

Acoustic Properties of Organic/Inorganic Composite Aerogels

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Abstract

Composite aerogels (with varying concentrations of silica and poly-dimethylsiloxane) were developed and their acoustic absorption coefficient as a function of composition and average pores size have been measured. The polydimethylsiloxane modified the ceramic structure of the silica aerogels, decreasing the material's rigidity while maintaining the high porosity of the aerogel structure. The composite aerogels were found to exhibit different modes of acoustic absorption than that of typical porous absorbers such as fiberglass. At some frequencies, the composite aerogels had 40% higher absorption than that of commercial fiberglass. Physical data show that these materials have a large surface area (> 400 m²/g) and varying pore sizes (d ~ 5 - 20 nm).

Introduction

When sound encounters a material, the pressure wave can be reflected, transmitted, or absorbed. Material properties such as elastic modulus and porosity directly influence how a material will interact with incident acoustic waves. For materials with a high elastic modulus, regardless of porosity, acoustic reflection is high. Materials with both a low modulus of elasticity and low porosity allow for acoustic transmission. In terms of absorption, the combination of low modulus and high porosity are ideal. (Table 1)

Table 1. Influence of elastic modulus and porosity on a material's acoustic response.

Modulus	Porosity	Example	Reflection	Transmission	Absorption
↑	↑	Concrete	High	Low	None
↑	↓	Steel	High	None	None
↓	↓	Water	Low	High	Low
↓	↑	Snow	Low	Low	High

In optimizing materials for architectural acoustic absorption, typically a high absorption coefficient across a wide range of frequencies is desired. Commercially available acoustic materials generally have high absorption at frequencies above 1000 Hz and negligible absorption below 800 Hz. The primary goal of this project is to develop materials with tunable modulus and porosity to prepare an adjustable acoustic absorber, specifically one that can absorb well below 800 Hz. A composite of silica aerogels and poly-dimethylsiloxane (PDMS) combines the low modulus and high porosity.

Silica aerogels have very high surface areas (300 – 1000 m²/g) and high porosity (80 – 99%) and can be synthesized through a liquid-precursor, room-temperature method. The high porosity leads to extremely low sound velocity (down to 90 m/s) through the silica aerogel. [1] However, the high elastic modulus also results in high reflection coefficients, especially at low frequencies. In order to modify the elastic modulus of the silica aerogels, PDMS is incorporated into the silica structure making Organically Modified Silicates (or ormosils). (Table 2) In contrast to traditional composites of alternating layers of mm-thick polymer and ceramic fibers, ormosils contain alternating polymer and ceramic groups at the molecular level giving a more homogeneous structure.

Table 2. It has been shown that the elastic moduli in silica/PDMS ormosils can be modified by controlling the concentration of PDMS. [2]

Mol % Polymer (PDMS)	0	7.9	12.8	19.5	23.7
Elastic Modulus (GPa)	20.7	18.6	16.0	15.0	13.0
Vickers Hardness (kg/mm ²)	186	160	140	110	88
Fracture toughness (Mpa/m ^{1/2})	0.50	0.49	0.48	0.47	0.46
Brittleness (mm^{-1/2})	3.63	3.19	2.86	2.32	1.88

Additionally, the concentration of PDMS can modify the pore size of the ormosil aerogel. It has been shown that pore sizes of 8 nm and greater produce a Rayleigh scattering of phonons. [3] [4] Increasing the mean pore size has been shown to increase the sound attenuation of silica

aerogels in addition to decreasing the longitudinal sound velocity. [4] The larger pore sizes also aid in better impedance matching with air, which leads to a lower reflectivity.

Experimental Procedures

The silica/PDMS ormosils were synthesized through combining 0.025 mol TEOS (Alfa Aesar), 0.0035 mol HCl, 0.042 mol THF (Acros Organics), 0.17 mol isopropanol (Acros Organics), 0.25 mol distilled water, and PDMS (Gelest Inc.). The PDMS:silica ratio varied between 0.05:0.95 to 0.40:0.60. The mixture was then sonicated (Branson Ultrasonics) for 60 minutes to achieve homogeneity. Gelation occurred within 20 minutes after sonication. The gels were allowed to age for 3 days and then washed with acetone to remove all liquid reaction byproducts. The acetone is then removed through CO₂ supercritical drying (Polaron ES300).

