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Thermal conductivity of fiber and opacifier loaded silica aerogel composite

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ABSTRACT

Due to pure silica aerogel is fragile and nearly transparent to 3–8 μm infrared beam, reinforced fiber and opacifier are composited with silica aerogel to optimize its performance. The suppression of radiative heat transfer is related to opacifier type, size and content. However, the addition of opacifier will enhance solid conduction inevitably. In addition, the influence of doping opacifier will also be affected by temperature since thermal radiation is temperature and wavelength dependent. Therefore thermal conductivities of 10 fiber and opacifier doped silica aerogel composites are studied experimentally and theoretically. Transmittances of silica aerogel doped with opacifier of different type, size and content are measured within 1.4–25 μm by FTIR. The spectral extinction coefficient of fiber/opacifier loaded silica aerogel composite is obtained by considering the practical absorption of KBr diluents. The thermal conductivity of silica aerogel composite is measured by transient plane source method at different temperature and gas pressure. Effective model with considering heat conduction via gaseous molecules, matrix skeleton, fiber, opacifier and thermal radiation via skeleton, fiber and opacifier is developed for fast predicting the thermal conductivity of fiber/opacifier doped silica aerogel composite. It is found that the silica aerogels doped with SiC or TiO_2 particles has lower radiative thermal conductivity than that doped with ZrO_2 particles with volume fraction of 3.75% and diameter of 3.5 μm. Silica aerogel doped with 3.75%, 3.5 μm SiC particles has the lowest thermal conductivity at temperature less than 1000 K.

1. Introduction

Aerogel manufactured by the sol-gel process and different drying process, such as supercritical drying, freeze drying and ambient pressure drying, has extremely low density, high porosity, high specific surface area, and random nano-porous structure. Aerogel and their composite have promising application prospects since these materials posses extremely low thermal conductivity, low sound velocity, high optical transparency or high adsorption capacity [1]. The high porosity, nanosized pores and particles are primarily responsible for the low thermal conductivity of aerogel [2]. Silica aerogel is the most widely studied aerogel for its relative low price and variety of exceptional properties, especially outstanding thermal insulation capability [3,4]. Although the thermal conductivity of silica aerogel can be as low as 0.013 W/m K at ambient pressure, pure silica aerogel is too fragile and nearly transparent to infrared light with wavelength ranging from 3 μm to 8 μm [5]. Thus reinforced fiber and opacifier particles are usually doped in silica aerogel to enhance its mechanical strength and high temperature thermal insulation performance. For example, Feng et al. prepared the SiO_2 aerogel composites using reinforced ceramic fibers, which could greatly decrease the radiative thermal conductivity and enhance the mechanical properties. The composite has thermal conductivity as low as 0.042 W/m K and high bending strength of 1.80 MPa at 800 °C [6–8]. They developed a new ambient-dried silica aerogel composites reinforced by smaller diameter microglass fiber mat, the composite has thermal conductivity of 0.022 W/m K at 650 °C and high bending strength of 1.4 MPa [9]. They also prepared thermally stable Al_2O_3–SiO_2 aerogels which could be used up to 1200 °C [10], and introduced an original method that pyrolyzing polycarbosilane to form SiC coatings on mullite fibers as reinforcement for Al_2O_3–SiO_2 aerogel composites. This creative and excellent method, for the first time, combined the reinforcement and opacifier together, and the resulting aerogel composites exhibited relatively high effective specific extinction coefficient of 56.3 m^2/kg (2.5–7.5 μm) and low thermal conductivity of 0.049 W/m K at 1000 °C [11].

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Considerable studies have been conducted to reveal the heat transfer mechanism, to predict the thermal conductivity and to optimize the thermal insulation performance of nano-porous insulating material. Tang [2] and Xie [12] reviewed the progresses of thermal transport in nano-porous aerogel materials, including mechanism of different influence factors, models and numerical methods for estimating effective thermal conductivity and constitutes' thermal conductivity.

Because the motion of gas molecules in nano-porous materials is suppressed by the nano-scale solid structure, the gaseous thermal conductivity in nano-porous materials is different from that in free space. Researchers gained the gaseous thermal conductivity in porous medium by modifying the mean free path, considering the practical pore size distribution and considering the solid-gas coupling effect [13–17]. By conducting the thermal conductivity measurement of open-porous materials at different gas pressure, the influence of gas heat conduction to the overall thermal conductivity can be decomposed by subtracting the effective thermal conductivity at ultimate vacuum circumstance from the values at different gas pressure [18–21]. However, the varied quantity in thermal conductivity with gas pressure is different from the gaseous thermal conductivity in the pores which has been proved and elucidated experimentally [19,20] while the difference between them was neglected in some studies [18,21].

The mean free path of phonons is on the order of 1 nm which is approximate to the size of skeleton particles [22]. Thus the size effect and interfacial resistance of nano-scale skeleton reduce the energy transmission via heat conduction and it is benefit for obtaining high thermal insulating capacity. It is difficult to obtain the solid thermal conductivity at nano-scale accurately regardless experiment or theoretical analysis. Some attempts have been made to predict and further decrease the solid thermal conductivity in aeorgel [23–25].

