

Lawrence Livermore National Laboratory

Porous Materials for Laser Target Applications

September 30, 2010

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This work performed under the auspices of the U.S. Department of Energy by
Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344

LLNL-PRES-454079

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

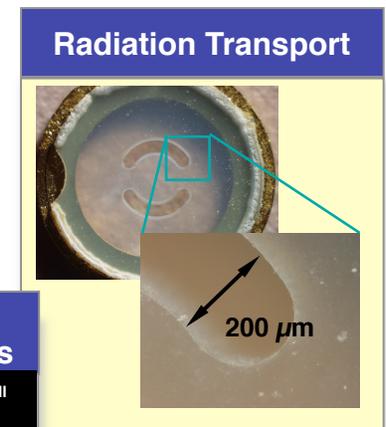
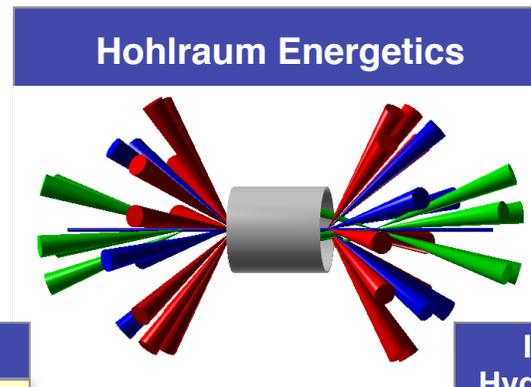
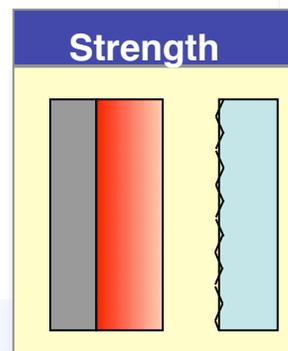
Current Development Example



Motivation

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

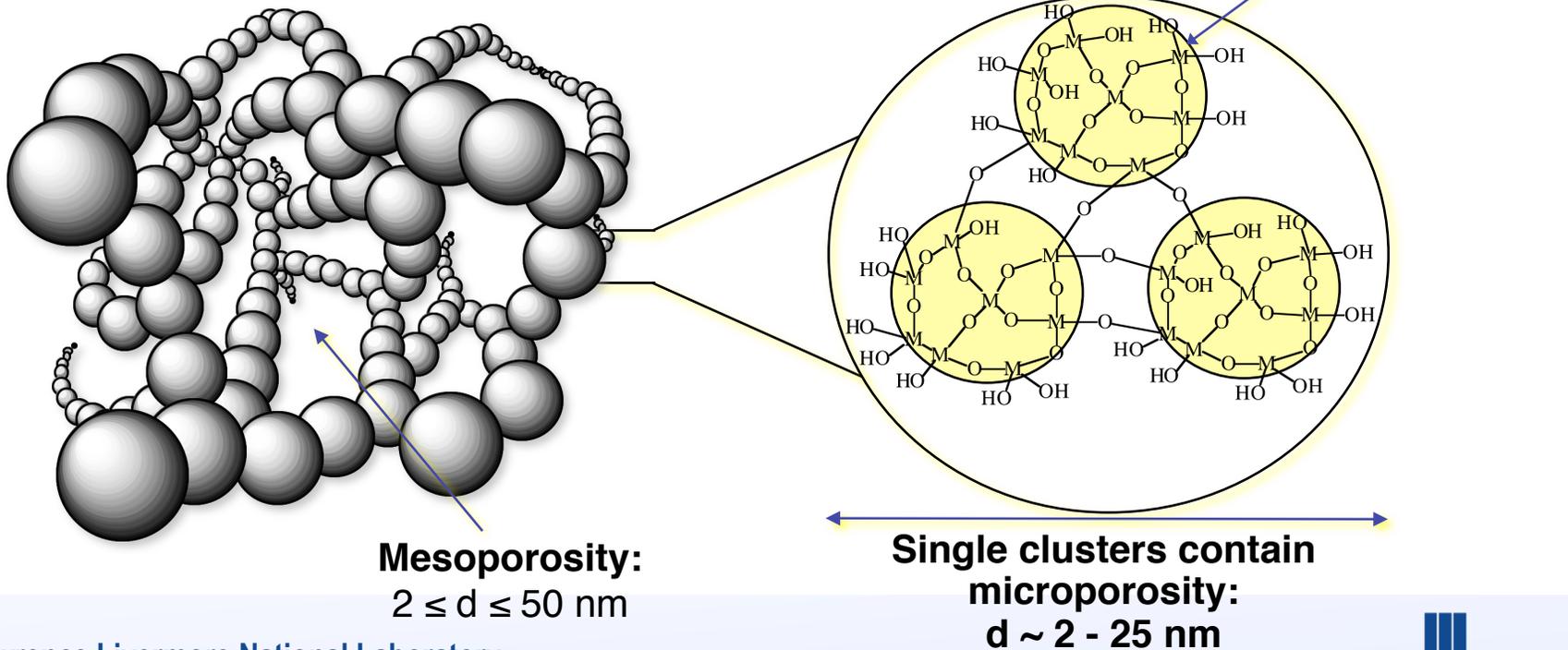
- Many designs require porous materials with challenging attributes:
 - *Machined or cast* high- and low-Z materials (inorganic and organic)
 - Range of densities, density gradients
 - Single component, composites, or doped materials
- To meet these challenges, we endeavor to develop reliable methods for preparation of materials with controlled:
 - Composition
 - Cell sizes
 - Uniformity/homogeneity
 - Dopant levels
 - *Fashioned into a target*



Aerogels

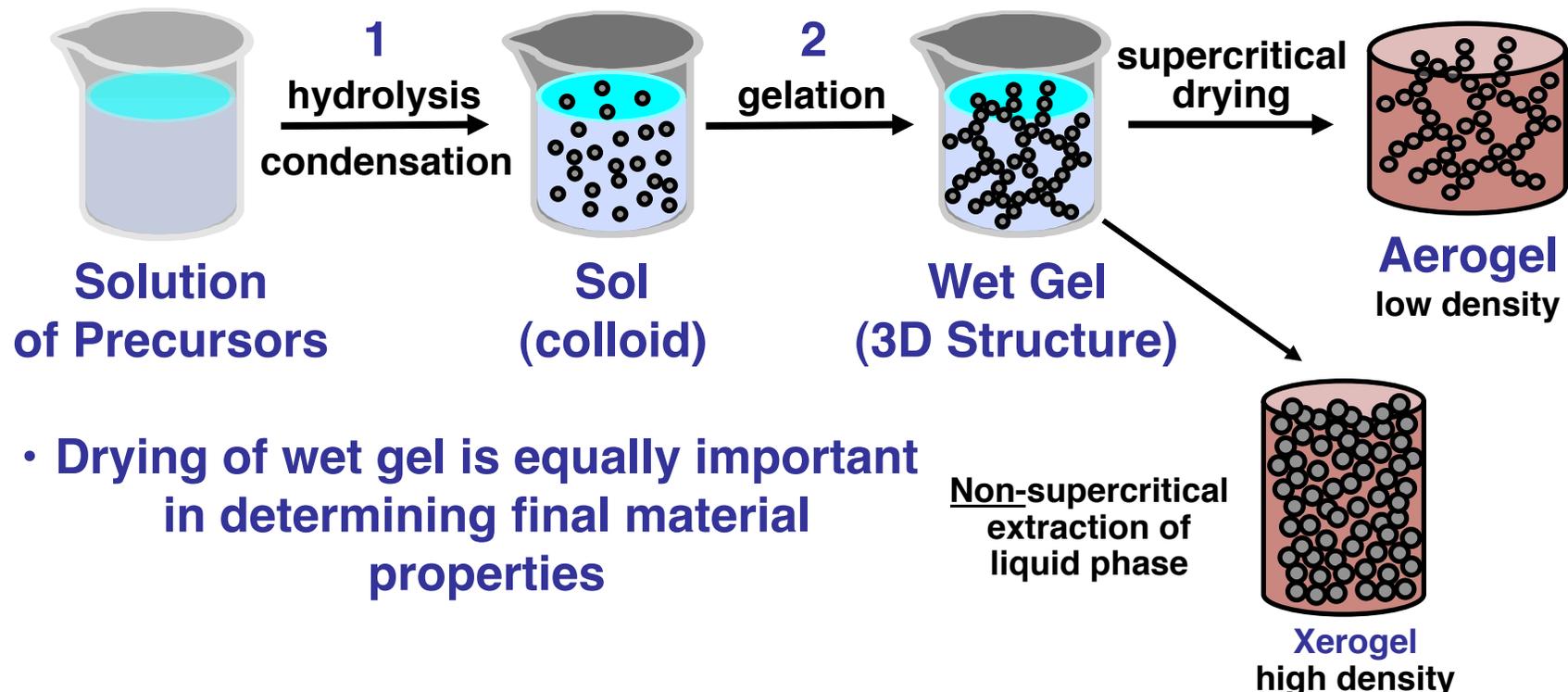
Aerogels possess a number of structural features that are desirable for laser target designs