The absorption coefficients of the ormosil aerogels were measured in a home-made impedance tube, based on ASTM standards (C 384 04). The impedance tube is an enclosed tube with a speaker on one end and the acoustic absorber sample on the other end. An acoustic standing wave can be set-up within the tube and the wavelength and amplitude of this standing wave is measured with a movable microphone within the tube. By measuring the locations of the wave maximums and minimums, the Standing Wave Ratio (SWR) and the absorption coefficient (α) of the absorber can be calculated. The SWR is the ratio of the maximum acoustic pressure and the minimum acoustic pressure within the tube. The absorption coefficient represents the percentage of acoustic energy absorbed by the material.

$$\alpha = 1 - \frac{(SWR - 1)^2}{(SWR + 1)^2}$$

Surface area and average pore size were measured with nitrogen gas adsorption (Micromeritics, ASAP 2010). Scanning electron micrographs (SEM, Zeiss, Evo) were obtained to view microstructure.

Results and Discussion

Figure 1 shows the acoustic absorption profile of the silica/PDMS ormosils as a function of PDMS concentration. Their absorption profiles are compared to two commercially available acoustic insulators – fiberglass and a composite acoustic pad.

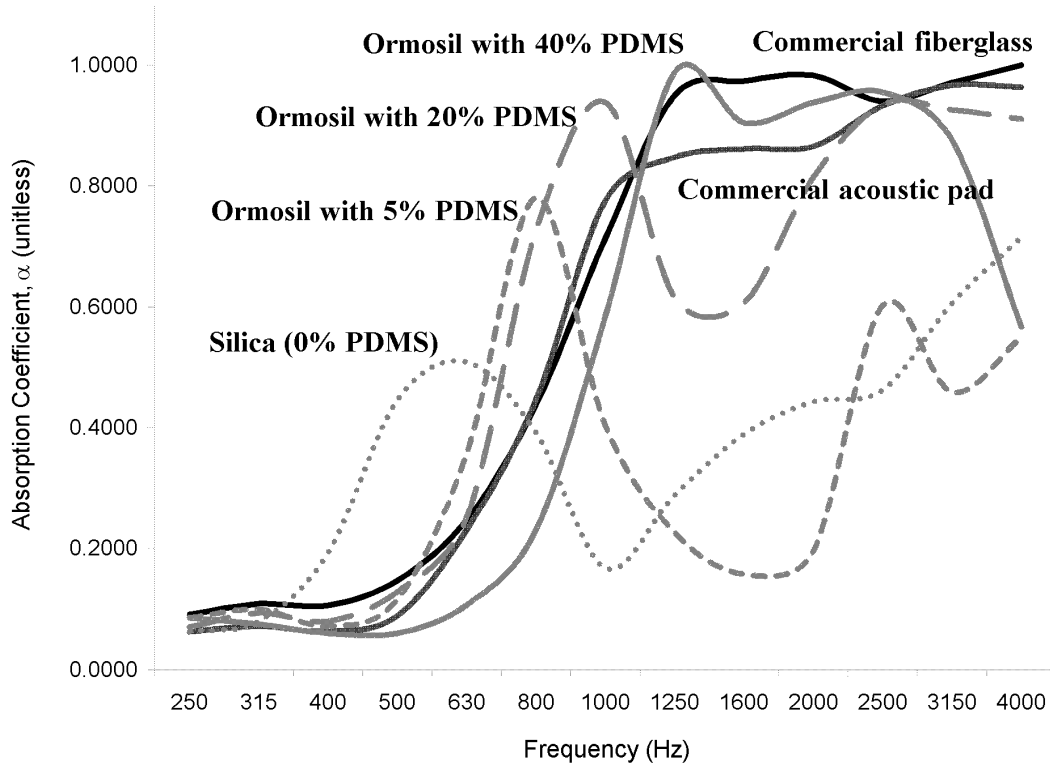


Figure 1. Adsorption coefficient as a function of the acoustic frequency. Silica/PDMS ormosils (with varying concentrations of PDMS) are compared with commercially available acoustic insulators.

