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A thermal conductivity study of double-pore distributed powdered silica aerogels



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ABSTRACT

Powdered nanoporous silica aerogel is a typical double-pore distributed material. This paper develops a total effective thermal conductivity model that considers the coupling effect between different forms of heat transfer for the microstructure features of powdered silica aerogel by using an ideal structure with nanoporous spheres arranged in a spatially periodic structure. Performance of the developed model is compared to experimental results, with the effects of various parameters on thermal conductivity analyzed. The results show that the thermal conductivity of powdered silica aerogel decreased with decreasing gas pressure, and stabilized at its lowest value at pressures below 20 Pa. A turning point existed at approximately 10³ Pa when looking at the logarithmic scale of gas pressure. The thermal conductivity decreased quite slowly with decreasing pressure when $p > 10^3$ Pa, but more quickly when gas pressure was lower than 10³ Pa. The thermal conductivity reduced with increased specific surface area, but did not change with powder diameter D when the pressure was higher than 10^3 Pa. On the other hand, the thermal conductivity increased with increasing powder diameter D, but did not change with specific surface area when the pressure was less than 10^3 Pa. As the powder diameter D decreased, the lowest stable point of the thermal conductivity increased. The radiation heat transfer contribution was very small when the temperature was less than 400 K. With elevating temperature, the thermal conductivity of powdered silica aerogel distinctly increased. Thus, silica aerogel samples with large macro pores achieve greater thermal conductivity at elevated temperatures.

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1. Introduction

Aerogel is a super insulating material made through a sol-gel chemical process and drying (supercritical or conventional drying). Aerogels possess excellent insulation performance, with a thermal conductivity lower than that of still air at ambient temperature due to its nanoporous structure and very high porosity. There is considerable application potential for aerogel in aeronautics and astronautics, energy savings in buildings, and transportation of off-shore oil and gas [1–6]. Aerogel is also an ideal potential core material for vacuum insulation panels because it can achieve vacuum insulation at medium vacuum conditions [7]. There are primarily three structured forms of aerogel: monolithic, powdered, and granular (i.e., a broken style of monolithic aerogel). The major disadvantage of monolithic aerogel for use as an insulating material is that it is brittle and easy to break up. Instead, powdered aerogels have been commercialized for their flexible arrangement in real

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http://dx.doi.org/10.1016/j.ijheatmasstransfer.2016.12.062 0017-9310/© 2016 Elsevier Ltd. All rights reserved. applications and lower manufacturing cost, despite their thermal conductivity being higher than monolithic aerogels due to micrometer pores between the particles.

The heat transfer mechanism and associated prediction of thermal conductivity by theoretical methods has been widely studied in recent years for monolithic aerogels [6,8-19]. Zeng [8] presented three regular structures for open-cell aerogel super insulators: intersecting square rods, intersecting cylindrical rods, and a cubic array of nano-spherical structures. By using a cubic array of nano-spherical structures, Wei et al. [6] deeply analyzed the thermal conductivity of monolithic aerogels and xonotlite-aerogel composite insulation materials. Lu et al. [10] analyzed the thermal conductivity of aerogel-based composite materials by using a similar cubic array of nano-spherical structures. Xie et al. [11] developed a fractal-intersecting sphere model for nanoporous silica aerogels. Spagnol et al. [20,21] used a periodic two-dimensional fractal Von Koch snowflake structure and a diffusion-limited cluster-cluster aggregation (DLCA) model to generate a twodimensional random structure for representing the real structure of an aerogel, then used numerical calculations to get the effective

Nomenclature			
а	contact diameter between two nano-particles	Т	temperature
a_1	ratio of diameters d/D_1	t	time
a_2	ratio of diameters a/d	V	volume
D	diameter of the aerogel particle	у	coordinate
D_1	diameter of the nanoparticle	α	Stefan-Boltzmann constant; ratio of thermal conductiv-
D_m	characteristic size		ity 1
d	diameter	α_1	energy accommodation coefficient
d_g	diameter of the gas molecules	β	specific ratio
e_b	blackbody emissive power	β_1	ratio of thermal conductivity 2
$e_{b\lambda}$	spectral blackbody emissive power	ρ	density
k_B	Boltzmann constant	ϕ	porosity
$K_{e,m}$	specific Rossland mean extinction coefficient	γ	adiabatic index of gases
$K_{\lambda,m}$	spectral extinction coefficient	λ	wavelength
Kn	Knudsen number		-
k	thermal conductivity	Subscripts	
l_0	mean free path	c	conductive
п	number	g	gaseous
Р	pressure	por	porous
P_0	power	r	radiative
Pr	Prandtl number	S	solid
Q	energy	t	total
S	specific surface area	λ	wavelength
S	contact diameter between two particles		

