



# Effect of carbon nanotube interfacial geometry on thermal transport in solid–liquid phase change materials



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## HIGHLIGHTS

- Nanoparticle constriction effect on the thermal conductivity of a PCM is quantified.
- Interfacial thermal resistance is found as a function of the constriction geometry.
- Contact area between nanoparticles has a significant impact on thermal transport.

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## ABSTRACT

For nearly two decades, research groups have attempted to increase the thermal conductivity of bulk materials by saturating them with different concentrations and types of nanoparticles. However, optimal enhancements in the thermal conductivity of these materials continue to go unrealized, primarily due to interfacial phenomena that impede phonon propagation from the bulk material to the nanoparticle or between individual, contacting nanoparticles. Though it is almost certain that interfacial resistances between contacting nanoparticles are responsible for the underwhelming thermal performance of nanoparticles in bulk materials, the physical mechanisms that limit thermal transport at nanoparticle junctions is not well understood. In this study, we investigate the effect of nanoparticle constriction size (i.e. the contact area between linked nanoparticles) on the bulk thermal conductivity of a surrounding paraffin-based phase change material. To this end, we measure the effective thermal conductivity of different Multi-walled Carbon Nanotube (MWCNT)/paraffin nanocomposites is measured with the transient plane source technique. Using a newly developed physical model, the interfacial thermal resistance between the contacting nanoparticles is determined as a function of the constriction geometry. Results suggest that the thermal conductance (i.e. the rate of heat transfer) between contacting MWCNTs can be increased by a factor of 27 by increasing their diameter by only 58 nm. This is in direct conflict with effective medium approximations, which predict an increase in the bulk thermal conductivity of MWCNT–PCM composites when MWCNT diameters are reduced. Such a result indicates that the contact area between individual, contacting nanoparticles has a significant impact on thermal transport within the PCM nanocomposite, and therefore its bulk thermal conductivity. It is expected that these results will strongly impact the design of nanoparticle-laden PCMs.

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## 1. Introduction

Organic phase change materials (PCMs) have the ability to remove and store heat passively, thereby making them excellent candidates for the thermal management of portable electronic devices [1] and electric vehicle batteries [2], as well as for storing and reusing thermal energy in residential solar-thermal and

HVAC systems [3–5]. The amount of heat that can be stored within a PCM is governed by its phase change enthalpy, which is typically high for commercial paraffin waxes (on the order of 200 kJ/kg [6,7]). As a result of their high phase change enthalpies, the amount of heat that can be stored in paraffins on a per unit mass basis is expected to be sufficient for meeting the performance expectations of the aforementioned applications.

Unfortunately, paraffin-based PCMs have exceptionally low thermal conductivities (on the order of 0.1 W/mK [8]). This is over four orders of magnitude lower than the thermal conductivity of

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conventional heat transfer materials (oxygen-free copper, for instance, has a thermal conductivity of  $\sim 400$  W/mK [9]). The poor thermal conductivities of organic PCMs result in extremely slow melt and solidification rates and thus very little utilization of the PCM mass in thermal energy storage applications [6,7,10]. Low PCM thermal conductivities can also result in severe thermal gradients in close proximity to sensitive electronics and thus failure of the primary electronic component(s) in critical energy infrastructure. Consequently, it is imperative that the thermal conductivity of organic PCMs be increased.

In this work, multi-walled carbon nanotubes (MWCNTs) are inserted into an organic paraffin PCM in order to improve its bulk thermal conductivity. MWCNTs are known to have remarkably high intrinsic thermal conductivities (as high as 6000 W/mK [11]), low densities and high surface area-to-volume ratios, making them excellent candidates for PCM thermal enhancement in applications with weight and volume constraints. Additionally, the thermal conductivity of MWCNTs can be nearly an order of magnitude greater than some of the conventional materials used in thermal management systems, primarily due to the strong  $sp^2$  bonds that are formed between neighboring carbon atoms and the long mean free path of phonons within the nanoparticle itself [12,13] ( $\sim 1$   $\mu\text{m}$  for CNTs and graphene [14]).

