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To: PSVRF Board

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## **Final Project Report: “Acoustic Absorption and Impedance of Composite Aerogels”**

### **Abstract:**

Composite aerogels (with varying concentrations of silica and polydimethylsiloxane) were developed and their acoustic absorption and impedance as a function of composition have been measured. The polydimethylsiloxane modified the ceramic structure of the silica aerogels, decreasing the material’s modulus of elasticity and bulk modulus while still maintaining the high porosity of the aerogel structure. The composite aerogels were found to have excellent absorption characteristics in the 1000 Hz frequency range, approximately 40% better than that of commercial fiberglass.

### **Introduction and Background:**

When sound energy encounters an absorbent material, the energy can be reflected, transmitted, or absorbed through viscous dissipation. Materials that have high acoustic absorption are generally characterized by high porosity, having many channels for the dissipation to take place. Silica aerogels are extremely light-weight ( $0.1 - 0.01 \text{ g/cm}^3$ ) and porous (> 99% porous) materials. They have been shown to be excellent thermal insulators and their porosity indicates that they have the potential to be good acoustical insulators as well. However, previous reports on the acoustical properties of monolithic silica aerogels have indicated that although the material’s high porosity, pore tortuosity, and surface area contribute to a low sound velocity in the material, the rigid matrix of the aerogel exhibit a high reflection coefficient. To improve the acoustic performance (reduce the rigidity of the matrix), the silica aerogels are combined with polydimethylsiloxane (PDMS), creating an organically modified silica (Ormosil) aerogel – a material that is both ceramic and polymer.

Silica aerogels are created using the sol-gel process, where the hydrolysis and condensation of liquid precursors produce a gel-like structure, a porous amorphous solid network surrounded by the liquid byproducts of the reaction. Typically, the removal of the liquid byproducts will result in the collapse of the porous network, resulting in a dense material, very similar to glass. However, when the liquid byproduct is removed through the supercritical process, the porous structure of the solid can be preserved.

One great advantage of the sol-gel process is that it is a room-temperature process, compared to the traditional process of synthesizing glass which requires significant heating  $\sim 2000^\circ\text{F}$ . The low-temperature enables the addition of polymers to

the silica structure. (Figure 1) This results in a porous structure allows for Rayleigh scattering of the phonons, while a lowered bulk modulus allows the sound waves to enter the material by lowering the reflection coefficient. The correlation between PDMS concentration and elasticity of a silica/PDMS aerogel can be seen in Table 1.

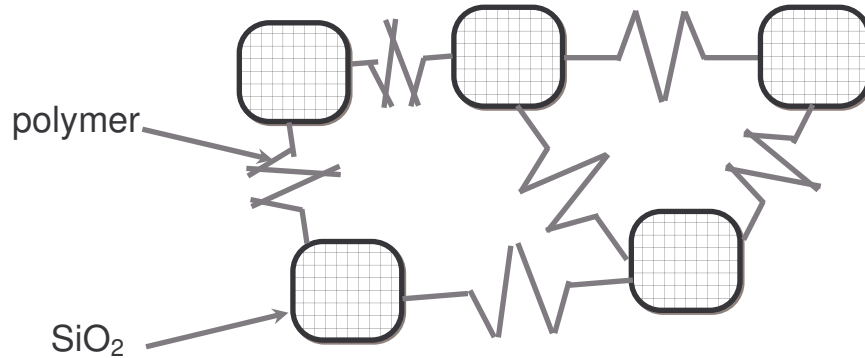


Figure 1. A schematic of the silica/PDMS composite on the molecular scale, producing a porous and elastic structure.

