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THERMAL AND SOLAR-OPTICAL PROPERTIES OF SILICA AEROGEL FOR USE IN INSULATED WINDOWS

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ABSTRACT

Silica aerogel is a porous insulating material that is transparent to solar radiation. To understand its insulating performance in a window system, it is necessary to first study component heat transfer paths. Aerogel's absorption coefficient, a measure of the attenuation of radiation heat transfer, was determined over the spectral range 1-200 μm. Although radiation heat transfer is negligible over much of this region, there is a transmission window between 3-6 μm. At ambient temperatures, for aerogel thicknesses of 0.5-5.0 cm, radiation heat transfer through an unmodified aerogel window is less than 15% of the total heat flux. For evacuated or high-temperature furnace windows, this contribution can be over 50%. Thermal radiative transfer can be somewhat decreased by allowing the aerogel to absorb moisture, but solar transmission and optical clarity are sacrificed. Absorption of water vapor over time causes irreversible structural changes that increase scattering in the solar spectrum. Aerogel's thermal performance can be improved by replacing the pore gas with one of lower conductivity or by evacuating the aerogel to pressures below 0.1 atm. A hypothetical evacuated aerogel window has a calculated U-Value of ≈ 0.5 W/m^2–K for a gap spacing of 12.5 mm, which is four times better than currently available low-emissivity gas-filled units of similar size.

1. INTRODUCTION

This paper summarizes the thermal performance properties of silica aerogel, a unique material that is both optically transparent and thermally insulating (1). Sandwiched between two panes of glass (i.e., placed in the air space of a conventional double-glazed window), aerogel can greatly reduce heat transfer without significantly reducing solar transmission or increasing the thickness of the window. Decreasing the pressure inside an aerogel window can lead to further reductions in the overall U-value of aerogel windows.

Silica aerogel is a microporous material made by supercritical extraction of an alcohol solvent from a colloidal gel of silica. Kistler discovered aerogel and studied its thermal properties in 1931 (2). Recently aerogel has been produced by replacing the alcohol with liquid CO_2, simplifying production by lowering the process temperature and pressure (3). The result is a network of short bonded chains of silica particles, roughly spherical but fused together (1). Particles on the order of 2-3 nm in diameter and pore sizes on the order of 50-100 nm produce a solid-gas matrix in which the solid fraction can be less than 5% (4). Because of this low solid fraction and fine structure, aerogel is an excellent thermal insulator. Also, unlike other insulating materials, aerogel is visually transparent because the silica particles are much smaller than visible wavelengths.

This study was undertaken in order to optimize the design of aerogel windows for residential applications. Both radiation and conduction heat transfer are greatly reduced and convection is non-existent in aerogel. In order to analyze radiation heat transfer through aerogel, we extended the measurements of the absorption coefficient (a measure of the attenuation of radiation heat transfer) to wavelengths that are longer than were previously measured and to aerogels made by the improved process mentioned above. A summary of heat transfer through aerogel follows with a presentation of calculated effective conductivities as a function of thickness and temperature. Immediately after the production process, when placed in ambient conditions, aerogel will absorb moisture. The effects of this water absorption on thermal and solar optical performance are examined. Finally, these results are used to predict the performance of aerogel-filled windows and to compare them to currently available window configurations.

2. SPECTRAL ABSORPTION COEFFICIENTS

Fundamental to an accurate analysis of heat transfer through aerogel is an understanding of its infrared transmitting and absorbing properties. In a conventional window, glass is opaque to thermal radiation from room temperature sources (98% of which is between 5 and 75 μm); however, radiation heat transfer by absorption and reradiation can be significant, especially if low-emittance coatings are not used. For the case of aerogel, where the infrared reflectance is close to 0, transmittance measurements by wavelength will also yield overall absorptance. In the spectral regions below 7.5 μm and above 40 μm, transmittance measurements were made on specially prepared bulk samples. However, aerogel in thickness greater than 0.1 mm is opaque to infrared radiation between 7.5 and 40 μm. It is not possible to cut a sample of aerogel thin enough to transmit in most of this wavelength region. Thus, we derive the infrared optical properties for aerogel between 7.5 and 40 μm from those of fully dense silica as outlined in Hartmann (5). From this information, we calculate the absorption coefficient, α (in units of 1/thickness). For a given wavelength, if d is the thickness of the sample, I_0 the incident flux, and I the transmitted flux, the absorption coefficient is defined in: $I = I_0 * e^{-d\alpha}$. Thus, the absorption coefficient is a measure of radiation attenuation in a medium; higher values of α correspond to less radiative heat transfer. Figure 1 presents our calculated and measured values of the absorption coefficient normalized by density over the infrared spectrum. Note the transmission window between approximately 3 μm and 6 μm and the fact that the absorption coefficient decreases steadily from wavelengths of 25 μm on up.

