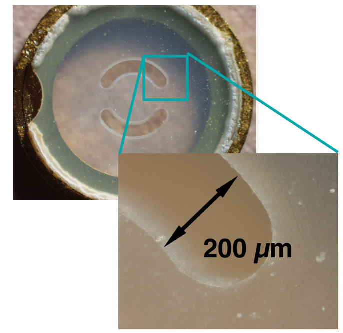
- Synthetic variables are manipulated to impact the mechanical properties of the aerogel:

Precision machined TiO_2 aerogels for hydrodynamic experiments



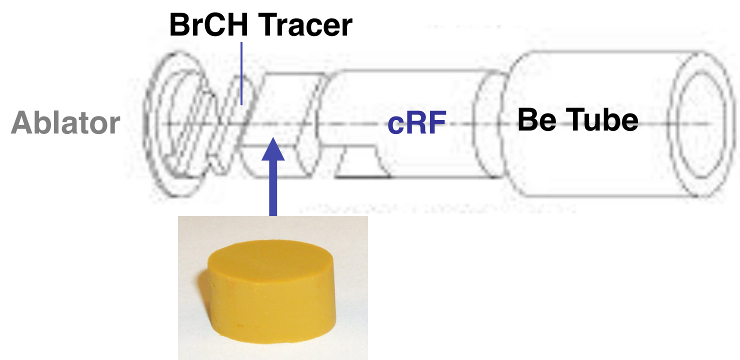
TiO_2 @ 1.8 g/cc

Machined divot
Depth: 5 μm



Precision machined Ta_2O_5 aerogels for radiation transport experiments

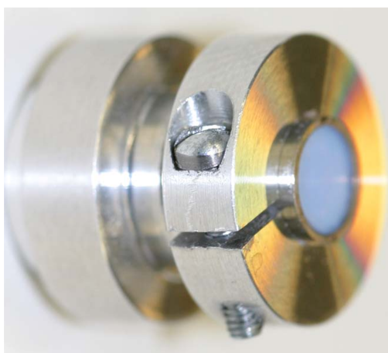
Precision machined iron(III) oxide aerogels for dynamic two color radiography experiments



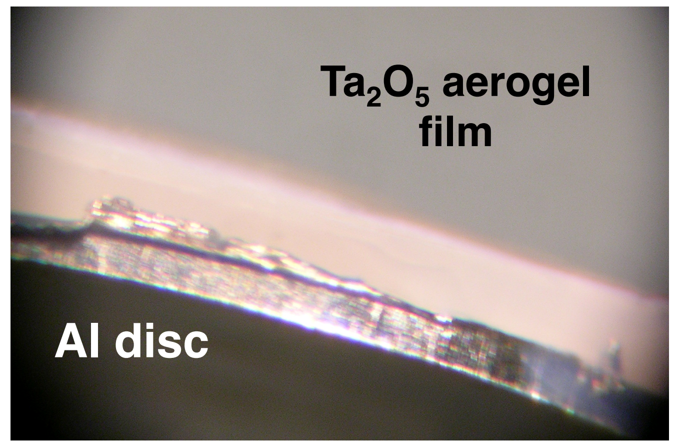
Gash, A. E. et al. *Chem. Mater.* **15**, 3268 (2003). Kucheyev et al. *Phys. Rev. B* **69**, 245102 (2004).

Example 2: In other designs, we can prepare net-cast aerogels for the fabrication of targets

- **Synthesis and processing conditions are controlled to produce materials with minimal shrinkage:**
  - **Wide range of densities**
  - **Cast in a variety of shapes (films, spheres, cylinders)**



Net shape cast **Ta_2O_5 aerogel (100 mg/cc) for Hohlraum development**

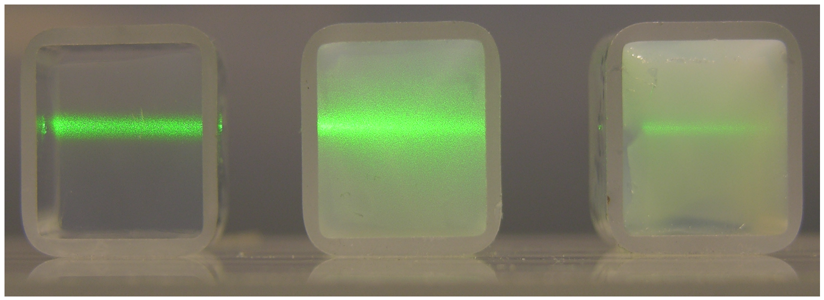


Ta_2O_5 aerogel (100 mg/cc) cast directly on Al disc for EOS targets (*surface adhesion*)

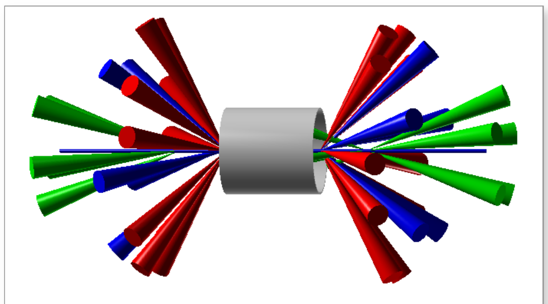
Example 3: We have also developed synthesis protocols for a series of multi-component sol-gel materials

- We have synthesized aerogels containing high-Z dopants with aggregate density below 0.2 n_c ~ < 5.0 mg/cc

Ti (hν = 4.7keV) Zn (hν = 8.9keV), Ge (hν = 10.0 keV))



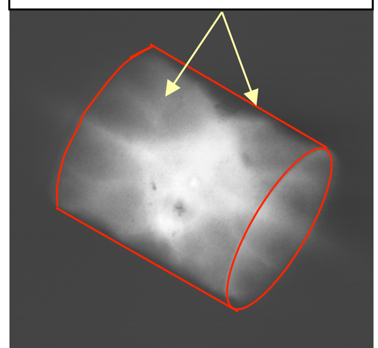
20 at % Ge/SiO_2 4 mg/cc | **3 at % Ti/SiO_2 3 mg/cc** | **GeO_2 5 mg/cc**



