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Thermal conductivity of copper and silica nanoparticle packed beds☆

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ABSTRACT

We report here thermal conductivity measurements of packed beds of nanoparticles both under vacuum and at ambient pressure and compare those measurements to theoretical predictions. The thermal conductivity measurements of the nanoparticle beds under vacuum were found to be as low as 0.018 W/m °C, among the lowest thermal conductivities of solid materials ever measured. Theoretical predictions agreed with the conductivity measurements under vacuum within experimental uncertainty. In contrast, thermal conductivities measured for nanoparticle beds at ambient pressure were an order of magnitude higher than the measurements under vacuum, and significantly higher than the theory predicted. These results indicate a serious limitation that could impact the use of nanoparticle beds as very low conductivity insulating materials.

1. Introduction

Nanoparticle packed beds have been identified as having extremely low thermal conductivities. They along with other nanostructured materials have been proposed as the basis for creating new insulating materials [1–4]. Hu et al. were the first to show experimentally that nanoparticle packed beds could demonstrate remarkably low thermal conductivities, among the lowest measured for solid materials [5]. Hu et al.'s measurements were motivated by theoretical work from Prasher whose analysis showed that the nanoscale contacts between particles in a packed bed would constrain ballistic phonon transport, resulting in high interparticle contact resistance and extremely low bed conductivity [6]. More recently, Voges et al., in an effort to create nanopowder composite insulation, measured the thermal conductivities of composite materials made by bonding together mixtures of nanoparticles consisting of 30 nm SiO_2, 50 nm Al_2O_3, and 30 nm carbon black. For the most part, they also found very low thermal conductivities in good agreement with Hu et al.'s results, with one significant exception, samples exposed to air for long periods had anomalously high thermal conductivities [7]. In addition to their use as insulating systems, packed beds of nanoparticles are also of interest because of their potential application as catalysts in microreactors, and fuel cells and as electrodes in batteries [8–11].

In spite of the potential applications of nanoparticle packed beds, their thermophysical properties have not been well explored. This is in contrast to the numerous studies of the thermal properties of nanoparticles in other forms. For example, many researchers have investigated the thermal conductivity of nanofluids or nanoparticles suspended in heat transfer fluids [12–14]. Likewise, a number of workers have investigated the conductivity of phase change materials (PCM) loaded with nanoparticles [15–17]. However, aside from Hu et al. and Voges et al., no other measurements of the thermal conductivities of nanoparticle packed beds have been published. In addition, while there has been significant work on the theory of heat transfer through nanoparticle beds, that theory has not yet been validated against experimental measurements.

The purpose of this paper is to report experimental measurements of heat transfer across packed beds of nanoparticles both under vacuum and at ambient pressure and to compare those measurements to theory. We show for first time that an analytical model, based on the theory developed by Prasher, is able to predict the thermal conductivity of a nanoparticle bed measured under vacuum within experimental uncertainty. However, we also find that the thermal conductivities of nanoparticle beds at ambient pressure are ten times higher than beds under vacuum. In addition, these measurements are significantly higher than what the analytical model predicts. These results point up a potential limitation on the use of nanoparticle packed beds as insulating systems.

2. Experimental methods

Characterization of the packed beds' thermal conductivity is accomplished via the guard-heated calorimeter shown schematically in Fig. 1. The apparatus, which has been used previously to characterize the effective thermal conductivity of thermal switches and their contacts, measures both the heat transfer rate, Q, and the temperature drop, ΔT, across a sample under steady state conditions [18,19]. A nanoparticle bed is placed between the guard-heated calorimeter, used to measure the heat transfer rate across the bed, and silicon carrier die. The guard-heated calorimeter consists of a heat flux sensor sandwiched

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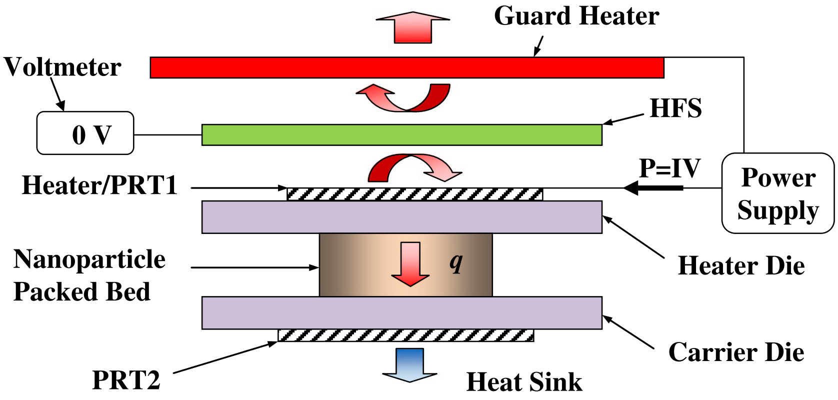

Fig. 1. Schematic of the guard-heated calorimeter with nanoparticle packed bed sample in place.