thermal conductivity at different densities. Using the DLCA method, Zhao et al. [17] generated a three-dimensional random particle accumulation structure to study the coupled radiativeconduction effective thermal conductivity. Bi et al. [13] used four kinds of three-dimensional regular structures and a numerical method to get the effective thermal conductivity of aerogels. However, a literature review reveals a lack of an appropriate prediction model for the thermal conductivity of powdered aerogels. The main difficulty in constructing a theoretical model for powdered aerogel is that both nanometer and micrometer pores coexist in the material. This means the gas conduction mechanism between these two dimensional pore sizes is different, and so the model must consider the disparate heat transfer mechanisms simultaneously. Even so, previous studies have proven that it is an effective and useful method for determining the thermal conductivity of porous material by model prediction.

This work develops a double-pore distributed effective thermal conductivity model based on the work of Zeng et al. [8] and Wei et al. [6] on monolithic aerogels that considers the microstructure characteristic of powdered aerogels, and which is then validated by experimental results. Using this model, we can comprehensively analyze the effects of corresponding parameters on the effective thermal conductivity of powdered silica aerogels.

2. Theoretical models

2.1. Unit cell structures and gas-solid coupled conductivity

There are two categories of pores in powdered silica aerogel insulation materials: open-cell nanometer pores in the particle and micrometer pores between the particles. For simplification, we can envision a powdered silica aerogel material as a periodic array of spheres in a spatial structure with a diameter of D, as shown in Fig. 1(a), such that a contact surface exists between two particles with a surface diameter of s. Fig. 1(b) gives the unit cell of this simplified structure. A silica aerogel particle is considered an aggregation of a cubic array of nanospheres, as shown in

Fig. 1(c). The cubic array of nanosphere structures in Fig. 1(c) has been successfully adopted to depict the gas-solid coupled thermal conductivity of monolithic silica aerogels [6,10]. Fig. 1(d) gives the contact pattern between two adjacent nanospheres. In the figure, d and D_1 are the structural parameters of the unit cell, a is the contact diameter between two nanospheres, and n is the nanosphere number in each column.

By considering the one-dimensional conduction from the bottom to the top surface of the unit cell in Fig. 1(c), the gas-solid coupled effective thermal conductivity was derived for monolithic silica aerogel in our previous study [6] as

$$\begin{aligned} k_{\rm cq} &= \left\{ \frac{\pi a_1^2 a_2^2}{4(1-\beta_1)} + \left(1-a_1^2\right) - \frac{\pi a_1^2 \left(1-a_2^2\right)}{2\beta_1^2} \left[\beta_1 + \ln\left(1-\beta_1\right)\right] \right. \\ &+ \frac{\pi}{\beta_1} \left(1/\sqrt{1-a_2^2} - a_1\right) \left[\frac{1}{\beta_1 a_1} \ln\frac{1-\beta_1 a_1 a_2}{1-\beta_1 a_1} - \left(1-a_2\right)\right] \right\} \cdot k_{\rm g1} \end{aligned}$$

where $a_1 = d/D_1$, $a_2 = a/d$, and $\beta_1 = 1 - k_{g_1}/k_{s_1}$ where k_{s_1} is the solid thermal conductivity in the nanospheres and k_{g_1} is the gaseous thermal conductivity in the nanoporous silica aerogel. If air is considered, k_{g_1} is calculated as [22]

$$k_{g1} = \frac{60.22pT^{-0.5}}{0.25S_s\rho_{\rm por}/\phi_1 + 4.01 \times 10^4 pT^{-1}}$$
(2)

Here, $\rho_{\rm por}$ is the density of a single silica aerogel particle such that $\rho_{\rm por} = \rho/(1-\phi)$, ρ is the accumulation density of the material, ϕ is the porosity of the micrometer pores between the particles, S_s is the measured specific surface area of the aerogel, and ϕ_1 is the porosity of a single particle. Note that ϕ_1 is also the percentage of nano-pore volume in a particle, calculated as $\phi_1 = 1 - \rho_{\rm por}/\rho_{\rm bulk}$ (where $\rho_{\rm bulk} = 2200 \text{ kg/m}^3$ is the density of bulk silica). Based on Fig. 1(c), the structural parameters *d* and D_1 in Fig. 1(d) can thus be determined as [6,23]

$$d = \frac{12(1-\phi_1)}{(2+a_2^2)\rho_{\rm por}S_{\rm s}}$$
(3)

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