



Enhanced specific heat capacity of high-temperature molten salt-based nanofluids

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ABSTRACT

Four different sized silicon-dioxide nanoparticles (5, 10, 30, and 60 nm in diameter) were dispersed in a molten salt eutectic (lithium carbonate and potassium carbonate, 62:38 by molar ratio) to obtain high temperature operating fluids (nanomaterials). A modulated differential scanning calorimeter was employed to measure the specific heat capacity of the molten salt eutectic and nanomaterials (molten salt/nanoparticle mixture). The specific heat capacity of nanomaterials was enhanced by evenly 25% over that of the base molten salt eutectic (base fluid), regardless of the size of embedded nanoparticles. The measurement uncertainty of experiments was less than 5%. Material characteristic analyses using electron microscopy show that the addition of nanoparticles into the molten salt eutectic induces nearby molten salts to form needle-like structures. These special structures were only observed within the nanomaterials whose specific heat capacity was significantly enhanced. The observed enhancements in specific heat capacity can be explained by the high specific surface energies that are associated with the high surface areas of the embedded nanoparticles and the needle-like structures induced by the nanoparticle addition.

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1. Introduction

General concentrated solar power (CSP) systems utilize solar thermal energy to generate electricity by using a general thermodynamic cycle (e.g., Rankine cycle, Stirling cycle, etc.). Hence, the overall thermodynamic efficiency depends on the operating temperature of the system, which is limited by the operating temperature of thermal energy storage (TES). Conventional TES materials are organic-based (e.g., paraffin wax, fatty acid, mineral oil, etc.), which are thermally stable only up to 400 °C. This limits the operating temperature of the system to around 400 °C [1]. Increasing the operating temperature from 300–400 °C to 560 °C, which is the creep starting temperature of stainless steel, can significantly enhance the theoretical thermodynamic efficiency (the Carnot efficiency can increase from 50% to 65%). However, very few materials are compatible for the high temperature applications.

Some molten salts (e.g., alkali nitrate salts, alkali carbonate salts, etc.) are thermally stable up to high temperatures (over 600 °C) [2]. Utilizing the molten salts as TES materials can promise several benefits. (1) High temperature stability of molten salts can allow CSP to operate at higher temperature. This can greatly enhance the thermodynamic cycle efficiency and as a result the cost of electricity by CSP can be significantly reduced. (2) Molten salts

are relatively cheap and naturally abundant compared with the conventional organic TES materials. This can dramatically reduce the cost. (3) The molten salts are environmentally safe and can reduce the potential environmental costs (e.g., material disposal). However, typical molten salts have poor thermo-physical properties, especially specific heat capacity. (c_p is less than 2 kJ/kg °C [2,3])

Nanoparticle dispersions in a liquid are termed as nanofluids [4]. Due to the very large specific surface area of nanoparticles, the effect of surface charge can be significantly increased and as a result enhance the stability of the resulting nanoparticle/liquid mixture. They have been reported in the literature for their significant enhancement in thermal conductivity [4–8]. Many attempts have been made to explain the mechanism of thermal conductivity enhancement, including: (1) Brownian motion effects [9–12], (2) Clustering of nanoparticles [13,14], (3) Liquid layering around nanoparticles [15,16], (4) Dipole–dipole interactions in nanoparticles [17], (5) Effects of motion of nanoparticles and structure of fluids around them [18], and (6) Self-ordered motion effects of nanoparticles in the host fluids [19]. However, none of them clearly explained the enhanced thermal conductivity of nanofluids and further investigations are in progress.

Similarly, specific heat capacity can be enhanced by doping nanoparticles especially when they are doped into a fluid mixture (mixture-based nanofluids). Nelson et al. [20] observed 50% enhanced specific heat capacity from polyalphaolefin (PAO)-based graphite nanofluids (0.6 wt.%). Shin and Banerjee [21,22] reported 15% and 26% enhanced specific heat capacity of molten salt

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Nomenclature

$c_{p,t}$	specific heat capacity of nanomaterial	ρ_f	density of base material
$c_{p,p}$	specific heat capacity of nanoparticle	V_p	volume fraction of nanoparticle
$c_{p,f}$	specific heat capacity of base material	V_f	volume fraction of base material
ρ_p	density of nanoparticle		

mixture-based silica nanofluids (1%). Bridge et al. [23] measured 30% enhanced specific heat capacity of ionic liquid mixture-based alumina nanofluids (less than 2.5%). However, almost no research has been reported to explore the enhancement mechanism for the specific heat capacity of the mixture-based nanofluids.

The aim of this study is to investigate the specific heat capacity mechanism of mixture-based nanofluids to explain the specific heat capacity enhancement of the nanofluids. We first synthesized several molten salt nanofluids with varying nanoparticle sizes (5 nm to 60 nm in diameter). We characterize their specific heat capacity using a modulated differential scanning calorimeter (MDSC; Q20, TA Instruments). Since specific heat capacity of a material is directly related to the phase or structure of the material, we performed material characterization analyses using a scanning electron microscopy (SEM; ZEISS Supra 55 VP) to explore the micro-structural change of the nanofluids.

