THERMAL TRANSPORT IN MONOLITHIC SILICA AEROGEL

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Résumé : Des mesures sur le transport thermique dans des briques d'aérogels de silice (épaisseur voisine de 8 mm) ont été réalisées pour deux densités (ρ = 110 et 270 Kg.m^{-3}) et pour des émissivités ϵ = 0,04 et 0,77 dans l'air et sous vide. L'analyse est réalisée dans un intervalle de température compris entre 100 et 650°K. De plus pour une densité de 70 Kg.m^{-3} les pertes thermiques ont été déterminées pour deux épaisseurs (l'une étant le double de l'autre). Pour bien interpréter les résultats nous avons réalisé des mesures de transmission IR radiative en fonction de la longueur d'onde. L'évaluation des données suit les méthodes développées pour les matériaux optiques de faible épaisseur et transparents.

Abstract - Measurements on the thermal transport in SiO_2 aerogel tiles (thickness about 8 mm) were performed for two densities (ρ = 110 kg·m^{-3} and 270 kg·m^{-3}), for emissivities ε = 0.04 and 0.77, in air and evacuated. The covered temperature range was 100 K to 650 K. In addition for density ρ = 70 kg·m^{-3} the thermal losses were determined for two thicknesses (differing by a factor of two). For proper interpretation wavelength dependent IR radiative transmission measurements were performed. Data evaluation proceeded along the methods developed for optically thin, non-grey materials.

1 - INTRODUCTION

In highly porous aerogels heat is transferred via infrared (IR) radiation and solid conduction along the fragile SiO_2-skeleton. For non-evacuated specimens in addition gaseous thermal conduction occurs. As already Kistler /1/ knew, the latter contribution is considerably smaller than would be expected for a free non-convecting gas. In earlier papers /2,3/ we have pointed out that aerogels are optically thin materials in layers of 10 - 20 mm which are typical thicknesses for window applications. An important consequence is that the thermal conductivity depends on thickness and boundary emissivity, and thus is not a material property. In aerogels the IR radiative extinction is provided by absorption

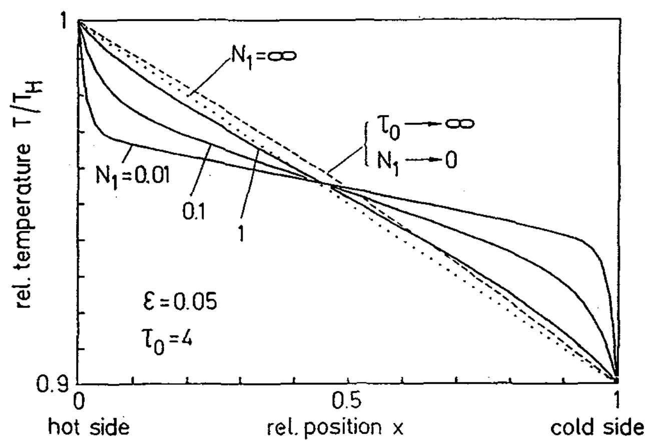

Fig.1. Temperature profiles for a grey medium with optical thickness τ_0 = 4 and boundary emissivity ε = 0.05 for different ratios N_1 of solid conduction to radiative heat transfer /4/.

(scattering only occurs in the visible spectral region). The absorption coefficient strongly depends on wavelength. Therefore an extremely complex interaction between solid conduction and IR heat transport occurs. In general under these conditions one expects a temperature gradient which is steep close to the low emissivity boundaries and rather flat within the bulk (fig.1).

2 - THEORETICAL REMARKS

In optically thin insulations the heat transfer for combined radiation and conduction is described by an integro-differential equation. This was performed recently by G. Döll /5/. It is very tedious to solve the exact equation numerically for a non-grey medium as aerogel, where the optical thickness $\tau_o(\Lambda)$ is a function of wavelength. Therefore the simplest way is to generalize an approximate three-flux solution for grey media to non-grey media /6/. Instead of the constant optical thickness τ_o one uses its spectral value $\tau_o(\Lambda)$ and integrates the heat flux over the thermal spectrum with the Rosseland distribution $\partial e_\Lambda(T)/\partial e_b(T)$ as a weight factor ($e_\Lambda(T)$ being the spectral and $e_b(T) = \sigma T^4$ the total blackbody emissive power).