There are three general types of acoustic absorption – porous absorbers, panel absorbers, and resonators. [5] The fiberglass and the acoustic pad exhibit typical porous absorber behavior. Their open cell structures allow air to flow through and the acoustic pressure wave is converted to heat as it passes through the material. This is the most common type of acoustic absorber with high α at frequencies above 1000 Hz and almost no absorption below 800 Hz. Common material characteristics considered by acousticians are open porosity (volume of free air per unit volume of porous medium) and surface area. [6] The silica aerogel (with no PDMS) and the ormosil sample with up to 20% PDMS exhibit very different absorption behaviors. The narrow absorption peak suggests that instead of behaving as a porous absorber, the aerogels are behaving as a resonator, with the pores acting as the resonating cavities.

Resonators are typically materials with discrete cavities. Air does not necessarily flow through the material but the vibration of the air in the cavities is what dissipates acoustic energy. The frequency at which resonators absorb depend strongly on the cavity dimensions and therefore, resonators only absorb over a very narrow frequency range. There are two models that approximate the type of porous structure of the aerogels. The Helmholtz model assumes a cavity with a neck. The resonance frequency is inversely proportional to the size of the cavity and also inversely proportional to the length of that neck.

$$f_H = \frac{v}{2\pi} \sqrt{\frac{A}{VL}}$$

f_H = Helmholtz resonant frequency

v = speed of sound

A = cross-sectional area of neck

L = length of neck

V = volume of the cavity

If the cavities are shaped more like a sphere, the resonance frequency is inversely proportional to the diameter of the cavity and directly proportional to the size of the opening.

$$f = Y \sqrt{\frac{d}{D^3}}$$

F = resonant frequency

Y = material constant

d = diameter of the cavity opening

D = diameter of the cavity

In both cases, as the cavity size increases, resonance based absorption occurs at lower frequencies. [7]

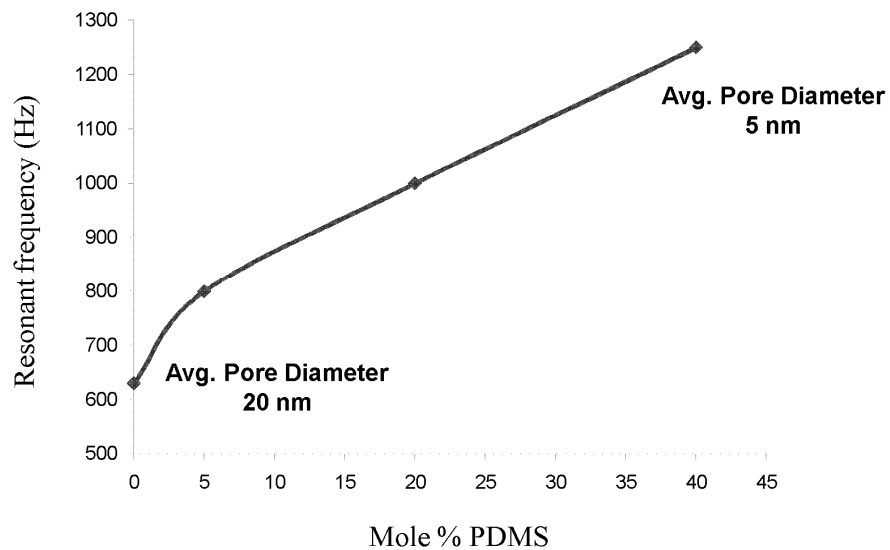


Figure 2. Resonant frequency as a function of PDMS concentration.

By increasing the PDMS concentration, the absorption peaks of the ormosils are shifted to higher resonant frequencies. This reflects the data that as the concentration of PDMS increases, the pore size of the ormosil aerogels decreases. As polymer is added, the average pore

size decreases from 20 nm to 5 nm. (Table 3) Therefore, peak absorption frequency of the ormosil aerogels is inversely proportional to pore size, which matches the resonator model.

Mol % PDMS	Surface Area (m ² /g)	Avg. Pore Volume (cm ³ /g)	Avg. Pore Diameter (nm)
0	800	8.0	20
40	450	0.5	5

Table 3. Surface area and average pore size of silica aerogels compared to an ormosil aerogel with 40% PDMS.

