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Multi-layer graded doping in silica aerogel insulation with temperature gradient

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ABSTRACT

Radiative heat transfer at high temperature deteriorates the insulating capability of silica aerogel due to its poor extinction characteristics for wavelength below 8 μm. Both the infrared opacifier doping and fiber doping can reduce the heat transfer and improve insulating capability of silica aerogel efficiently at high temperature. We determine the optimal temperature-dependent size for typical opacifiers and silica fibers by combining the spectral extinction coefficient with blackbody radiation. The optimal temperature-dependent doping amount is obtained by minimizing the effective thermal conductivity. Based on the obtained temperature-dependent optimal parameters and the graded temperature distribution in silica aerogel, four solutions of multi-layer graded doping are presented and the insulating capability is improved effectively. The measured back temperature curves of doped silica aerogel in the experiment qualitatively verify the optimization predictions.

1. Introduction

Silica aerogel as one of typical super-insulating materials has been particularly focused on in industry and research recently [1–10]. However, at high temperature, the significant radiative heat transfer deteriorates the insulating capability of silica aerogel because the pure silica aerogel is almost transparent for infrared radiation in such spectrum range [11], which limits its applications. The doping infrared opacifiers in silica aerogel can essentially eliminate the infrared thermal transport and therefore suppress the radiative heat transfer. On the other hand, the aerogel structure is made of nanoporous network of SiO_2 nanoparticles with quite high porosity, which decreases its mechanical strength. The reinforcing fibers need to be doped for improving the mechanical properties of aerogel, and the doped fibers not only increase the strength of silica aerogel but also reduce the infrared radiative transfer as well.

For opacifier particle doping, several studies have been conducted to minimize the heat transfer in silica aerogel. Zeng et al. [12] presented an approach to determine the carbon content in silica aerogel to minimize the total energy transfer. Wang et al. [13] investigated the silica aerogel doped with TiO_2 powder, and the measured thermal conductivity of doped silica aerogel at 800 K is 0.038 W m^{-1} K^{-1}. Feng et al. [14] studied the effects of particle size, doping amount and doping material for silica-opacifier composites by experiments, the results showed that the SiC opacifier can suppress high temperature radiation effectively, and the optimal diameter and mass fraction for the SiC opacifier are 3 μm and 25% at 773.15 K, respectively. Wang et al. [15] studied the radiative characteristics of the opacifier-loaded silica aerogel and discovered that a small size of opacifier was more appropriate at high temperature. Zhao et al. [16] applied the Mie scattering theory and Kramers–Kronig relation to calculate the complex refractive indexes of opacifiers and found that the carbon black was the best opacifier for $T < 600$ K while the SiC was the best opacifier for $T > 600$ K, and the optimal diameter for the SiC opacifier was 4 μm below 400 K and 3 μm above 400 K. Zhao and Tang [17] applied the Monte Carlo method to simulate radiative energy transfer in microstructures of the silica aerogel doped with carbon black and obtained the optimal temperature-dependent particle size (3.5–1.4 μm) and doping amount (26–39% mass fraction) in temperature range of 300–1300 K to minimize the thermal radiation, and a graded doping solution is designed and proved effective.

For fiber doping, Lee and Cunnington [18] modeled the radiation heat-transfer based on the diffusion approximation and used a modified Rosseland mean coefficient to correct the effect of scattering by fibers and absorption of the matrix medium. Daryabeigi [19] combined the radiation/conduction heat transfer in fibrous insulations and modeled the radiative heat transfer by the

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modified two-flux approximation. Zhang et al. [20] investigated the high temperature characteristics of fibrous insulating materials numerically and experimentally. Feng et al. [21] experimentally studied the carbon fiber reinforced carbon aerogel. Results indicated that the doped fibers can overcome the brittleness and the pyrolysis shrinkage of carbon aerogel, thus expand the application ranges of carbon aerogel greatly. Zhao et al. [22] applied the anomalous diffraction theory (ADT) to study the radiative properties and heat transfer in fiber-loaded silica aerogel composites and found that the diameters of 4–6 μm silicon fibers were best for high-temperature thermal insulation. Xie et al. [23] applied the Mie scattering theory for doping fibers and investigated the doping concentration, size, orientation and temperature on thermal conductivity of silica aerogel with fibers.

In spite of the existing studies for opacifier particle and fiber doping, effective approach is still demanded to guide doping for silica aerogel in practical applications. Since (1) different types of opacifiers have different extinction characteristics and temperature tolerances, (2) the extinction characteristics of opacifier and fiber relate closely with the size, wavelength and temperature, and (3) the optimal doping amount for the opacifier and fiber depends on the suppression of both heat conduction and radiation, it indicates that the doping solution of opacifier and fiber requires above considerations to ensure adequate insulating capability to resist high temperature. The present article takes all these factors into account. We discuss different types of opacifiers and fibers, combine the spectral extinction coefficient with blackbody radiation to determine the optimal size, evaluate the optimal doping amount between heat conduction and radiation, and finally present four solutions of multi-layer graded doping considering temperature gradient. This work can play a guiding role in design of the doped aerogel in engineering applications.

