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Effective thermal conductivity of the solid backbone of aerogel

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ABSTRACT

Based on the superlattice nanowire model, a modified model to predict the effective thermal conductivity of aerogel solid backbone is presented. We study both the size effect and interfacial resistance effect on the thermal conductivity of interconnected particles of the aerogel backbone. The effective thermal conductivity of the aerogel backbone is significantly lower than that of the pure nanowire and the single particle due to the interfacial thermal resistance. The solid thermal conductivity of the aerogel calculated by the present model is in good agreement with that by the minimum thermal conductivity model, and also agrees well with available experimental data when the aerogel density is above 100 kg/m^3.

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1. Introduction

Aerogel is a typical nano-porous material and its backbone structure represents a three-dimensional network consisting of interconnected spherical nano-particles (see Fig. 1(a)) [1,2]. This kind of structure can reduce the heat transfer via the gas filled in the pores and via the solid backbone, which results in excellent thermal insulation performance [3–6]. Studies have shown that the heat transfer via the solid backbone, which is called solid thermal conductivity, has great effect on the aerogel thermal insulation [7,8]. Due to the amorphous structure of the aerogel backbone, the heat transfer via the solid phase depends on the localized atomic vibrations and is limited by the scattering of the atoms at a distance of the inter-atomic spacing, and thus the solid thermal conductivity can be described by the theory of minimum thermal conductivity [9]:

$$\lambda_{\min} = (\pi/6)^{1/3} k_B n^{2/3} \sum_i v_i (T/\Theta_i)^2 \int_0^{\Theta_i/T} x^3 e^x/(e^x-1)^2 dx \quad (1)$$

where, k_B is the Boltzmann's constant, n the number density of atoms, i the summation index of the three sound modes (two transverse and one longitudinal) with speed of sound v_i, T the temperature, and Θ_i is the cut off frequency for each polarization expressed in Kelvin, given by: $\Theta_i = v_i(\hbar/k_B)(6\pi^2 n)^{1/3}$, where $\hbar$ is the reduced Planck's constant. Once the parameters of n, T, and v_i are given, the thermal conductivity can be obtained. In the aerogels, the pores will cause a reduction in the sound velocity compared with that in the bulk material, and Hopkins et al. [10] point that the sound velocity in Eq. (1) should be scaled by the atomic density:

$$v/v_{\text{bulk}} = (n/n_{\text{bulk}})^c \quad (2)$$

where, v is the sound velocity in aerogel, v_{bulk} the sound velocity in bulk material, and c the correction factor. For silica aerogel, we have $c = 1.4$ [10].

On the other hand, the solid thermal conductivity λ_s can also be derived by measuring the sound velocity in the aerogel [11]:

$$\lambda_s = \lambda_0 \frac{\rho}{\rho_0} \frac{v}{v_0} \quad (3)$$

where ρ is the density of aerogel, ρ_0 the density of solid backbone, v the sound velocity in aerogel, v_0 the sound velocity in solid backbone, and λ_0 the thermal conductivity of solid backbone. Compared with Eq. (1), Eq. (3) is more convenient to calculate λ_s. However, the thermal conductivity of the aerogel backbone λ_0 is not easy to measure, and we often replace λ_0 with the bulk thermal conductivity λ_{bulk} to simplify the calculation of the solid thermal conductivity λ_s, which will lead to larger errors. To obtain accurate λ_0, we can use the kinetic theory:

$$\lambda_0 = C_V v_0 \Lambda_0/3 \quad (4)$$

where C_V is the volume specific heat, v_0 is the mean sound velocity of the solid backbone and Λ_0 is the average inter-atomic spacing in solid backbone.