Omega irradiation geometry

Underdense multi-keV x-ray sources for large-area backlighters and weapons effects testing

2 propagating laser-heating waves

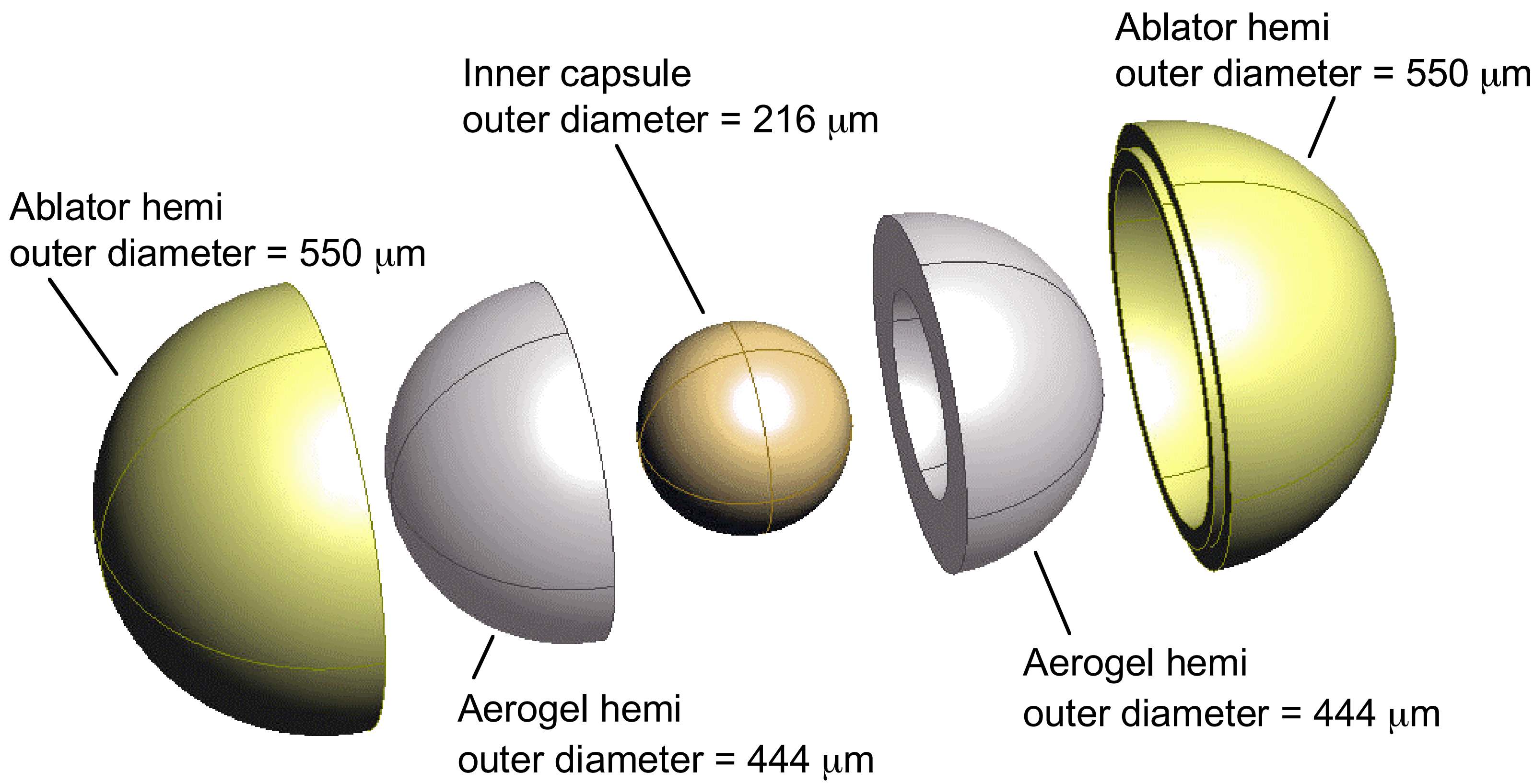


OMEGA x-ray image of Ti-doped aerogel

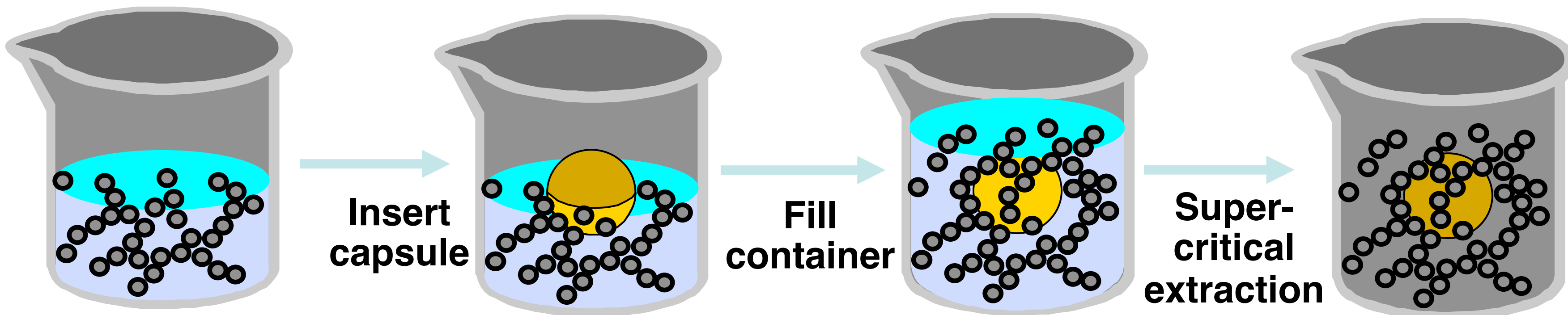
Fournier et al, *Phys. Rev. Lett*. **92**, 165005 (2004)

Process Considerations: Double shell targets were originally machined

Carbon vs. silica aerogel design



Current approach: glass capsule is cast in a monolithic piece of 50 mg/cc SiO_2 aerogel