- 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 m²/g)
 - Low mass densities (1.8 to 0.001 g/cm³)
 - Ultrafine cell/pore sizes (≤ 0.1 micron)



Aerogels are prepared using the sol-gel process

- Sol-gel chemistry involves two basic steps:



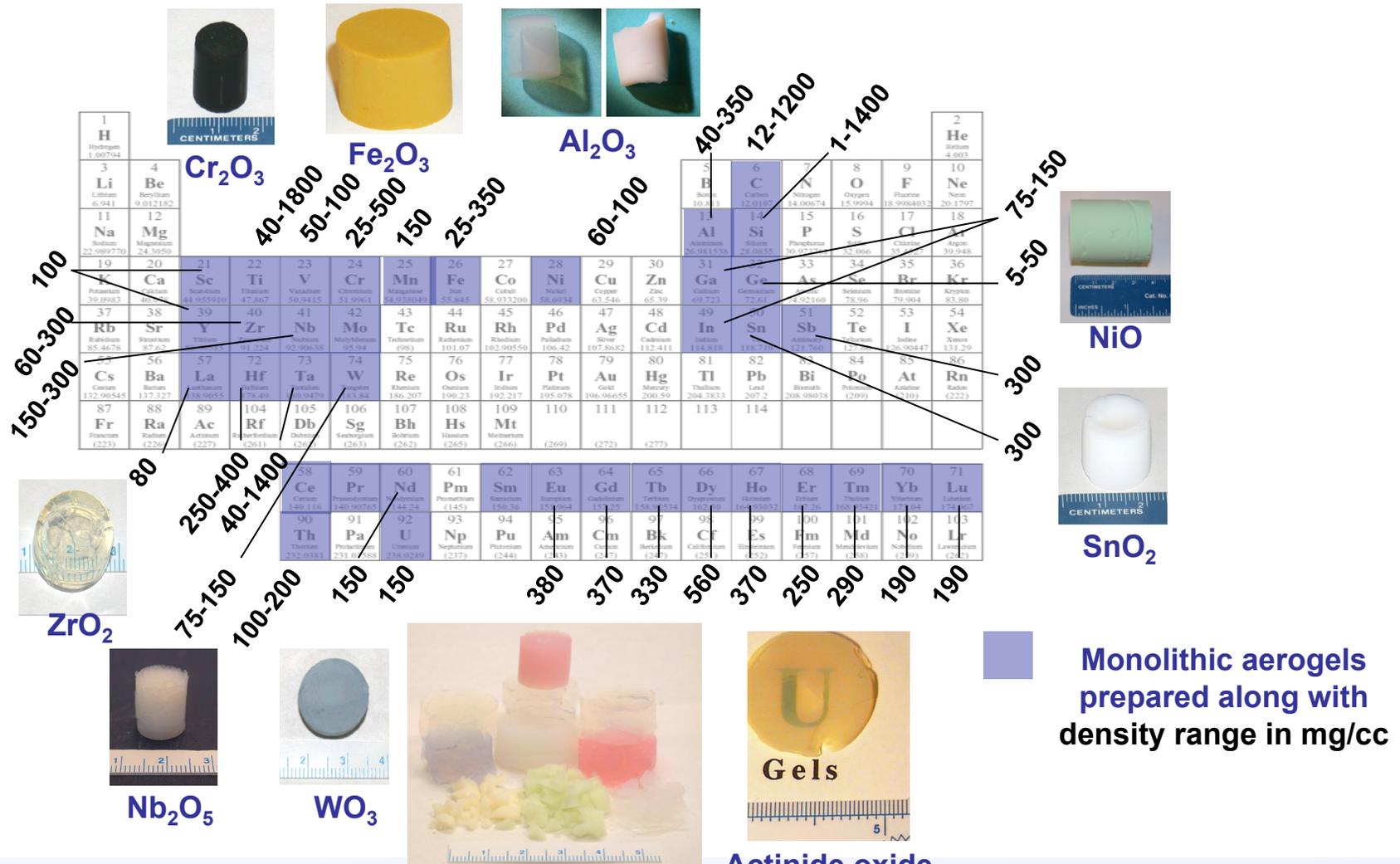
- Drying of wet gel is equally important in determining final material properties

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



Aerogels

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



Lawrence Livermore National Laboratory aerogels

Lanthanide oxide aerogels Actinide oxide aerogels

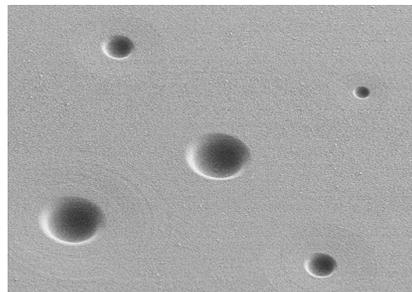


Experimental Platforms

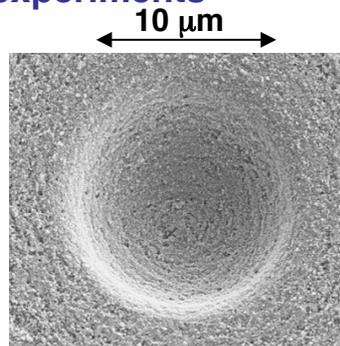
Example 1: In some designs, machineable inorganic aerogel materials are required

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

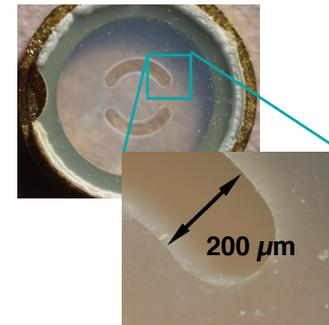
Precision machined TiO₂ aerogels for hydrodynamic experiments



TiO₂ @ 1.8 g/cc

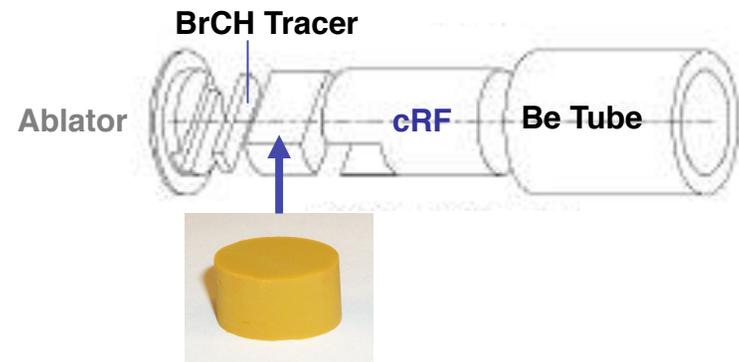


Machined divot
Depth: 5 μm



Precision machined Ta₂O₅ 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).