between a silicon heater die below it, and a guard heater above it, all mounted on a rigid aluminum plate. Thermal resistance measurements are made by controlling the power delivered to the heater die and guard heater until they are at the same temperature and the voltage output of the heat flux sensor is zeroed. When the heat flux sensor output is zeroed, all the electrical power dissipated in the silicon heater die is transferred as heat down from the heater die, through the nanoparticle packed bed to the silicon carrier die. The heat transfer rate across the packed bed is thus equal to the input power to the heater die. The temperature difference across the packed bed is measured with platinum resistance thermometers (PRT's) micromachined on the top silicon heater die and the bottom carrier die. The measured thermal resistance across a packed bed is the ratio of the temperature difference to the heat transfer rate across the packed bed:

$$R_m = \frac{(T_{top} - T_{bot})}{Q} = \frac{\Delta T}{Q} \tag{1}$$

The bottom carrier die is epoxied on an aluminum support with a cooling water loop and mounted atop a z-axis stage. A stepper motor and load cell control the vertical force applied to the packed bed sample. The entire apparatus is enclosed in a bell jar, enabling control of gas pressures from 10^{-2} Torr to ambient.

Before any thermal conductivity measurements were made, the apparatus was calibrated. This involved first calibrating the two PRTs in the top heater die and bottom carrier die. The uncertainty in temperature measurements made by the calibrated PRT's was estimated to be ±0.03 °C. The guard-heated calorimeter was then calibrated to account for any systematic bias due to a heat leak in the apparatus. To calibrate the calorimeter, the apparatus was set up with the top heater die and the bottom carrier die separated by a gas gap of known thickness L. In this configuration, heat transfer across the gas gap was by conduction only. The gas, heated from above, was stably stratified, so convection was suppressed and radiative transfer was calculated to be negligibly small. As a result, the measured thermal resistance across the air gap was known to be composed of only two parallel resistances, the resistance due to conduction across the air gap, R_{gas}, and the resistance due to any heat leak from the heater die, R_{leak}. The thermal resistance due to conduction across the gas gap could be determined from the known thermal conductivity, k_{gas}, gas gap thickness, L, and cross-sectional area, A_{gap}, of the gas gap. The parallel heat leak resistance, R_{leak}, could be determined from the measured thermal resistance and known properties of the gas gap. The parallel leak resistance was characterized for each nanoparticle bed, over the same conditions used in the thermal conductivity measurements. Details about the calibration can be found in Ref. [20].

Packed beds assembled from two types of nanoparticles were characterized in this work. Dry copper nanoparticles, obtained from SkySpring Nanomaterials Inc., were nominally 300 nm in diameter, and had a purity of 99.5%. Dry silica nanoparticles (NanoXact nanoparticles) obtained from NanoComposix had diameters of 200 ± 7 nm with nonfunctionalized surfaces. In both cases, the particles were used as received from the vendors.

To avoid any contamination, the packed beds were assembled by pouring the nanoparticles directly from the sealed vendor packaging into a machined steel mold, and a micrometer spindle inserted in the mold. The micrometer dial was rotated until the nanoparticles were pressed in the mold with an applied pressure of 15.4 MPa. The diameter of the mold, 6.75 mm, determined the diameter of the disk-shaped packed bed. The extension of the micrometer spindle into the mold determined the height of the packed bed disk. Once the bed was packed tightly together, the steel mold was opened and the nanoparticle bed extracted using clean tweezers. Fig. 2 illustrates the molding process.

The thermal conductivity of a nanoparticle packed bed was measured by inserting it between the heater die of the guard-heated calorimeter and the bottom carrier die. The stepper motor holding the bottom carrier die was adjusted to apply a set force of 3.5 N or a load pressure of 98 kPa. The bell jar was placed over the apparatus, and the gas pressure around the packed bed reduced to the desired pressure. When electrical power was supplied to the top silicon heater die and

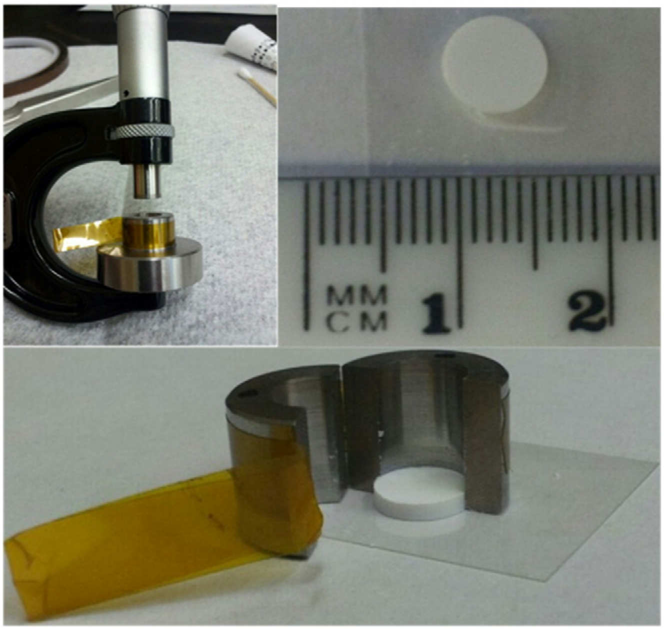

Fig. 2. Molding a silica nanoparticle packed bed.