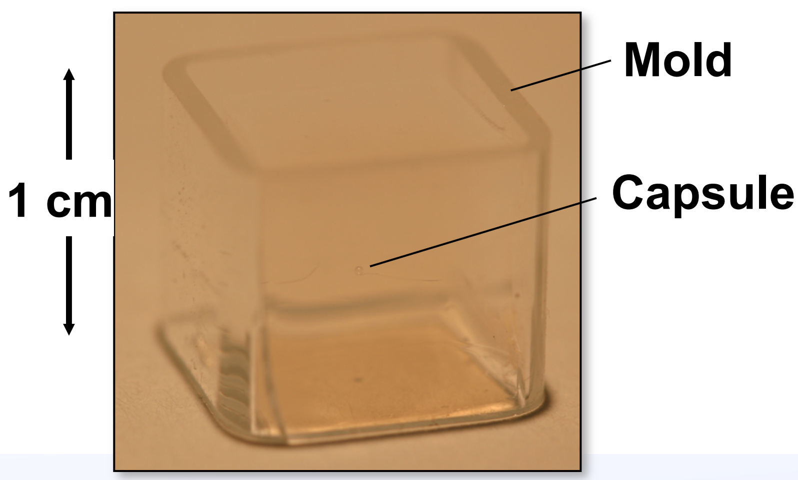


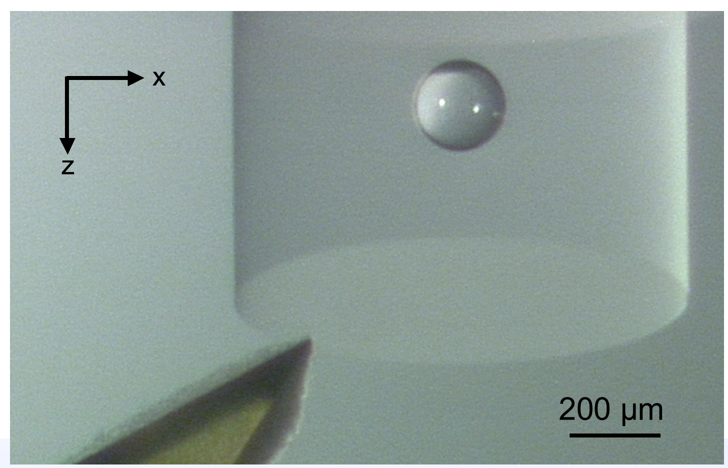
Container partially filled with solution

