



# Thermal conductivity enhancement of lauric acid phase change nanocomposite in solid and liquid state with single-walled carbon nanohorn inclusions



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## ABSTRACT

We prepared lauric acid based phase change nanocomposite embedded with chemically functionalized single-walled carbon nanohorns and measured its thermal properties. We report contrasting enhancements in thermal conductivity of such nanocomposites in the solid and liquid phase for the same loading of nanohorn inclusions. Maximum thermal conductivity enhancement in solid and liquid phase at 2 vol% is found to be  $\sim 37$  and  $\sim 11\%$ , respectively. The nanocomposites' thermal conductivity enhancement is compared with calculations of effective medium theory considering the role of interfacial thermal transport. Model calculations show that Kapitza resistance is an order of magnitude lower at the solid–solid interface compared to the solid–liquid interface. Differential scanning calorimetry study of the nanocomposites shows that the phase change temperature and enthalpy marginally increases to that of pristine material. Such a nanocomposite with enhanced thermal transport and phase change enthalpy makes it a promising candidate for thermal energy storage applications.

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## 1. Introduction

Nanomaterials offer a facile way to enhance the thermophysical properties of heat transfer fluids, latent heat storage and thermal interface materials [1–3]. Manipulating the thermal transport properties of conventional materials using nano inclusions has several practical applications of technological importance in the field of thermal energy storage and thermal management of microelectronic devices [4–6]. Organic phase change materials, often employed in thermal energy storage applications possess low thermal conductivity which limits the energy storage and release rate [7]. This has resulted in intense efforts to increase the thermal conductivity of phase-change materials (PCM) through the addition of high thermal conductivity nanomaterials [8–13].

High conductive metallic and metallic oxide nanoparticles and carbon allotropes have been used for enhancing the thermal conductivity of PCMs as reviewed by Khodadadi et al. [14]. However, most of the thermal conductivity results reported in the literature correspond to the solid phase of the PCM. Recently,

Zheng et al. [15] reported that the thermal and electrical conductivity of *n*-hexadecane based phase change nanocomposites can be manipulated by solid–liquid phase transition process using graphite nanoparticles. Schifferes et al. [16] with 1 vol% of *n*-hexadecane/graphene nanocomposites showed that by varying the cooling rate of the phase change material from  $10^{-3}$  °C/min to  $10^2$  °C/min, the aspect ratio of crystal can vary from micrometers to several millimeters. For such nanocomposites, they reported that the electrical and thermal conductivity contrast ratio (solid state conductivity to the liquid state conductivity) can vary by a factor of 5 and 2 respectively. Harish et al. [17] reported that for *n*-octadecane/single walled carbon nanotube nanocomposite the thermal conductivity enhancement in liquid state is nominal and in good agreement with the predictions of classical theoretical models, while in solid state, the thermal conductivity enhancement is anomalous and beyond the predictions of classical theories. Recently, Angayarkanni and Philip [18] reported that the thermal conductivity of *n*-hexadecane can be significantly enhanced using inexpensive cationic and anionic surfactants. They reported that the linear chain surfactant micelle entrapped within the grain boundaries upon solidification enhance the thermal conductivity of phase change material significantly. However, surfactant micelles possess lower thermal conductivity than many

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conventional PCMs. Hence, the role of surfactant micelles in enhancing the thermal transport of PCMs upon solid–liquid phase transition remains unclear.

Based on the previous studies, it is evident that limited thermal conductivity enhancement results exist in the literature about the effect of nanomaterials on both the solid and liquid phase of PCMs which is important to understand the melting and freezing characteristics of such materials. Also, a significant amount of literature reported about the effect of several nanoparticles on paraffins [14]. Experimental results on the effect of nanoparticles on fatty acid based PCMs remains scant [8] and none have explored the potential of single walled carbon nanohorns as a filler candidate to enhance the thermal properties of phase change materials.

Single walled carbon nanohorns (SWCNH) are an attractive new form of nanocarbon, which is similar to single walled carbon nanotubes. The tubular nanostructure has a horn shaped cap with a cone angle of  $20^\circ$  at one end [19]. In this work, the authors utilize chemically functionalized SWCNH nano inclusions to investigate the thermal conductivity enhancement of phase change nanocomposites. We demonstrate that the thermal conductivity enhancement varies significantly based on the state of the material for the same loading of nanomaterial. For SWCNH/organic fatty acid nanocomposite, the thermal conductivity contrast ratio (solid state thermal conductivity to liquid state thermal conductivity) can be increased by a factor of  $\sim 2$  during the solid–liquid phase transition process.