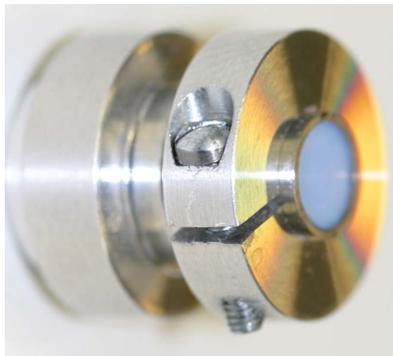
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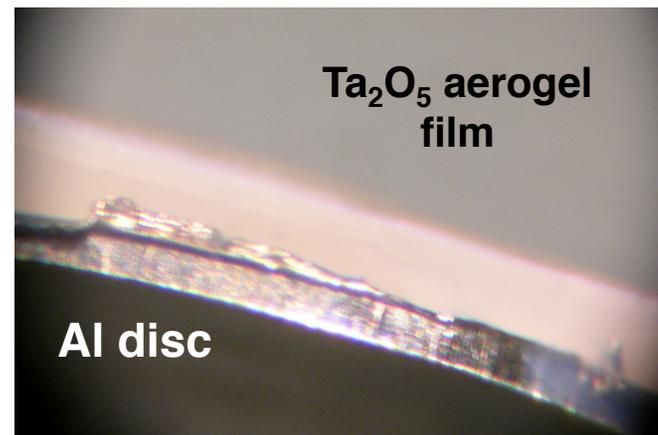
Experimental Platforms

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 Ta₂O₅ aerogel (100 mg/cc) for Hohlräum development



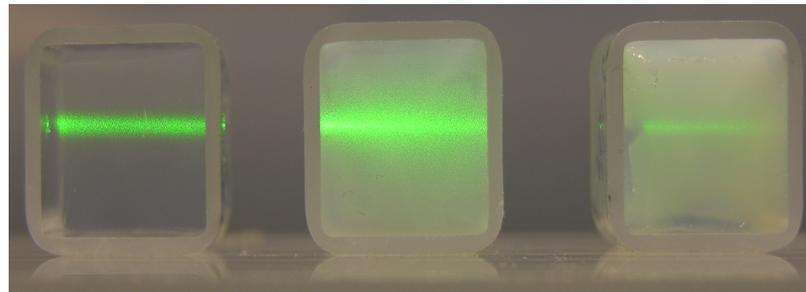
Ta₂O₅ aerogel (100 mg/cc) cast directly on Al disc for EOS targets (surface adhesion)

Experimental Platforms

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 $0.2 n_c \sim < 5.0 \text{ mg/cc}$

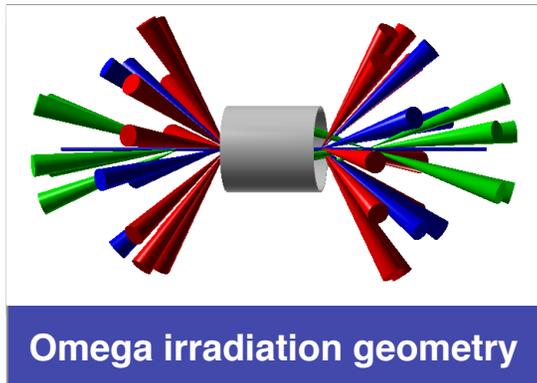
Ti ($h\nu = 4.7\text{keV}$) Zn ($h\nu = 8.9\text{keV}$), Ge ($h\nu = 10.0 \text{ keV}$)