3 - EXPERIMENTAL SET-UP

The caloric data were derived with the evacuable, load-controlled guarded hot plate system LOLA III (fig.2). A special cylindric guard (temperature controlled) allows to cover an unusually large temperature range (T = 100 to 900 K). The size of the specimens was 20 cm Ø. Infrared transmission between 2.5 and 45 µm was measured with a fast FTIR spectrometer. A typical IR transmission spectrum is shown in fig.3. Clearly visible is the large extinction above 10 µm and the very small extinction between 3 and 5 µm.

1 vacuum chamber
   a. upper part
   b. middle part
   c. lower part
2 hot plate with two guard-rings
3 upper cold plate
4 lower cold plate
5 tested specimen
6 heat sinks
7 cylindric guard (temperature controlled)
8 high-temperature insulation
9 ceramic support
10 piston
11 ground plate
12 flange for pumps
13 small flanges
14 sensor for thickness
15 fluid pipes

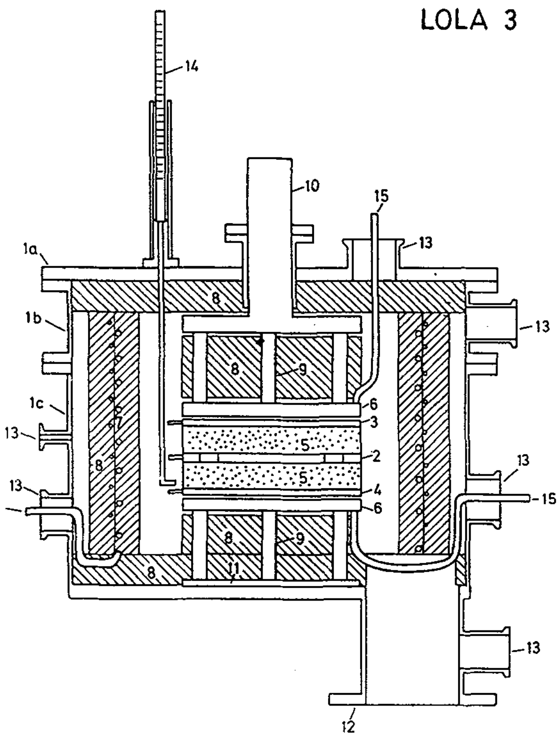

Fig.2. Guarded hot plate apparatus LOLA III, which can be evacuated and externally pressure loaded. The metering section is 12 cm Ø.

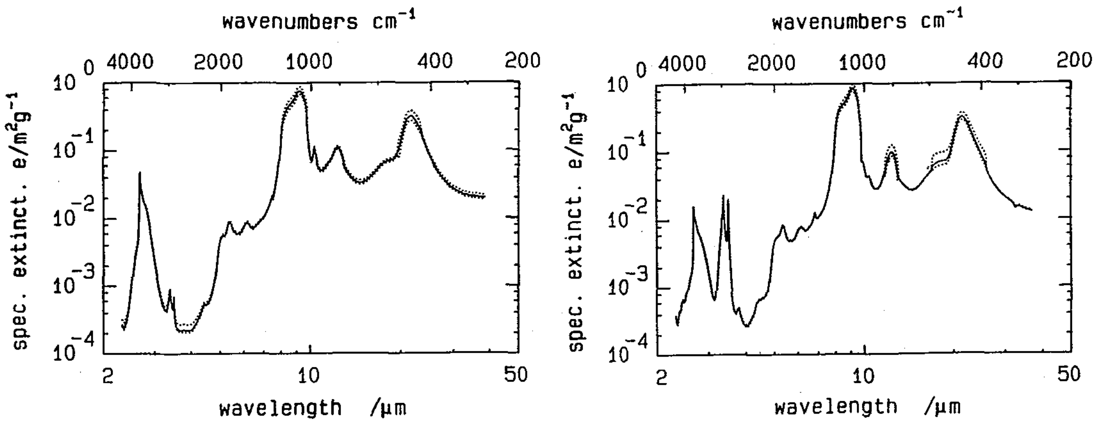

Fig.3. Specific extinction coefficient e versus wavelength
a) for hydrophilic, b) for hydrophobic aerogel.