Capsule partially submerged

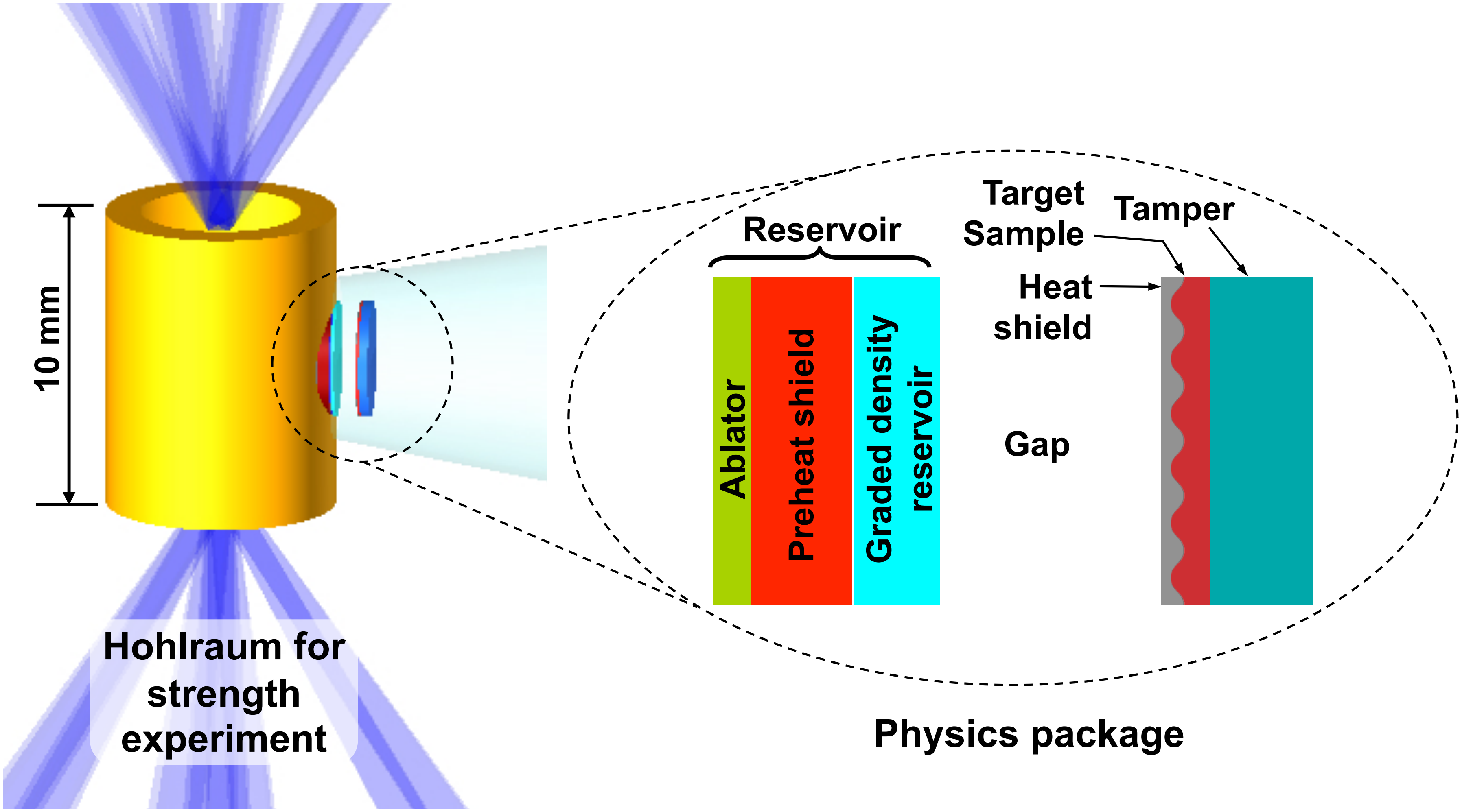
Capsule sub-merged in gel

Capsule embedded in aerogel





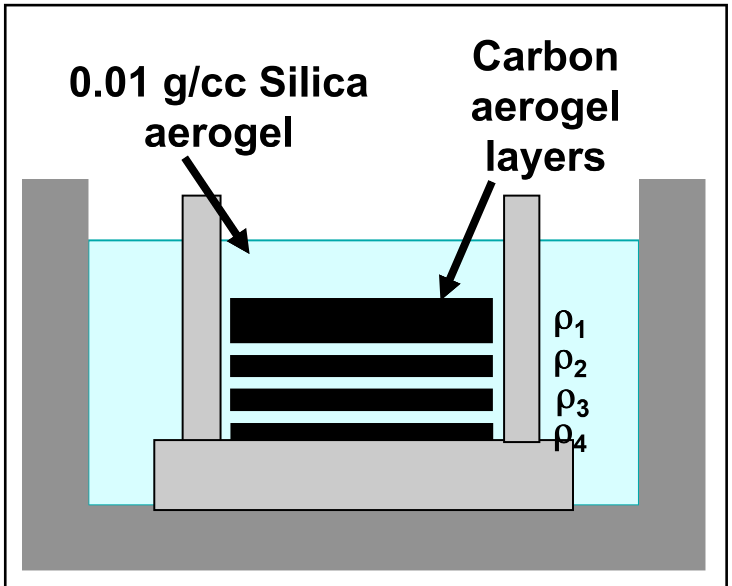
Proceess Considerations: Materials dynamics experiments measure the strength of samples at high pressure



Critical target fabrication challenges include:
— Low Z graded density structures from 1 to 0.001 g/cc

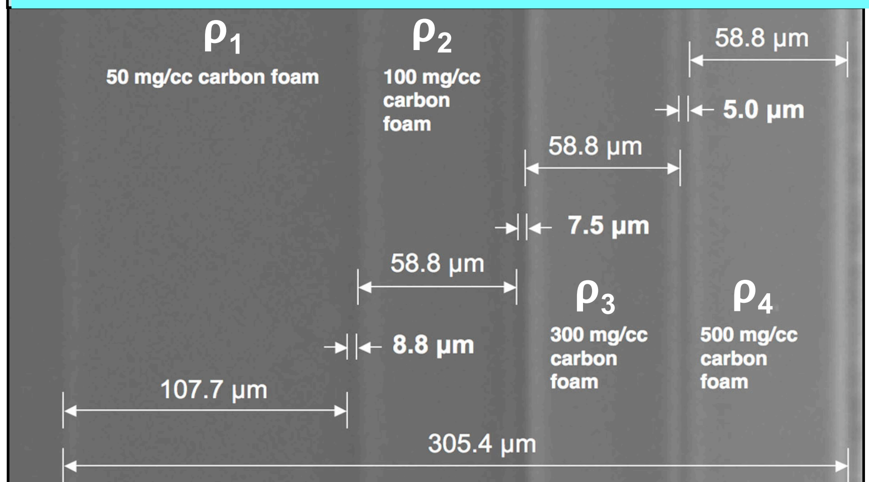
Proess Considerations: Gradient density reservoirs are made using an aerogel "glue"

Issue: Traditional bonding would introduce high density interfaces



Supercritical extraction to produce low density "adhesive" was key to success

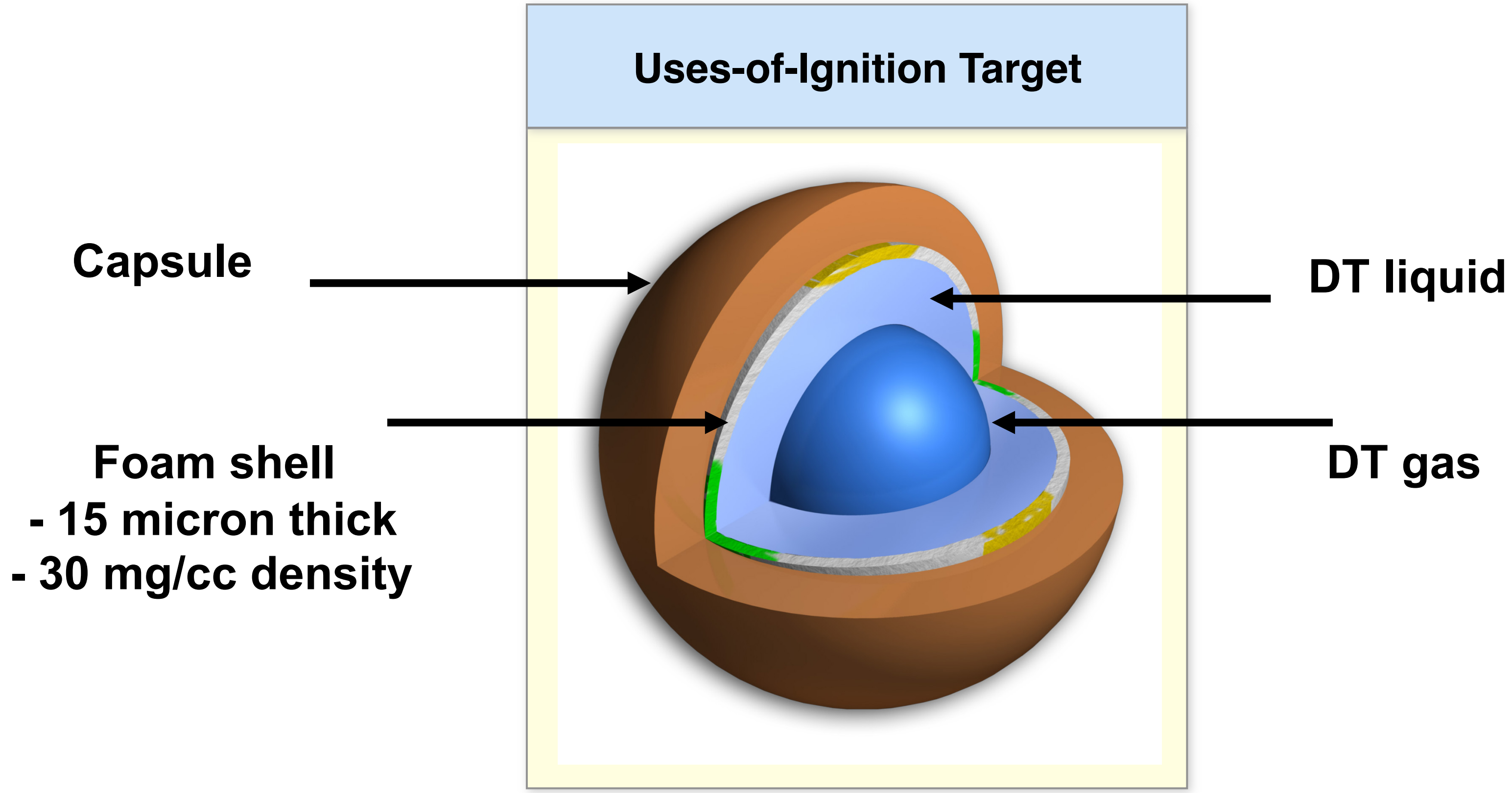
Reservoir for 2007 OMEGA Graded Density Campaign



