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Thermal conductivity and characterization of compacted, granular silica aerogel

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ABSTRACT

Monolithic silica aerogels are well known for their low thermal conductivity (approximately 15 mW/(m K)) (Aegerter et al. (Eds.), 2011. Aerogels Handbook, first ed., Springer-Verlag New York, LLC, New York, NY). Their low relative density (typically less than 5%) reduces conduction through the solid and their small pore size, typically less than one hundred nanometers, on the order of the mean free path of air, reduces conduction through air, as well as convection and radiation. As they are fragile and brittle, they are often used in a granular form in thermal insulation, with some increase in their thermal conductivity from the air between the granules. Here, we describe a technique for compacting a bed of granular silica aerogel that reduces the thermal conductivity from 24 mW/(m K) (when uncompacted) to 13 mW/(m K) (after compaction). We find that there is an optimum level of compaction to minimize the thermal conductivity: at higher levels of compaction, the contact area between the granules increases and the granules densify, increasing conduction through the solid.

1. Introduction

With the ever-increasing importance of energy efficiency, improved products and techniques for thermal insulation are needed. Aerogels are one such cutting-edge material with significant potential to meet this need. [1,2] Their thermal conductivities are typically around 15 mW/(m K) [3], much lower than that of conventional insulations, such as standard fiberglass batts (40 mW/(m K) [4]) and closed-cell polyurethane foams (25 mW/(m K) [5]). After cost, one of the main limitations that has prevented aerogels from becoming more widely used as a high-performance insulation product is their fragility, which results from their low density and inefficient distribution of solid for resisting mechanical loads.

The solid matrix of the nanoporous microstructure of aerogels can be described as "beads on a string" or "pearls on a necklace" [3], referring to their structure of roughly spherical particles connected by small necks or thin strands. For a given mass of material, this structure is much less stiff and strong than that of an open-cell foam (or a fully dense solid). For instance, at a density of 100 kg/m^3, the modulus and flexural strength of silica aerogels are typically 1 MPa and 0.02 MPa, respectively [6]; by comparison, rigid, closed-cell polyurethane foams of similar density have moduli and strengths of about 10 MPa and 0.6 MPa, respectively [5]. The Young's modulus and strength of aerogels decreases with decreasing density much faster than those of open-cell foams: the Young's modulus of aerogels varies with relative density raised to the power 3.7 [6] while that of open-cell foams varies with relative density squared [5]; and the flexural strength of aerogels varies with relative density raised to a power of between 2.5 and 3 [6,7] while that of open-cell foams varies with relative density raised to the power 1.5 [5]. (Relative density is the density of the porous material divided by that of the solid from which it is made).

The microstructure of aerogels also gives rise to their extremely low thermal conductivities [8]. Heat transfers from one object, surface or substance to another in three main ways: by conduction, convection and radiation [9]. Conduction is minimized by reducing the volume fraction of solid in the material (as solids have higher thermal conductivities than gases), which is why most insulations have relatively low densities. The remainder of the insulation is filled with a gas, such as air (thermal conductivity of 25 mW/(m K)), but which is susceptible to convection and radiation. With large voids, these two mechanisms can provide much higher heat

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transfer than solid conduction. Large voids allow radiation to quickly bypass significant lengths of the material and convection currents to be set up. Therefore, to minimize these heat transfer modes, the air (or gas) is typically contained in small pockets throughout the material; this is why most insulation materials are designed to have small pore sizes. In closed-cell foams with pore sizes less than a few millimeters, convection is negligible, as the buoyancy forces associated with a temperature gradient across the cell are countered by viscous drag of the fluid against the walls of the foam. In aerogels, the pores are at the scale of 2 to 50 nm [10], less than the mean free path of air, significantly reducing conduction through the gas, too. Insulation materials can also decrease heat transfer by replacing the air within the pores with a lower conductivity gas (such as argon) or by decreasing the air pressure inside the insulation (as in vacuum insulated panels).

As a result of their fragility, silica aerogels used in commercial products are often in a granular form, so that handling the material becomes easier and varying shapes of cavities can be more readily filled. However, since there is air in the interstitial spaces between the silica aerogel granules and the thermal conductivity of air is higher than that of monolithic aerogel (25 mW/(m K) for air versus typically 15 mW/(m K) for silica aerogel at ambient conditions [3]), a bed of granular aerogel has a higher thermal conductivity than monolithic aerogel. Here, we describe and characterize a means of compacting silica aerogel granules, reducing the interstitial volume fraction of air and decreasing the thermal conductivity of the granular bed from 24 mW/(m K) (when uncompacted) to 13 mW/(m K) (after compaction), almost a 50% reduction in thermal conductivity. In a companion paper [11], we describe the use of compacted granular aerogels in a sandwich panel with a three-dimensional truss core, which combines structural support with low thermal conductivity, suitable for building applications; the panel, which is opaque, is intended for insulation of walls rather than fenestration.

