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Modeling of the apparent solid thermal conductivity of aerogel

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ABSTRACT

This paper provides an insight into the heat transfer in the solid backbone of aerogel and an effective approach to model the apparent solid thermal conductivity of aerogels. First, a model for the thermal conductivity of aerogel solid backbone is developed based on thermal constriction resistance between interconnected nano-particles by taking into account the size effect of a single particle, and validated by the numerical solutions of the gray Boltzmann transport equation (BTE). Then, combined with the analytical expression derived based on the Laplace heat conduction equation, the proposed model is used to predict the apparent solid thermal conductivity of aerogels, and the predictions are in good agreement with the available experimental data of different kinds of aerogels.

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1. Introduction

Aerogel renowned for its low density, high porosity and high specific surface area, has been particularly focused on in industry and research recently [1–3]. It has a complex structure with a three-dimensional network skeleton consisting of interconnected spherical nanoparticles and micro- or meso-scale pores (Fig. 1(a)). Such a special structure results in the extremely low thermal conductivity of aerogel because of the very large particle-to-particle thermal resistance between two connected nanoparticles and the reduction of gaseous thermal conductivity caused by the restriction of molecular motion in these nano-pores, which makes aerogel an excellent insulating material.

There are different heat transfer modes which contribute to the total thermal conductivity of aerogel, including heat conduction via solid, heat conduction through gas phase and thermal radiation. Even though the radiation heat transfer in high temperature and the heat conduction through gas phase in aerogel are important [4–7], the heat conduction through the solid backbone is worth investigating [8–10]. For aerogel, the apparent solid thermal conductivity can be experimentally determined under vacuum condition and at low temperature by neglecting the radiation heat transfer. However, an accurate theoretical prediction was not publicly reported so far due to the nanometric dimensions and the complexity of the solid backbone structure.

The most commonly used model for predicting the apparent solid thermal conductivity of aerogel was derived by way of an analogy conclusion measuring the sound velocity of the porous material [8,10]

$$\lambda_s = \lambda_0 \frac{\rho}{\rho_0} \frac{v}{v_0} \tag{1}$$

where ρ is the density of aerogel, ρ_0 is the density of solid backbone, v is the sound velocity in aerogel, v_0 is the sound velocity in solid backbone and λ_0 is the thermal conductivity of solid backbone which may be much smaller than that of bulk material λ_{bulk} due to the size effect and phonon filtering between contacting particles [10]. It is worth noting that the product of the density and the sound velocity is the acoustic impedance, and this product for aerogel can be affected by the trapped gas in pores and the microstructures [8]. So, the value of ρv is difficult to be determined analytically.

On the other hand, solutions for the apparent solid thermal conductivity of porous materials and packed bed have been developed [11–14]. Among these models, the analytical model derived from the Laplace heat conduction equation by Bauer [12] has shown its validity in a wide range of porosities with randomly distributed pores. This model is effective especially for relatively high porosity materials and can be expressed as

$$\lambda_s = \lambda_0 \left(\frac{\rho}{\rho_0}\right)^{3\varepsilon/2} \tag{2}$$

where ε is a shape factor of a randomly oriented pore, which is semi-empirical constant. According to Xu [15] and Yang [16], for the 3-D porous materials with complex pore structures, Eq. (2) with $\varepsilon = 1$ could fit the experimental data and numerical results well. Furthermore, experiments showed that, for aerogels, the

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Nomenclature

a	contact radius, m
e	phonon energy density, J/(m^3 Sr)
f	distribution function of phonons
l	mean free path, m
q	heat flux, W/m^2
v	the sound velocity, m/s
v_g	phonon group velocity, m/s
A	area, m^2
C	specific heat
D	particle diameter, m
D_p	the phonon density of state
R	thermal resistance, K/W
T	temperature, K
γ	dimensionless coefficient
ε	shape factor of pores
λ	thermal conductivity, W/m K
ρ	density kg/m^3
σ	electrical conductivity, Ω^{-1} cm^{-1}
ω	angular frequency (m^{-1})

Subscripts

0	solid backbone
c	constriction
g	group, gas phase
p	particle
p-p	particle-particle
s	apparent solid
t	total

relationship between the apparent solid thermal conductivity and density can be expressed as $\lambda_s \propto \rho^{\alpha}$ where $\alpha \approx 1.5$ [17], which is consistent with $\varepsilon = 1$ in Eq. (2). Compared with Eq. (1), Eq. (2) is more convenient to calculate λ_s without measuring the sound velocity in aerogel, so we will employ Eq. (2) to predict the apparent solid thermal conductivity of aerogel in this work. However, determining the value of λ_0 in Eq. (2) is a challengeable task due to the size effect of aerogel particles and the phonon filtering between two contacting aerogel particles, and quite few studies have been performed on modeling λ_0. Wei et al. [18] calculated the thermal conductivity of aerogel grains based on the kinetic theory by modifying the phonon mean free path. Bi et al. [10] developed a modified model to predict the thermal conductivity of aerogel solid backbone based on the super-lattice nanowire model. However, these models were developed with some uncertain assumptions and were not verified.