4 - RESULTS AND DISCUSSION

From the temperature difference $\Delta T = T_h - T_c \approx 30$ K across the specimen and the power input per unit area P/A the thermal transfer coefficient Λ can be derived according to $P/A = 2\cdot\Lambda\cdot\Delta T$. The factor 2 takes care of the double-sided measurements. In all plots shown Λ is depicted versus third power of mean temperature for radiation T_r.
From fig.4a it is evident that optically thin specimens are investigated: A doubling of thickness d only reduces Λ marginally, while with optically thick insulations a reduction of Λ by a factor of 2 would be expected. This holds down to temperatures of about 150 K (fig.4b), where the measured 14.2 mm-curve is about one half of the 7.4 mm curve. The extraordinary large influence of surface emissivity ε on Λ (see fig.5) is another proof for the small optical thickness of the investigated aerogel tiles in the IR. This effect again shows down to about 150 K. One should point out, that above T_r = 350 K the differences in Λ for low and high emissivities are clearly visible even for non-evacuated specimens.

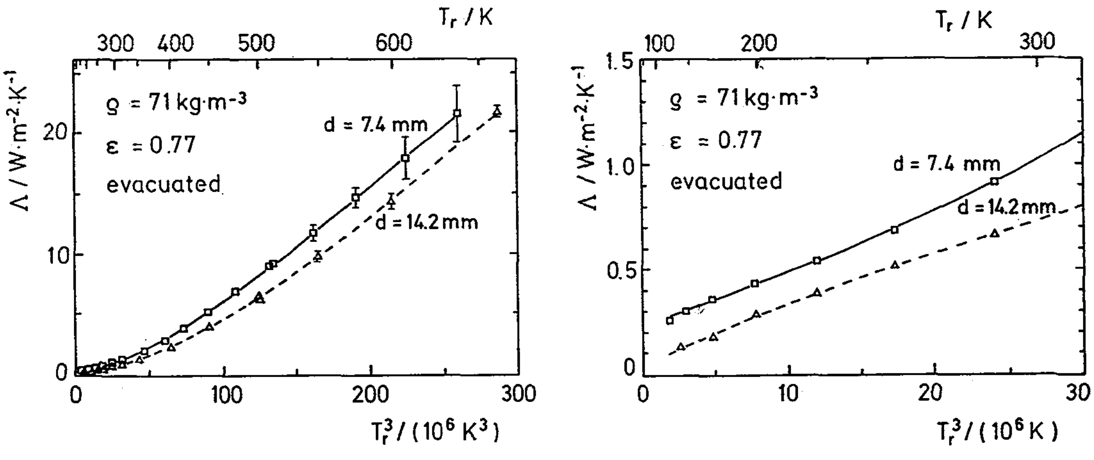

Fig.4. Thermal transfer coefficient Λ versus T_r^3 for aerogel tiles (ρ = 70kg·m^{-3}) with thicknesses d = 7.4 (——) and 14.2 mm (---); a) temperature range 100 K - 670 K, b) temperature range 100 K - 300 K.

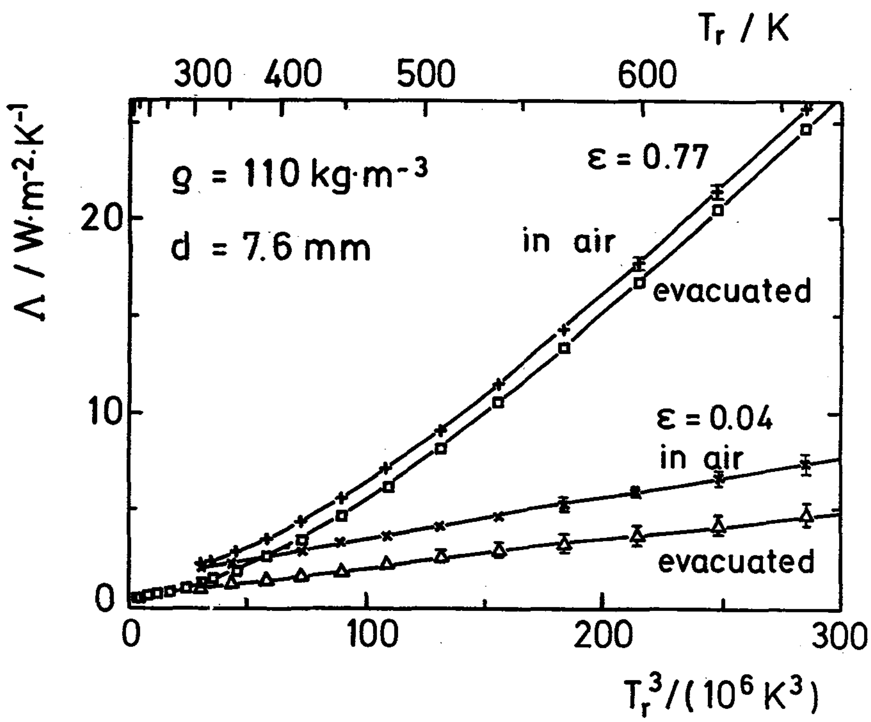

Fig.5. Thermal transfer coefficient Λ versus T_r^3 for an aerogel tile with ρ = 110 kg·m^{-3}, d = 7.6 mm; parameter is boundary emissivity ε; data for evacuated and non-evacuated specimens are shown.