In most cases, Eq. (4) can directly calculate the thermal conductivity of the bulk material, thin film (even for metallic film [12]), and single particle. But the solid particles of the aerogel backbone are interconnected, and their contact diameters (assuming spherical particles) are less than the particle size, which leads to an interfacial resistance when the heat transfers through the interconnected

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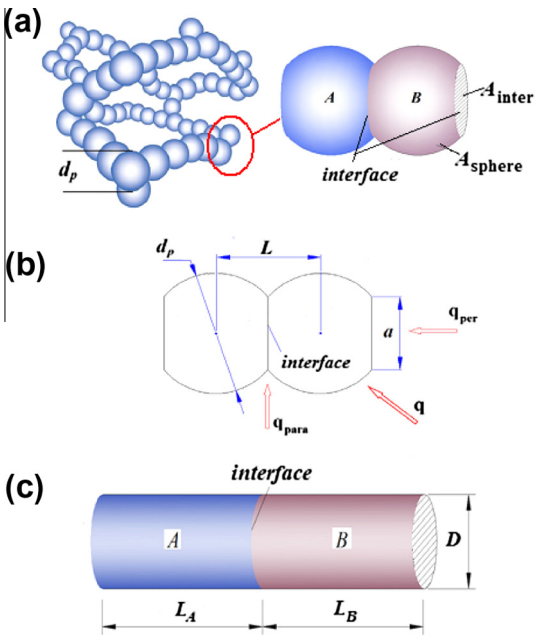

Fig. 1. Schematic of aerogel and supperlattice nanowire. (a) Aerogel backbone and interface between aerogel particles. (b) Definition of geometry size of aerogel backbone. (c) Interface between two segments of superlattice nanowire.

particles [13–15]. Therefore, the effective thermal conductivity λ_0 of the aerogel backbone must be different from that of the single particle. The purpose of this study is to establish a model with the consideration of the size effect and interfacial resistance effect of the interconnected particles, and then obtain the thermal conductivity of the aerogel backbone for further predicting the solid thermal conductivity of aerogels.

2. Theoretical model

According to Dames and Chen [16], the phonon thermal resistance in crystalline solids can be divided into three parts: (1) volumetric resistance R_V due to mass difference scattering, Umklapp scattering, resonant scattering, and other phonon scattering mechanisms according to solid state physics, (2) transmission resistance R_T due to the interfacial resistance, and (3) space resistance R_S due to the boundary scattering on the side walls. And then the phonon mean free path in the crystal solid can be obtained in the form of Matthiessen's rule:

$$1/\Lambda_0 = 1/\Lambda_V + 1/\Lambda_S + 1/\Lambda_T \tag{5}$$

where the mean free path Λ_V, Λ_S, and Λ_T are indirectly proportional to the corresponding thermal resistances R_V, R_S, and R_T, respectively. Although the concept of phonons fails in amorphous solid, we can still borrow the idea in [16] to divide the thermal resistance of the amorphous solid into three parts similar to that of the crystalline solid. But the differences are that (1) R_V in amorphous solid results from the localized atomic vibrations and inter-atomic scattering and (2) R_S is due to the atomic scattering on the boundaries of the solid. Then Eq. (5) can also be used to solve the average inter-atomic spacing in the aerogel backbone, but the symbols Λ_V, Λ_S and Λ_T present the average inter-atomic spacing corresponding to the volumetric resistance, transmission resistance and space resistance of the amorphous solid, respectively.

For the volumetric resistance, we can calculate Λ_V from Eq. (4) with the parameter of the bulk material by

$$\Lambda_V = \Lambda_{bulk} = 3\lambda_{bulk}/(C_V v_{bulk}) \tag{6}$$

For cylindrical nanowires, the computational model of Λ_S is given by [17]

$$\Lambda_S \approx D(1+p)/(1-p) \tag{7}$$

where p is the boundary specularity, and D is the diameter of the nanowire. For completely diffused boundary with $p = 0$, we have:

