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# Theoretical model of radiative transfer in opacified aerogel based on realistic microstructures

Hai-Tong Yu [a], Dong Liu [a], Yuan-Yuan Duan [a,*], Xiao-Dong Wang [b,c,*]

[a] Key Laboratory for Thermal Science and Power Engineering of MOE, Beijing Key Laboratory for $CO_2$ Utilization and Reduction Technology, Tsinghua University, Beijing 100084, China
[b] State Key Laboratory of Alternate Electrical Power System with Renewable Energy Sources, North China Electric Power University, Beijing 102206, China
[c] Beijing Key Laboratory of Multiphase Flow and Heat Transfer for Low Grade Energy, North China Electric Power University, Beijing 102206, China

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ABSTRACT

Opacified silica aerogels are composite insulating materials containing silica nanoparticles and microsize opacifier grains. The radiative heat transfer in this dispersed medium was analyzed using a realistic microstructure model to calculate the opacified aerogel's optical properties. The aerogel matrices were simulated using aggregates generated by a DLA algorithm, where the particle sizes and numbers were determined from the basic physical parameters. The theoretical predictions of the aerogel's optical parameters agreed well with experimental data. A geometric unit containing one opacifier particle and a large number of aerogel particles was then built to study the coupled radiation effect between the aerogel and the opacifier. The optical parameters were computed using a multi-sphere T-matrix code with comparisons with Mie scattering solutions. The results show how the opacifier's modified optical properties reduce the aerogel's radiative conductivity.

## 1. Introduction

Opacified silica aerogels are some of the best known thermal insulating materials. Over 90% of the composite's volume is silica aerogel, a highly porous solid matrix composed of packed silica nanoparticles with various silica aerogel parameters [1]. The aerogel's microstructure greatly reduces the heat transfer in the solid and gas phases; however, the infrared thermal radiation is not effectively blocked due to the low absorbance of silica in the 1–8 μm wavelength region. The strong radiative transfer at high temperatures can be reduced by opacifying the aerogels with microsize mineral powders such as carbon black, silica carbide, titania, or zirconia. These opacifier grains strongly absorb infrared thermal radiation [2]. The opacifier particles in opacified aerogels are very uniformly distributed in the nanoporous aerogel matrices with mass fractions of 10–30%.

Opacified aerogels are ideal as lightweight, high performance thermal insulation for buildings, automobiles, thermoelectric devices, and cryogenic applications [3–5]. The use of aerogels to improve building energy efficiencies was reviewed by Bouquerel et al. [6] who discussed the heat transfer in vacuum insulating panels. Space applications are especially exciting [7]. For high temperature insulation applications, the thermal radiation is usually the main heat transfer mechanism; thus, there have been many experimental studies on aerogel radiative transfer [2,8]. However, there have been only a few quantitative theoretical investigations due to the complexity of both the aerogel's microstructure and the radiation mechanism. Xie et al. [9] evaluated the radiative extinction due to monodispersed opacifier particles in silica aerogels using Mie theory to calculate the radiative conductivity as a function of temperature. Wang et al. [10] also used Mie theory to calculate the opacified aerogel's radiation heat transfer and recommended gradients of the opacifier sizes to minimize the radiative transfer in aerogel slabs. Zhao et al. [11] considered the irregular shapes of the opacifier grains when calculating the extinction coefficient of the opacifier by Mie theory and anomalous diffraction theory (ADT).

In these previous studies [9–11], the total extinction coefficient of the opacified aerogel was assumed to be the sum of the separate extinction coefficients of the aerogel and the opacifier, while ignored the interactions between the micro opacifier particles and the surrounding aerogel nanoparticles. Also, the optical properties of the aerogel matrices have mostly been based on experiment rather than calculation. The theoretical difficulties are partly due to the opacified aerogel's complicated microstructure with the scattering of multiple $SiO_2$ particles interacting with each other in the aerogel matrices so the properties cannot be described

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[*] Corresponding authors. Tel./fax: +86 10 62796318 (Y.Y. Duan). Addresses: Department of Thermal Engineering, Tsinghua University, Beijing 100084, China Tel./fax: +86 10 62321277 (X.D. Wang), State Key Laboratory of Alternate Electrical Power System with Renewable Energy Sources, North China Electric Power University, Beijing 102206, China.

*E-mail addresses:* yyduan@mail.tsinghua.edu.cn, 13651223062@163.com (Y.-Y. Duan), wangxd99@gmail.com (X.-D. Wang).