Scanning electron micrographs also confirm the initial pore size measurements. The SEMs of a 40% PDMS ormosil and that of a 20% PDMS ormosil look very similar at the 100 nm length scale. Both are composed of small, 10 nm sized, spherical particles. (Figure 3)

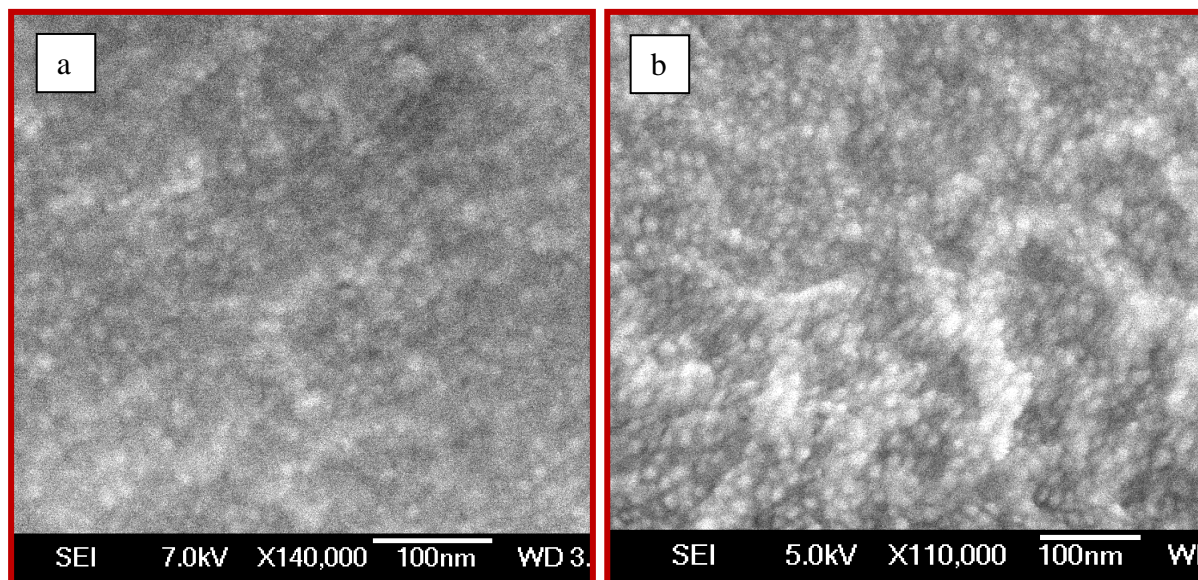


Figure 3. SEMs at the 100 nm-range of ormosil aerogels with a) 40% PDMS and b) 20% PDMS.

However, at the 10 μm length scale the two materials look very differently. The 40% PDMS ormosil is much smoother with fewer pores whereas the ormosil with 20% PDMS has larger pores and a rougher structure. (Figure 4) The higher concentration of PDMS results in a smoother sample with fewer or smaller pores and the sample with less PDMS is more porous with larger pores. This further confirms that the ormosils exhibit resonator absorption behavior and that the larger the pores, the lower the resonant frequency.

