



Enhanced specific heat capacity of molten salt-based nanomaterials: Effects of nanoparticle dispersion and solvent material

Byeongnam Jo ^{a,b,*}, Debjyoti Banerjee ^b

^a Nuclear Professional School, The University of Tokyo, 2-22 Shirakata, Tokai-mura, Ibaraki 319-1188, Japan

^b Department of Mechanical Engineering, Texas A&M University, College Station, TX 77843, USA

Received 22 February 2014; received in revised form 28 April 2014; accepted 5 May 2014

Available online 29 May 2014

Abstract

This study investigated the effect of nanoparticle dispersion on the specific heat capacity for carbonate salt mixtures doped with graphite nanoparticles. The effect of the solvent material was also examined. Binary carbonate salt mixtures consisting of lithium carbonate and potassium carbonate were used as the base material for the graphite nanomaterial. The different dispersion uniformity of the nanoparticles was created by employing two distinct synthesis protocols for the nanomaterial. Differential scanning calorimetry was employed to measure the specific heat capacity in both solid and liquid phases. The results showed that doping the molten salt mixture with the graphite nanoparticles significantly raised the specific heat capacity, even in minute concentrations of graphite nanoparticles. Moreover, greater enhancement in the specific heat capacity was observed from the nanomaterial samples with more homogeneous dispersion of the nanoparticles. A molecular dynamics simulation was also performed for the nanomaterials used in the specific heat capacity measurements to explain the possible mechanisms for the enhanced specific heat capacity, including the compressed layering and the species concentration of liquid solvent molecules.

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Keywords: Nanomaterial; Specific heat; Molten salts; MD simulation; Differential scanning calorimetry

1. Introduction

Nanofluids, as first referred to by Choi [1], have received great attention over the last decade due to their anomalously increased thermal conductivity compared to base fluids [2,3]. Their significant enhancement in thermal conductivity and pool boiling characteristics, such as a 200% enhancement of critical heat flux, have enabled nanofluids to be considered as a new working fluid in diverse fields [4,5]. Owing to these outstanding features, a number of theoretical and experimental studies have been performed

to understand the effect of nanoparticles on thermal properties and heat transfer performance [6–8]. In addition, the viscosity of nanofluids has been reported to be significantly higher than their base fluids, and existing literature has also shown their non-Newtonian rheological behavior [9–12].

Previous studies on the increased viscosity of nanofluids, attribute the increase in viscosity to agglomerations of the nanoparticles [9,10,12,13]. While the effect of particle clustering on increased viscosity is well accepted, there are controversial issues concerning the enhanced thermal conductivity of the nanofluids, particularly on the effect of uniformity in nanoparticle dispersion. Keblinski et al. [14] reported that the linear agglomeration of particles that could serve as extended paths were responsible for thermal conductivity. They also showed that previous experimental

* Corresponding author at: Nuclear Professional School, The University of Tokyo, 2-22 Shirakata, Tokai-mura, Ibaraki 319-1188, Japan. Tel.: +81 29 287 8423; fax: +81 29 287 8488.

E-mail address: jo@vis.t.u-tokyo.ac.jp (B. Jo).

results for thermal conductivity were bounded in a Hashin and Shtrikman correlation [2,3,15–18]. However, Murshed [19] noted that it was difficult to determine the percolation network through particle aggregations – a major mechanism for the thermal conductivity enhancement of nanofluids. The author also noted that surfactants, sonication duration, and particle sizes favorably affected the thermal conductivity enhancement of nanofluids, which is seemingly contradictory to nanoparticle agglomerations [20–24]. As previously noted, nanoparticle dispersion may be one of the key parameters for enhancing thermal conductivity and further augmenting heat transfer performance.

This study investigates the effect of the nanoparticle dispersion on the specific heat capacity of nanomaterials with a base material consisting of a binary mixture of molten salts with a high melting point of 490 °C. Well-dispersed nanoparticles essentially result in an increased surface to volume ratio, which can lead to enhanced thermal conductivity – a transport-related property. Conversely, specific heat capacity may generally be considered unrelated to an increase in surface to volume ratio. However, previous literature experimentally and computationally showed that a compressed liquid creates a layer adjacent to the interface with the solid [25,26]. According to a suggested mechanism that the specific heat capacity enhancement in a nanomaterial can result from a compressed liquid layer near the nanoparticles, an increased interface area between the surrounding liquid and the nanoparticles can significantly enhance the specific heat capacity of nanomaterials [27,28]. Moreover, dispersants (surfactants) and the short elapsed time of nanoparticles were also proven to have a positive effect on the enhancement of specific heat capacity [29]. It is therefore thought that the dispersion of nanoparticles affects the specific heat capacity of nanomaterials.

