Acoustic Properties of Silica Aerogels from $400mK$ to $400K$

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Abstract

Using acoustic resonant spectroscopy, we have studied the low frequency elastic properties of silica aerogels with densities between $0.045g/cm^3$ and $0.267g/cm^3$ over a temperature range of $400mK$ to $400K$. We observe a large change in the elastic moduli between $20K$ and $100K$, accompanied by a damping peak, and attribute this to a thermally activated relaxation process. Above $100K$, the elastic moduli increase linearly with temperature, a phenomenon normally associated with elastomers such as rubber, and indicative of a significant entropic contribution to the stiffness of the aerogels.

Key words: aerogel; acoustics; elastic moduli

1. Introduction

Aerogels are best thought of as a network of strands of nanometer size particles connected by sparse weak links.[1] As a result the elastic moduli of aerogels are orders of magnitude smaller that those of the corresponding bulk solids. There is some similarity in the structure of aerogels and elastomers such as rubber, in the sense that both consist of randomly linked mobile segments. In contrast to most solids, elastic deformation of a rubber leads to significant changes in the available configuration phase space, and thus the entropy.[2] Among other things, this leads to a shear modulus that is linearly proportional to the temperature. To investigate the extent of the entropic contributions to the elastic properties of aerogels, we determined the temperature dependence of the sound speeds in aerogel using acoustic resonant spectroscopy.

We prepared three samples, with properties summarized in table 1. The two denser samples were made in a single step process, with TMOS as the silica precursor and ammonia as catalyst. The lowest density sample was made using the two step method described by Tillotson and Hrubesh.[3] The acoustic resonators consist of a copper block with a cylindrical cavity. The aerogel samples fit the cavity but are approximately $0.5mm$ longer. The cavity is capped off with two acoustic transducers that slightly compress the samples. This ensures that the samples are firmly held in place in the cavity over the full temperature range of the experiments. The transducers are brass caps, at the center machined down to membranes with a thickness of approx. $0.25mm$ over a diameter of $6mm$, supporting small piezoelectric (PZT) disks. The caps are attached to the copper body using an epoxy adhesive. The resonators are evacuated through a hole in the side of the cavity.

Table 1
Sample densities and dimensions, and the low-temperature values of the transverse sound speed, $c_{t,0}$ and the shear modulus, μ_0.

$\rho(g/cm^3)$	$d(cm)$	$l(cm)$	$c_{t,0}(m/s)$	$\mu_0(MPa)$
0.267	0.795	1.81	156	6.50
0.124	1.32	4.02	86	0.92
0.045	0.787	1.87	91.3	0.375

Figure 1 shows the resonance frequency of the fundamental mode, f_1, and the corresponding damping, Q^{-1}, as a function of temperature for the lowest density

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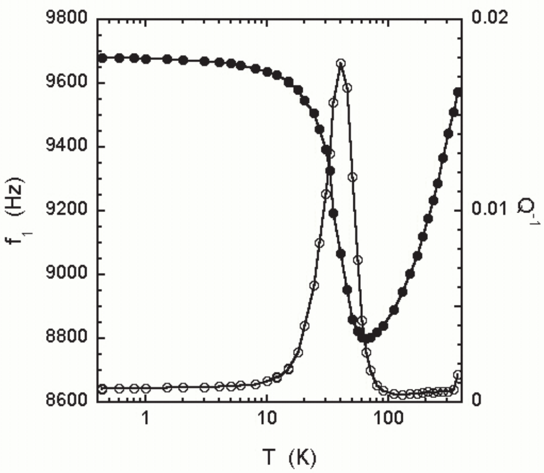

Fig. 1. The resonance frequency of the lowest mode (solid symbols) and the corresponding damping (open symbols) for an aerogel sample with a density of $0.045 g/cm^3$. The up turn in the damping around $400K$ is due to softening of the epoxy that seals the caps to the body.

sample. The higher density samples show very similar behavior. To analyze the data we have written a program to calculate the resonance frequencies of an elastic cylinder. The calculation is based on a variational method commonly used in resonant ultrasonic spectroscopy, but with a choice of basis functions adapted to meet fixed boundary conditions.[4] The numerical model shows that the resonance frequencies of a cylinder with fixed boundary conditions are mainly determined by the transverse sound speed, c_t. Using the geometry and density of the samples as input to our program, and assuming a Poisson ration of 0.22 we obtain c_t from f_1.

Similar to the ultrasonic results of Xie and Beamish [5] , we observe a drop in the sound speed and an accompanying damping peak between $20K$ and $100K$. The magnitude of the damping peak is consistent with an otherwise unspecified thermally activated relaxation process. In addition, we find that on further increasing the temperature the sound speed increases again. Both the strength of the relaxation process and the increase in sound speed at higher temperature are more pronounced in the denser, one-step samples.

From the transverse sound speed we can obtain the shear modulus, $\mu = \rho c_t^2$. To be able to compare the temperature dependence of μ between samples, we show in figure 2 the change in the modulus scaled by its low-temperature value, μ_0. The behavior of the two denser samples is quite similar, with a drop in shear modulus of nearly 50% between $20K$ and $100K$, followed by a linear increase at higher temperature, and a damping peak centered at $60K$. The lower density sample shows qualitatively similar behavior, but both the damping peak and the corresponding decrease in the modulus are significantly smaller and take place at lower temperature, with the damping peak at $40K$. We take the linear temperature dependence of μ as indicative of the importance of the entropic contribution to the aerogel stiffness.

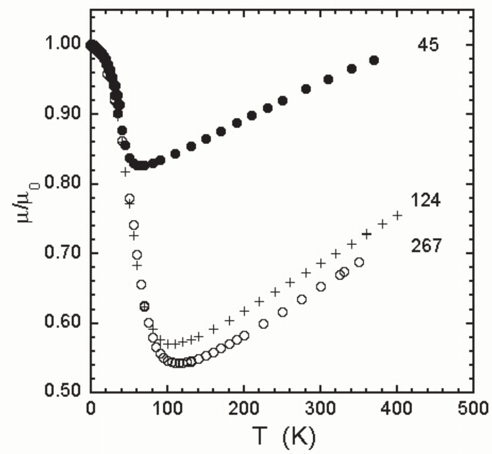

Fig. 2. The relative change in shear modulus. The numbers in the figure indicate the density of the samples in kg/m^3.

Acknowledgements

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