



Effect of solvent on specific heat capacity enhancement of binary molten salt-based carbon nanotube nanomaterials for thermal energy storage



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ABSTRACT

The specific heat capacity of a binary molten salt-based carbon nanotube nanomaterial was experimentally measured in both solid and liquid phases. The effect of the solvent on the specific heat capacity was examined for a wide range of chemical compositions for a binary carbonate salt mixture. The binary salt mixture consisted of lithium carbonate and potassium carbonate. Multi-walled carbon nanotubes were dispersed in the solvent to synthesize the nanomaterials. A surfactant, gum arabic, at 1% mass concentration with respect to the solvent was added to disperse the carbon nanotubes homogeneously in the salt mixture. The results indicated that the specific heat capacity of the carbonate salt mixtures was significantly enhanced by adding the carbon nanotubes in the solid and liquid phases. Moreover, the enhanced specific heat capacity was affected by the chemical composition of the salt mixtures. Furthermore, molecular dynamics simulations were used to examine the effects of different chemical compositions of the binary salt mixtures with nanotubes on the specific heat capacities of the nanomaterials.

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1. Introduction

Since nanofluids have been firstly shown to significantly increase the thermal conductivity of solvents [1], the thermal conductivity of the nanofluids has been extensively studied for a variety of nanoparticles (as summarized in Refs. [2,3]). Additionally the drastically increased critical heat flux in pool boiling was also observed by You et al. [4]. These exceptional features of nanofluids have spurred research into diverse heat transfer applications and have promoted the use of nanofluids as working fluids. A number of experimental and numerical studies utilizing nanofluids in diverse fields have been reported in the past two decades [5–12]. Initially, conventional liquids, such as water, alcohol, and ethylene glycol, were used as nanofluid solvents. Over time, other solvent materials, such as alkali molten salts, have been utilized in unique nanofluid applications, such as thermal energy storage. Molten salts and their

eutectics have been considered to be thermal energy storage media for concentrating solar thermal power plants and heat transfer fluids in heat exchangers.

Recently, studies have shown that the specific heat capacity of the molten salt-based nanofluids or nanomaterials was significantly enhanced in the solid and liquid phases by the dispersion of nanoparticles in salts [13–20]. However, contradictory observations have been made on the specific heat capacity of the nanofluids and nanomaterials (summarized in Ref. [21]). In 2009, Nelson et al. [22], reported that the specific heat capacity of poly-alphaolefin–graphite nanofluids was enhanced up to 50% compared with that of the solvent and further increased with temperature. Following that study, other reports observed increased specific heat capacities of other nanofluids and nanomaterials. However, the mechanism responsible for the increased specific heat capacity remained unknown. A previous study suggested several enhanced specific heat capacity models between nanoparticles and solvent molecules [14] and reported the existence of dense liquid layering adjacent to nanoparticles. This phenomenon was confirmed experimentally and numerically [22–24]. However, there remains insufficient experimental evidence to support a definitive conclusion. Therefore, additional work is

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required to examine the effects of different experimental conditions on the specific heat capacity of different solvent materials.

The aim of this study is to investigate the effect of solvent composition on the specific heat capacity enhancement for binary carbonate salt-based nanomaterials mixed with multi-walled carbon nanotubes. As performed in a previous study [17], which tested two chemical compositions, the effects of the chemical composition of two constituent salts for the solvent phase on the specific heat capacity were examined for a wide range of compositions. To verify the effects of the solvent phase composition on the specific heat capacity, measurements of the specific heat capacity were carried out for a variety of carbonate salt chemical compositions. These measurements help determine how the thermal and chemical properties of pure solvents influence the specific heat capacity of nanomaterials formulated from nanoparticles mixed with solvent.

In the present study, the specific heat capacity of the nanomaterials was measured in the solid and liquid phases, which have different solvent chemical compositions. The chemical composition of each nanomaterial was obtained using inductively coupled plasma-mass spectrometry (ICP-MS) after specific heat capacity measurements were performed using a differential scanning calorimeter. Based on the results for the specific heat capacity and the chemical composition, the effect of the chemical composition on the thermal properties of the solvent was resolved. Finally, molecular dynamics simulations were also conducted to numerically investigate the characteristics of the compressed layers, which have been implicated in enhancing specific heat capacity, as a function of the chemical composition of the solvent material.