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**Nomenclature**

| | |
|---|---|
| $C$ | optical cross section |
| $d$ | aerogel particle diameter, nm |
| $d_a$ | aggregate's circumscribing diameter, nm |
| $D$ | opacifier particle diameter, μm |
| $D_g$ | circumscribing diameter of one geometric unit, μm |
| $f_v$ | solid volume fraction of aggregates |
| $g$ | scattering asymmetry factor |
| $L$ | distance between opacifier particles |
| $k_r$ | effective radiative conductivity, W m$^{-1}$ K$^{-1}$ |
| $m$ | optical constant ($m = n + i\kappa$) |
| $n$ | real part of the optical constant |
| $N$ | number of particles in an aggregate |
| $Q$ | optical efficiency |
| $T$ | temperature, K |
| $x$ | optical size parameter ($x = \pi d/\lambda$) |

*Greek Symbols*

| | |
|---|---|
| $\beta$ | extinction coefficient, m$^{-1}$ |
| $\kappa$ | imaginary part of the optical constant |
| $\lambda$ | wavelength, μm |
| $\omega$ | albedo |

*Subscripts and superscripts*

| | |
|---|---|
| a | aggregate |
| abs | absorption |
| eff | effective |
| ext | extinction |
| g | geometric unit |
| sca | scattering |
| tr | transport coefficient |
| $\lambda$ | spectral |
| 0 | medium properties |

by the independent scattering Mie theory [12] or dependent scattering models based on ideal geometric formulations [13]. Even numerical methods applied to complex geometries are not effective for opacified aerogels due to the large size difference between the opacifier and silica particles so that each geometric unit must contain thousands of particles.

Recently, Dlugach et al. [14] modeled the optical properties of aerosols with similar geometries where 49–249 smaller particles were attached to each larger particle with diameter ratios between the larger and smaller spheres of 5–10. Their numerical calculations showed that the small particles affected the optical parameters of the larger particle. However, actual opacified aerogels differ from Dlugach's model with even larger size differences between the opacifier and aerogel particles with the opacifier grains having diameters that are 50–500 times the diameter of the silica nanoparticles. Moreover, while the larger particles in the Dlugach et al. [14] model were covered by only a few smaller particles, the opacifier grains in the aerogels are completely embedded in the nanoparticle networks, with much larger numbers of small particles in realistic geometries.

This work characterizes the influence of the opacified aerogel's unique microstructure on its radiative transfer. The optical properties of the opacified aerogel are modeled using realistic microstructures. The aerogel matrix optical properties are characterized by aggregates formed using diffusion limited aggregation (DLA), with the particle sizes and numbers determined from the material's basic physical parameters. Then, a representative geometric unit containing one opacifier grain surrounded by many silica nanoparticles is generated based on the material's actual microstructure to calculate the opacifier's optical properties. The optical parameter calculations used the efficient, numerically accurate T-matrix code [15].

## 2. Microstructures and radiation modeling of aerogel matrices

### 2.1. Microstructures of aerogel matrices

A proper geometric model for realistic opacified aerogel microstructures requires insight into the material's actual structural features. Opacified aerogels are usually prepared using the sol–gel process with supercritical drying [1]. A suspension of opacifier powder is mixed with a silica source solution with a catalyst then added to trigger the gelation process. Aging and supercritical drying then create a solid monolithic opacified aerogel. The resulting microstructure of the opacified aerogel is shown in Fig. 1.

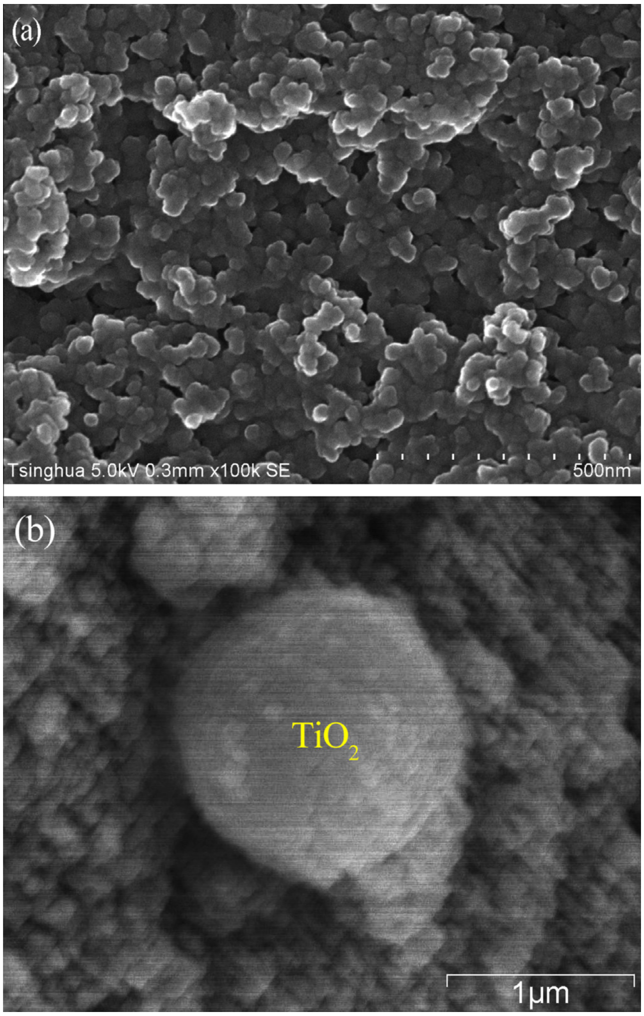

**Fig. 1.** SEM pictures of an opacified aerogel microstructure: (a) silica aerogel matrix and (b) $TiO_2$ opacifier powder embedded in the aerogel.

The monolithic aerogel is supported by the silica matrices, which are complicated 3D networks of $SiO_2$ nanoparticles with diameters of 5–20 nm and open pores with equivalent diameters