2. Experiments

2.1. Nanofluid synthesis

Eutectic of lithium carbonate and potassium carbonate ($\text{Li}_2\text{CO}_3\text{-K}_2\text{CO}_3$, 62:38 by molar ratio) were procured by Spectrum Chemical Mfg. Corp. and silicon dioxide nanoparticles were procured by Meliorum Technologies, Inc. Four different nominal sizes of silicon dioxide nanoparticles (5, 10, 30 and 60 nm) were used in the experiments. Photon correlation spectroscopy was used for pre-analysis of the size of nanoparticles. The result is shown in Fig. 1. To make 200 mg of nanofluid sample (at 1% concentration by weight), 198 mg of pure eutectic salt mixture of lithium carbonate and potassium carbonate (62:38 by molar ratio) and 2 mg of silica nanoparticles were precisely mixed using a microbalance (Sartorius CPA225D). The mixture was then poured into a 25 ml glass bottle filled with 20 ml distilled water. The solution underwent an ultra-sonication process for 200 min (Branson 1510, Branson Ultrasonics Corporation) to ensure uniform dispersion and minimal agglomeration of nanoparticles. After that, the solution was put on a hot plate (Fisher Scientific, Isotemp) maintained at 200 °C to evaporate water from the solution for 3 h. The remaining nanomaterials were heated up to 300 °C for 1 h to remove any remaining water molecule (i.e., chemically bonded to the salt eutectic).

2.2. Specific heat capacity measurement

Specific heat capacity of samples was measured using a modulated differential scanning calorimeter (MDSC; Q20, TA Instruments, Inc.). To have a reliable result and a reasonable comparison, three pure samples and twelve nanomaterial samples (three of each nanoparticle size (5, 10, 30, and 60 nm) were independently prepared and tested. Standard Tzero aluminum hermetic lid/pan (TA Instruments, Inc.) was used to mount a sample in the MDSC. To remove any absorbed moisture, each sample was kept at 300 °C for 30 min on a hot plate before each experiment and each sample pan was hermetically sealed (No moisture was confirmed by the MDSC heat flow signal). A custom MDSC protocol was

programed to measure the specific heat capacity of samples according to the standard MDSC method. First of all, each sample was heated up to 560 °C to remove moisture in the MDSC cell before testing. Then, the sample was heated up to 140 °C and held at this temperature for 5 min to obtain a stable heat flux signal. The samples were then ramped up to 560 °C at a fixed ramp rate of 2 °C/min. After that, the samples were maintained at 560 °C for 5 min to reach a steady state heat flow condition. This procedure was applied to measure the specific heat capacity of pure eutectics and all nanomaterials (pure eutectic doped with nanoparticles). The sample weight was measured before and after the experiment to ensure no mass loss of the sample.

3. Results and discussion

3.1. Specific heat capacity results

The specific heat capacity of the pure eutectics and the nanomaterials in the solid phase and the liquid phase are shown in Tables 1 and 2. Figs. 2–5 are presented to show the variation of specific heat capacity of the pure eutectics and the nanomaterials in the solid phase, in the liquid phase, and in the full temperature range. As it is shown in the tables and figures, the average values of specific heat capacity of pure eutectic samples are 1.25 J/g K and 1.59 J/g K in the solid and in the liquid phases, respectively. It makes a very good agreement with the literature value [3]. The average specific heat capacity of nanomaterials is 1.54–1.61 J/g K in the solid phase and 1.95–2.01 J/g K in the liquid phase. The average enhancements of specific heat capacity are 23–28% in the solid phase and 22–26% in the liquid phase, respectively. Considering the measurement uncertainty of 3%, the average enhancement in the specific heat capacity is 25% regardless of the nanoparticle size (5, 10, 30, and 60 nm). The values of enhanced specific heat capacity of nanomaterials agree with the reported specific heat capacity enhancement of 26% (with 35 nm nanoparticles) [21]. The results from MDSC tests were compared with the classical heat capacity model based on the thermal equilibrium (i.e., simple mixing theory) as follows:

$$C_{p,t} = \frac{V_p \rho_p C_{p,p} + V_f \rho_f C_{p,f}}{V_p \rho_p + V_f \rho_f} \quad (1)$$

where c_p is specific heat capacity, V is volume fraction, and ρ is density. The subscripts, t , p , and f denote nanofluid, nanoparticle, and pure eutectic, respectively. The specific heat capacity of nanoparticle and pure eutectic is about 1.0 J/g °C and 1.6 J/g °C, respectively. Moreover, the nanoparticle concentration was very small (1% by weight). Therefore, based on the theoretical approach by Eq. (1), the specific heat capacity of nanofluids should be slightly less than pure eutectic (1.6 J/g °C). However, it was found in the experiments the specific heat capacity was enhanced by 25%. This implies that the observed enhancement in the specific heat capacity cannot be explained by the conventional macro-scale heat transfer.

3.2. Material characterization

Generally heat capacity of a material is closely related to the phase and structure of the material. Hence, we performed material

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