20 at % Ge/SiO₂
4 mg/cc

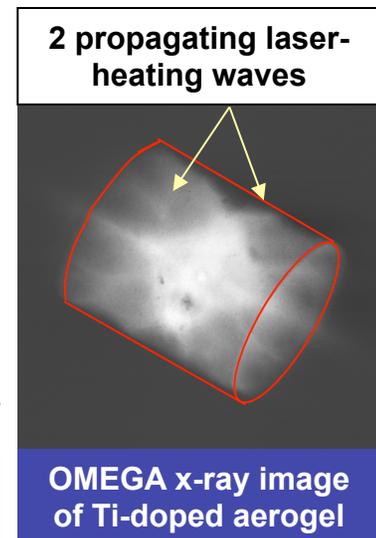
3 at % Ti/SiO₂
3 mg/cc

GeO₂
5 mg/cc



Omega irradiation geometry

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

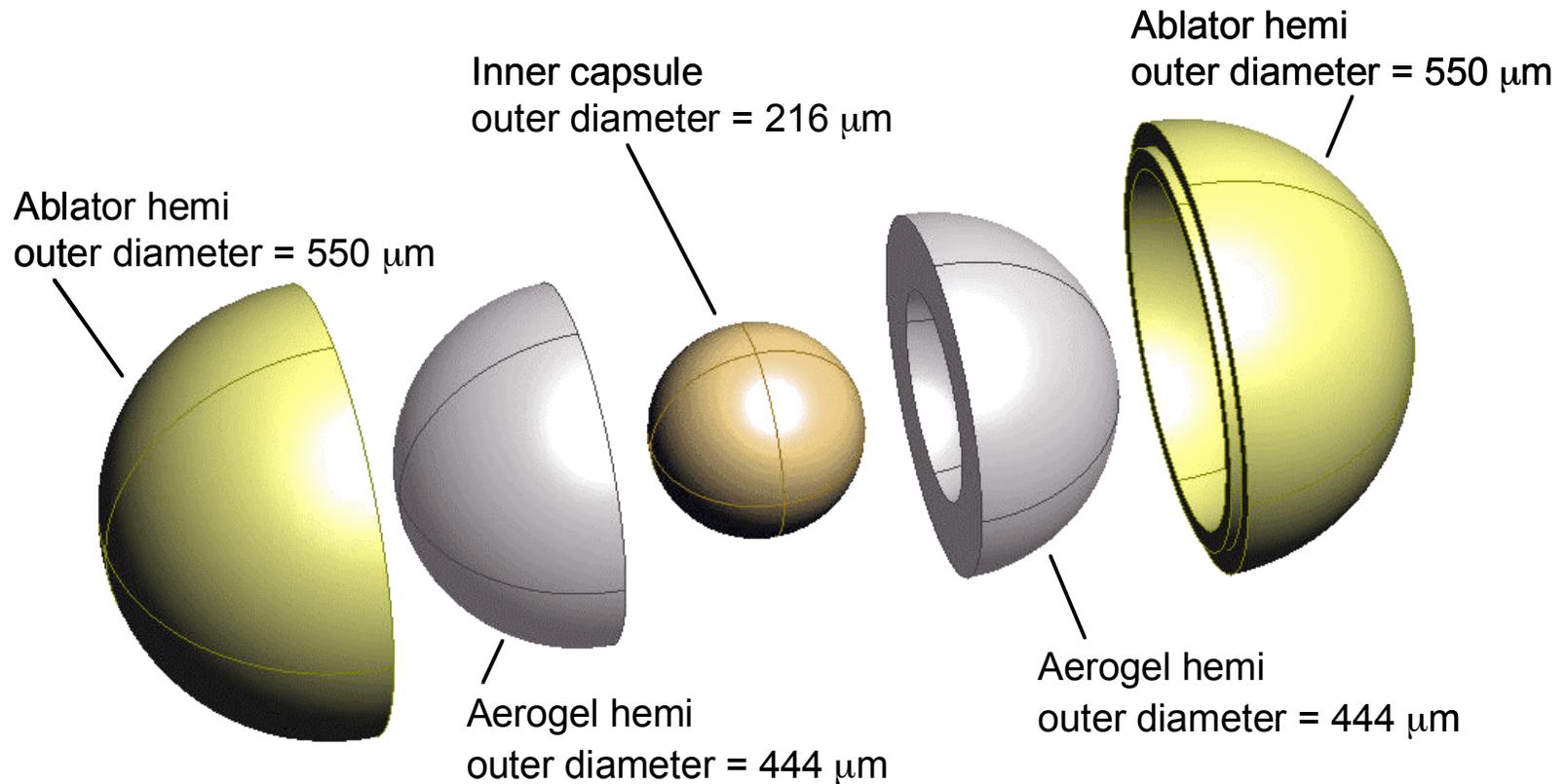


OMEGA x-ray image of Ti-doped aerogel

Experimental Platforms

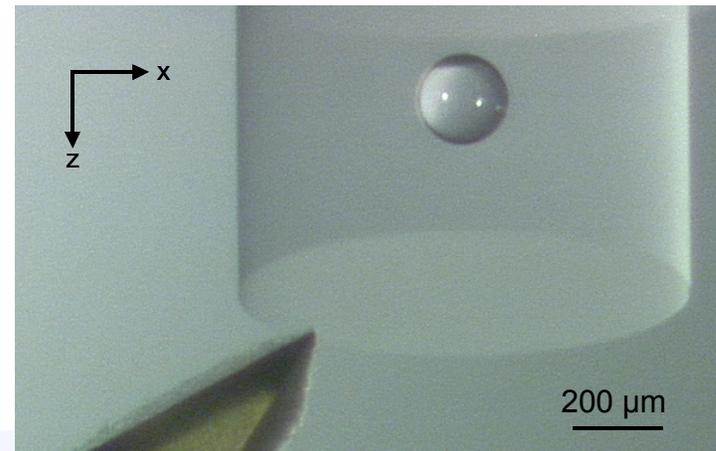
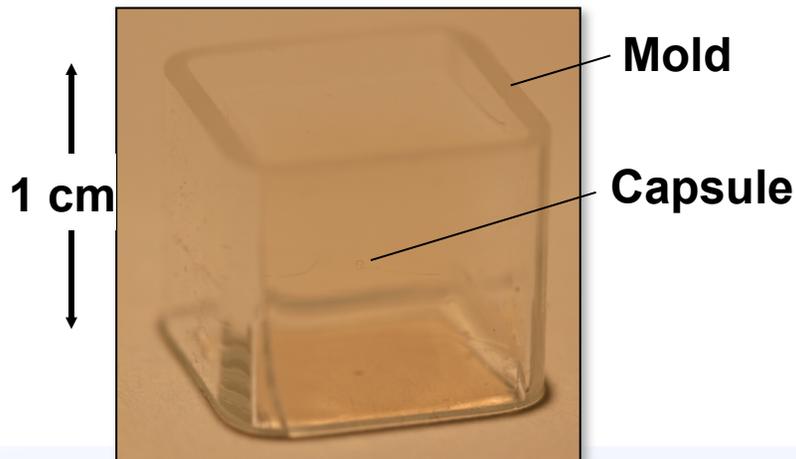
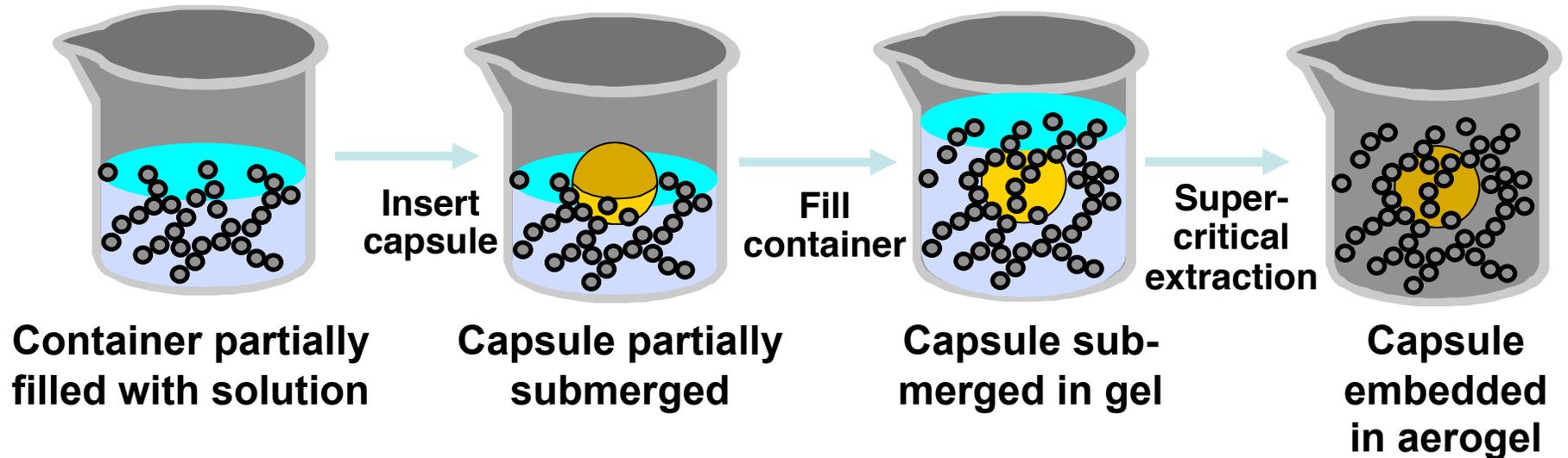
Process Considerations: Double shell targets were originally machined