For optical thick medium, the radiation can be approximated as a diffusion process and the radiative thermal conductivity can be calculated by the Rosseland diffusion model. Since silica aerogel materials have very low absorption within some wavelength region, the porous medium cannot be simply regarded as an optical thick one. Zeng et al. proposed an approximate formulation for the coupling conduction and radiation in porous medium with arbitrary optical thickness [26]. The addition of fiber and infrared opacifier such as carbon, silicon carbide and tanium oxide will enhance the absorption of radiation energy. Theoretical model, numerical study and experiment measurement have been conducted to obtain the radiative properties of fiber/opacifier doped aerogels. For example, Zeng developed a theoretical model to determine the optimal carbon content doped in silica aerogel to minimize the thermal conductivity [5]. Lots of numerical studies have been made to reveal the radiative properties of fiber/opacifier loaded aerogels [27–32]. The microstructure of aerogel composite is reconstructed and simulation is conducted to analyze the influence of opacifier and fiber based on the Mie scattering theory, anomalous diffraction theory or Monte Carlo method. Since the spectral extinction coefficient of opacifier is also depending on the particle size and temperature, multi-layer graded doping in consistent with the temperature distribution in silica aerogel also can be made to further improve the thermal insulating ability [33]. In practical test, the spectral extinction coefficient of fiber/opacifier loaded silica aerogel is usually measured by the Fourier transform infrared (FTIR) spectrometer with KBr as the diluents [34–38].

In order to fast predict and optimize the effective thermal conductivity of fiber/opacifier doped aerogel, regular structures are developed to represent the random microstructure to obtain its effective thermal conductivity. Both unit cell model [39–42] and fractal model [22,43,44] can be used to calculate the thermal conductivity via conduction of the matrix. Then geometric parameter model or effective model is adopted to calculate the coupling solid and gaseous thermal conductivity of fiber loaded aerogel by treating the matrix as a uniform medium [45]. Then the effective thermal conductivity can be obtained by adding the radiative thermal conductivity to the coupling thermal conductivity via conduction [41,45].

Numerical simulation could reveal more microscopic features than the effective regular structures and was also conducted to analyze the heat transfer properties of fiber/opacifier doped aerogel composite. The micro-structure of the nano-scale matrix is reconstructed first to calculate the coupling solid and gas thermal conductivity of the aerogel matrix [46–48]. Then the practical micro-structure of the doped fiber in micro-scale is generated to calculate the coupling thermal conductivity via conduction of fiber doped aerogel by regarding the matrix as uniform medium [30,48]. Finally, combined the total extinction effect of aerogel, fiber and opacifier to the radiation, the effective thermal conductivity of the composite can be obtained.

Experimental investigation on the thermal conductivity of aerogel composite not only could be used for validating the result predicted by theoretical model and numerical simulation, but also the most important and finalized toll for evaluating the thermal insulation performance. Lu et al. measured the effective thermal conductivity of organic aerogel at different density and gas pressure to determine the contributions of solid conduction, gaseous conduction and thermal radiation to the thermal conductivity as a function of density [21]. Wei et al. measured the thermal conductivities of silica aerogel, xonotlite-type calcium silicate and xonotlite–aerogel composite at temperature from 300 K to 970 K and gas pressure from 0.045 Pa to atmospheric pressure [41,49]. Liu et al. measured the effective thermal conductivity of nano-porous silica aerogel composite at temperature ranging from 280 K to 1080 K and gas pressure between 0.01 Pa and 100 kPa [50]. The influences of temperature and humidity on the thermal insulation performance of silica aerogel composite also have been experimentally investigated [51].

Upon the current studies on the thermal properties of silica aerogel composite, thermal conductivity of fiber and opacifier loaded silica aerogel composite still lack of experimental investigation, especially the influences of additions under different environment factors. Therefore, the purpose of present study is to experimentally investigate the thermal conductivities of different fiber/opacifier doped silica aerogel composites at different temperature and gas pressure. In addition, to reveal the influences of opacifier type, size and content on effective thermal conductivity, the radiative properties of silica aerogel composited with different opacifier are studied by conducting transmittance measurement. The thermal conductivities of silica aerogel composited with opacifier of different type, size and content are measured using transient plane source method at different temperature and gas pressure. The transmittances of the silica aerogel composite are measured by FTIR to obtain the extinction coefficient. Effective thermal conductivity model is also applied to calculate the effective thermal conductivity of the fiber/opacifier doped silica aerogel composite and compared with experiment result.

2. Theoretical models

Heat transfer within nano-porous materials via three modes, heat conduction via phonons (solid skeleton) and gaseous molecules, heat convection via gaseous molecules, thermal radiation via photons. Due to the macroscopic motion of gas molecules is suppressed by the nano-porous structure, heat convection can be neglected in the heat transfer process [47]. The effective thermal