Table 1. The material rigidity (Young's Modulus), hardness, and brittleness all decrease with increasing the concentration of the PDMS. (Mackenzie, Huang *et al.* 1996)

Mol % Polymer (PDMS)	0	7.9	12.8	19.5	23.7
Young's Modulus – material rigidity (GPa)	20.7	18.6	16.0	15.0	13.0
Hardness (kg/mm <sup>2</sup> )	186	160	140	110	88
Fracture toughness (Mpa/m <sup>1/2</sup> )	0.50	0.49	0.48	0.47	0.46
Brittleness (mm <sup>-1/2</sup> )	3.63	3.19	2.86	2.32	1.88

### Experimental:

Tetraethoxysilane (TEOS), polydimethylsiloxane (PDMS), tetrahydrofuran (THF), and half of the total amount of isopropanol (ISO) is weighed and combined. The mixture is slowly agitated to form a homogeneous reactant solution. The remaining amount of isopropanol, hydrochloric acid, and distilled water are then mixed together and combined with the reactant solution. The amounts of the precursors were varied to determine the best recipe for synthesizing a homogeneous, elastic aerogel. A schematic of the sol-gel process is represented in Figure 2.

Once a homogeneous sol was obtained, several methods were used to facilitate and speed up the gelation process. The first strategy was to add heat using a hot water bath (40°C) while stirring. At low PDMS concentrations (below 10 mol %) and high HCl concentrations (above 30 wt % of total solution), gelation occurred within 24 hours in the water bath, with no stirring. At higher PDMS concentrations (above 30 mol %), sonication is needed for gelation. In general, gelation time was inversely proportional to

HCl concentration and directly proportional to PDMS concentration. Figure 3 is a diagram showing the relationship between PDMS concentration and the method required to form a homogeneous gel.

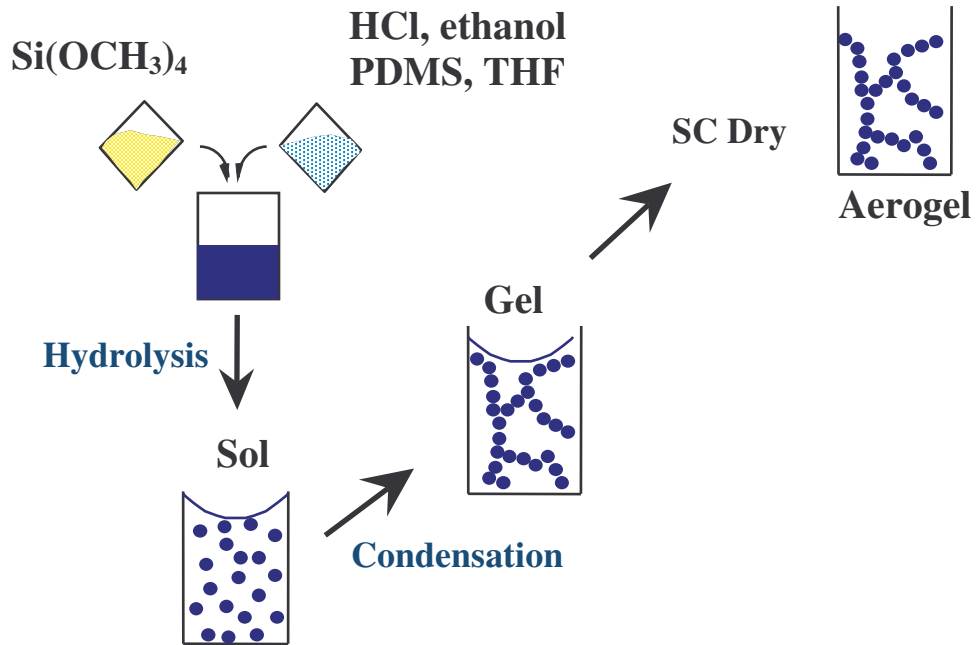


Figure 2. Schematic of the sol-gel process. The precursors are combined, and after hydrolysis (aided by stirring), a sol is formed. The condensation process (aided by heat, stirring, and/or sonication) produces a gel which, after supercritical drying, becomes an aerogel.