Carbon vs. silica aerogel design



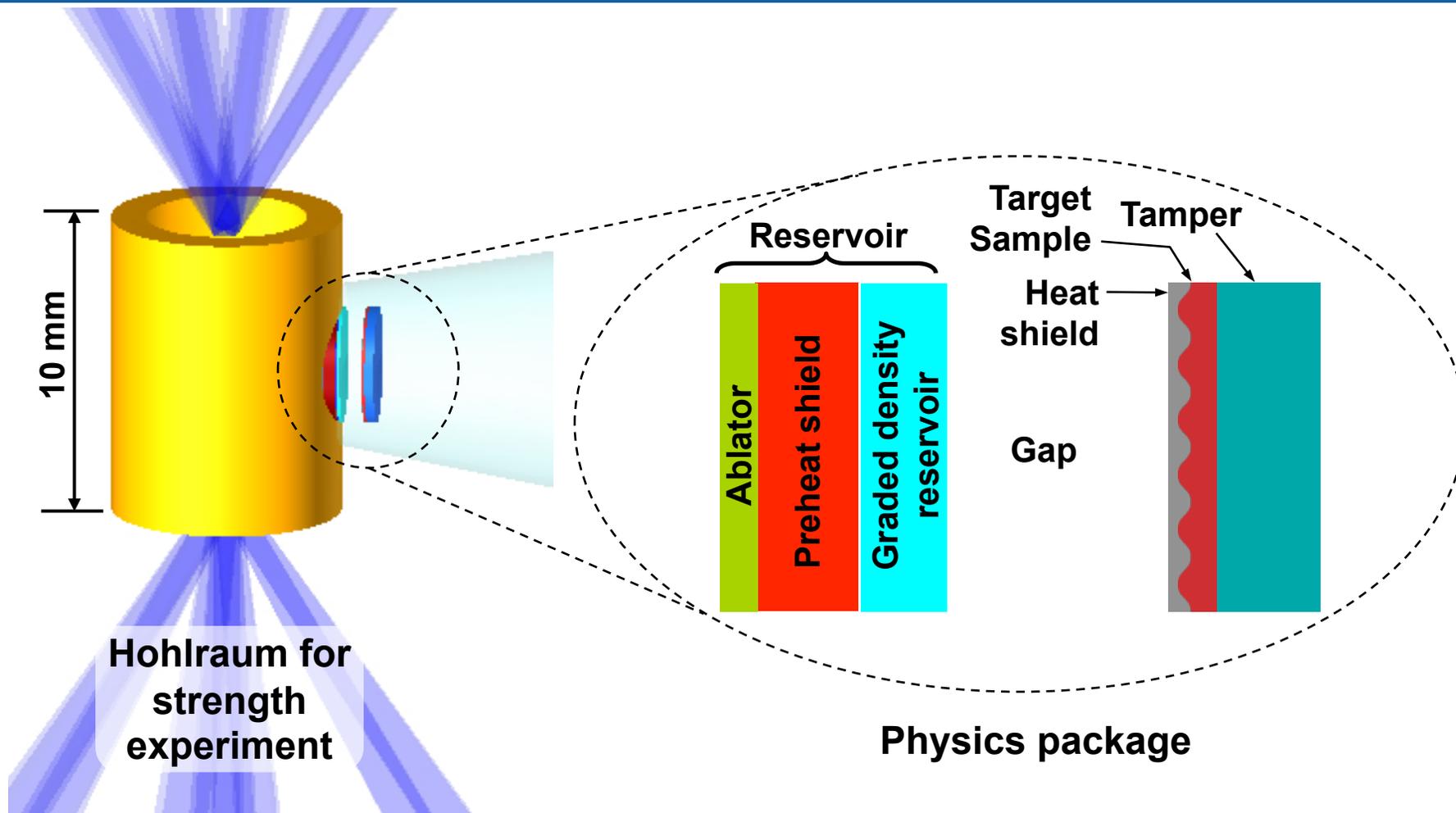
Experimental Platforms

Current approach: glass capsule is cast in a monolithic piece of 50 mg/cc SiO₂ aerogel



Experimental Platforms

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



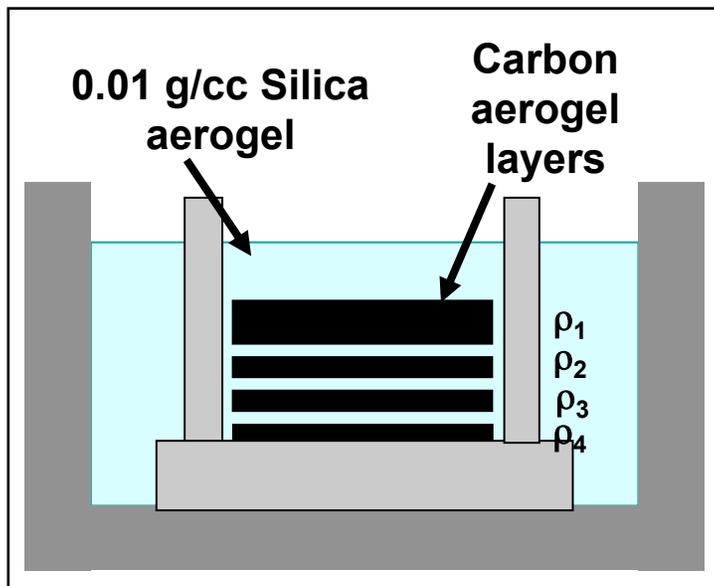
Critical target fabrication challenges include:
— Low Z graded density structures from 1 to 0.001 g/cc