In the region below 7.5 μm, absorption is strongly dependent upon the water content of the aerogel. To study this in detail, aerogel samples were first heated to 400 C for 2 to 4 days and placed in a dry nitrogen environment to displace as much moisture as possible. Following this, the aerogel was allowed to age under ambient conditions for approximately 3 months. Figure 2 shows the results of the absorption measurements taken over the three-month period. Note that the depth of the window between 3 and 6 μm decreased by more than an order of magnitude as a result of water absorption. This decrease in the absorption coefficient will reduce radiative heat transfer for wavelengths in this range.

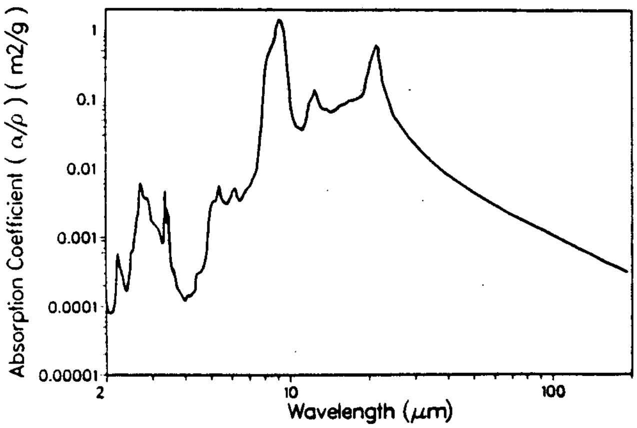

Fig. 1. Absorption coefficient of dry aerogel normalized to density as a function of wavelength, 2-200 μm.

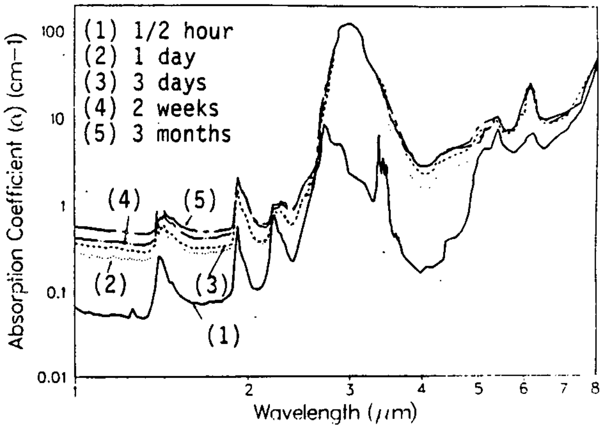

Fig. 2. Absorption coefficient of aerogel dried and then placed under ambient conditions for three months as a function of wavelength, 1-8 μm.

3. HEAT TRANSFER IN AEROGEL

As with all porous insulating materials, aerogel derives much of its high thermal resistance from the dead air space within it. Due to its fine microstructure, which restricts molecular transport (and thus heat transfer), aerogel's thermal conductivity under most conditions is lower than still air (6). However, for some wavelengths, the high void fraction allows for significant radiative heat transfer through the medium. As a result, "conductivity" is no longer an intrinsic property, but rather is dependent on the thickness of the material and the boundary conditions of the specific application. Significant radiative transfer within the medium causes a coupling of radiation and conduction and significantly increases the complexity of the heat transfer solution. A rigorous solution demands an involved numerical treatment. For incorporation into our window heat transfer models, we used a semi-empirical method. Results of this method are within 10% of the more complex numerical calculations. Derivations are well beyond the scope of this paper and the interested reader is referred to Hartmann (5) for a more detailed discussion.