Digital radiograph of step graded density reservoir shows gaps less than 10 µm

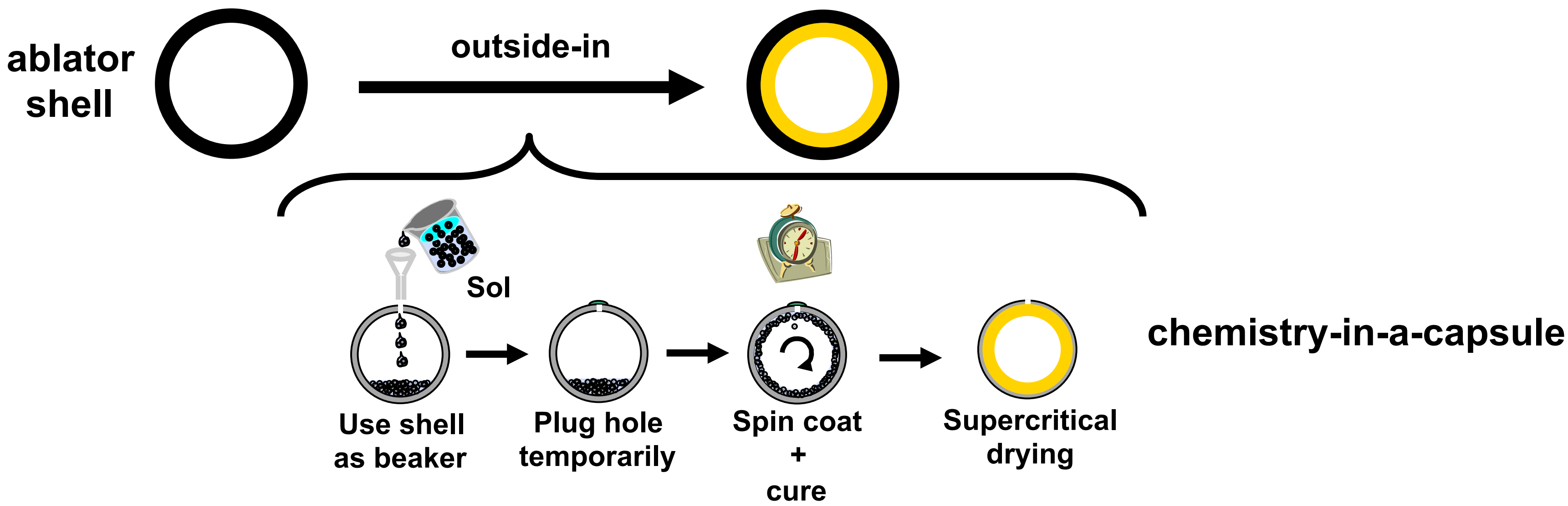
Using 0.01 g/cc interpenetrating network of silica aerogel enabled the fabrication of step function graded density reservoirs down to 0.05 g/cc

Foam shells inside a capsule



- Low density, open cell, hydrocarbon foam shell
- High-Z doping of foam, potentially spatially controlled