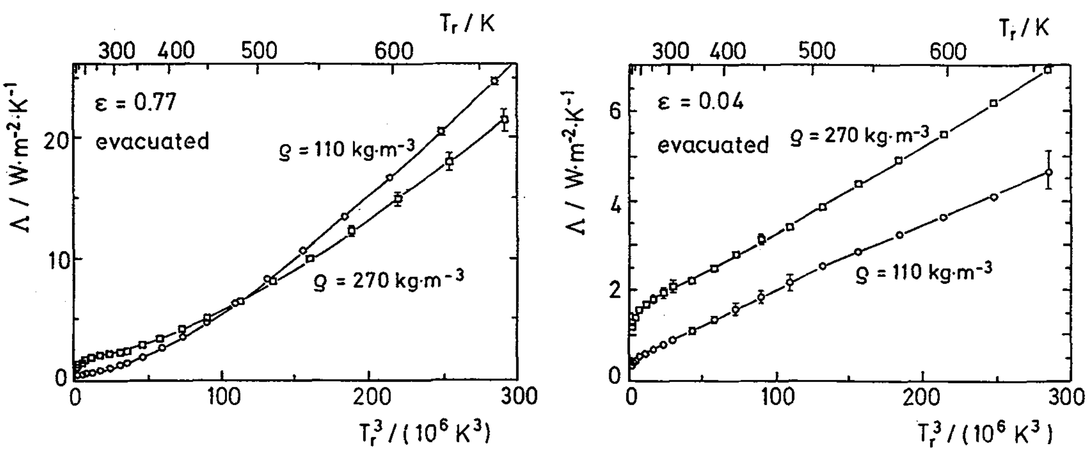

Fig.6. Thermal transfer coefficient versus T_r^3 for two tiles with different densities. a) With ε = 0.77 the expected curve crossing is clearly to be seen, b) with ε = 0.04 non-linear coupling between radiation and conduction masks this effect.

Another important quantity influencing the thermal transport is the aerogel density. It is plausible that at low temperatures, where solid conduction gives the major contribution, Λ is larger for higher densities. With rising temperature more and more IR radiation is emitted. For high emissivitiy boundaries heat transfer then is governed by the aerogel IR extinction properties, leading to lower Λ-values for higher densities (see fig.6a). For lower emissivities the situation is not as clear: Though an increasing density increases the IR extinction, non-linear coupling effects close to the low emissivity boundaries mask this effect, possibly leading to higher Λ-values for higher densities (fig.6b).

In order to study the influence of the gas (N_2) on the thermal transport, the transfer coefficients with air Λ_{total} and for evacuated specimens Λ_{evac} are subtracted from each other. In a naiv consideration one would expect this difference to be determined by the free N_2 conductivity (λ_N(300 K) $\approx$ 0.026 $W \cdot m^{-1} \cdot K^{-1}$), corresponding to a value $\Lambda_{gas} \approx 3.5\ W \cdot m^{-2} \cdot K^{-1}$ for d = 7.6 mm. Firstly we recognize from fig.7 that we get $\Lambda_{total} - \Lambda_{evac} \approx 1\ W \cdot m^{-2} \cdot K^{-1}$ for $\varepsilon = 0.77$ over the whole temperature range investigated. This small contribution can be explained by the microporous aerogel structure, in which gas conduction is only partially developed even at ambient pressure /7/. For $\varepsilon = 0.04$ the difference $\Lambda_{total} - \Lambda_{evak}$ is larger and shows a strong temperature dependence. The reason again is the non-linear coupling between radiative transport and conduction (caused by the skeleton and the gas) within the radiative boundary layer in front of the low-ε-surfaces.