The rest of this paper is outlined as follows. In Section 2, both the optimal doping size and amount of opacifier particles against the temperature are investigated. The optimal doping size and amount of fibers against the temperature are discussed in Section 3. Section 4 presents the multi-layer graded doping solutions of opacifier particles and fibers with temperature gradient. The back temperature of doped silica aerogel is measured to verify the optimization predictions in Section 5, followed by a brief conclusion in Section 6.

2. Optimization for opacifier particle doping in silica aerogel

The mean extinction coefficient of opacifier closely depends on the opacifier material, particle size, amount and application temperature. Since the optimal parameters are difficult to be obtained from experiment, the correlation theory must be provided to establish the quantitative relations between the above parameters and the mean extinction coefficient of opacifiers. The carbon black, SiC, ZrO_2 and TiO_2 powers are usually doped into aerogel [16]. Therefore, these four types of opacifiers are investigated whose complex refractive indexes in the wavelength range of 2.5–25 μm are taken from Refs. [16,17,24]. The doped opacifier particles are distributed in pure silica aerogel homogeneously.

2.1. Optimal particle size

Suppose that the opacifier particles are spherical. According to the Mie scattering theory, the extinction efficiency factor Q_{ext} of single opacifier particle is given by [25]

$$Q_{ext}(m,\chi) = \frac{2}{\chi^2}\sum_{n=1}^{\infty}(2n+1)\mathrm{Re}\{a_n + b_n\} \tag{1}$$

where m is the particle's complex refractive index, χ is the size factor defined as $\pi d/\lambda$ with particle diameter d and wavelength λ, and a_n and b_n are Mie coefficients which are functions of particle complex refractive index. When Q_{ext} is known, with the assumption of homogeneous particle size distribution, the extinction coefficient for opacifier particles of volume fraction f_V is given by [26]

$$\beta_\lambda = \frac{3Q_{ext}f_V}{2d} \tag{2}$$

where d is the particle diameter. The calculations refer to the diameters of opacifier particles ranging from 0.1 to 30 μm for λ = 2.5–25 μm, since the optimal size of particle is normally at several microns in diameter and the infrared radiation energy mainly concentrates in the wavelength range of 2.5–25 μm. Here the temperature-dependent mean extinction coefficient is obtained by averaging the spectral extinction coefficient over the blackbody radiation distribution [27]:

$$\beta = \left[\int_0^\infty \frac{1}{\beta_\lambda}\frac{\partial e_{b\lambda}}{\partial T}d\lambda \Big/ \int_0^\infty \frac{\partial e_{b\lambda}}{\partial T}d\lambda\right]^{-1} = \left[\int_0^\infty \frac{1}{\beta_\lambda}\frac{\partial e_{b\lambda}}{\partial e_b}d\lambda\right]^{-1} \tag{3}$$

where e_b is the blackbody emissive power, $e_{b\lambda}$ is the spectral blackbody emissive power, T is the temperature and β_λ is the spectral extinction coefficient of material. When we obtain all the mean extinction coefficients at a certain temperature within given particle size range, the diameter at which the maximum mean extinction coefficient achieves is the required optimal doping particle diameter at that temperature. The extinction characteristics of opacifier particle include the scattering extinction and absorption extinction, but the effect of particle size is mostly manifested by the scattering extinction because its characteristic scale is comparable to the wavelength of infrared radiation. It is known that the thermal radiation wavelength corresponding to the maximum blackbody emissive power changes with temperature and shifts to the short-wavelength with temperature increasing. However, the scattering extinction of opacifier particle is inversely proportional to the fourth power of the wavelength, and the scattering extinction reaches the maximum when the particle diameter approximates to the wavelength [28]. Therefore, as shown in Fig. 1, the optimal particle diameter decreases with the increase in temperature, and smaller particle is more appropriate for higher temperature. The optimal particle diameter for carbon black is much lower than the others due to the difference in optical constants. Above all, the radiation energy at high temperature concentrates on the

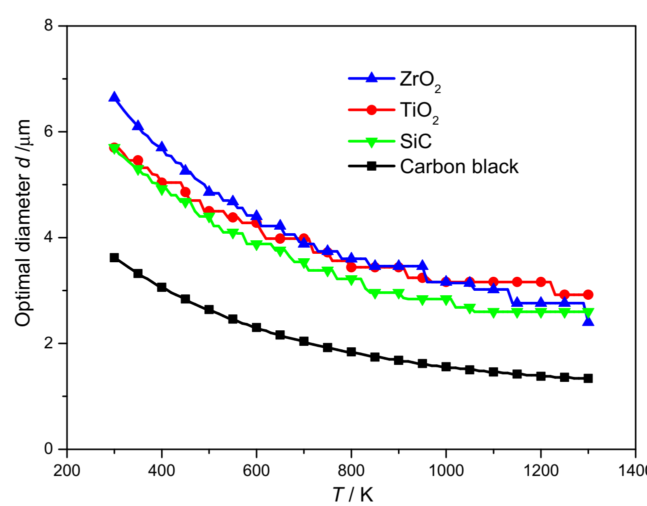

Fig. 1. Optimal diameter of opacifier particles against temperature.