$$\Lambda_S \approx D \tag{8}$$

However, for the aerogel, see Fig. 1(a), the backbone composed of the interconnected particles does not present a cylindrical shape as the nanowire, so that Λ_S of the aerogel backbone should be re-approximated. For this purpose, Liu [18] introduced Zeng's unit cell model [19], and assumed that the heat flux is perpendicular or parallel to the interfaces of the particles in such a cubic structure, and then considered two cases: (1) when the heat flux, like $\mathbf{q}_{per}$ in Fig. 1(b), is perpendicular to the interface, $\Lambda_S \approx a$, in which a is the contact diameter of interconnected particles. Because only the atoms scattering on the interface has the chance to transfer heat through the interconnected particles, and the contact diameter will be the maximum distance in which an atom scattering on the interface can move; and (2) when the heat flux, like $\mathbf{q}_{para}$ in Fig. 1(b), is parallel to the interface, $\Lambda_S \approx d_p$, in which d_p is the particle diameter of the backbone. However, due to the complex structure of the actual aerogel backbone as schematic in Fig. 1(a), the heat flux, like $\mathbf{q}$ in Fig. 1(b), usually is not exactly perpendicular or parallel to the interfaces of the particles. Therefore, the above model is no longer suitable. We believe that the Λ_S of the aerogel backbone depends on both the contact diameter a and particle diameter d_p, and we assume that the Λ_S also can be obtained in the form of Matthiessen's rule:

$$\frac{1}{\Lambda_S} = \frac{1}{a}\frac{2A_{inter}}{A_{eff}} + \frac{1}{d_p}\frac{A_{sphere}}{A_{eff}} \tag{9}$$

where A_{inter} and A_{sphere} are the areas of the interface and spherical cap, respectively. A_{eff} is the effective atomic scattering area of a single particle of the aerogel backbone, i.e. $A_{eff} = A_{sphere} + 2A_{inter}$, see Fig. 1(a). Eq. (9) denotes that the Λ_S of the aerogel backbone can be calculated from the contact diameter a and particle diameter d_p by the weighted average in accordance with the atomic scattering area of the boundaries.

For the transmission resistance in the cylindrical supperlattice nanowire, the corresponding Λ_T is [16]:

$$\Lambda_T = 3t_{AB}L/2 \tag{10}$$

where L is the segment length, and t_{AB} is the energy transmissivity from segment A to B, see Fig. 1(c). The energy transmissivity of a diffused scattering interface of the nanowire t^n_{AB} can be calculated by [20]:

$$t^n_{AB} = \frac{C_{V,B}v_B}{C_{V,B}v_B + C_{V,A}v_A} \tag{11}$$

For the aerogel backbone, we can assume that L is equal to the distance between the interconnected particle centers, see Fig. 1(b). But we cannot use Eq. (11) to calculate the energy transmissivity of the aerogel backbone, unless that the contact diameter of the interconnected particles is equal to the particle diameter, i.e. $a/d_p = 1.0$, see Fig. 1(b), which means that the backbone recovers to the nanowire. However, in the real aerogel, the contact diameter is always less than the particle diameter, i.e. $a/d_p < 1.0$, so that the energy transmission area is $\pi a^2/4$ instead of $\pi d_p^2/4$ corresponding to the case of $a/d_p = 1.0$. Therefore, the energy transmissivity of the aerogel backbone t^a_{AB} should be modified with the energy transmission area, leading to:

$$t^a_{AB} = \frac{C_{V,B}v_B}{C_{V,B}v_B + C_{V,A}v_A}\frac{a^2}{d_p^2} \tag{12}$$

Moreover, the interconnected particles of aerogel almost have the same physical properties, and on the assumption of rough interfaces we can obtain $t^a_{inter} = t^a_{AB} = t^a_{BA} = 0.5a^2/d_p^2$.

From the above analysis, we can refer to Eqs. (5), (6), (8), (10), and (11) as Dames and Chen's model, which is employed to calculate the phonon mean free path of the superlattice nanowire. If using Eqs. (9) and (12) to replace Eqs. (8) and (11) in Dames and Chen's model, we can calculate the average inter-atomic spacing of the aerogel backbone, and we refer to Eqs. (5), (6), (9), (10), and (12) as the present modified model.