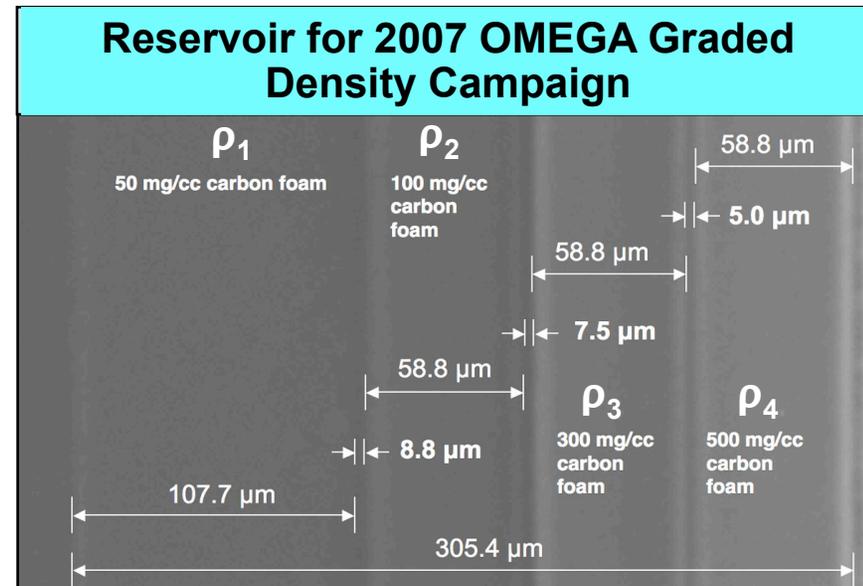
Experimental Platforms

Process 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

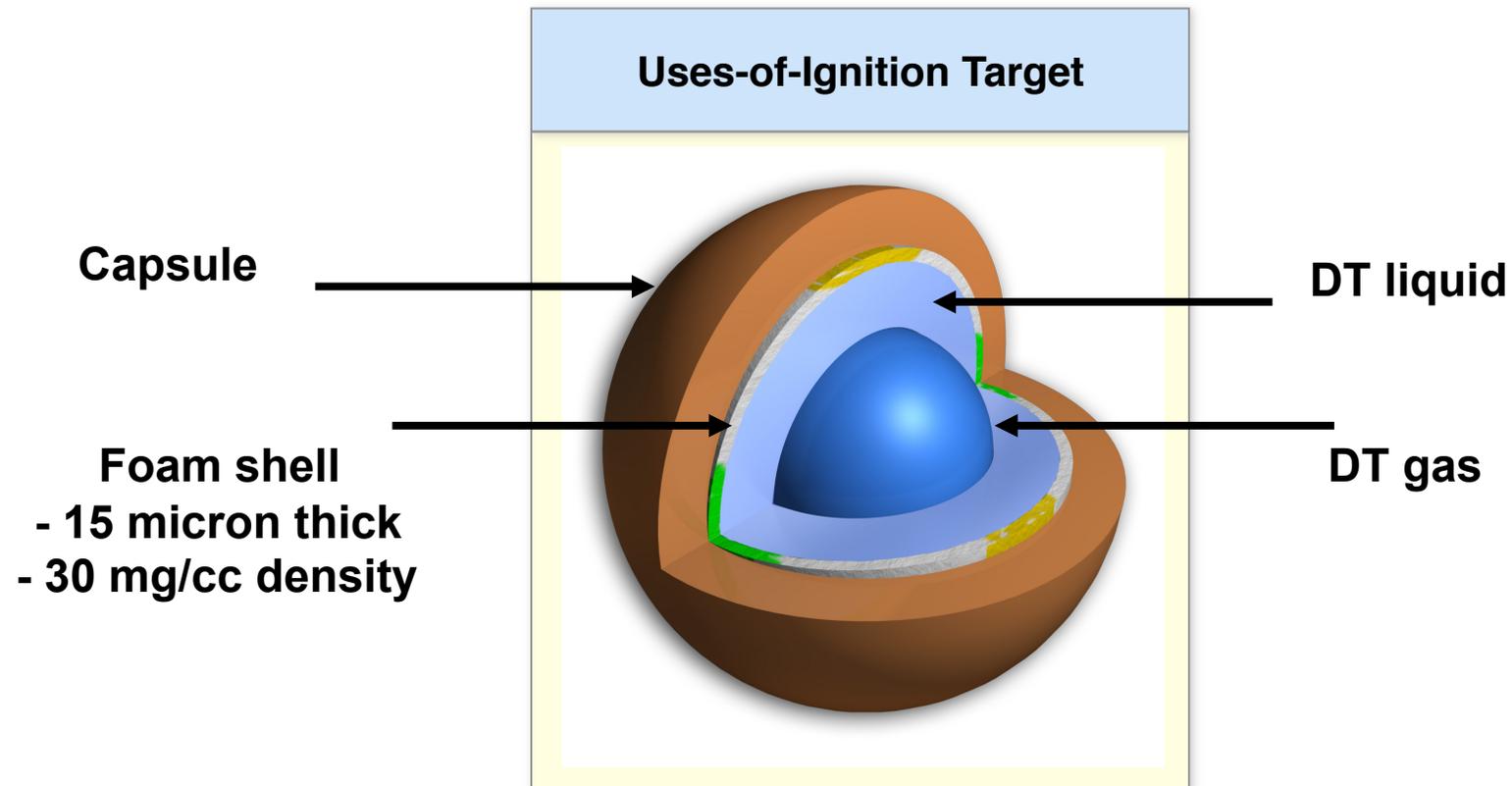


Digital radiograph of step graded density reservoir shows gaps less than 10 μm

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

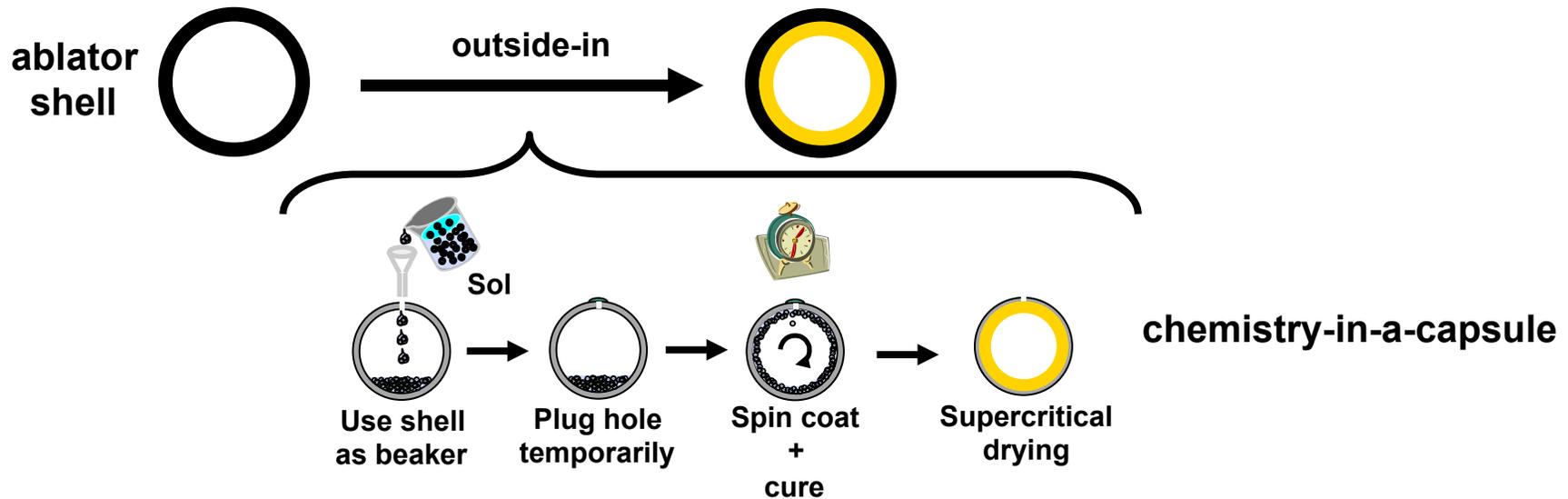


Foam shells inside a capsule



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

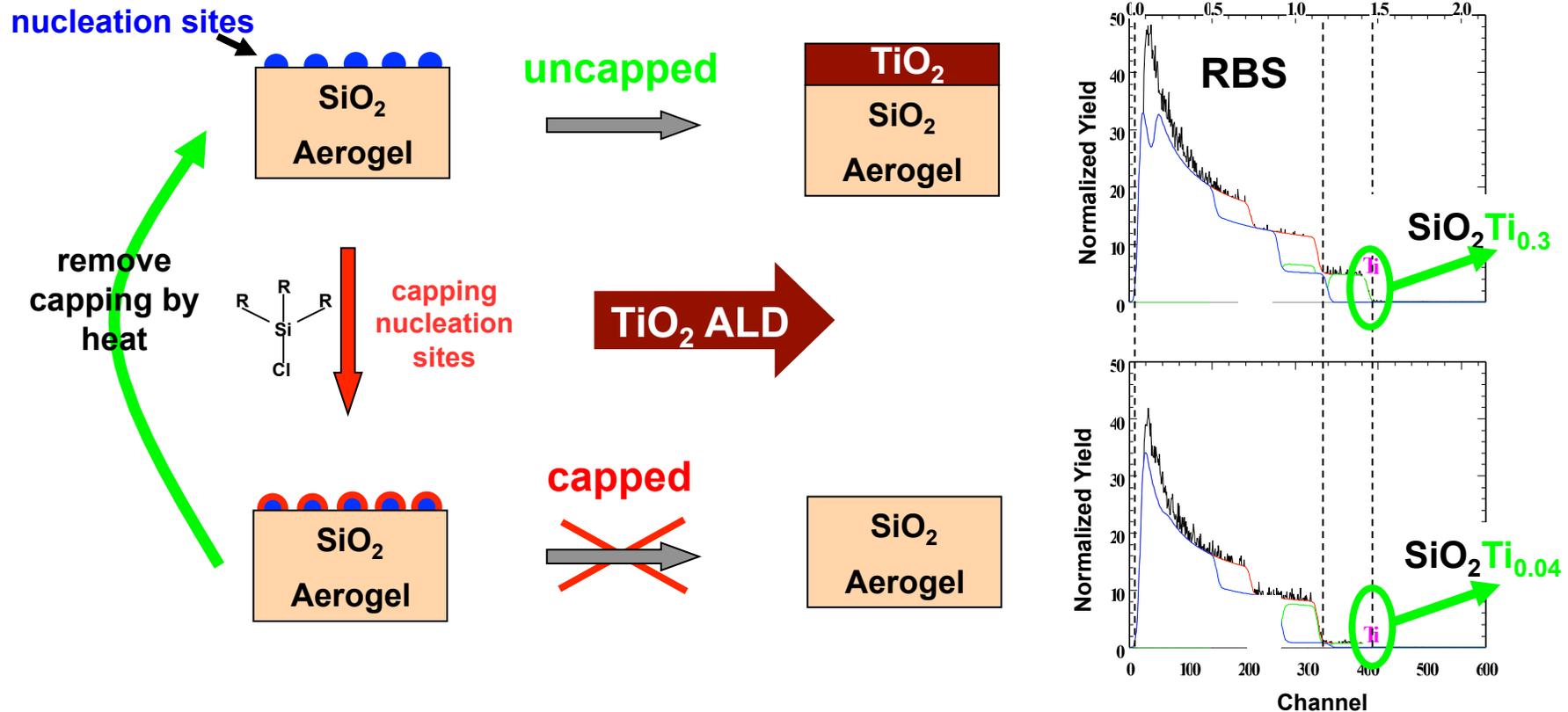
Our current approach: Chemistry-in-a-capsule



