Physica B 456 (2015) 237–243

Contents lists available at ScienceDirect

Physica B

journal homepage: www.elsevier.com/locate/physb

Near-field radiative heat transfer across a pore and its effects on thermal conductivity of mesoporous silica

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ARTICLE INFO

Article history:
Received 1 August 2014
Received in revised form
2 September 2014
Accepted 6 September 2014
Available online 18 September 2014

Keywords:
Near-field radiation
Cylindrical pore
Spherical pore
Thermal conductivity
Mesoporous silica

ABSTRACT

Mesoporous silica substrate consists of uniformly distributed, unconnected cylindrical or spherical pores. Since the diameters of the pores are less than the wavelength, near-field radiative heat transfer across a cylindrical or spherical pore was simulated by employing the fluctuation dissipation theorem and Green function. Such factors as the diameter of the pore, and the temperature of the material were analyzed. It turned out that when the diameter is greater than 1 nm and less than 50 nm, the radiative heat flux at the mesoscale is 2–6 orders higher than the value at the macroscale, and decreases exponentially with the pore radius increasing for both cylindrical and spherical pore. The thermal conductivity of the mesoporous silica was modified with consideration of near-field radiation. It was concluded that the combined thermal conductivities of mesoporous silica which considering near-field radiation can agree with the experimental results more properly than non-considering near-field radiation. The smaller the pore diameter, the more significant the near-field radiation effect. The combined thermal conductivities of mesoporous silica decrease gradually with the pore diameter increasing, while increase smoothly with the temperature increasing.

1. Introduction

With rapid development of micro-electro-mechanical systems (MEMS), the integration of various devices is getting higher and higher, research attentions have gradually been turned to the nano-micro-scale. Micro-scale heat transfer including the near-field radiation has got more and more concern [1–18]. Different from the far-field radiation (macroscale thermal radiation) by propagating the transmissible electromagnetic waves, the near-field radiation is by photon tunneling of the evanescent waves [2,3].

Stefan–Boltzmann law is no longer applicable, when the characteristic size of the medium, e.g. the distance d between two surfaces, is comparable with or shorter than the characteristic wavelength of thermal radiation (λ_T). The near-field radiative heat transfer can exceed blackbody radiation by 5–6 orders of magnitude due to wave interference effect and photon tunneling effect [4–6].

As early as 1971, Polder and Van Hove [7] initially deduced a formula, based on the fluctuation-dissipation theorem (FDT), for the radiative heat flux between two parallel metallic plates with micro-scale space. Since then, researchers launched a large number of studies on the near-field radiative heat transfer between two semi-infinite bodies [8–11], between a spherical particle and a semi-infinite body [12,13], or between two spherical particles [14–16]. Although the theoretical research of the near-field radiation mechanism has experienced four decades, and achieved certain results, but so far, there are still a lot of questions yet to be resolved. For example, computational requirements are usually prohibitive once the computational domain reaches a span of several wavelengths [10]. For complex geometries, the dyadic Green's functions cannot be readily obtained [6]. Since the definition of a temperature is questionable for nanoscale materials, FDT might not be applicable [11]. In order to apply FDT, we have to assume that the media are in local thermodynamic equilibrium to define an equilibrium temperature.

Shen et al. [17] improved cantilever technology of atomic force microscope (AFM) to experimentally study the near-field radiation between SiO_2 microspheres and substrate with different materials. It was found to be several orders of magnitude higher than the Planck blackbody radiation law predicted. Chapuis et al. [18] designed the experimental device of a dielectric and the conductor to obtain the near-field radiative heat transfer depending on the spacing between the microsphere and the planar base. Hu et al. [19] used small polystyrene particles as spacers to maintain a micro-scale gap between two parallel glass flats; the experiment is designed to ensure that the radiative heat transfer can exceed the far-field upper limit, which dominates heat transfer between two

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http://dx.doi.org/10.1016/j.physb.2014.09.005

glass surfaces. Although experimental research achieved certain progress, the test results are usually unstable with uncontrollable measurement error. Accordingly, theoretical research is the main method to study the near-field radiation.

In this paper, we focus on the near-field radiation across cylindrical and spherical (2D/3D) pores and its influences on the thermal conductivity of the mesoporous silica. Recently, there has been a dramatic increase in the literature dealing with design, synthesis, characterization and property evaluation of the mesoporous silica for catalysis [20,21], adsorption [22,23] and separation [24–26]. Although some investigations on the thermal conductivity of the mesoporous silica were reported [27,28], near-field radiation effects has not been considered yet. In this paper, we use porosity weighted dilute medium(PWDM) model [29] to combine the near-field radiation effect with the thermal conductivities of gas confined in the pore of mesoporous silica substrate. The fluctuating electromagnetic field is described by the Maxwell equations with the fluctuating current density. The radiative heat transfer across the pore is gained by using fluctuation-dissipation theorem and dyadic Green's functions to calculate the Poynting vector. The combined thermal conductivity of the mesoporous silica is finally obtained and compared with the measurement.

2. Near-field radiative heat transfer across a pore in mesoporous silica

2.1. The structure of mesoporous silica

We focus on the near-field radiative heat transfer across a pore in mesoporous silica. The schematic nanostructure of mesoporous silica was shown in Fig. 1(a). Silica substrate, consists of uniformly distributed, unconnected cylindrical or spherical pores with diameters that can be tailored within the range 7–10 nm [30] or 1.33–2.01 nm [31], respectively.