This study measures the specific heat capacity of carbonate salt-based graphite nanomaterials in both solid and liquid phases by using a differential scanning calorimeter. To create well-dispersed and relatively poorly-dispersed nanomaterials, two distinct synthesis protocols were employed where the water evaporation duration was drastically different between the two methods. The effect of the base fluid composition (i.e., solvent consisting of a binary mixture of carbonate salt) was examined to explore the possible reasons for the enhanced specific heat capacity that was observed in the experiments. Finally, molecular dynamics simulations were performed to explore the nature of the compressed layers as a function of the chemical properties of the solvent material.

2. Materials and methods

2.1. Molten salt nanomaterials

A binary carbonate salt mixture composed of lithium carbonate (Li_2CO_3 , Sigma Aldrich) and potassium carbonate (K_2CO_3 , Sigma Aldrich) was used as a base material for the nanomaterials. Graphite nanoparticles with a diameter

of 50 nm were dispersed in the base material. The specific heat capacity of each pure carbonate salt in liquid phase is 2.51 [J/g K] and 1.51 [J/g K] for the lithium carbonate and the potassium carbonate, respectively [30]. Additionally, the specific heat capacity of the graphite nanoparticles obtained from literature 1.66 [J/g K] at the maximum temperature employed in this study [31]. Fig. 1 shows images of the graphite particles taken by a transmission electron microscope (TEM, JEOL JEM-2010). The images show that the diameter of the graphite nanoparticles was larger than 50 nm. The two pure salts—lithium carbonate and potassium carbonate – were initially mixed with a eutectic composition of 62:38 in a molar ratio, but the composition of the base material was varied to examine the effect of the solvent on the specific heat capacity enhancements. This will be discussed later.

Fig. 2 illustrates the two synthesis procedures for the molten salt nanomaterials used in this study. Graphite and gum arabic (GA, Sigma Aldrich) were initially added to distilled water, and this suspension was subjected to sonication in an ultra-sonic bath for 2 h. The carbonate salt eutectic was added to the aqueous graphite nanofluid prepared in the previous step, and the aqueous salt–graphite nanofluid was sonicated again for 3 h to avoid agglomerations of nanoparticles. To obtain the dehydrated powder of the nanomaterial, water was evaporated on a hot plate set at 100 °C. In the last step, the evaporation method was varied, resulting in differing dispersion qualities of the graphite nanoparticles. This was achieved with a large petri dish (Method 1) and a vial (Method 2). It was expected that Method 1 would yield better uniformity in nanoparticle dispersion because the petri dish method substantially reduces the duration for complete evaporation. The shorter duration for evaporation is conducive for GA to prevent the agglomeration of the graphite. In addition, the large surface area of the petri dish is also beneficial for maintaining the homogeneity of the nanoparticle dispersion. Images of the dehydrated nanomaterials on the petri dish are shown in Fig. 3. As for the nanomaterials prepared by Method 1, a finer salt powder formed on the periphery of the petri dish (referred to as Nanomaterial 1) and a coarser salt powder

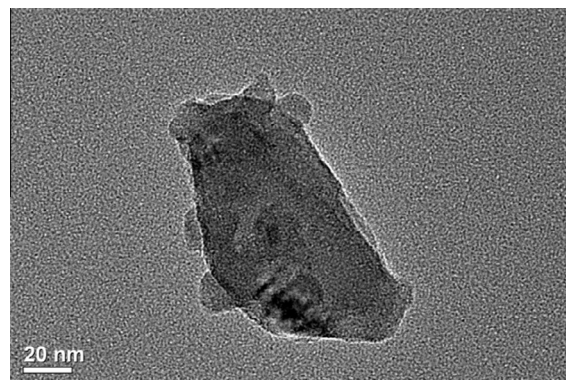


Fig. 1. Transmission electron microscope image of a graphite nanoparticle.

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