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

1 H Hydrogen 1.00794																	2 He Helium 4.003														
3 Li Lithium 6.941	4 Be Beryllium 9.012182																	5 B Boron 10.811	6 C Carbon 12.0107	7 N Nitrogen 14.00674	8 O Oxygen 15.9994	9 F Fluorine 18.9984032	10 Ne Neon 20.1797								
11 Na Sodium 22.989770	12 Mg Magnesium 24.3050																	13 Al Aluminum 26.981538	14 Si Silicon 28.0855	15 P Phosphorus 30.973761	16 S Sulfur 32.066	17 Cl Chlorine 35.4527	18 Ar Argon 39.948								
19 K Potassium 39.0983	20 Ca Calcium 40.078	21 Sc Scandium 44.955910	22 Ti Titanium 47.867	23 V Vanadium 50.9415	24 Cr Chromium 51.9961	25 Mn Manganese 54.938049	26 Fe Iron 55.845	27 Co Cobalt 58.933200	28 Ni Nickel 58.6934	29 Cu Copper 63.546	30 Zn Zinc 65.39	31 Ga Gallium 69.723	32 Ge Germanium 72.61	33 As Arsenic 74.92160	34 Se Selenium 78.96	35 Br Bromine 79.904	36 Kr Krypton 83.80														
37 Rb Rubidium 85.4678	38 Sr Strontium 87.62	39 Y Yttrium 88.90585	40 Zr Zirconium 91.224	41 Nb Niobium 92.90638	42 Mo Molybdenum 95.94	43 Tc Technetium (98)	44 Ru Ruthenium 101.07	45 Rh Rhodium 102.90550	46 Pd Palladium 106.42	47 Ag Silver 107.8682	48 Cd Cadmium 112.411	49 In Indium 114.818	50 Sn Tin 118.710	51 Sb Antimony 121.760	52 Te Tellurium 127.60	53 I Iodine 126.90447	54 Xe Xenon 131.29														
55 Cs Cesium 132.90545	56 Ba Barium 137.327	57 La Lanthanum 138.9055	72 Hf Hafnium 178.49	73 Ta Tantalum 180.9479	74 W Tungsten 183.84	75 Re Rhenium 186.207	76 Os Osmium 190.23	77 Ir Iridium 192.217	78 Pt Platinum 195.078	79 Au Gold 196.96655	80 Hg Mercury 200.59	81 Tl Thallium 204.3833	82 Pb Lead 207.2	83 Bi Bismuth 208.98038	84 Po Polonium (209)	85 At Astatine (210)	86 Rn Radon (222)														
87 Fr Francium (223)	88 Ra Radium (226)	89 Ac Actinium (227)	104 Rf Rutherfordium (261)	105 Db Dubnium (262)	106 Sg Seaborgium (263)	107 Bh Bohrium (262)	108 Hs Hassium (265)	109 Mt Meitnerium (266)	110 (269)	111 (272)	112 (277)	113	114																		
																		58 Ce Cerium 140.116	59 Pr Praseodymium 140.90765	60 Nd Neodymium 144.24	61 Pm Promethium (145)	62 Sm Samarium 150.36	63 Eu Europium 151.964	64 Gd Gadolinium 157.25	65 Tb Terbium 158.92534	66 Dy Dysprosium 162.50	67 Ho Holmium 164.93032	68 Er Erbium 167.26	69 Tm Thulium 168.93421	70 Yb Ytterbium 173.04	71 Lu Lutetium 174.967
																		90 Th Thorium 232.0381	91 Pa Protactinium 231.03588	92 U Uranium 238.0289	93 Np Neptunium (237)	94 Pu Plutonium (244)	95 Am Americium (243)	96 Cm Curium (247)	97 Bk Berkelium (247)	98 Cf Californium (251)	99 Es Einsteinium (252)	100 Fm Fermium (257)	101 Md Mendelevium (258)	102 No Nobelium (259)	103 Lr Lawrencium (262)

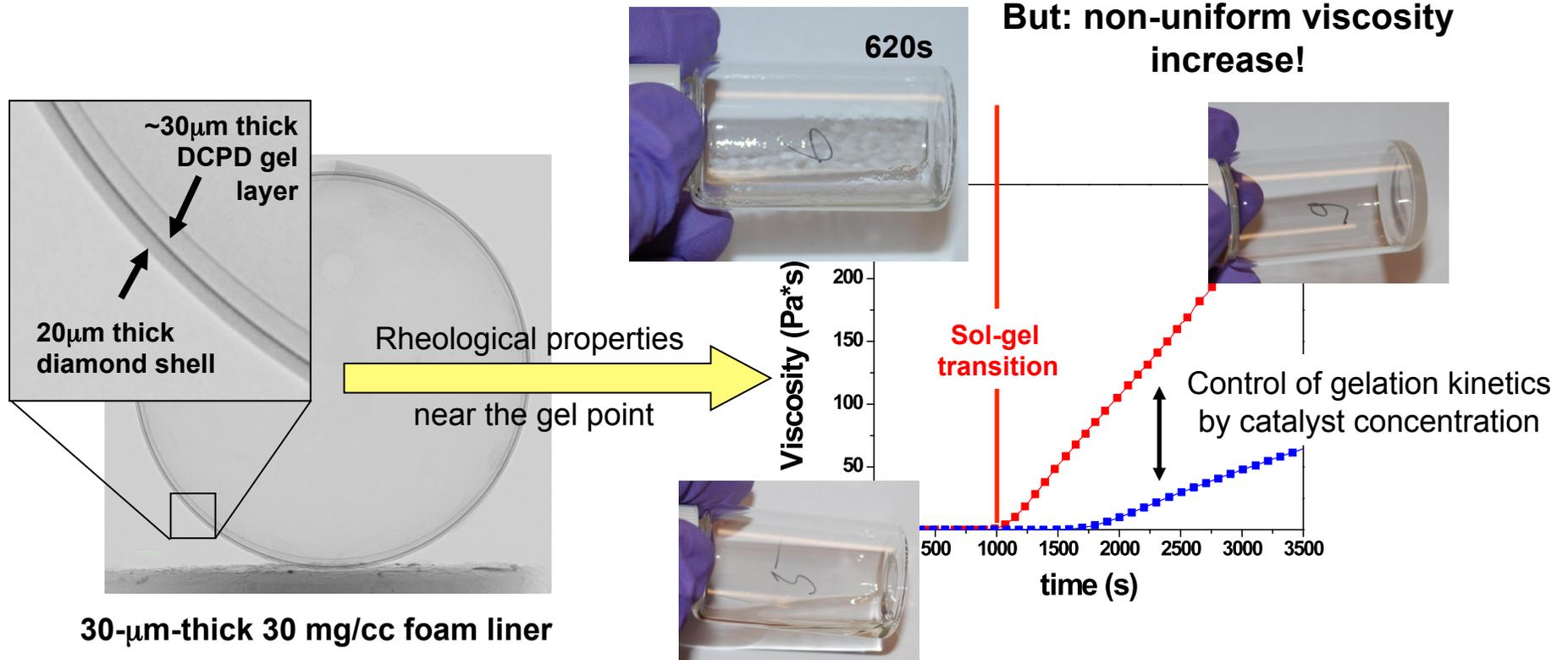
- = Organic/carbon aerogels
Doped polymers
- = Metal oxide aerogels
- = Composite foams
- = Metal foams