As previously discussed, the absorption coefficient of aerogel is weak in the spectral region between 1 and 7 μm and beyond 50 μm (Figure 1). At room temperature (e.g. for a conventional window application), the blackbody emission peak occurs around 10 μm, well into the highly absorbing region of aerogel, and radiative heat transfer will only account for 10-15% of the total. However, as the temperature increases (as in furnace applications) or decreases (for cryogenics) the blackbody emission peak shifts into the less absorbing regions of the spectrum and radiative heat transfer becomes a more significant part of the overall heat transfer. In cases where conduction heat transfer is greatly reduced (i.e., by reducing the pressure), radiation heat transfer becomes more important (up to and sometimes over 50%).

Conductive heat transfer in aerogel can be broken into two components, that through the solid matrix and that through the air- or gas-filled pores; convection is minimal. We therefore define two corresponding components of the conductive conductivity (k_c):

$$k_c = k_s + k_g. \qquad (1)$$

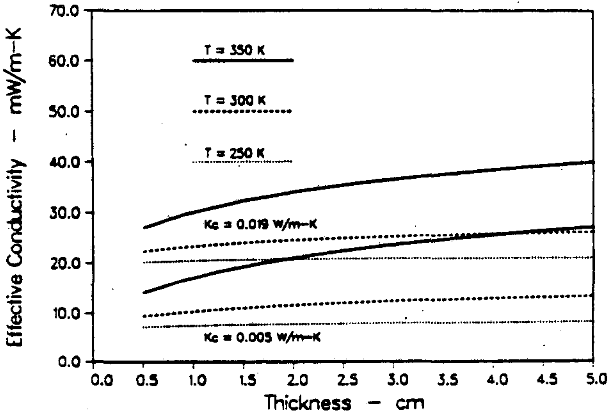

Fig. 3. Effective conductivity (including radiation) of both aerogel at atmospheric pressure (k_c = 0.019 W/m-K) and evacuated aerogel (k_c = 0.005 W/m-K) as a function of thickness for three different temperatures. Boundary emissivities are 1.0.

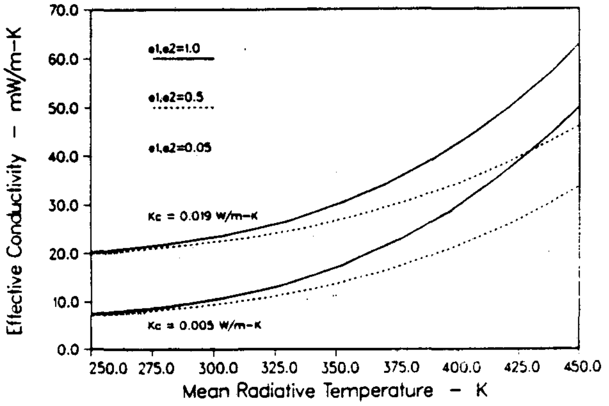

Fig. 4. Effective conductivity (including radiation) of both aerogel at atmospheric pressure (k_c = 0.019 W/m-K) and evacuated aerogel (k_c = 0.005 W/m-K) as a function of mean radiative temperature for three different boundary emissivities. Samples are 10-mm-thick.

The thermal conductivity of the solid matrix (k_s) is determined by measuring the total effective conductivity (including radiation) of evacuated aerogel (such that k_g is 0) and then subtracting off the analytically calculated radiative conductivity. The "thermal conductivity" of a gas within a porous matrix (k_g) is a function of the mean free path of the gas molecules and the effective pore diameter of the matrix. The mean free path is the average distance a gas molecule travels before it collides with another molecule and is a function of the molecular properties of the gas and temperature.

Kistler measured an effective overall thermal conductivity, including radiation ($k_{eff} = (q_{tot}d)/(\Delta T)$) for aerogel slabs under ambient conditions to be k_{eff} = .0203 W/m–K (6). This is lower than pure conduction through still air (k_{air} = .026 W/m–K), since the size of the pores constricts the mean free path of the air molecules. For air filled aerogel at atmospheric pressure, k_s is small (≈.005 W/m–K) and k_g dominates (≈.014 W/m–K).