3. Results and discussion

The silica aerogel and carbon aerogel are two of the most common insulation materials. In this section we predict the thermal conductivity of the backbone of the silica and carbon aerogels. The parameters of the bulk silica in the present calculation are $C_v = 1.687 \times 10^6 J/m^3 K$, $v_{bulk} = 4400$ m/s, $\Lambda_{bulk} = 0.558$ nm, $\rho_{bulk} = 2278$ kg/m^3, and $\lambda_{bulk} = 1.38$ W/m K [21]. For the bulk carbon, the parameters of $C_v = 1.562 \times 10^6 J/m^3$ K, $\rho_{bulk} = 2200$ kg/m^3, and $\lambda_{bulk} = 5.6$ W/m K are obtained from graphite in Ref. [22], $\Lambda_{bulk} = 2.689$ nm are calculated from Eq. (4), and $v_{bulk} = 4000$ m/s are obtained from Ref. [8].

To calculate the effective thermal conductivity of aerogel backbone λ_0, Eq. (4) was employed, where v_0 was approximated by the bulk one v_{bulk} since v_0 is not easy to measure, and Λ_0 was calculated by means of the modified model, i.e. Eqs. (5), (6), (9), (10), and (12), and the other parameters were taken from the bulk materials. Fig. 2 shows the relation between λ_0 and particle size. We can find that the contact diameter of the interconnected particles has significant effect on the effective thermal conductivity of the aerogel backbone. As the contact diameter increases, the interfacial resistance becomes lower and therefore the thermal conductivity gets larger. But when $a/d_p > 0.5$, the effect of the contact diameter on the thermal conductivity is no longer significant. The size effect is also notable, especially in the range of $d_p < 10$ nm. For larger contact diameter, say, $a/d_p > 0.5$, as the particle sizes of the silica and carbon backbones are greater than 100 nm and 300 nm, respectively, we can approximately ignore their size effects.

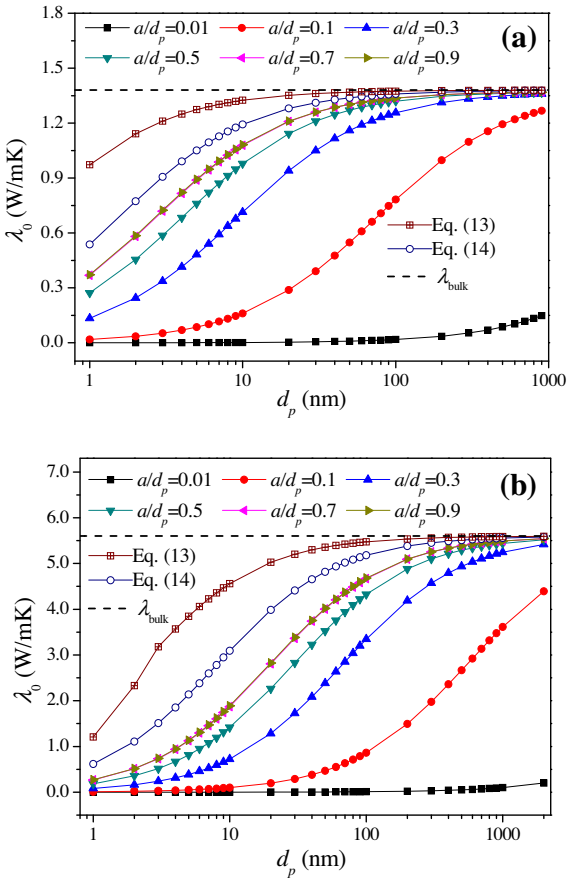

Fig. 2. Effective thermal conductivity of aerogel backbone, pure nanowire (Eq. (13)) and single particle (Eq. (14)). (a) Silica aerogel. (b) Carbon aerogel.

In addition, for the case of $a/d_p = 0.01$, we think that the connections of the particles are in point contact, which means that it is extremely difficult for the heat to transfer between the interconnected particles. So the effective thermal conductivity of the backbone is almost close to zero. We can imagine that an ideal structure of the aerogel used as the insulation material should require the connection of solid particles by the point contact to completely suppress the thermal conduction via the solid phase of the aerogel.