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





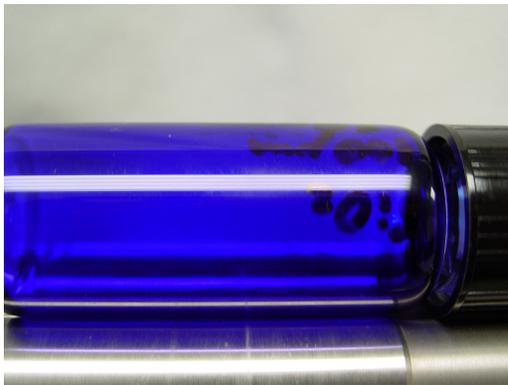
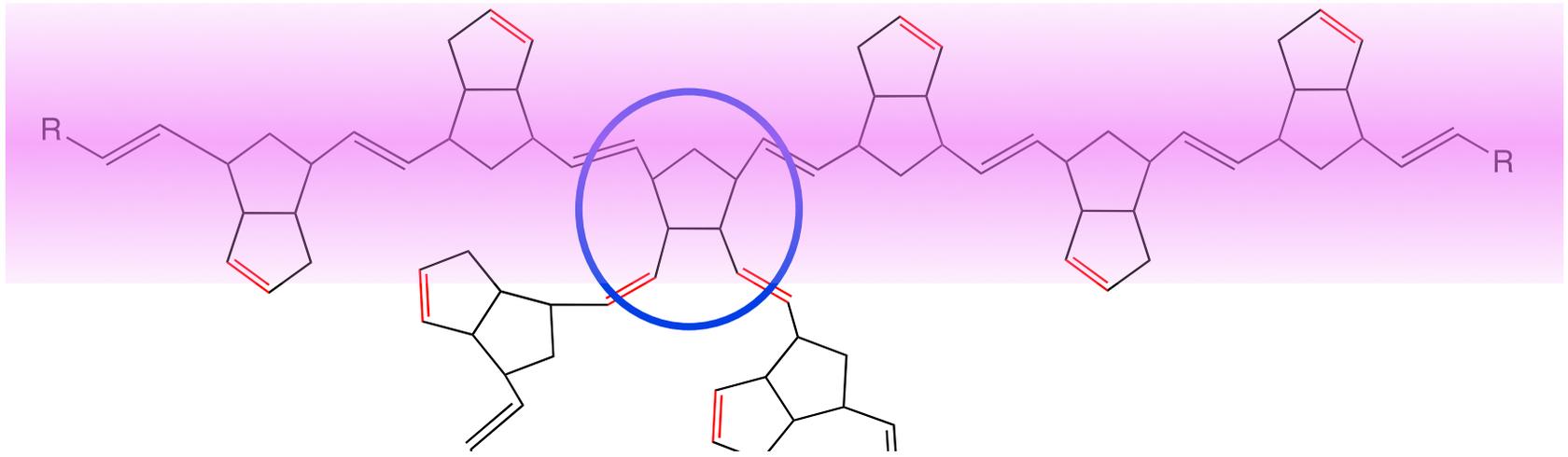
Ultrathin nanoporous foam liner



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

Current Development

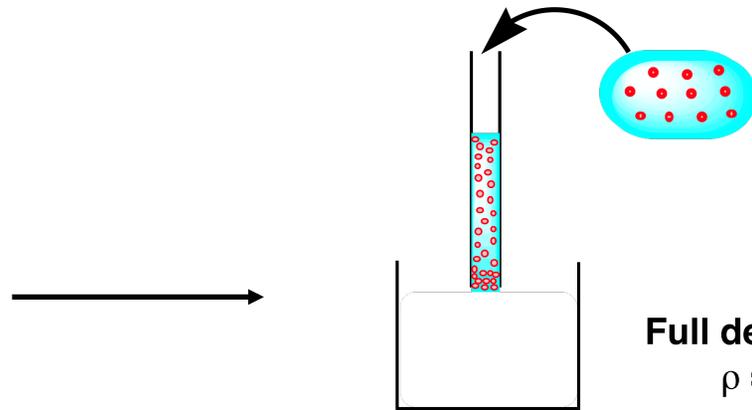
Modifying cross-link density to impact viscosity change



Progression to Produce Low Density Porous Metals



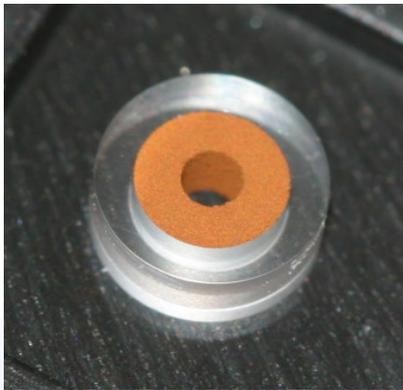
full density gold
 $\rho = 19.3 \text{ g/cc}$



Slip casting
Full density Au nanoparticles
 $\rho = 5.8 \text{ g/cc}$ (30 % rd)



Templating and Slip Casting
Hollow full density gold shells
 $\rho = 1.7 \text{ g/cc}$ (9% rd)

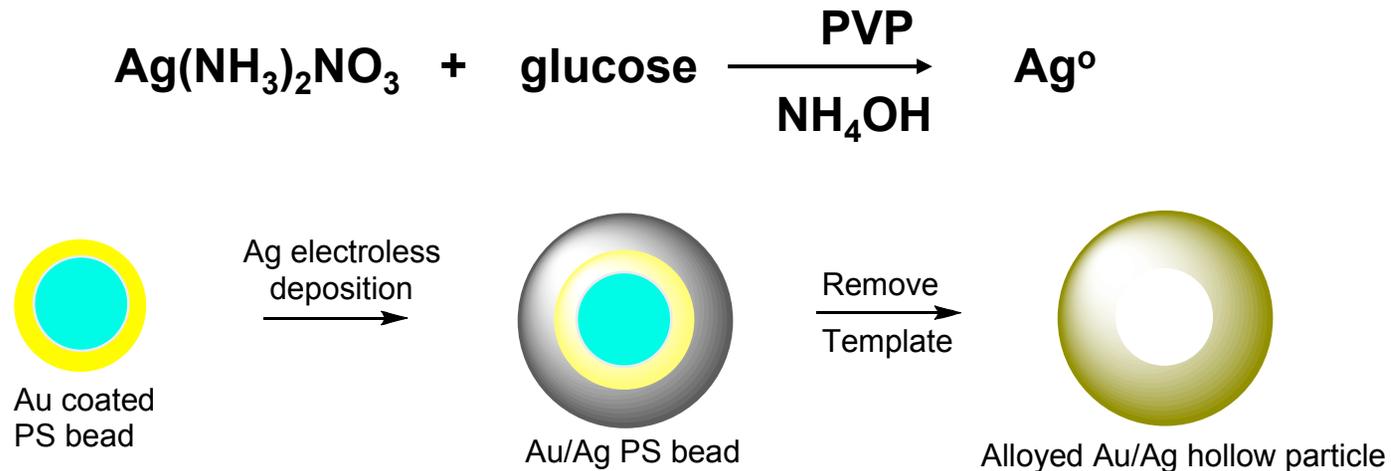


Slip Casting, Templating, and Dealloying
Hollow porous gold shells
 $\rho = 0.4 \text{ g/cc}$ (2% rd)



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

Synthesis of hollow silver/gold alloy shells



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