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A novel phase change material containing mesoporous silica nanoparticles for thermal storage: A study on thermal conductivity and viscosity ☆

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ABSTRACT

In this research, mesoporous silica ($MPSiO_2$) nanoparticles were dispersed in n-octadecane as an organic phase change material (PCM) in order to produce a novel composite for thermal storage. Stable PCMs containing 1 wt.%, 3 wt.% and 5 wt.% $MPSiO_2$ nanoparticles (PCM/$MPSiO_2$) were fabricated by dispersing $MPSiO_2$ in PCM. $MPSiO_2$ particles were investigated by SEM and TEM techniques, which showed high order of porosity and spherical particles of ca. 300 nm. The thermal conductivity in both solid and liquid phases was measured by transient plane source (TPS) technique in the temperature range of 5–55 °C. A maximum thermal conductivity enhancement of 5% for 3 wt.% $MPSiO_2$ at 5 °C, and 6% for 5 wt.% $MPSiO_2$ at 55 °C was experimentally obtained. Moreover, it was observed that enhancement in thermal conductivity is non-monotonic in solid phase with increasing $MPSiO_2$ particle loading. The viscosity results showed that for mass fractions of nanoparticles greater than 3% in liquid PCM, the behavior of liquid is non-Newtonian. Also, the viscosity of PCM containing $MPSiO_2$ nanoparticles was measured to be increased up to 60% compared to the liquid PCM for 5 wt.% $MPSiO_2$ at 35 °C.

1. Introduction

Phase change materials (PCMs) are extensively employed as thermal energy storage media in many applications because of their capacity of storing and releasing a large amount of thermal energy during melting and solidifying at phase change temperature. These materials have relatively low thermal conductivity (TC) that reduces the melting–solidification (charging–discharging) rate [1]. Several methods such as using fins, metal foams, and graphite matrix [2,3] have been proposed to enhance the heat transfer performance of PCM storage. Dispersing a small amount of nanostructured materials (nanoparticles, nanowires, nanotubes, nanofibers etc.) in a heat transfer media has received increasing attention in recent years. Nanofluids provide an excellent example of suspending such materials in heat transfer fluids to improve thermal performance of system by increasing fluid TC [4]. The addition of highly conductive nanostructured material to PCMs creates new composite materials possessing enhanced TC which shows promise for thermal energy storage. A review of studies focused on enhancing the thermal conductivity of PCMs for thermal energy storage upon introduction of nanostructures was recently presented by Khodadadi et al. [5]. The advantages of PCMs containing nanostructured materials compared to conventional TC enhancement methods include their lighter weight, no contact heat transfer issues, availability of natural convection to enhance heat transfer during melting, and their easy recycling [5].

Numerous types of nanostructured materials such as carbon nanotubes [6–14], carbon nanofibers [12,15], graphite nanofibers [16,17], graphite nanoplatelets [18,19], graphene [14,20], oxide nanoparticles (Al_2O_3 [21–23], Fe_3O_4 [24], TiO_2 [25,26], SiO_2 [27,28], ZrO_2 [29], CuO [30–32]), metallic nanoparticles (Ag [33], Al [34], Cu and C/Cu [35]) and Ag nanowires [36] were explored as dispersed phase in synthesis of PCM-nanostructured materials. Paraffin wax [8,14–17,23,28,33,34], palmitic acid [9–11], n-docosane [18,19], n-octadecane [22], n-eicosane [30] and water [20,25] were often used as PCM, or continuous medium for dispersion. Generally, a two-step method is used for preparation and dispersion of nanostructured materials in PCM liquid phase [5]. Evaluation of the TC of PCM by dispersing nanostructured materials has received significant attention in recent years [6,8–10,14,16,21,22,30,31,35]. Generally, all investigators emphasized TC enhancement by adding nanostructured materials to PCMs. For example, Ho and Gao [22] measured the relative enhancements of more than 2% and 6% in the TC for the paraffin containing 5 wt.% and 10 wt.% of alumina

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nanoparticles at 30 °C, respectively. They found a relative increase of more than 17% in TC when temperature was increased up to 60 °C. Fan [30] indicated that the enhancement of TC is not generally monotonic with the mass concentration of nanoparticles, moreover the experiments showed that the greatest enhancement of about 9% relative to pure eicosane is observed for the 2 and 10 wt.% samples at 20 °C. The enhancement of TC by dispersing carbon based nanomaterials in PCM has been higher than that of oxide and metallic nanoparticles, because of their superior TC of the order of 1000 W/m·K. Wang et al. [11] reported that the largest TC enhancements of the palmitic acid are 46.0% at 25 °C and 38.0% at 65 °C for the composite with 1 wt.% treated multi-walled carbon nanotubes (TCNT) in solid state and liquid state, respectively. Yavari et al. [20] achieved the enhancement of 140% in TC by dispersing 4 wt.% graphene in 1-octadecanol as PCM. Transient hot wire technique (THW) [7,8,10,11,13,14,22,23,25] and transient plane source (TPS) [6,9,30,31,35] have been the most common two techniques for TC measurement. In the available literature, attention has rarely been paid to other thermophysical properties, like viscosity of PCMs. Nonlinear dependency on the mass fraction of nanoparticles for the effective dynamic viscosity of the nanoparticle-in-paraffin emulsion was displayed by Ho and Gao [22]. Moreover, at a temperature of 30 °C, relative increases of nearly 20% and more than 28% in the dynamic viscosity were found for the paraffin containing 5 wt.% and 10 wt.% of alumina particles, respectively. Kumaresan et al. [13] experimentally showed the shear thinning behavior at the lower shear stress range (0–1 Pa) and the Newtonian behavior at the higher shear stress range (1–10 Pa) for the nanofluid phase change material. Also, the abnormal increase in viscosity of nanofluid PCMs with increase in volume fraction of multi-walled carbon nanotubes (MWCNT) was observed.

