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The influence of gaseous heat conduction to the effective thermal conductivity of nano-porous materials[☆]

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ABSTRACT

A thermal conductivity test apparatus based on transient plane source method is built and developed to measure effective thermal conductivity of open porous materials at different gas pressures. The effective thermal conductivity of open nano-porous silica materials with porosity of 88.5% is measured under gas pressures ranging from 0.001 Pa to 1 MPa. The contribution of gaseous heat conduction to the effective thermal conductivity of materials is decomposed by subtracting the effective thermal conductivity at ultimate vacuum from that at different gas pressure. It is found that the contribution of gaseous heat conduction is much different with the gas thermal conductivity in nano-porous materials and that in free space. The result is also demonstrated by theoretical analysis and numerical simulation.

1. Introduction

Aerogel, which is manufactured by applying a sol–gel process and supercritical drying technology, is a typical nano-porous material with open-cell and random structure [1]. It has merits such as high porosity, high specific area and super thermal insulation performance. Aerogels have the lowest thermal conductivity in solid or porous materials. Because aerogels are very fragile due to its extremely low density and near transparency to radiation of wavelengths of 3–8 μm, reinforced fibers and opacifiers are usually embedded and doped in the aerogels to maintain high mechanical strength and to ensure high thermal insulation performance at high temperature [2]. The composite still has high porosity, open porous random spatial structure and high thermal insulation performance. So we call aerogel and their composites nano-porous materials since their nano-scale pores with random size are the key factor of reducing their effective thermal conductivity. Owing to the outstanding thermal insulation property, they have a broad application prospect from the viewpoints of saving energy and thermal protection such as the thermal insulation materials in buildings and thermal protection system of shuttle and aircraft [3]. Therefore, great attention has been paid to analyze the heat transfer mechanism and to optimize the thermal insulation property [2,4–12]. The heat transfer paths in nano-porous materials are mainly divided into three types: solid heat conduction, gaseous heat conduction and thermal radiation. In nano-porous materials, the size effect is significant for both heat conduction via solid and gas and thermal radiation due to the size of solid skeleton and pore is close to or even smaller than the characteristic size of the energy carriers such as gas molecules, phonons and photons [13–15].

There are three kinds of heat transfer models to predict the effective thermal conductivity of porous materials in literatures [4,13]. The first model regarded the effective thermal conductivity as the superposition of solid (λ_s), gas (λ_g) and radiation (λ_r) thermal conductivities with the form of $\lambda_e = \lambda_g + \lambda_s + \lambda_r$ [5–7,10,12–15]. Gas convection is neglected because the pore size is less than 1 mm at ambient pressure [5]. The second model treated the effective thermal conductivity as the sum of radiation and a combined solid and gas conduction (λ_c): $\lambda_e = \lambda_c + \lambda_r$ [2,8,9,15,16]. The combined thermal conductivity of solid and gas, λ_c, can be calculated from effective structure models simplified from the practical structure or from empirical models. The third model is to conduct numerical simulation by generating the actual structure to calculate the effective thermal conductivity [4,17]. In this paper, we focus on the gas heat conduction in nano-porous materials because the gaseous heat conduction plays a significant or even dominant role on reducing heat transfer in nano-porous materials [6]. The gas thermal conductivity in nano-porous materials is lower than that in free space because the motion of gas molecules in nano-pores is suppressed by the nano-porous structure [14,15]. The gas thermal conductivity in nano-porous materials varies with gas pressure as well as the effective thermal conductivity (λ_e). By subtracting the effective thermal conductivity of materials at ultimate vacuum ($\lambda_{e,0}$) from that at different gas pressures, the contribution of gaseous heat conduction to the effective thermal conductivity ($\lambda_{g,0}$) could be obtained. In most of the previous works, the effective thermal conductivities of porous materials were experimentally investigated at the gas pressures of 1 bar or lower [5,6,9,10]. However, the decomposed $\lambda_{g,0}$ was usually regarded as the gas thermal conductivity in porous materials without distinguishing their differences by applying the first heat transfer model [5,10]. Recently,

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Nomenclature

a	diameter of contact area, nm
c_p, c_v	isobaric/isochoric specific heat capacity, J/mol · K
d	diameter, nm
D	diameter of spheres on each edge, nm
k	$k = 1 - \lambda_g / \lambda_s$
k_B	Boltzmann constant, 1.38×10^{-23} J/K
l	mean free path, nm
m	mass, g
p	gas pressure, Pa
S_s	specific surface area, m^2/g
T	temperature, K
Greek symbols	
γ	$\gamma = c_p / c_v$
λ	thermal conductivity, W/m·K
ρ	density, kg/m^3
Π	porosity, %
Subscripts	
c	conductive
e	effective
$e,0$	effective at vacuum
g	gas
$g,0$	contribution of gas heat conduction
r	radiative
s	solid

Reichenauer et al. found that $\lambda_{g,0}$ is about seven times higher than the gas thermal conductivity in a glass sphere bed with sphere diameter of 1 mm at ambient pressure [6]. They believed that the difference is caused by the local thermal shortcuts where spheres are touching; however, no further explanation was given. Swimm et al. studied the thermal conductivity of aerogels with average pore sizes of about 600 nm and 7 μm, respectively, under the gas pressures ranging from 10 Pa to 10 MPa [7]. A significant difference between $\lambda_{g,0}$ and the gas thermal conductivity was also observed, which is believed to be caused by the coupling heat transfer effect between solid and gas. A simple unit cell of two accumulated spherical particles was proposed to explain the coupling heat transfer effect; however, only qualitative agreement was obtained.

