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Effect of metallic nanoparticle fillers on the thermal conductivity of diatomaceous earth

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ABSTRACT

Thermal conductivity of solid nanoparticles (aluminum) in a nanoporous solid matrix (diatomaceous earth) is examined to understand the effect of conductive fillers on the thermal properties of a porous material. We find that thermal conductivity is strongly dependent on load applied to prepare the mixture compacts, while porosity is influenced by the composition of the mixture. The addition of nanoparticles contributes to limited increases in thermal conductivity of the mixture by (1) increasing contact area between the mixture constituents and (2) reduction of porosity that leads to enhanced solid–gas coupling contribution. Thermal conductivity increases exponentially with external gas pressures due to the coupling effect between the solid particles and the entrapped air.

1. Introduction

Diatomaceous earth (DE) is a naturally occurring soft powdered material composed of fossilized remains of unicellular algae called diatoms; up to 100,000 species of diatoms can be found in a single sample [1–3]. The diatom skeletons are primarily made of amorphous silica. Each of the different diatom species has a unique frustule structure, making DE an extremely heterogeneous material with notable variations in physical properties [2,4–6]. DE finds applications as filters, building material, abrasive, catalyst carrier, acoustic and thermal insulators, and even as ingredients in some medicines [7–12]. DE has a low thermal conductivity (∼0.1 W/mK) because of its nanoporous structure, and has recently received interest as a potential candidate for core insulation material in superior performance vacuum insulation panels (VIPs) [8,13,14]. The vacuum inside VIPs degrades with time due to pressure difference, ultimately resulting in gradual loss of thermal resistance. A major challenge is to increase the lifetime of VIPs for use in energy efficient buildings, and one possible solution is to tune the thermal conductivity of DE with fillers. While thermal conductivity (k) of pure DE has been reported in the literature [7,15], the effect of impregnating the nanoscale pores with nanoparticles has not been explored. In particular, the presence of thermally conductive nanoscale fillers in the DE porous matrix can facilitate application-specific modification of k for insulation materials. Furthermore, DE has been traditionally used as a high temperature insulator (>1000 °C) whereby radiation heat transfer becomes especially pronounced in porous material [3,14]. Since metallic nanofillers could function as an opacifier without significantly increasing the thermal conductivity of the mixture, we examine the influence of Al nanoparticles (AlNPs) dispersed in bulk DE on the k of the mixture.

The thermal conductivity of porous materials depends on porosity [16–20]. In addition, k of granular materials like DE is strongly influenced by the contact resistance between particles, leading to a significant change in k with gas pressure [8]. In this letter, we discuss the contribution of nanoparticles and porosity on the k of AlNP/DE mixture. The addition of AlNPs, as expected, increases the overall k of the mixture. However, as discussed below in details, this increment is only marginal even though a highly conductive material such as Al is used. Our results aid in understanding how the fillers influence k by modifying the contact between the mixture components.

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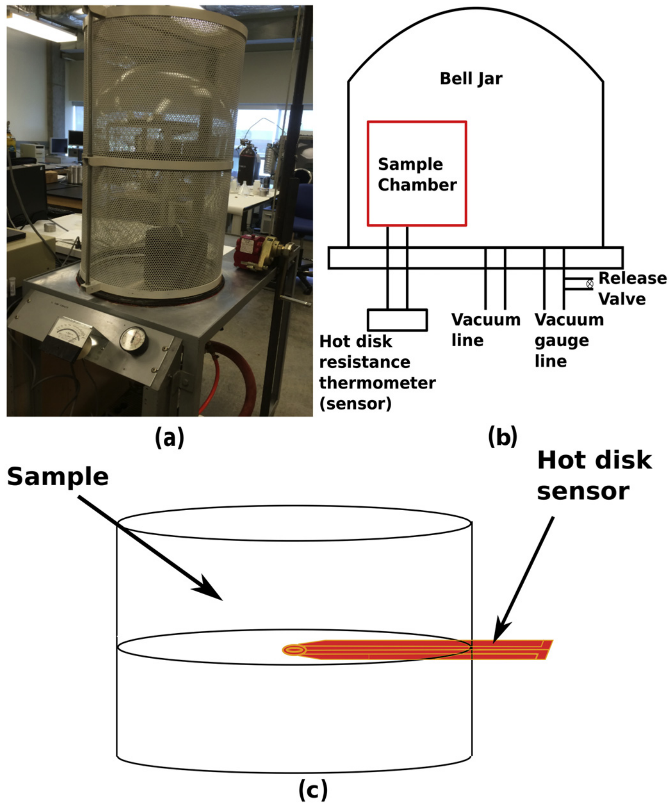

Fig. 1. The experimental setup (a) comprises of a sample chamber placed inside an evacuated bell jar, as shown in the schematic (b). The chamber contains a Hot Disk™ thermal conductivity probe, which is sandwiched between the dry pressed cylindrical DE samples, as illustrated in (c), during thermal conductivity measurement.