2. Nanomaterial synthesis

Binary carbonate salt mixtures were used as solvents for the molten salt nanomaterials. The salt mixture was composed of lithium carbonate (Li_2CO_3 , Sigma–Aldrich, St. Louis, MO) and potassium carbonate (K_2CO_3 , Sigma–Aldrich, St. Louis, MO). Multi-walled carbon nanotubes (MWCNTs, Meliorum Technologies, Rochester, NY) were dispersed in the carbonate salt mixtures ($\text{Li}_2\text{CO}_3 + \text{K}_2\text{CO}_3$). The diameter and length of the MWCNTs were 10–30 nm and 1.5 μm , respectively. All chemicals were used as received. Fig. 1 shows images of the MWCNT taken by a transmission electron microscope (TEM, JEOL JEM-2010). Fig. 2 shows the nanomaterial synthesis procedure. First, the MWCNTs were dispersed in distilled water with 0.1 wt.% gum arabic (Sigma–Aldrich, St. Louis, MO). The concentration of the MWCNTs was fixed at 1% mass ratio with respect to the base salt mixture. Gum arabic was used as a surfactant because it was reported that the gum arabic is appropriate to prevent the agglomeration of the MWCNTs during the dehydration step shown in Fig. 2 [19]. A water-MWCNT suspension was sonicated in an ultrasonic bath for 2 h

(Step 1). Afterward, the carbonate salt mixture was added to the aqueous MWCNT suspension (nanofluid), and the resultant aqueous salt-MWCNT nanofluid was subjected to sonication again for 3 h (Step 2). Finally, the aqueous salt-MWCNT nanofluid was evaporated on a large petri dish on a hot plate set at 100 °C to recover the dehydrated powder of the molten salt-based MWCNT nanomaterials (Step 3).

3. Specific heat capacity measurements

A differential scanning calorimeter (DSC, TA Instruments-Q20) was used to measure the specific heat capacity of the pure salt mixtures and nanomaterials. Based on the heat flow measured by the DSC instrument, the specific heat capacity was determined by following a standard method, American Standard Test Method (ASTM E1269) [26]. The heat flow difference between an empty pan (Tzero aluminum pan, TA Instruments) and a reference sapphire disk was recorded over the temperature. The measurements were repeated for all samples (pure salt mixture or nanomaterials) under the same thermal cycling conditions. Because this study only focused on the specific heat capacity of the molten salt-based MWCNT nanomaterials, during the heating process, the temperature was rapidly ramped at 20 °C/min. This ramp rate is fast in comparison with those used in other DSC studies [27–29], but increased the measurement accuracy of the specific heat capacity [30], because the measured heat flow was proportional to the sample mass, specific heat capacity, and heating rate, as shown in Eq. (1). A slower heating rate would have been considered if phase change-related thermal properties, such as transition temperature or latent heat, were of the primary focus.

$$\frac{dQ}{dt} = m \cdot c_p \cdot \beta \quad (1)$$

where dQ/dt is the heat flow, m is the sample mass, c_p is the sample specific heat capacity, and β is the heating rate.

The thermal cycle of the DSC was conducted between 150 °C and 560 °C. Due to thermal equilibrium issues, the first heat flow curves in all measurements for the empty pan, the sapphire disk, and the samples (pure salt mixtures and nanomaterials) were excluded in the calculation of the specific heat capacity. For the samples, the thermal cycles in the DSC were repeated 5 times to determine the reproducibility of the measurements and the variations (i.e., degradation) in the specific heat capacity. The enhancement of the specific heat capacity was examined for both solid (at 400 °C) and liquid (average from 525 to 555 °C) phases.

As shown in Fig. 3, after the evaporation step (Step 3), two different patterns (a fine salt region and a coarse salt region) formed on the petri dish. It was expected that, in the fine salt region, lithium carbonate was more concentrated, and in the coarse

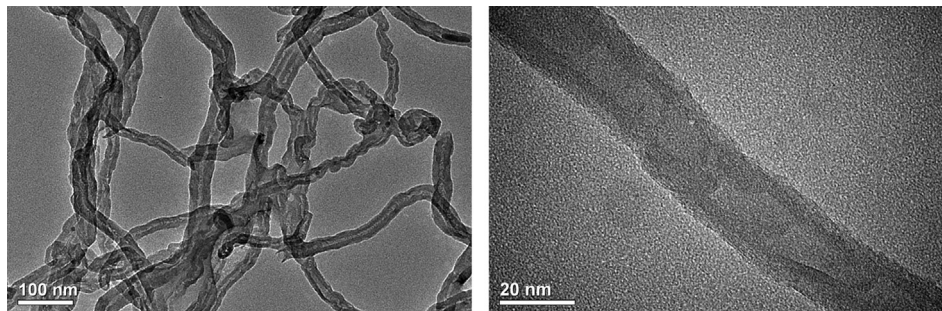


Fig. 1. Transmission electron microscope images of multi-walled carbon nanotubes.

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