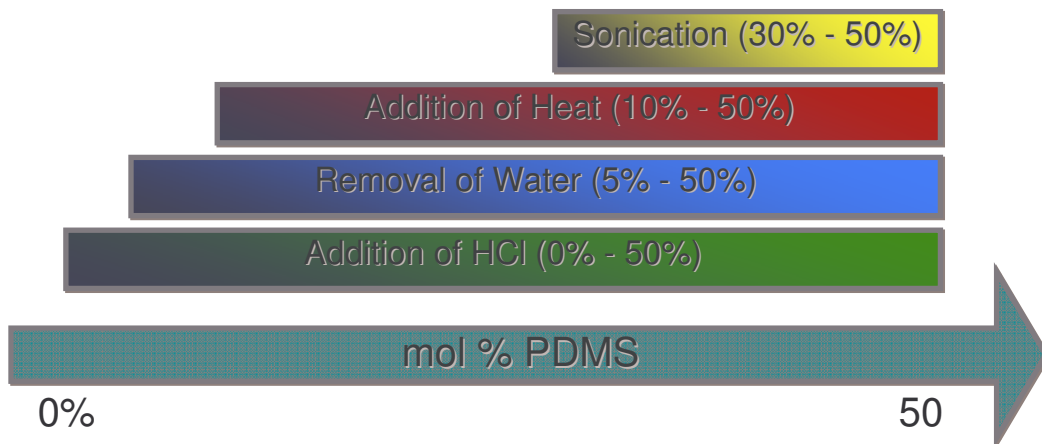


Figure 3. This diagram shows the steps needed in order to increase the concentration of PDMS in the composite aerogel. The more PDMS, the more energy is needed during the gelation process (through heating, sonication, etc.)

Once the gel has formed, it allowed to age for at least three days. This allows the PDMS/silica network to more fully form. A solvent exchange process is conducted with acetone and the gel is dried under supercritical conditions. Carbon dioxide is the supercritical fluid used.

The acoustic absorption and impedance data were gathered in a horizontally mounted impedance tube. Aerogel particles of sizes between 1 mm – 2 cm in diameter were tightly packed (but not compressed) into a cardboard sample holder. The cylindrical sample holder was 10 cm in length and fit snugly into the impedance tube. Testing were performed according to ASTM, C 384-04 procedures.



Figure 5. Impedance tube.

Surface area, porosity, and pore size distribution data were gathered with nitrogen adsorption. Scanning electron microscopy was also used to determine structure at the sub-micrometer range.

## Results and Discussion:

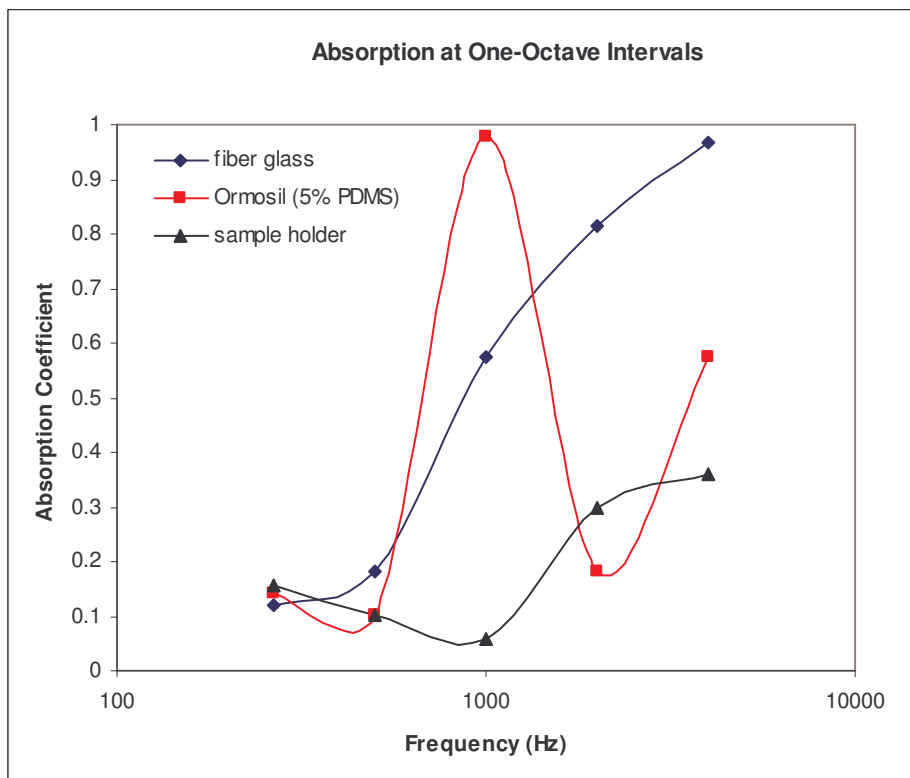


Figure 6. Acoustic absorption of the composite aerogel (with 5 mol% PDMS) compared to that of fiberglass. The absorption of an empty sample holder is also shown for comparison.