We calculated effective overall conductivities as a function of mean temperature and thickness for both unmodified and evacuated aerogel. Details are given in Hartmann (5). Figure 3 presents the effective conductivity of aerogel as a function of slab thickness. Two cases are considered for each of three mean radiative temperatures. The top curves are for air-filled aerogel at atmospheric pressure (k_c=0.019 W/m-K) while the bottom three curves are for aerogel evacuated to 0.1 atm pressure (k_c=0.005 W/m-K). The boundary emissivities are constant at 1.0. As temperature rises, radiation heat transfer becomes more important, increasing the effective conductivity. Also, as the temperature rises, a thin aerogel sample will become more transparent to infrared radiation. Thus, the total heat flux and effective conductivity will be higher. Once the sample becomes thick enough and thus virtually opaque to all infrared radiation, the heat flux will rise linearly (or the conductivity will be constant) with thickness. Figure 4 shows effective conductivities as a function of boundary emissivities (e_1 and e_2) and temperature for the cases of k_c = 0.019 W/m-K (unevacuated aerogel) and k_c = 0.005 W/m-K (evacuated aerogel) for a thickness of 10 mm. At higher temperatures (above room temperature), effective conductivities can vary greatly with boundary emissivity. For the case of an aerogel window, where the mean radiative temperature might vary between 275 and 300 K, boundary emissivities will not have a significant effect on effective conductivities. In such a window, with aerogel sandwiched between two layers of glass, the boundary emissivities will be those of the glass. The emissivity of uncoated glass is approximately 0.84; adding a good low-E coating could cause it to approach 0.05.

4. SOLAR-OPTICAL PROPERTIES AND WATER ABSORPTION

Solar transmission (0.25-3 μm) measurements were made in conjunction with the previously described infrared measurements. The effects of water absorption and aging were studied to determine the water content for the optimum performance of aerogel as a transparent insulating material. Figure 5 shows transmittance throughout the solar spectrum for a 20-mm-thick sample immediately after the production process and for the same sample heated for two days at 400 C. Much of the absorbed and bound water can be driven off by heating, improving the overall transmission. In this case, heating increased solar transmittance, τ_s, from 68% to 75% and visible transmittance, τ_v, from 56% to 65%. The fall in transmission below 0.7 μm is due to scattering. (As a result, aerogel appears slightly yellow when viewed against a bright background and shows a blue haze when viewed against a dark background due to backscattering (4).)

Aerogel is extremely hydroscopic because of its large surface area. When dried aerogel is exposed to the atmosphere, surface water absorption is extremely rapid, with an immediate decrease in transmission between 2.8 and 5 μm, and a slower progressive decrease in solar and visual performance caused by water absorption and the resulting structural changes that cause scattering. Figure 2 shows the effect of water absorption on the absorption coefficient and Figure 6 shows the effect on the τ_s (thickness=9.7mm). For about two weeks, the initial value of τ_s can be regained by heating and driving out the moisture. After this, structural changes are irreversible.

In the near-middle infrared (2.5 - 7 μm), however, absorbed water (which decreases radiative heat transfer) is easily displaced from aged aerogel when placed in a dry N_2 environment. Figure 7 shows infrared transmission curves for a 3-mm piece of aged aerogel before and after being placed in a dry nitrogen environment for thirty minutes. Much of the water is easily displaced by the dry gas. Evacuating an aerogel sample would produce similar results and thus one cannot reduce radiative heat transfer in evacuated aerogel by allowing it to absorb moisture.

5. AEROGEL WINDOWS

The thermal performance of windows has significantly improved over the last few years. Low-emissivity coatings and gap gases of low conductivity have greatly increased the thermal resistance between the panes (7). However, the insulating potential for combinations of multiple low-E layers and gas-filled gaps is limited by decreasing solar transmittances and increasing glazing thickness.