The porosity ϕ was introduced to stand for the volume ratio of pores in mesoporous silica. For cylindrical pore, $\phi=\pi r^2/2\sqrt{3}(r+t)^2$, for spherical pore, $\phi=\pi r^3/6(r+t)^3$, where r is the pore radius, d is the mesopore diameter and t is half value of the wall thickness. Since the diameter of the pore is much smaller than λ_T, the electromagnetic field across the pore is dominated by the near-field radiation.

Since the mesoporous silica for both cylindrical and spherical pore had periodical structure, one structure element containing a pore can be suggested, as shown in Fig. 1(b). The pore is characterized by the free-space permittivity ε_0, and is bounded by silica with relative permittivity ε_1. We can see from Fig. 1(b) that, the temperature distribution can be expressed as $T(\rho,\theta)=1/2[(T_1-T_2)\rho\cos\theta/r+T_1+T_2]$ [32]. It drives radiative heat transfer across the pore, where T_1 is the high temperature at $\theta=0^\circ$, T_2 is the low temperature at $\theta=180^\circ$ on the pore boundary, and r is the pore radius.

2.2. Mathematical model for radiative heat transfer

For a monochromatic wave in a dielectric, isotropic, nonmagnetic body, according to the electromagnetic wave theory, the Maxwell equations corresponding to radiative heat transfer, can be shown as

$$\nabla\times\vec{E}=-\mu_0\frac{\partial\vec{H}}{\partial t}$$
$$\nabla\times\vec{H}=\varepsilon_0\varepsilon_1\frac{\partial\vec{E}}{\partial t}+\vec{j} \tag{1}$$

where E is the electric field, H the magnetic field, ε_1 the relative permittivity (dielectric constant) of silica, ε_0 the vacuum permittivity,

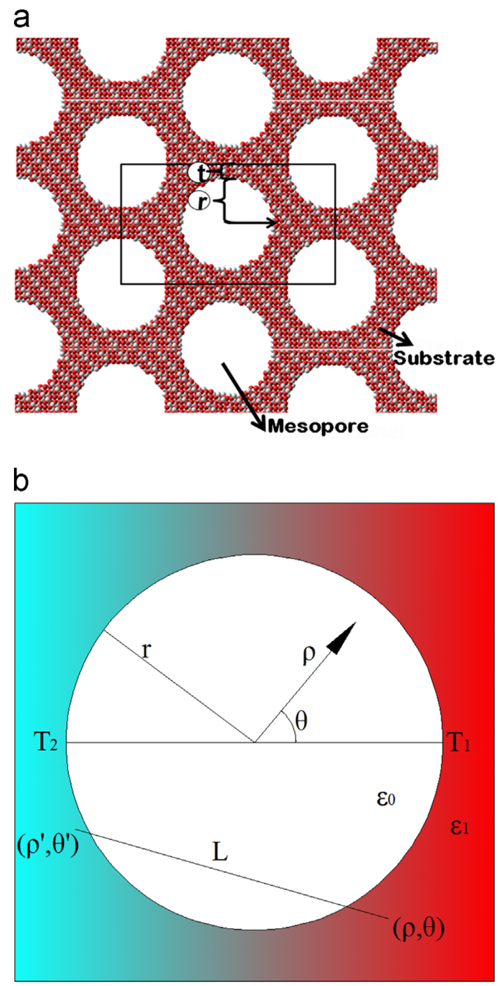

Fig. 1. (a) Structure of mesoporous silica: red spheres represent oxygen atoms, white spheres represent silicon atoms [20]. (b) Simplified a pore for near-field radiation calculation.

μ_0 the magnetic permeability of vacuum, j the fluctuating current density. Considering boundary conditions, we can get the fluctuating electromagnetic field, then obtain the frequency-dependent spectral Poynting vector, i.e. spectral heat flow across the pore, as given by

$$\begin{aligned}&\langle\vec{S}_{c\rho}(\omega)\rangle=\langle\vec{E}_\theta(\omega)\times\vec{H}^*_z(\omega)\rangle, && \text{for cylindrical pore}\\ &\langle\vec{S}_{c\rho}(\omega)\rangle=\langle\vec{E}_\theta(\omega)\times\vec{H}^*_z(\omega)\rangle, && \text{for spherical pore}\end{aligned} \tag{2}$$

where * denotes the complex conjugate, ω is the angular frequency of the electromagnetic wave.

Assuming that the Maxwell equations are with implicit $\propto e^{-i\omega t}$ time-dependent factors and there only exists fluctuating current souce j within the materials, the Maxwell equations become

$$\begin{cases}\nabla\times\vec{E}=i\omega\mu_0\vec{H}\\ \nabla\times\vec{H}=-i\omega\varepsilon_0\varepsilon_1\vec{E}+\vec{j}\end{cases} \tag{3}$$

The electric field can be given by

$$\nabla\times\nabla\times\vec{E}-\left(\frac{\omega}{c_1}\right)^2\vec{E}=i\omega\mu_0\vec{j} \tag{4}$$

where c_1 is the speed of light in the material. The complex wave vector in a vacuum can be expressed as $\kappa_0=\omega/c$, where c is the speed of light in vacuum, and κ_1 is the wave vector in silica, $\kappa_1=\omega/c_1=\sqrt{\varepsilon_1}\kappa_0$.