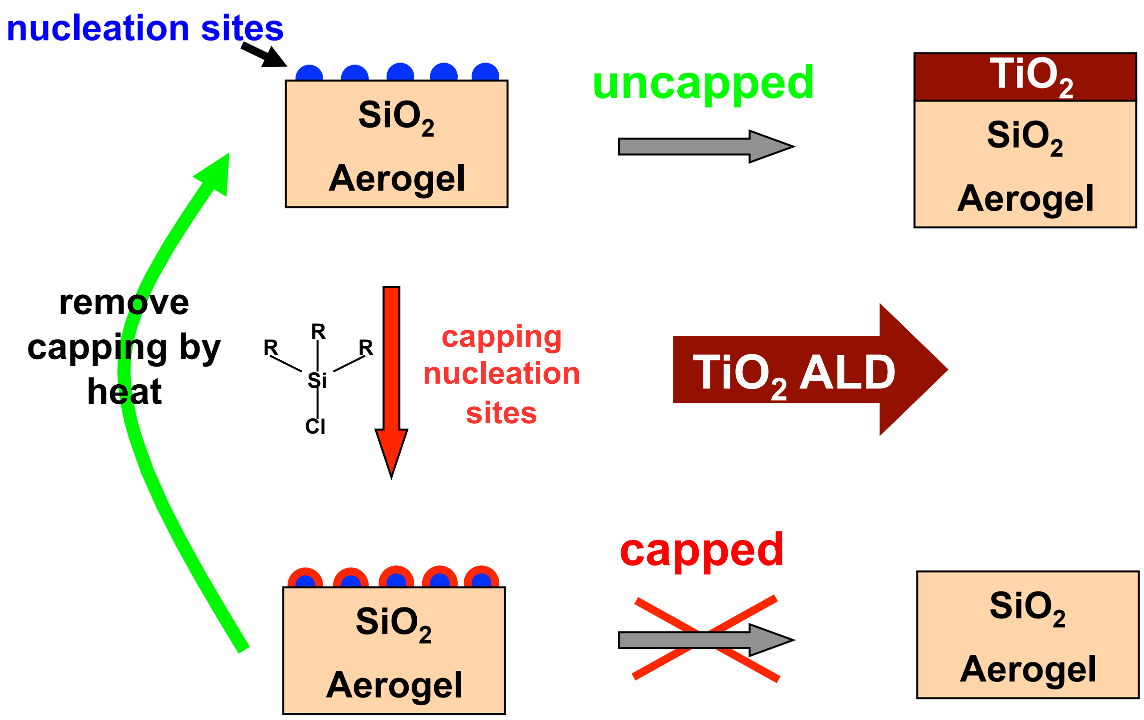
Our current approach: Chemistry-in-a-capsule

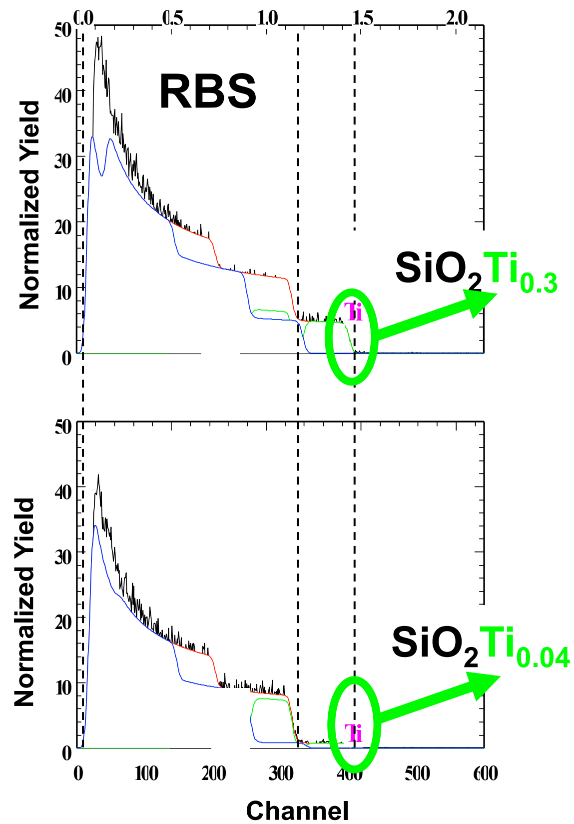


challenges	Effect/problem	solution
Picoliter volumes	Evaporation, accurate delivery	Ionic solvents, microfluidics
Mechanically robust hydrocarbon aerogel liner	Shrinkage can induce cracking Aerogel must survive hydrogen wetting	Develop new aerogels ROMP of DCPD (CH)
Controlled gel time	Catalyst deactivation by surface groups	Surface engineering
Uniform layer under shear	Viscoelastic properties at sol-gel transition	Tune aerogel chemistry

Current Development

Surface engineering enables controlled doping of aerogels via Atomic Layer Deposition (ALD)





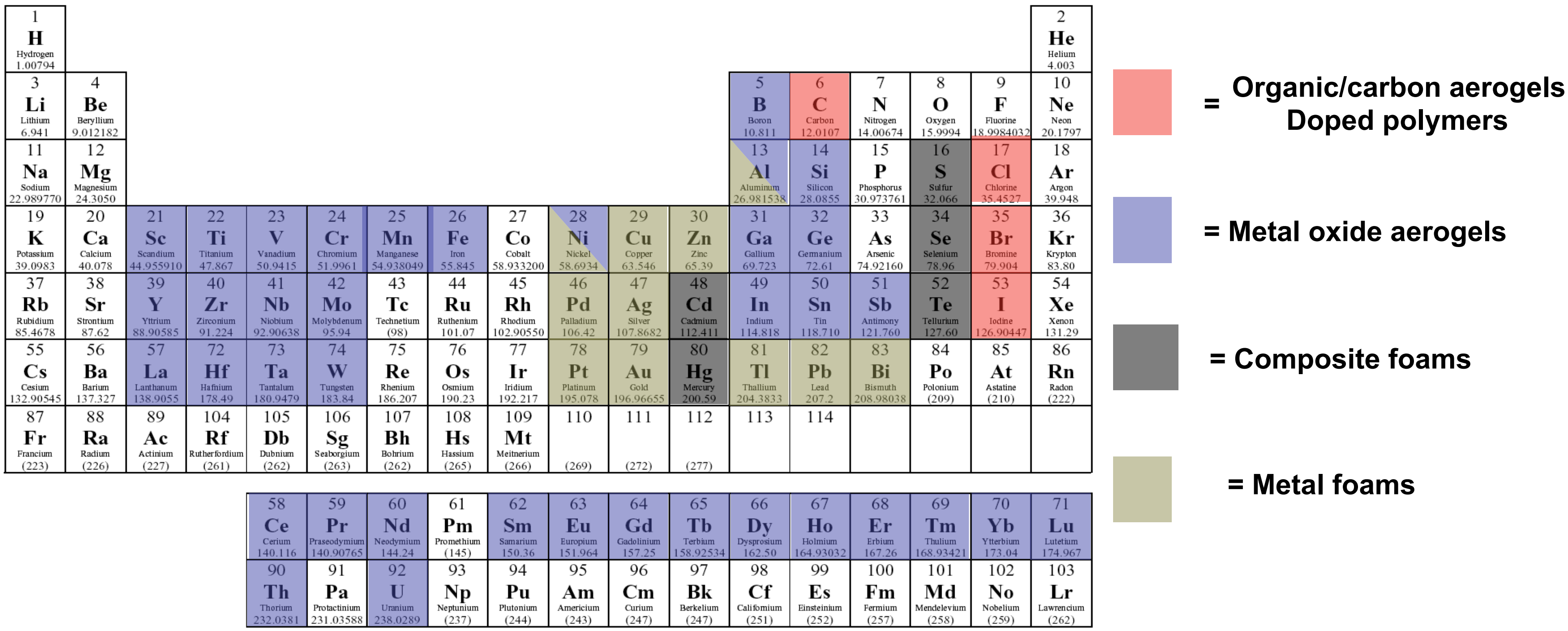
Ghosal S. *et al. Chemistry of Materials* 21, 2009, 1989