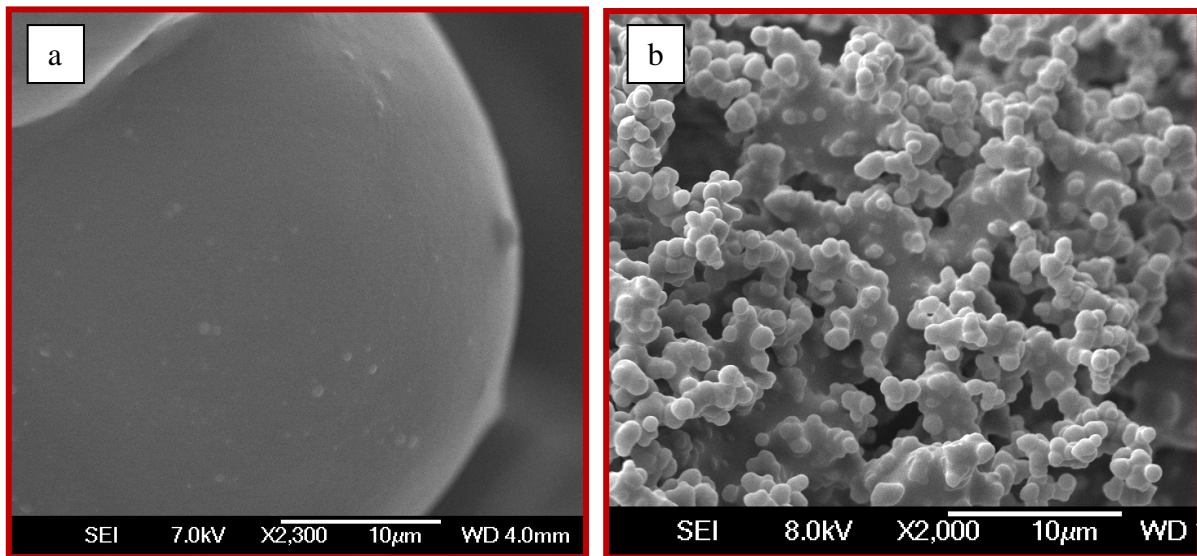


Figure 4. SEMs at the 10 μm -range of ormosil aerogels with a) 40% PDMS and b) 20% PDMS.

In terms of the amount of acoustic energy absorbed, the absorption coefficient also varies as a function of PDMS concentration. (Figure 5) The higher the concentration of PDMS, the higher the level of absorption. This can be explained by the tuning of elastic moduli of the ormosil aerogel with the addition of PDMS. An increased concentration of PDMS results in lower elastic modulus, which results in higher levels of acoustic absorption.

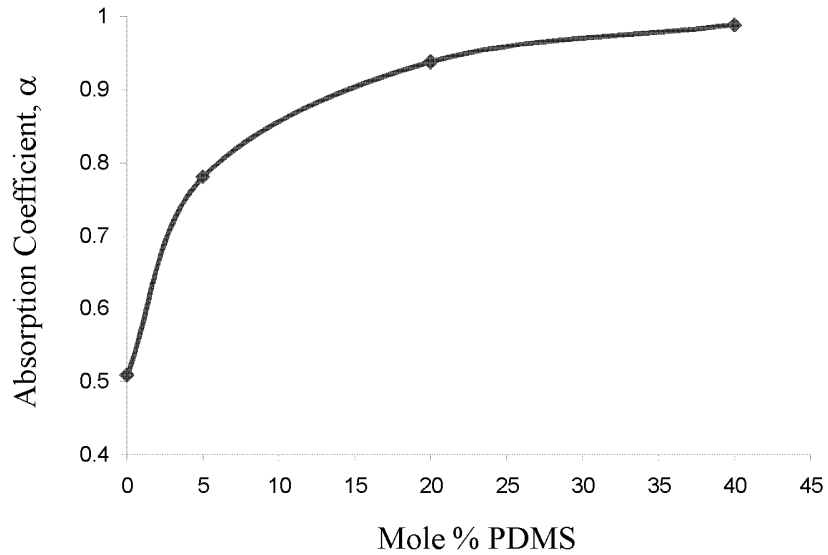


Figure 5. Peak absorption coefficient of silica/PDMS ormosil aerogels as a function PDMS concentration.

Conclusions

The elastic moduli of silica aerogels can be modified and lowered through the incorporation of PDMS to form an ormosil aerogel. The mode of absorption of the aerogels is different than that of commercially available acoustic insulators. Whereas commercially available material exhibited porous open-cell absorption, the aerogels' behavior resembled that of resonators. The moduli and the pore size of the resultant aerogel are functions of the concentration of PDMS, the higher the concentration of PDMS, the smaller the average pore diameter and the lower the elastic modulus. The resonant frequency is generally considered a function of cavity size, higher resonant frequencies are correlated to smaller pore sizes, which in this case is a higher concentration of PDMS. The level of absorption is a function of the elastic modulus of the material where the lower the modulus (higher concentration of PDMS) the higher the absorption.

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