2. Materials and methods

The granular silica aerogel used in this study was commercially available Cabot P100 aerogel (Cabot Corporation, Billerica MA). According to Cabot literature and correspondence with a Cabot representative, the thermal conductivity of an “untapped” particle bed of this material at room temperature was about 22 mW/(m K) and the density of individual particles ranged from 120 to 180 kg/m^3 [12,13]. The bulk granular aerogel was uniaxially compressed in modified graduated cylinders. The microstructure of the as-received and compacted granular aerogel was characterized by micro-computed tomography imaging, as well as measurement of the particle size distribution by sieving, and the pore size distribution and internal surface area by gas absorption/desorption analysis, described in more detail below. The volume fraction of air in the interstitial spaces between the particles was calculated. The thermal conductivity was measured on bulk samples of the as-received and compacted granular aerogel.

2.1. Compaction technique

Since air has a higher thermal conductivity than monolithic aerogel, the conductivity of a bed of aerogel granules can be reduced by decreasing the volume of air in the interstitial spaces between the granules. In this study, we achieved this by compacting the granules uniaxially in rigid molds.

The initial naturally-settled bed (or bulk) density of the Cabot aerogel was approximately 68 kg/m^3. The compression percentages mentioned in this research reference the compaction from this initial bed density. These percentages and their approximate corresponding bed densities can be found in Table 1. Compaction was performed in modified graduated syringes marked in 1 mL increments, since they provided a convenient means of tracking the changes in height and volume of the samples during the experiment. A flat plastic base was inserted into the bottom of the cylinder of the syringe to prevent granules from exiting through the hole at the tip of the syringe. The volumetric scale of the syringe was consequently offset by the thickness of this base. A flat base was also attached to the end of the plunger, such that a cylindrical chamber was formed to hold the granules. These bases were designed to fit loosely enough in the syringe such that the granules are maintained but air is free to move in and out of the sample column. An Instron mechanical testing machine (Model 4201, Instron, Canton, MA) was used to compact the granules to strains ranging from 10% to 70% at a nominal compaction speed of 10 mm/min.

Table 1
Cabot granular aerogel compressive strain and corresponding bed density.

Compressive strain (%)	Bed density (kg/m^3)
0	68
10	76
20	85
30	97
40	113
50	136
60	170
70	227

2.2. Micro-computed tomography

Micro-computed tomography (micro-CT) uses x-rays to scan a three dimensional object and a computer to process the data from the scan to allow users to see cross sections of solid objects. Aerogel granules were first compressed in modified cylindrical chambers to different bed densities, and then scanned with a micro-CT scanner at the David H. Koch Institute for Integrative Cancer Research at MIT (GE eXplore CT 120, 25 μm resolution; Little Chalfont, UK). These scans provided a three-dimensional digital image of the sample which could be viewed, slice by slice, in planar cross sections in a grayscale format. Objects that appeared in a lighter shade of gray corresponded to a higher density, while darker shades of gray represented lower density materials. Due to the similarity in density, and consequently in the gray color between the aerogel granules and the ambient air, it was difficult to distinguish between the granules and the interstitial air in the initial scans. To improve the contrast between the granules and the interstitial spaces, the samples were submerged and compressed in water. During compression the water was free to flow out of the cylindrical chamber through a filtered opening, to minimize any additional stresses from the water pressure, while keeping the hydrophobic granules in the chamber. One sample with an initial uncompressed granular volume of 20 mL was scanned at each level of compression. Images of the compressed granules in air and in water are shown in Fig. 1.

2.2.1. Determining the grayscale threshold and the volume fraction of air

The digital planar cross sections obtained by the scanner were then analyzed using ImageJ, an image-processing program developed at the National Institutes of Health. A histogram of the grayscale level of all voxels in a sample was first plotted (Fig. 2). By specifying an appropriate grayscale threshold value, the granules were identified, allowing the volume fraction of air to be computed at each compression level. The grayscale threshold that optimally distinguishes the aerogel granules from the interstitial water between the granules corresponds to the deepest part of the trough between the peak for each phase in the grayscale histogram