ALD will enable area-selective doping of surface modified of aerogels

Summary

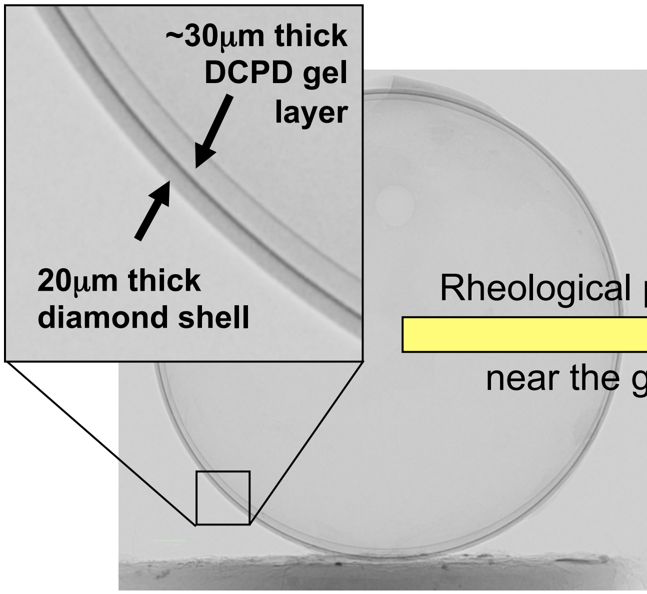
We are developing materials synthesis capabilities for a broad range of porous materials with tailored properties

Flexibility in all synthetic approaches



Gash, A. E. et al. *Chem. Mater.* **15**, 3268 (2003); *J. Non-Cryst. Solids* **350**, 145 (2004); Reibold, R. A. et al. *J. Non-Cryst. Solids* **319**, 241 (2003); *J. Non-Cryst. Solids* **341**, 35 (2004); Baumann et al. *Chem Mater.* 17, 395 (2005); *Adv. Mater.* (2005), Nyce et al. Chem Mat. V19 (3), 344-346, 2007