Heat transfer across the gap between two glazing layers may be greatly reduced by evacuating the

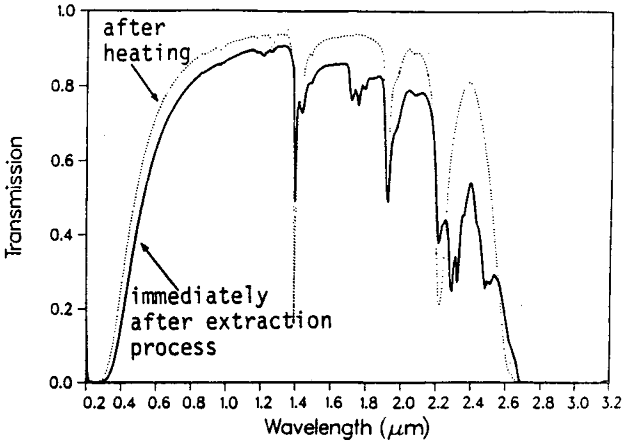

Fig. 5. Transmittance throughout the solar spectrum for a 20-mm-thick aerogel sample immediately after production and for the same sample heated at 400 C for two days to drive out absorbed water.

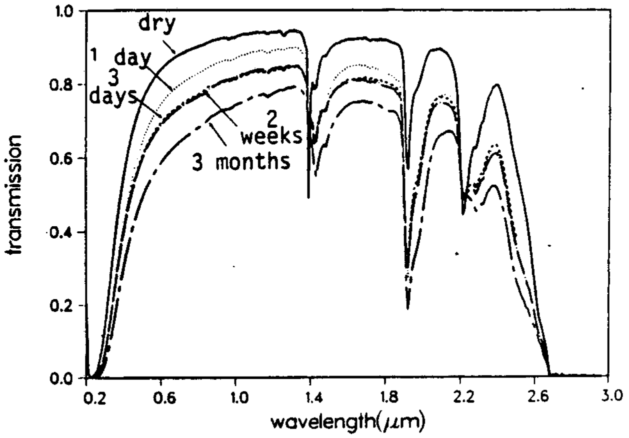

Fig. 6. Transmittance throughout the solar spectrum for a dry 9.7 mm thick aerogel sample and for the same samples aged in ambient conditions for one day, three days, two weeks, and three months.

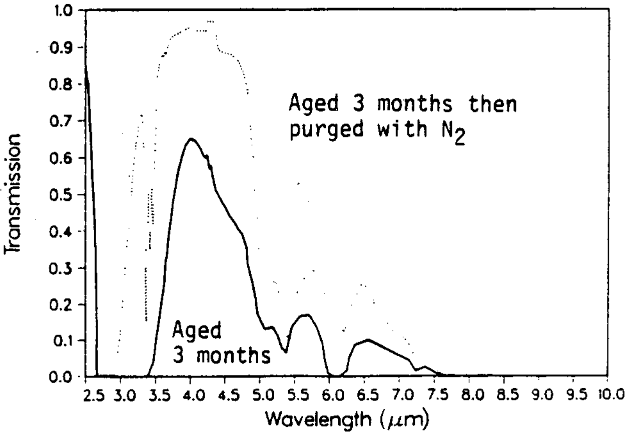

Fig. 7. Transmittance in the infrared spectrum between 2.5 and 10 μm for a 3-mm-thick aerogel sample aged three months and then for the same sample purged with dry N_2 for thirty minutes.

space or by filling it with a transparent insulating material such as aerogel. Measurements of the effective thermal conductivity of aerogel have shown it to be an excellent transparent insulator at atmospheric pressure; furthermore, decreasing the pressure to $\approx$0.1 atm greatly reduces conductive heat transfer. This is a relatively weak vacuum when compared to the pressures of $\approx 10^{-6}$ atm required for effective evacuated windows. Although weak in shear, aerogel is relatively strong in compression and can easily withstand such pressures. The practical difficulties of developing edge seals for maintaining a vacuum of 0.1 atm should be significantly fewer than for a vacuum of 10^{-6}atm.

