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Effective structure of aerogels and decomposed contributions of its thermal conductivity

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HIGHLIGHTS

- Deviation of effective thermal conductivity via effective models is analyzed.
- An effective model based on the structures of aerogels is proposed and verified its adaptability.
- Thermal conductivity of two materials at different gas pressure is measured.
- Contribution of gas conduction, solid conduction and thermal radiation is decomposed.

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ABSTRACT

In this paper, a new effective model, spherical hollow cube model, is proposed based on the structures of aerogels and the prediction equation for the apparent thermal conductivity is theoretically derived. The effective thermal conductivity of different types of aerogels is estimated by the present model, and the predicted results are more agreeable with experimental data than that of the previous models. In addition, the thermal conductivity of two nano-porous materials at different gas pressure is investigated experimentally. The contributions of gas conduction, solid conduction and thermal radiation are obtained by decomposition method.

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1. Introduction

Aerogels are typical nano-porous materials with open-cell structure manufactured through sol–gel process and supercritical drying technology [1,2]. The high porosity (85%–99%), large specific surface area (700–1300 m²/g) and low density (3–150 kg/m³) of aerogels result in their outstanding thermal insulation performance. The effective thermal conductivity of aerogels can reach as low as 0.012 W/m K [3] at ambient temperature and atmospheric pressure.

The heat transfer in aerogels includes collisions between gas molecules, gas convection in the pores, conduction through the solid skeleton and thermal radiation. However, gas convection could be

neglected in porous materials when the pore size is less than 1 mm at ambient pressure [4]. Heat transfer in nano-porous materials has strong size effect for their nanoscale structure. The nano-porous skeleton restricts the motion of gas molecules and thus decreases the gaseous thermal conductivity [5]. The thermal resistance of the solid matrix is greater than that of the bulk materials with the same thickness due to the extremely long heat transfer path introduced by the porous structure. In addition, in aerogels the solid skeleton size is in the range of 2–5 nm, for which the phonon boundary scattering occupies a main part and lowers phonon mean free path greatly. Thereby, the thermal conductivity of solid skeleton is less than that of bulk materials. As far as the thermal radiation is concerned, plenty of heat shields are formed in aerogels which prevent the thermal radiation by reflection, adsorption, transmission and re-radiation at the numerous gas–solid interfaces. The difficulties of analyzing the effective thermal conductivity of aerogels also came from the fact that defects, opacifier and reinforced fibers with

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micron size may exist in aerogels and its composites. All these complexities make the heat transfer in these materials multiscale in nature, i.e., heat transport phenomena in the aerogels include microscale, mesoscale and macroscale processes. Therefore, the effective thermal conductivity of aerogels and their composites depends on their microscale, mesoscale and macroscale structures, and the determination of their effective thermal conductivity is actually a multiscale problem [6].

For a fast engineering evaluation of the effective thermal conductivity, regular structures of aerogels are developed widely to replace the random structure in order to analyze the heat transfer characteristics and obtain some important average quantity. In the past two decades, many researchers proposed various effective models to estimate the effective thermal conductivity of porous materials. For example, Verma et al. [7] derived an expression for the prediction of effective thermal conductivity with spherical inclusions. Hsu et al. [8] developed a lumped-parameter model for the effective thermal conductivity of some two-dimensional and three-dimensional spatially periodic media. Gori et al. [9] used a cubic cell model to evaluate the thermal conductivity of an ablative composite material. Zeng et al. [10] proposed three effective models to calculate the thermal conductivity of pure aerogels. Although the thermal conductivity obtained from these models could fit well with experimental data qualitatively, how to improve quantitative agreement is still a big challenge and needs more theoretical and experimental studies.

As indicated above, the effective thermal conductivity of aerogel is composed of the contributions of gas conduction, solid conduction and thermal radiation. One way to understand the transmission mechanism in depth is to decompose the contributions of gas conduction, solid conduction and thermal radiation from the effective thermal conductivity and this will help to find the major heat transfer mechanism in aerogels and its composites. Some works have been conducted in this aspect [4,11,12], in which the gas pressure is lower than 1 bar.