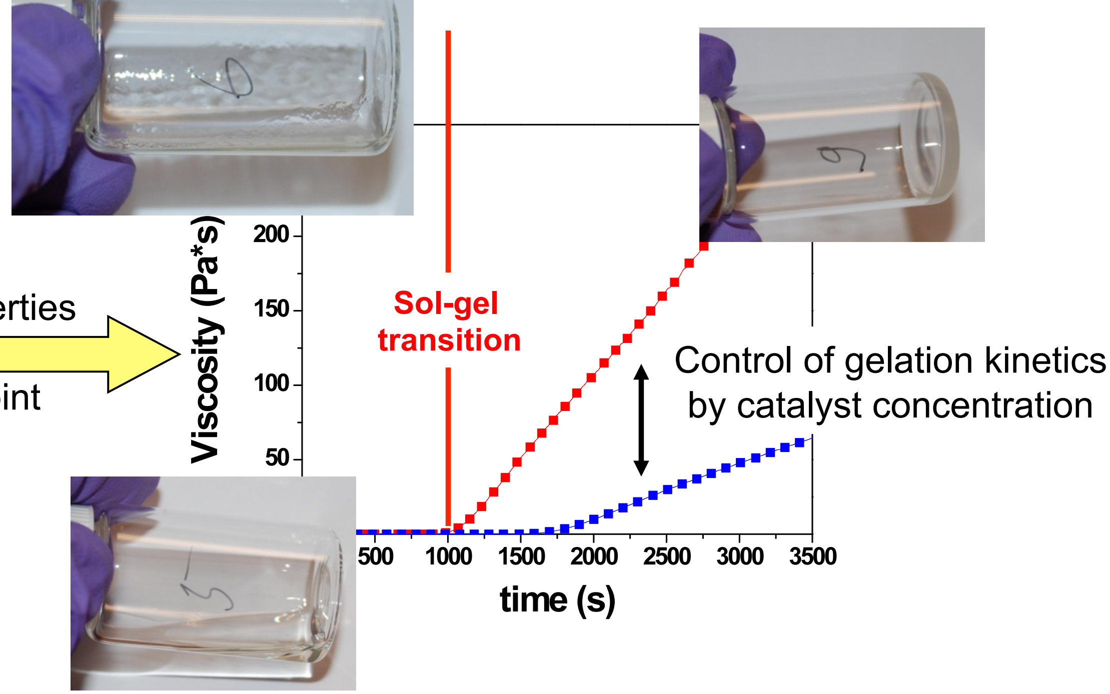
Ultrathin nanoporous foam liner



30-µm-thick 30 mg/cc foam liner

Rheological properties near the gel point

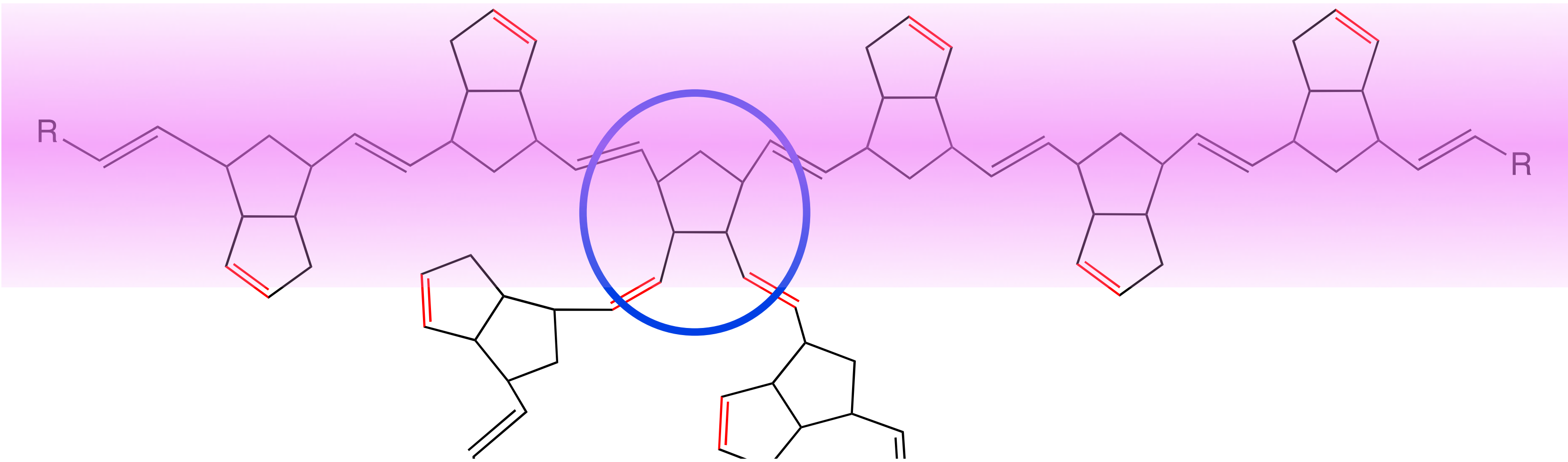
But: non-uniform viscosity increase!

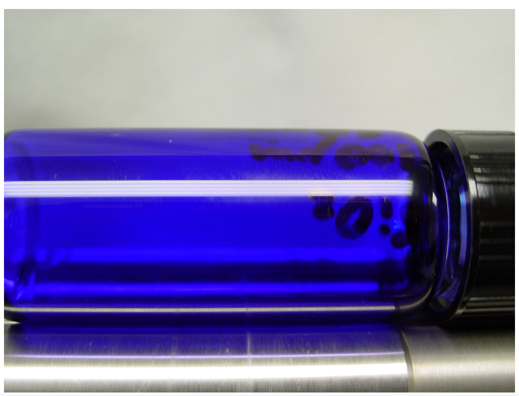


We need to understand and improve the rheological properties near the gel point

Current Development

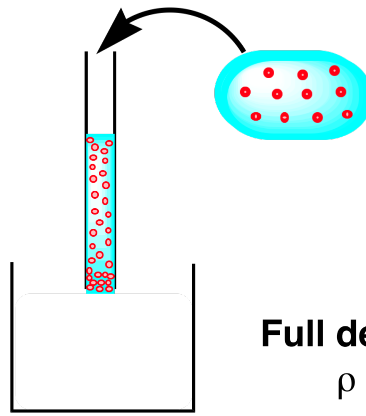
Modifying cross-link density to impact viscosity change



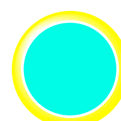


Progression to Produce Low Density Porous Metals

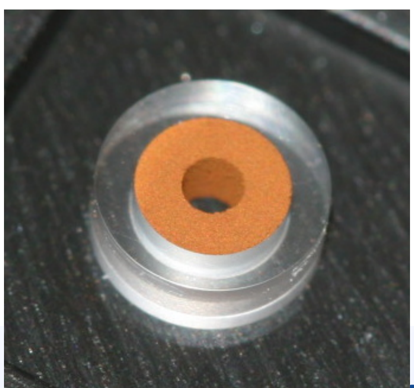
full density gold
19.3 g/cc

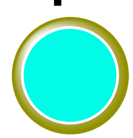


Slip casting
Full density Au nanoparticles
ρ = 5.8 g/cc (30 % rd)



Templating and Slip Casting
Hollow full density gold shells
ρ = 1.7 g/cc (9% rd)



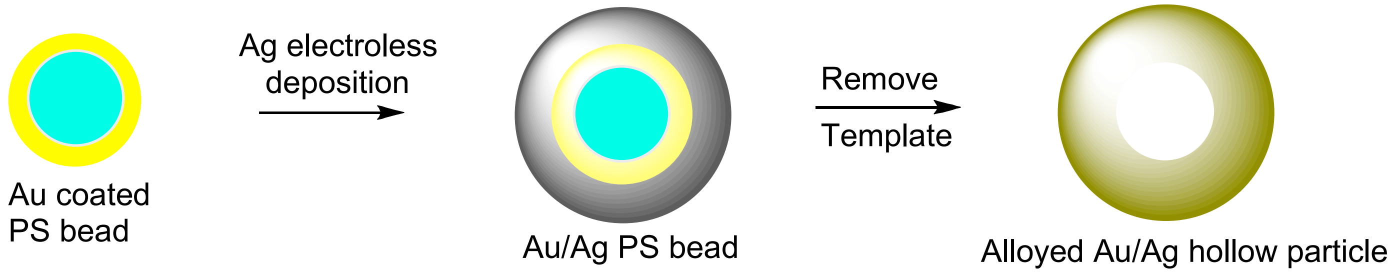


Slip Casting, Templating, and Dealloying
Hollow porous gold shells
ρ = 0.4 g/cc (2% rd)

Plating Silver on a Gold Plated PS Bead Is a Step to Porous Gold Hollow Particles

Synthesis of hollow silver/gold alloy shells

$$Ag(NH_3)_2NO_3 + \text{glucose} \xrightarrow[NH_4OH]{PVP} Ag^o$$



Using this approach, we can readily control the relative proportions of Ag and Au by adjusting plating conditions

We have also observed that subsequent heat treatment to remove the polystyrene template produces an alloyed Au/Ag hollow shell