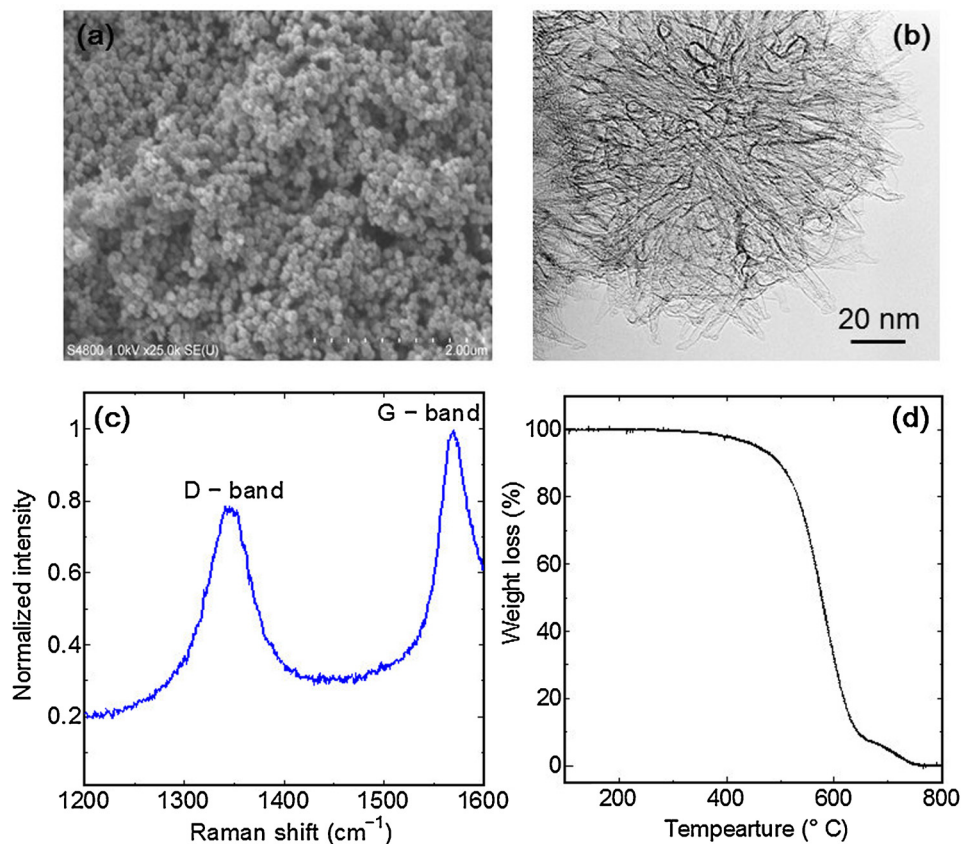
## 2. Materials and methods

### 2.1. Sample preparation

In the present work, we made use of *n*-dodecanoic acid (lauric acid,  $C_{12}H_{24}O_2$ ) as the phase change material with a phase transition temperature of  $\sim 43^\circ\text{C}$ . Oxidized single walled carbon nanohorns (SWCNH) were purchased from NEC Corporation, Tokyo with a diameter of 2–5 nm, length 40–50 nm, density  $1.1\text{ g/cm}^3$  and specific surface area of  $\sim 1300\text{ m}^2/\text{g}$ .

Fig. 1(a and b) shows the scanning electron microscopy (SEM) and transmission electron microscopy (TEM) visualization of the SWCNH used in this study. Fig. 1(c) shows the typical Raman spectra of the SWCNH obtained with a laser wavelength of 488 nm. The G-band observed around  $1582\text{ cm}^{-1}$  corresponds to the in-plane vibrations of the carbon atoms. The large D-band (disorder band) observed around  $1350\text{ cm}^{-1}$  corresponds to the defects in the graphitic structure due to oxygen functionalization. Fig. 1(d) shows the thermogravimetric analysis (TGA) results of the nanomaterial. The TGA results show the absence of amorphous carbon impurities and the absence of metallic impurities.

The raw materials were used as-such in this study without any further purification. The SWCNH samples were pre-dried in vacuum at  $150^\circ\text{C}$  to remove the moisture content. The nanocomposite samples were prepared by adding SWCNH inclusions with molten lauric acid kept on a hot plate maintained at a temperature of  $100^\circ\text{C}$  under rigorous stirring using a magnetic



**Fig. 1.** (a) SEM visualization of SWCNH nano inclusions used in this study. (b) TEM visualization of “dahlia-like” SWCNH nano inclusions used in this study. Image captured at an acceleration voltage of 200 kV. (c) Typical Raman spectra of SWCNH inclusions. The G-band at  $1582\text{ cm}^{-1}$  correspond to the in-plane vibrations while the D-band represent the structural defects due to chemical functionalization. (d) TGA analysis of functionalized SWCNH nano inclusions. The results show the nanomaterials burn uniformly at  $\sim 600^\circ\text{C}$ . The results demonstrate the absence of amorphous carbon and metallic impurities.

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