The semi-empirical model developed as part of this study (5) has been incorporated into the recently released 1-D heat transfer program WINDOW 2.0 (8) in order to calculate heat transfer through aerogel windows. U-Values were calculated as a function of gap width for aerogel-filled and currently available window configurations under standard ASHRAE winter conditions. Figure 8 shows that using aerogel to fill the gap greatly improves the thermal performance of standard double-glazed windows. Further, evacuated aerogel windows show a four-fold decrease in U-Value (0.5 W/m^2–K) when compared to a state-of-the-art low-emissivity unit filled with argon with a gap width of 12.5 mm. For this aerogel window configuration, $\tau_s \approx 71\%$ and $\tau_v \approx 59\%$. Unlike gas-filled units, convection induced by larger gap widths does not limit the thermal performance of aerogel windows. However, the increased amount of scattering for gap thicknesses greater than a few centimeters would significantly reduce transmission and degrade the visible qualities of the window. Low-emissivity boundaries have little effect on U-values at standard ASHRAE winter conditions, as seen in Figure 9 ($T_m \approx$ 270 K). Because radiation heat transfer is more significant with evacuated aerogel, a slightly larger although still minimal effect is seen by adding low-E coatings. The effect (not shown) is more noticeable, though still small, under standard ASHRAE summer conditions ($T_m \approx$ 300 K).

6. CONCLUSIONS

The measurement of the absorption coefficient of aerogel in the range of 1-200 μm has shown that there is a transmission window between 3 and 6 μm, which can permit significant radiation heat transfer even at moderate temperatures. Heat transfer in aerogel can be calculated using a semi-empirical diffusion solution that was found to be within 10% of the more complex numerical calculations (5). Radiation heat transfer through aerogel is not a significant problem in the temperature range for insulating windows, but it does become a more critical factor at elevated (i.e., furnace) or greatly reduced (i.e., cryogenic) temperatures. Allowing aerogel to absorb water improves the thermal preformance only slightly (1-5%), while causing a larger decrease in the solar and visible transmission over time (10-20%). Aerogel's thermal performance can be improved by replacing the pore gas with one of lower conductivity or by evacuating the aerogel to pressures below $\approx$ 0.1 atm. Evacuated aerogel windows have a calculated U-value of $\approx$ 0.5 W/m–K for a gap spacing of 12.5 mm, which is four times better than currently available low-emissivity gas-filled units of similar size.

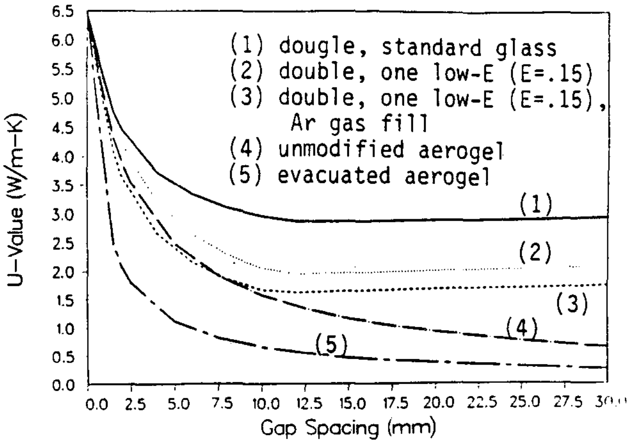

Fig. 8. U-value as a function of gap spacing for conventional and aerogel windows.

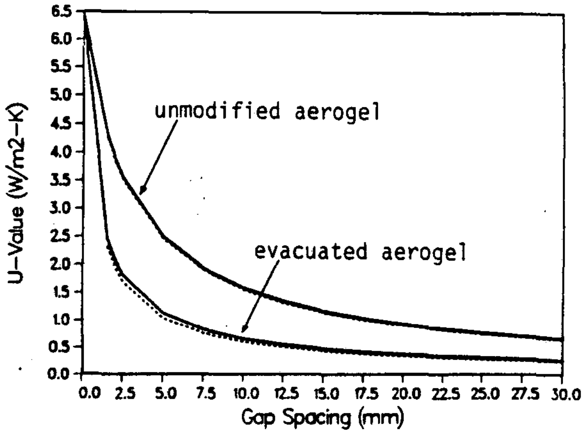

Fig. 9. U-value as a function of gap spacing for aerogel windows with (dashed lines) and without (solid lines) low-emittance boundaries.

7. ACKNOWLEDGEMENTS

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