Once the contact diameter of the particle is equal to the particle diameter, i.e. $a = d_p$, the aerogel backbone recovers to the cylindrical nanowire. For the pure nanowire (only one material), we can believe that these particles are in perfect contact and the interface resistance disappears, i.e. $R_T = 0$, so the thermal conductivity can be calculated by the nanowire model [23]:

$$\lambda_0 = \lambda_{bulk}(1 + AKn)^{-1} \tag{13}$$

where Kn is the ratio of the bulk mean free path to the nanowire diameter. For $Kn < 1$, $A = 3/4$, and for $Kn > 5$, $A = 4Kn/(4Kn - 1)$. For $1 < Kn < 5$, the thermal conductivity can be approximated based on a simple interpolation of the formula between $Kn < 1$ and $Kn < 5$. In addition, for a single particle, the thermal conductivity can be obtained by Chen' model [24]

$$\lambda_0 = 3\lambda_{bulk} d_p/(3d_p + 8\Lambda_{bulk}) \tag{14}$$

The thermal conductivities of the pure nanowire and single particle are also shown in Fig. 2 for comparison. We can find that the effective thermal conductivity of the aerogel backbone is lower than that of the single particle and much lower than that of the pure nanowire when the size effect cannot be ignored, since the existence of the interface resistance in the aerogel backbone suppresses the heat transfer via the solid phase.

When the aerogels are used as the insulation material, we usually concern the solid thermal conductivity λ_s instead of the backbone thermal conductivity λ_0, see Eq. (3). Available experimental data of λ_s for carbon aerogel [25] and silica aerogel [26] are shown in Fig. 3 to validate the present model. For the silica aerogel, the sound velocity is predicted by the fitting formula $v \approx 48.9 + 0.797\rho + 0.00129\rho^2$, which is fitted by the measured data in [27], and v_0 is approximated by v_{bulk} in the calculation. The mean particle size of the backbone d_p is assumed 8 nm according to [4], and the contact diameter is assumed to be $a/d_p = 0.7$ due to the independence between the thermal conductivity of the backbone and the contact diameter of $a/d_p \geqslant 0.7$. The thermal conductivity of the silica aerogel backbone is $\lambda_0 \approx 1.05$ W/m K according to Fig. 2(a). In Fig. 3(a), we can see that the present model underestimates the solid thermal conductivity of the silica aerogel slightly, but the results from the present model have the same trend as the experimental data, while the use of bulk thermal conductivity overestimates the solid thermal conductivity largely when the density is larger than 150 kg/m^3. For the carbon aerogel, the sound velocity is fitted from the data in [27]: $v \approx 66.3 + 3.48\rho - 0.00228\rho^2$, and v_0 is also approximated by v_{bulk}. The particle size is assumed an intermediate value, i.e. $d_p = 150$ nm compared with $d_p \approx 140$–190 nm in [28] and $d_p \approx 108$ nm in [29]. The contact diameter is also set to be $a/d_p = 0.7$ for the same reason as that of the silica aerogel. We can obtain $\lambda_0 \approx 4.7$ W/m K in