The mesoporous silica ($MPSiO_2$) has been considered as a fascinating material for many technological applications due to its porous and morphological characteristics [37]. $MPSiO_2$ nanoparticles were employed previously by Nikkam et al. [38] to produce nanofluids for heat transfer enhancement. In the present research, to our knowledge, $MPSiO_2$ nanoparticles were used as nanostructured material for dispersing in n-octadecane as PCM for the first time. After characterization and dispersion of $MPSiO_2$ in PCM, the TC of PCM/$MPSiO_2$ was investigated experimentally in both solid phase and liquid phase. Due to the dependence of natural convection on the Rayleigh number and therefore on viscosity, the rheological behavior of the PCM containing $MPSiO_2$ nanoparticle was examined.

2. Experimental

2.1. Materials preparation

$MPSiO_2$ particles used in this study were supplied by Nanologica AB, Sweden. The PCM was n-octadecane ($C_{18}H_{38}$ of 99% purity) with a nominal melting point of 27.5 °C and it was obtained from Alfa Aesar®, Germany. The materials were used as-received, without further purification. A certain amount of $MPSiO_2$ nanoparticles was dispersed in PCM to obtain PCM/$MPSiO_2$ with different mass fractions of $MPSiO_2$ (1%, 3% and 5%). Since liquid n-octadecane has high dissolved air content, the PCM should be degassed carefully prior to measurements. For this purpose, the PCM was melted under vacuum. PCM/$MPSiO_2$ samples were prepared by a two-step method. $MPSiO_2$ particles were added to liquid PCM at a temperature around 50 °C. Then the mixture was stirred by a mechanical stirrer at 1000 rpm and sonicated in an ultrasonic bath (VWR, USC2100D, Germany) for 30 min. This ensured uniform dispersion of $MPSiO_2$ particles in the liquid PCM.

2.2. Characterization techniques

Scanning Electron Microscopy (SEM) analysis of $MPSiO_2$ particles was performed by using FEG-HR SEM (Zeiss-Ultra 55). Transmission Electron Microscopy (TEM) analysis of the particles was performed using JEOL 2100 at 200 kV acceleration.

The TC of PCM/$MPSiO_2$ samples was measured by a thermal constants analyzer (TPS 2500, Hot Disk AB, Sweden) that is based on the transient plane source (TPS) technique. The method is based on the use of a transiently heated plane sensor. The Hot Disk sensor consists of an electrically conducting pattern in the shape of a double spiral, which has been etched out of a thin Nickel foil. This spiral is sandwiched between two thin sheets of an insulating material (Kapton). When performing a thermal transport measurement, the plane Hot Disk sensor is fitted within the sample. By passing an electrical current, high enough to increase the temperature of the sensor between a fraction of a degree up to several degrees, and at the same time recording the resistance (temperature) increase as a function of time, the Hot Disk sensor is used both as a heat source and as a dynamic temperature sensor. A cylindrical aluminum block with a hole (diameter of 25 mm and height of 15 mm) drilled on top surface, was fabricated to provide a constant temperature sample holder. The sensor of the equipment was fixed on the sample holder to submerge in liquid phase or sandwich in solid phase PCM. The sample holder block was placed within the reservoir of a constant temperature bath (Thermo Haake, C50P, Germany) to control measurement temperature. For measuring TC in solid phase, bath temperature was set to 0 °C and the holder was filled with liquid PCM layer-by-layer to prevent the formation of internal voids or air bubbles.

The rheological properties of PCM/$MPSiO_2$ samples in liquid phase were measured by a viscometer (LVDV-II + Brookfield programmable viscometer, USA) with a temperature-controlled bath. Viscosity measurements were performed at 35 °C, 45 °C and 55 °C while fresh sample was used for each measurement. Spindle ULA was used in this viscometer and was calibrated by using Brookfield viscosity standard fluids. All the viscosity measurements were recorded at steady state conditions.

2.3. Calibration experiments

In order to provide reasonable results, the Hot Disk Thermal Constants Analyzer was calibrated against known values of TC prior to experiments. Thermal conductivities of distilled water (DW) and a mixture of ethylene glycol and distilled water (EG/W) (50% by weight) were measured by the instrument (with the same as sample holder and sensor of other measurements) at 20 °C and 0.1 MPa. The measurements were repeated five times. Table 1 shows the mean value ($\bar{k}$) and standard deviation (SD) of measured thermal conductivities as well as reference values [39,40]. The SD value less than 0.001 shows a small value of scatter around the mean values. Moreover, the deviation of measured thermal conductivities from reference values is less than 1% that successfully verified the accuracy of 5% that is specified by the manufacturer.

Table 1
Measured and reference values of thermal conductivity of DW and EG/W (50 wt.%).

Fluid	Temperature (°C)	$\bar{k}$ (W/m·K)	SD	k_{ref} (W/m·K)	$\left(\bar{k}-k_{ref}\right)/k_{ref}$ (%)
DW	20	0.603	0.001	0.598 [39]	0.836
EG/W 50 wt.%	20	0.390	0.0006	0.389 [40]	0.222