Thus, the aim of this paper is to establish a heat transfer model for predicting the thermal conductivity of aerogel solid backbone (λ_0) theoretically and numerically, and then obtain the apparent

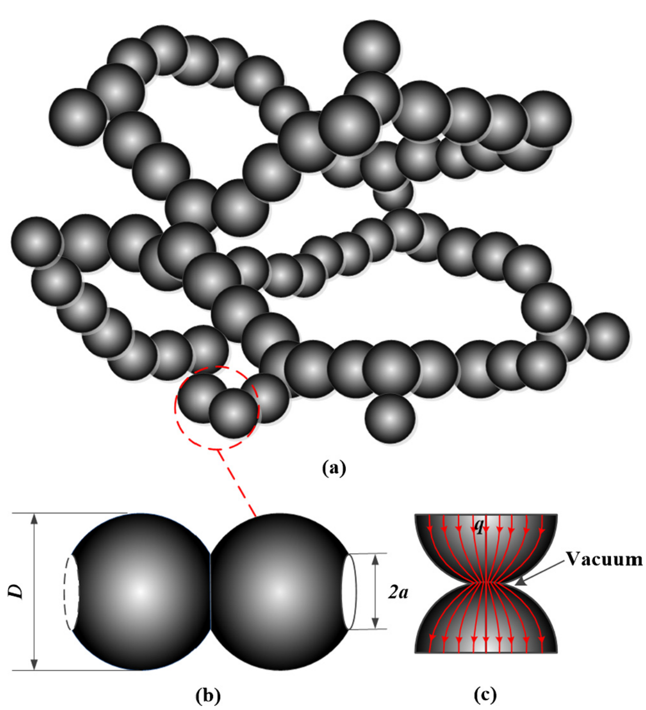

Fig. 1. Schematic of aerogel and the heat transfer unit.

solid thermal conductivity of aerogel (λ_s). We begin this work by developing a model to predict the effective thermal conductivity of two touching nanospheres (λ_{p-p}). Next, a numerical study is conducted to validate the developed λ_{p-p} model by solving the gray relaxation-time approximation Boltzmann transport equation. Finally, based on the present thermal conductivity model of solid backbone and Eq. (2), the apparent solid thermal conductivity of aerogel is obtained and compared with the available experimental data for different kinds of aerogels.

2. Heat transfer model

As mentioned in the introduction, the solid backbone of aerogel consists of random interconnected spherical nanoparticles (Fig. 1(a)). For simplicity, the particles in aerogel are also assumed to be spherical with homogeneous diameter denoted by D and the diameter of contact area between two contacting particles denoted by $2a$ [10,18–20]. For aerogel under ultimate vacuum, the region of two contacting hemispheres separated by vacuum is the basic heat transfer cell (Fig. 1(c)) in which heat is transferred by conduction through the contact area. The effective particle-particle thermal conductivity (λ_{p-p}) can be approximated as the thermal conductivity of solid backbone (λ_0).

On the basis of one dimensional steady-state heat transfer, we can obtain the effective particle-particle thermal conductivity as

$$\lambda_{p-p} = \frac{2\sqrt{r^2 - a^2}}{R_t \pi r^2} \tag{3}$$

where r is the radius of the nanosphere, R_t is the overall thermal resistance consisting of the thermal resistance of two hemispheres and constriction. The expressions of thermal constriction resistance have been developed for a circular contact between two infinite reservoirs as shown in Fig. 2. When the contact radius a is much larger than the mean free path of phonon, the heat transfer is in diffusive limit and the thermal constriction resistance in the diffusive limit regime is given as [21,22]

$$R_{cd} = 1/(2\lambda_{bulk} a) \tag{4}$$

where λ_{bulk} is the bulk thermal conductivity. If the contact radius a is much smaller than the mean free path of phonon, the heat transfer is in ballistic limit and the thermal constriction resistance can be expressed as [22,23]

$$R_{cb} = 4l/(3\lambda_{bulk} A) \tag{5}$$

where l is the phonon mean free path of the bulk and A is the contact area defined as $A = \pi a^2$.

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