The acoustic absorption data show that the composite aerogel has a significantly different frequency response compared to commercial fiberglass. Unlike the fiberglass which exhibited higher absorption at higher frequencies, the composite aerogel has a absorption peak (close to 95%) centered around 1000 Hz. (Figure 6) The addition of PDMS also changed the absorption characteristic of the aerogel, shifting the absorption peak from 800 Hz to 1000 Hz. Although the amount of PDMS did not have as large of an effect as just the presence of PDMS in the aerogel. (Figure 7)

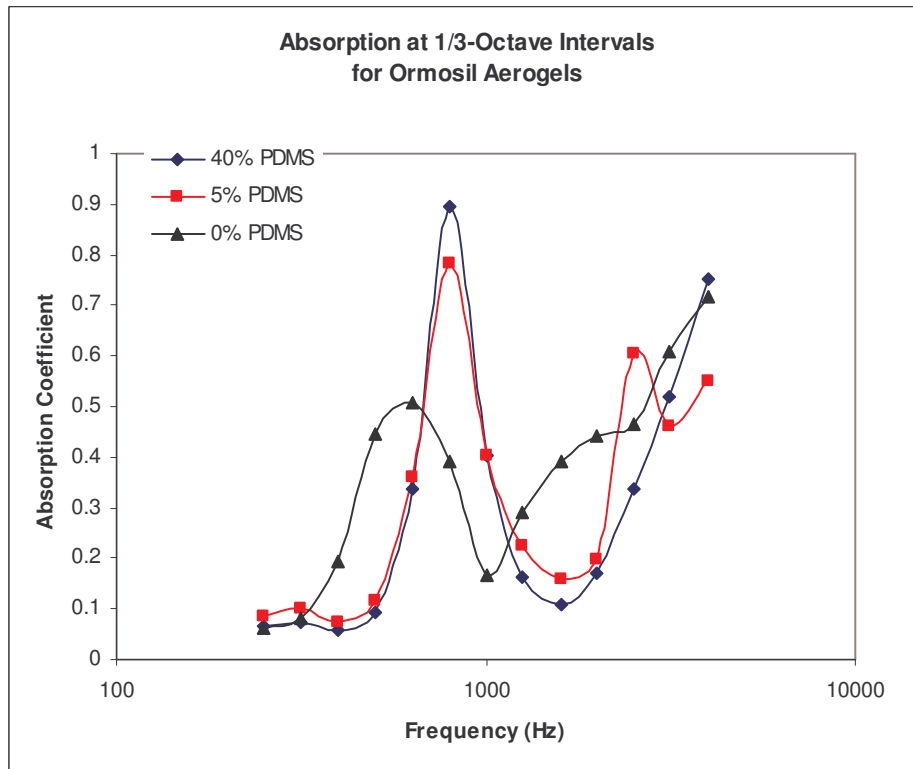


Figure 7. Absorption data for composite aerogels (5 mol% and 40 mol% PDMS) compared to that of an undoped silica aerogel (0 mol% PDMS).

Since aerogels are comprised of > 98% air, they can be modeled acoustically with an equivalent fluid. At normal incidence, the impedance of an unknown point can be found using the relationship:

$$Z_2 = Z_c \frac{-jZ_2 \cot kd + Z_c}{Z_2 - jZ_c \cot kd}$$

where,  $Z_c$  is the characteristic impedance,  $k$  is a material property

c is the velocity of propagation defined as:  $c = \sqrt{\frac{K}{\rho}}$

j is the imaginary quantity:  $j = \sqrt{-1}$

and d is the distance between 2 points,  $x_1$  and  $x_2$ .