2. Materials and methods

AlNPs of ∼800 nm diameter (US Research Nanomaterials, Inc.) are mixed with DE having a pore size of ∼1 μm (Perma-Guard™, Inc.) by shaking in a vibratory mill for 6 h, followed by dry pressing to form cylindrical compacts. The density of each compact is determined by measuring the mass and volume. In total, twenty-four samples are prepared each with varying AlNP mass fraction (0%, 10%, 20%, 30%, 40%, and 50%) and compaction load (0.45 MPa, 1.13 MPa, 1.70 MPa, 2.26 MPa). k is measured by the transient plane source technique (Hot Disk™ TPS 1500) that employs a sensor acting as both a heat source and a resistance thermometer. The change in temperature is measured as a function of time, and subsequently k is obtained using the Gustafsson equation [21]. The microstructure of the samples is examined under a scanning electron microscope (SEM, JEOL JSM-6060LV). For two of the samples, viz., pure DE and 50% (w/w) AlNP/DE mixture, we measure k as a function of gas pressure between 27 Pa (∼ vacuum) to 10^5 Pa (∼1 atm.), by evacuating the sample in a bell jar (schematic shown in Fig. 1).

3. Results and discussion

Effect of nanoparticle concentration: Air occupying the nanopores in pure DE at atmospheric pressure contributes to the overall heat conduction [22]. With the addition of AlNPs, these air pockets are replaced by highly conductive metallic particles. We neglect the influence of nanoscale size effects since both the electron mean free path ($\lambda_{electron} \sim 22$ nm) [23] and the phonon mean free path ($\lambda_{phonon} \sim 8.1$ nm) [24] of Al are much smaller than the average particle diameter of 800 nm. Thus, k_{Al} for the nanoparticle used

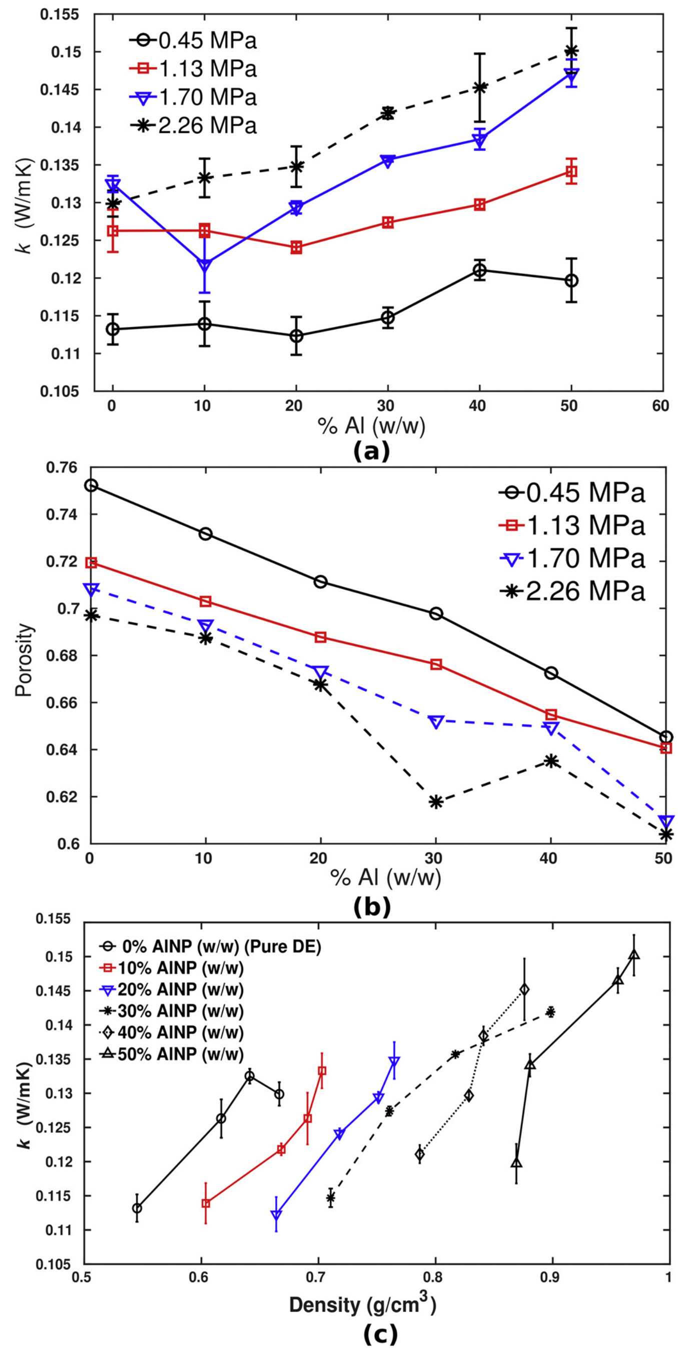

Fig. 2. (a) The thermal conductivity of AlNP/DE mixtures increase with increases in nanoparticle concentrations and compaction pressures. (b) The strong reduction in porosity with increasing AlNP concentration implies a weak dependence of the overall mixture thermal conductivity on porosity. (c) Thermal conductivity variation with density is shown for all the samples. The cases with the same AlNP concentrations are connected by lines to aid data visualization. Despite the large increase in density with addition of AlNP, thermal conductivity is more sensitive to the compaction.

in this investigation is similar to the bulk Al thermal conductivity ($k_{bulk} = 237$ W/m K).

We present the change in effective k of the mixture with AlNP concentration in Fig. 2a. Though k_{Al} is significantly higher than that of pure DE, k for the mixture increases only by 15% even for 50% nanoparticle mass fraction under a compaction of 2.26 MPa. We conjecture that this small increase in k is due to impedance to heat transfer by interfacial thermal resistance arising from the increase in interfacial areas between dissimilar surfaces