In this paper, both theoretical and experimental studies are performed for the prediction of the effective thermal conductivity. Firstly, the reasons brought in the discrepancy of the effective thermal conductivity from different existing models are analyzed and a new effective model is proposed based on the structures of aerogels. The adaptability of the developed model is verified by comparing the predicted results with existing experimental data. Secondly, the transient plane source method is adopted to measure the effective thermal conductivity of some nano-porous materials. The Hot Disk thermal constant analyzer is combined with a molecular pump group, adjustable valves and a high pressure source to carry out the experiments at different gas pressure. The test range of gas pressure is extended to 1 MPa for thermal conductivity

measurement of porous materials. The thermal conductivity of two nano-porous materials is measured at different gas pressure to decompose the contributions of gas heat conduction, solid heat conduction and thermal radiation.

In the following presentation, an improved model for the effective thermal conductivity is proposed based on the analyzing of the disadvantages of previous models in Section 2, then theoretical analysis is further made for gas, solid and radiative conductivity in Section 3. In Section 4 both theoretical and experimental results are presented and compared. Finally, some conclusions are presented in Section 5.

2. Effective thermal conductivity model of aerogel

2.1. Analyzing some previous effective models of aerogel

Zeng et al. [10] proposed intersecting square rod, intersecting cylindrical rod, and intersecting spherical structure as the simplified model for aerogel, with representative unit cells shown in Fig. 1. For the carbon-opacified aerogels (density 0.11 g/cm^3 , porosity 0.94, specific surface area $797 \text{ m}^2/\text{g}$), the calculated effective thermal conductivity from the three models are 0.0419, 0.0414 and 0.0416 W/m K , respectively, which are nearly two times higher than the experimental value (0.0137 W/m K). The discrepancy in Zeng's analysis mentioned above is conjectured to be size effects to some extent. However, the intersecting spheres structure with the highest contact resistance (smallest contact ratio; i.e., $a/d = 0$) still results in a large predicted thermal conductivity of 0.0226 W/m K which is still higher than the experiment value, which indicates that other reasons also exist and have a significant influence.

According to the present authors' analysis, the discrepancy of the effective model is mainly introduced by the following reasons:

- (1) The effective thermal conductivity derived from the models is based on the assumption of one dimension heat conduction while in fact three-dimension heat conduction occurs in aerogels as shown in the scanning electronic microscope image of silica aerogels in Fig. 2 by the present authors. Therefore, the effective thermal conductivity calculated from the effective model would be underestimated.
- (2) Due to the random porous structure of aerogels, the actual solid heat transfer path of solid is much longer than that in the simplified periodic models. In this aspect, the thermal conductivity predicted by the effective model would be overestimated.
- (3) The thermal conductivity of bulk materials was used as the thermal conductivity of solid skeleton. However, size effect exists in the solid skeleton of aerogels, so it is unreasonable

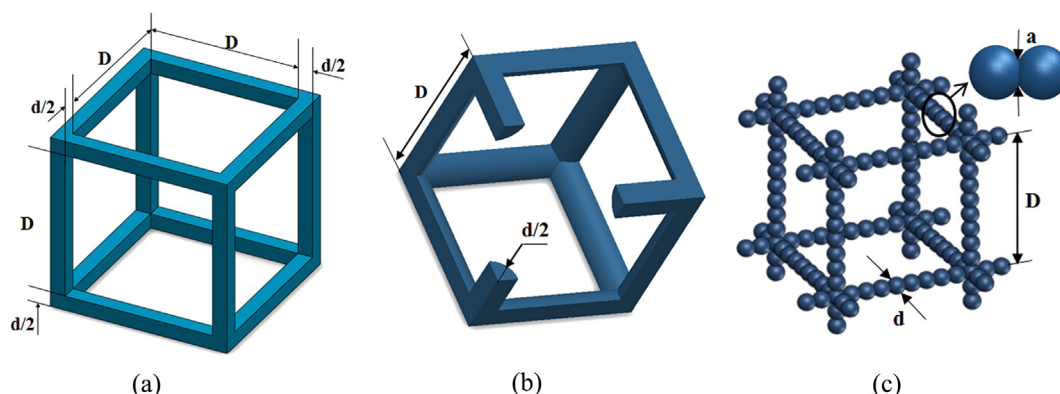


Fig. 1. Unit cells: (a) intersecting square rods; (b) intersecting cylindrical rods; (c) intersecting spheres.

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