In Propagation of Sound in Porous Media, the characteristic impedance ( $Z_c$ ) is defined as:

$$Z_c = \rho_0 c_0 \left[ 1 + 0.0571X^{-0.754} - j0.087X^{-0.732} \right]$$

X is dimensionless and can be expressed as  $X = \frac{\rho_0 f}{\sigma}$  and  $0.01 < X < 1.0$

f is the frequency,  $f = \frac{\omega}{2\pi}$

$\sigma$  is the flow receptivity:  $\sigma = \frac{p_2 - p_1}{Ve}$

where V is the mean flow of air per unit area of the material, e is the thickness, and p is pressure. At normal incidence, the reflection coefficient is related to impedance by:

$$R_2 = \frac{Z_2 - Z'_c}{Z_2 + Z'_c}$$

where  $Z'_c$  is the characteristic impedance of the material that adjoins the sample.

It is likely that the rigidity of the aerogel (0% PDMS) causes high sound velocities, high impedance, high reflectivity, and therefore low absorption. By lowering the modulus of elasticity of the aerogel with PDMS, the bulk modulus is also lowered and a corresponding increase in absorption is observed. However, when comparing the composite aerogels to fiberglass, it is less clear as to the difference in absorption behavior. It has been suggested that the peak at 1000 Hz could be the result of Helmholtz resonance of the aerogel. This is unlikely due to the high tortuosity of the pores and the small pore sizes, described below.

The composite aerogels, even with 40 mol% PDMS, retained a high surface area,  $\sim 430 \text{ m}^2/\text{g}$ . The average pore diameter was  $\sim 5 \text{ nm}$  with a narrow pore size distribution. (Figure 8) Pores greater than 8 nm in diameter has been shown to have a Rayleigh scattering effect for phonons. ("Acoustical Attenuation in Silica") The small pore size and relatively uniform pore distribution suggests that the sound attenuation of the composite aerogels are largely unaffected by the pores themselves.

Another area of consideration is the homogeneity of the composite on the nano-scale. Scanning electron microscopy show that there are may be some phase separation between the silica and the PDMS. (Figure 9) The interaction of each individual phase and the interaction between the phases also need to be explained in connection with the acoustic behavior.

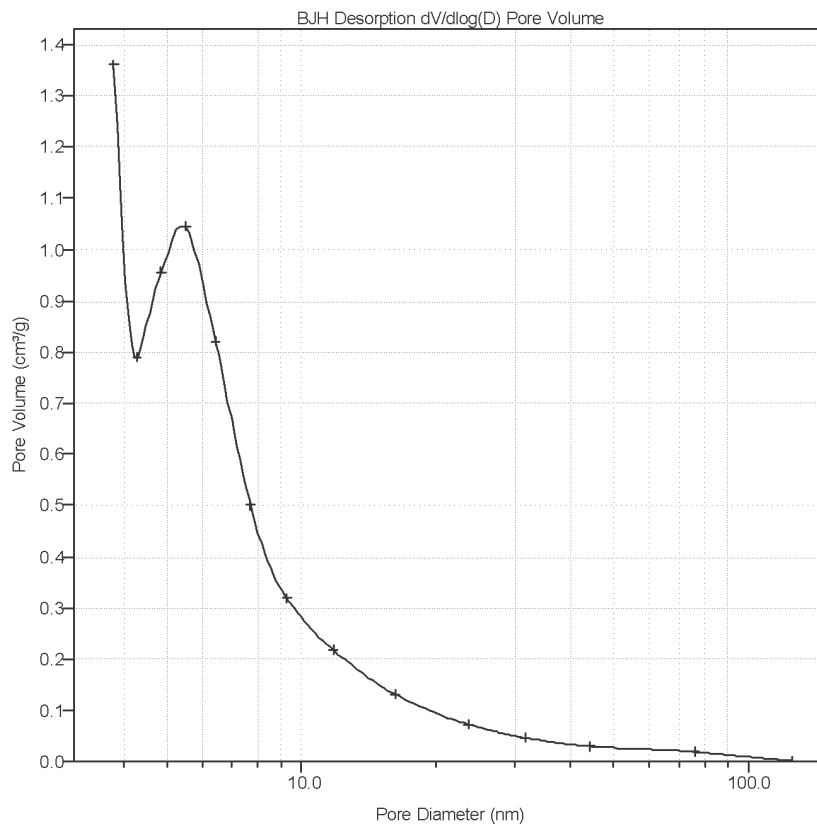


Figure 8. Pore size distribution of the composite aerogel with 40 mol% PDMS. The narrow peak around 5 nm is evidence for uniform pores with diameters around 5 nm.