However, the incorporation of nanoparticles into PCMs for thermal augmentation is not straightforward. This is primarily due to the fact that standard effective medium approximations often over predict the thermal conductivity of nanocomposite materials. Instead, the thermal conductivity enhancement of a nanocomposite is generally inhibited by a thermal resistance that occurs across nanoparticle interfaces [15]. This thermal resistance is governed by a variety of geometric, physical and chemical phenomena that occur at the nanoparticle–nanoparticle junction(s), and can be substantial when the nanoparticles have rough surfaces or extend in multiple dimensions [14] (as high as  $10^{-4}$   $\text{m}^2$  K/W for carbon nanofibers, for instance [6]). On the other hand, recent molecular dynamics simulations suggest that the presence of carbon-based nanoparticles with ‘smooth’ surfaces (such as CNTs and graphene) results in the alignment of alkane molecules across carbon nanoparticle–paraffin interfaces. In turn, a significant reduction in the interfacial thermal resistance at this type of interface can be achieved [16]. Consequently, the use of carbon-based nanoparticles for enhancing the thermal conductivity a paraffin-based PCM offers the most promising method for their thermal conductivity enhancement to date.

Several groups have decreased the thermal resistance at nanoparticle–nanoparticle junctions by increasing the adhesion energy across contacting interfaces. This technique generally results in mechanically strong, percolating networks [17–20]. In these studies, assortments of chemical modifications are made at the nanoparticle junction(s) in order to achieve higher rates of thermal transport and increased mechanical stability. However, it is not known whether the thermal conductivity of these percolating nanocomposite PCMs can be further tuned by adjusting the size of the constriction at the interface. Preliminary molecular dynamics simulations suggest that the size of the constriction does affect thermal transport at an individual nanoparticle–nanoparticle junction [21,22], but the range of nanoparticle sizes used for these predictions is too small to make accurate estimations about its influence on both nano-scale and macro-scale thermal transport within PCMs. Additionally, it is unknown to what degree ballistic and diffusive transport affect heat flow rates at these junctions. As a result, additional work is needed to determine the effect of nanoparticle contact area on the thermal performance and associated nano-scale and macro-scale thermal characteristics of nanocomposite PCMs.

In this study, four different MWCNTs with varying diameters and lengths are embedded within an organic paraffin PCM at concentrations above and below the percolation threshold. The objectives of this study are to determine the effect of CNT contact area on: (1) the interfacial thermal resistance between a CNT and another CNT and (2) the ballistic and diffusive components of heat transfer on heat flow at the interface. The effective interfacial thermal resistance between two linked CNTs is calculated by saturating the PCM with different volume concentrations of CNTs and applying the effective medium model for thermal conductivity of networked composites developed by Foygel et al. [23]. The procedure for this method is described in [15]. The insight gained from this study will allow for the design of nanocomposite PCMs with tunable thermal conductivity. In this case, thermal engineers will be able to tune the thermal properties of disordered, bulk materials based on the phonon physics that occur at sub-micron CNT interfaces. Such insight will allow for the thermal optimization of next-generation CNT-based composite materials.

## 2. Experimental and analytical methods

### 2.1. Physical properties of materials

The geometric properties for each of the different nanoparticles that are used in this study are listed in Table 1. In total, four different types of CNTs are used to produce the nanocomposites for this work. The values for the diameter, length and density of the nanoparticles are obtained from the manufacturer (US Research Nanomaterials, Inc.).

In Table 1,  $\phi_c$  is the volume fraction at which the nanoparticles begin to percolate and  $A_o$  is the average overlap area between nanoparticles (which is distinctly different from contact area, mentioned later). Eq. (1), developed by Foygel et al. [23] (and later confirmed by Wemhoff [24]), is used to determine  $\phi_c$ .

$$\phi_c = \frac{0.6}{p} \quad (1)$$

In Eq. (1),  $p$  represents the aspect ratio of the nanoparticle. The average overlap area between nanoparticles can be calculated according to Wemhoff [24] and is determined using Eq. (2).

$$A_o = \frac{2D^2}{\pi} \zeta(p) \quad (2)$$

where

$$\zeta(p) = \ln \left[ \frac{\sqrt{1+p^{-1}} + \sqrt{1-p^{-1}}}{\sqrt{1+p^{-1}} - \sqrt{1-p^{-1}}} \right] \quad (3)$$

In Eqs. (2) and (3),  $D$  is the diameter of the nanotube, and  $p$  is the aspect ratio of the nanoparticle ( $p = L/D$ , where  $L$  is the length of the nanoparticle).

The thermophysical properties of the commercial paraffin (IGI 1230A) used in this work are listed in Table 2 and are supplied by the manufacturer.

**Table 1**  
MWCNT physical properties.

$v$	Density (kg/ $\text{m}^3$ )	Diameter (nm)	Length ( $\mu\text{m}$ )	Aspect ratio	$\phi_c$ (%)	$A_o$ ( $\text{nm}^2$ )
1	2100	7	20	2857.0	0.022	270
2	2100	10	50	5000.0	0.012	587
3	2100	25	15	800.0	0.078	2390
4	2100	65	15	230.8	0.269	16,500

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