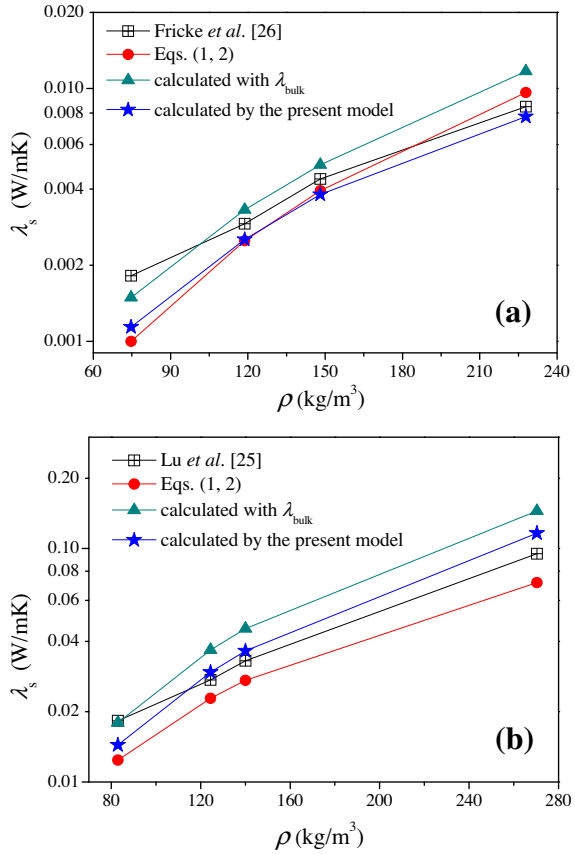

Fig. 3. Solid thermal conductivity of carbon aerogel at room temperature. (a) Silica aerogel. (b) Carbon aerogel. "Calculated with λ_{bulk}" denotes that the solid thermal conductivity is calculated by the bulk thermal conductivity, i.e. $\lambda_0 = \lambda_{bulk}$ in Eq. (3); "calculated by the present model" denotes that λ_0 is calculated by the present model of Eqs. (5), (6), (9), (10), and (12).

Fig. 2(b). In Fig. 3(b), we can observe that the results predicted by the present model are close to the experimental data when the density is above 100 kg/m^3, while the direct use of the bulk thermal parameters fails to predict the solid thermal conductivity. Therefore, the above comparison demonstrates that the present model has a good performance to predict the solid thermal conductivity of the aerogel.

In addition, the present model is also validated by the theory of minimum thermal conductivity (Eqs. (1) and (2)). In the calculations, the relation between the atomic density and the density of the material is $n = x\rho N_A/M$, where N_A is the Avogadro's number, M is the molar mass of the solid material, and x is the atomic number in a molecule. In this study, we use $x = 1$ for the carbon aerogel and $x = 3$ for the silica aerogel, and then the bulk atomic densities of the silica and carbon are about 6.859×10^{28}/m^3 and 1.104×10^{29}/m^3, respectively. The sound velocity in silica meets the formula of $v_{bulk} = 2/3v_t + 1/3v_l$. Then, on the assumption of $v_t \approx 0.6v_l$, we have $v_t = 3700$ m/s and $v_l = 5800$ m/s for the silica material, and $v_t = 3270$ m/s and $v_l = 5450$ m/s for the carbon aerogel. In Eq. (2), $c = 1.4$ is employed to predict the sound velocity in the silica aerogel [10], and $c = 0.8$ is used in the carbon aerogel since the sound velocity in the carbon aerogel agrees better with the scaling law $v \propto \rho^{0.8}$ instead of $v \propto \rho^{1.4}$ according to the experimental data in [27]. For the silica aerogel shown in Fig. 3(a), good agreement is achieved between the present model and the minimum thermal conductivity model. For the carbon aerogel, Fig. 3(b) shows some deviations between the two models, but compared with the experimental data, we can still believe that the present model has good performance to predict the solid thermal conductivity of the aerogel.

4. Conclusions

We have proposed a modified model to predict the thermal conductivity of aerogel backbone. In the modified model, the atomic scattering area of the boundaries and contact diameter of the interconnected particles are introduced to take into account the size effect and interfacial resistance effect. Both the particle size and contact diameter affect the effective thermal conductivity of the aerogel backbone significantly. The interfacial resistance is inversely proportional to the square of the contact diameter between the particles. But as the ratio of the contact diameter to the particle diameter is larger than 0.5, the effect of the contact diameter on the thermal conductivity is no longer significant. Due to the interfacial thermal resistance between the interconnected particles of the aerogel backbone, the effective thermal conductivity of the aerogel backbone is lower than that of the pure nanowire and single particle. The present model is validated by the experimental data and the minimum thermal conductivity model, and the comparison demonstrates that the present model is able to improve the accuracy for predicting the solid thermal conductivity of aerogels.

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