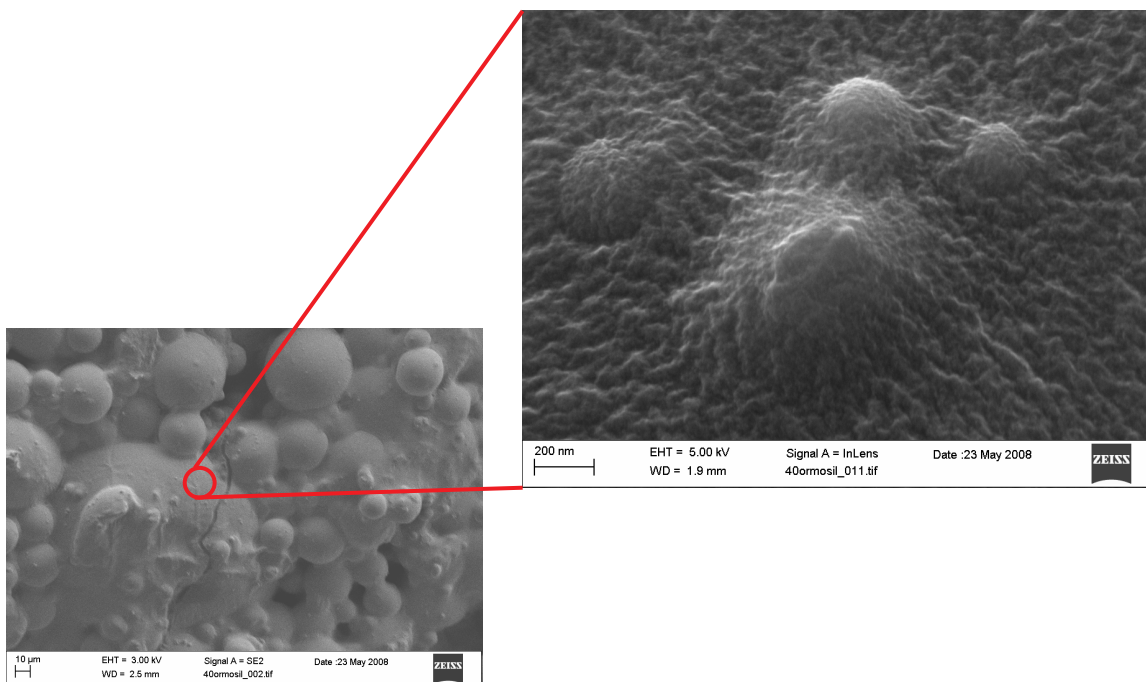


Figure 9. SEM of composite aerogel with 40 mol% PDMS.

**Conclusions:**

Silica/PDMS composite aerogels with varying concentrations of PDMS were synthesized and initial measurements of acoustic properties were conducted. The incorporation of the PDMS into the silica aerogel successfully modified the structure to reduce the rigidity of the aerogels. The results show that these materials have a large surface area ( $> 400 \text{ m}^2/\text{g}$ ) and small pore sizes ( $d \sim 5 \text{ nm}$ ). Although these composites appear homogeneous, SEM images also suggest phase separation at the nano-scale. The composite aerogels have a significantly different acoustic absorption characteristic compared to both fiberglass and undoped silica aerogels. The absorption peak (95% absorption at  $\sim 1000 \text{ Hz}$ ) is clearly a result of the addition of PDMS into the silica structure. However, the cause of this peak, whether it is due to resonance effects, interaction between silica and PDMS phases, or attenuation of sound at this frequency, is still under investigation.

The results of this project has been presented at the Cal Poly Pomona College of Engineering Research Symposium (May 2008) and the ASA International Meeting in Paris (July 2008), and will be presented at the Southern California Council on Undergraduate Research (Nov. 2008). This project is on-going and proposals to federal agencies such as the National Science Foundation for further funding is under preparation.

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