This study aims at investigating the influence of gaseous heat conduction on the effective thermal conductivity of nano-porous materials within a wide range of gas pressure. Experimental measurement, theoretical analysis and numerical simulation are conducted to clarify the relationship between the gas thermal conductivity and $\lambda_{g,0}$ in nano-porous materials.

2. Experimental investigations

An apparatus based on the Transient Plane Source (TPS) method [18] is established to measure the effective thermal conductivity of porous materials under the gas pressures ranging from 0.001 Pa to 1 MPa. It's the first time that the thermal conductivity has been measured at a different gas pressure using TPS method. The accuracy of the apparatus is validated by using NIST1453, an expanded polystyrene board with thermal conductivity of 0.032 W/m·K at room temperature with a deviation within 3%. We studied the theoretical accuracy of TPS method and measured thermal conductivity of nano-porous materials in our previous works [19–22]. In the experimental investigations, the nano-porous materials are initially vacuumized exhaustively for at least 3 times to remove the absorbed water vapor or impurity gas, and the gas pressure is adjusted from low to high. The effective thermal conductivity of Super-G, a commercial silica aerogel composite that was manufactured by Microtherm® with a density of 240 kg/m^3, porosity of 88.5% and claimed thermal conductivity of 0.0258 W/m·K at 373 K, is measured in nitrogen atmosphere under gas pressures ranging from 0.001 Pa to 1 MPa at 297 K.

3. Results and discussion

3.1. Effective thermal conductivity at different gas pressures

The effective thermal conductivity of Super-G (λ_e) measured at different gas pressures at 297 K is shown in Fig. 1. The results clearly show that the effective thermal conductivity varies greatly with gas pressure. The effective thermal conductivity keeps constant when the gas pressure is less than 0.01 kPa, which stands for the overall contribution of solid conduction and thermal radiation and it remains unchanged with gas pressure. However, the effective thermal conductivity increases sharply when the gas pressure is higher than 0.1 kPa due to the enhanced gaseous heat conduction.

3.2. Contribution of gaseous heat conduction decomposed from the effective thermal conductivity

The contribution of gaseous heat conduction to the effective thermal conductivity of Super-G ($\lambda_{g,0}$) is decomposed from the experiment results as shown in Fig. 1. The gas thermal conductivities in both free space and nano-porous materials are also theoretically depicted, as shown in Fig. 1. The gas thermal conductivity in free space is obtained from NIST database with taking the effect of actual gas into account [23]. The gas thermal conductivity in super-G used for comparison is calculated from Zeng's equation [14]. The mean free path (l_{m_0}) in free space:

$$l_{m_0} = \frac{k_B T}{\sqrt{2}\pi d_g^2 p}. \tag{1}$$

Due to the suppression of nano-porous structure, the mean free path of gas molecules in porous materials

$$l_m = \frac{1}{\sqrt{2}\pi d_g^2 p / k_B T + 0.25 S_s \rho_{por} \prod^{-1}}. \tag{2}$$

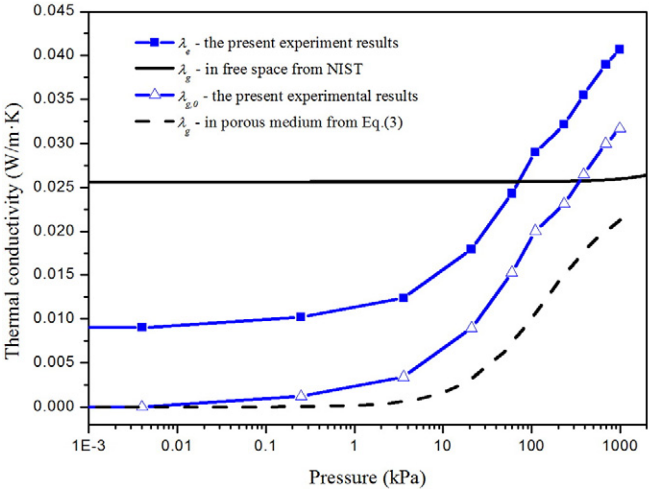

Fig. 1. Effective thermal conductivity of Super-G and comparison between gas thermal conductivity and contribution of gaseous heat conduction.