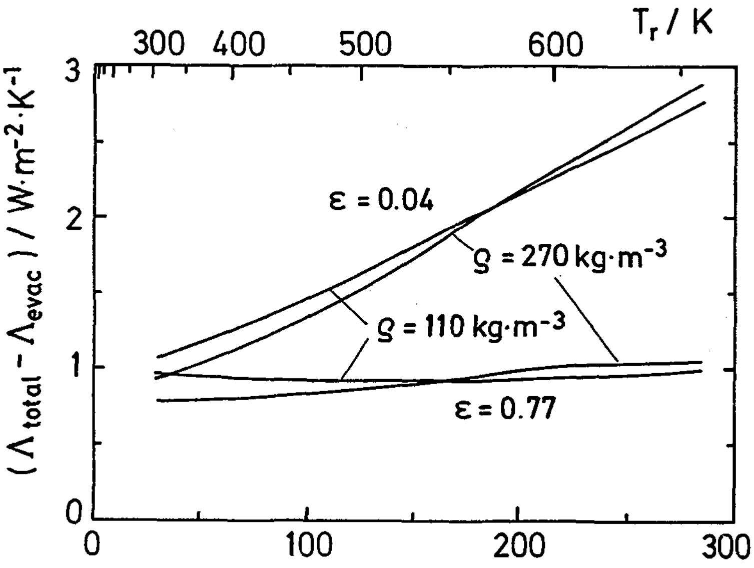

Fig.7. Difference of thermal transfer coefficients for non-evacuated and evacuated tiles versus T_r^3. The dramatic increase in $\Lambda_{total} - \Lambda_{evac}$ with temperature for low emissivity boundaries is caused by the non-linear coupling between radiation and conduction.

In order to evaluate the solid conductivity in aerogels as a function of density we tried to extend the measurements to as low temperatures as possible (fig.8). Below 150 K the influence of ε on Λ vanishes, indicating that the specimens become optically thick. If we neglect the radiative contributions we can derive upper values for the solid conductivities λ_{sc} for 150 K: For the 70 and 110 $kg \cdot m^{-3}$ tiles a value $\lambda_{sc} \approx 0.003\ W \cdot m^{-1} \cdot K^{-1}$ and for the 270 $kg \cdot m^{-3}$ tile $\lambda_{sc} \approx 0.01\ W \cdot m^{-1} \cdot K^{-1}$ is estimated. These values have to be compared with $\lambda_{sc} = 1\ W \cdot m^{-1} \cdot K^{-1}$ for fully dense silica glass. These large discrepancies can be explained in terms of the density ratios (2200:70 and 2200:270) and the sound velocity ratios (5000:120 and 5000:340).

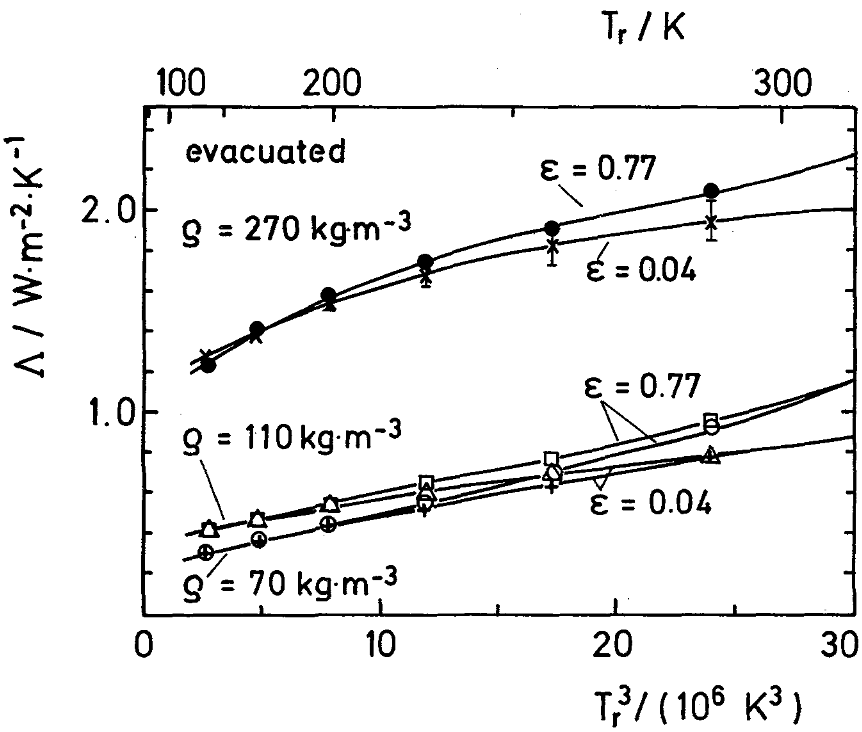

Fig.8. Low temperature behaviour of thermal transfer coefficient versus T_r^{3} for 3 densities and 2 emissivities. At the lowest temperatures achieved, the dominant component ought to be solid conduction.

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