The Prediction of thermal properties of single walled carbon nanotube suspensions

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ABSTRACT

The present work is a systematic numerical study of the thermal properties of single walled carbon nanotubes (SWNTs) in suspensions. A computational model, based on the simulation of the random movement of Brownian thermal walkers in aqueous and in oil suspensions of SWNTs, was used to investigate the effect of the SWNT aspect ratio, weight fraction and of the interfacial thermal resistance on the suspension effective thermal properties. The dependence of the effective thermal conductivity on the temperature for aqueous suspensions was also investigated.

Keywords: SWNT-in-suspension, Random walk, effective thermal conductivity

1. Introduction

Recent studies show that single walled carbon nanotubes (SWNTs) could enhance mechanical and electrical transport properties¹ and thermal transport properties^{2,3} of materials that incorporate SWNTs. Investigations have not focused much on thermal conductivities of fluids containing dispersed SWNTs. The classic models that describe the effective conductivity of suspensions are based on microscopic rather than nanoscale considerations. They have been derived assuming that the continuum approximation holds and they do not account for ballistic heat transfer. The variables that determine the effective conductivity are the particle shape and the volume fraction^{4,5}. The thermal conductivity of nanofluids also depends on possible epitaxial layering of the fluid molecules in the molecular layers adjacent to the suspended nanoparticles⁶ and on the temperature. The existence of a thermal resistance⁷⁻¹¹ to the transfer of heat between the nanoscale inclusions (SWNTs) and the surrounding matrix (suspending liquid) can also result in anomalous heat transfer behavior. As there are no accurate and reliable theoretical formulas currently available to predict the thermal conductivity of nanofluids satisfactorily, it is quite valuable to systematically exploit the thermal properties of SWNT-in-fluid suspensions by a numerical method.

Previous studies of the thermal conductivity of SWNT suspensions and of the thermal resistance when heat is crossing the SWNT-matrix interface include the work of Maruyama *et al.*¹⁰,

who studied the thermal conductance between a SWNT and confined water by molecular dynamics (MD) simulations. A (10, 10) SWNT with a length of 20.1 nm and a diameter of approximately 1.0 nm was simulated in a 20.1 x 10 x 10 nm fully periodic simulation cell. The SWNT contained 192 water molecules into its hollow space. Initially water molecules and the SWNT were equilibrated at 300°C and then the SWNT was suddenly heated up from 300°C to 400°C. By observing the heat transfer from the heated SWNT to the water, and using the lumped capacity method, Maruyama et $al.^{10}$ found that the thermal boundary resistance R_{bd} between the SWNT and the water was 2.0 x 10⁻⁷ $m^{2}K/W$ (thermal boundary conductance, $K_{bd} = 1/R_{bd} = 5 MW/m^{2}K$). Huxtable *et al.*¹¹ used picosecond transient adsorption to measure the interface thermal conductance of carbon nanotubes suspended in surfactant micelles in water. Their experimental results showed that the interface thermal conductance did not depend critically on the surfactant as long as the surfactant was not covalently bonded to the nanotube. The thermal boundary resistance was measured to be 0.83×10^{-7} $m^{2}K/W$ (K_{bd} = 12 MW/m²K). Huxtable *et al.*¹¹ also conducted MD simulations of heat flow from (5,5) SWNTs of various lengths to a surrounding octane liquid kept at standard conditions. Through the equilibration simulations, the thermal boundary resistance between the SWNTs and octane was calculated to be 0.4 x 10^{-7} m²K/W (K_{bd} = 25 MW/m²K). Even though both MD and experimental results are very rough estimates, one could assume that the typical thermal boundary resistance between SWNTs and fluids is $0.4 - 2.0 \times 10^{-7} \text{ m}^2\text{K/W}$ (K_{bd} = $5 - 25 \text{ MW/m}^2\text{K}$).

The simulation and experimental data reviewed above were used to study the thermal conductivities of SWNTs in water and oil engine suspensions in the present work. Table 1 shows the technical data of engine oil and water at different temperatures, and the velocity of sound used for the simulations. The thermal conductivity of SWNTs dispersed randomly in water and engine oil, and its temperature dependence, were studied by the random walk algorithm of Duong *et al.*¹². This computational model is an improvement of previous Monte Carlo-based models¹³⁻¹⁵ and has been validated with experimental data of SWNT-polymer composites.¹² The random walk algorithm is much faster than an MD algorithm. Since the carbon nanotube thermal conductivity is several orders of magnitude larger than the thermal conductivity of the fluid surrounding the SWNTs, there is no need to model random walks within the nanotubes. One can instead assume a uniform distribution of thermal walkers inside each SWNT, which is equivalent to the assumption of an infinite thermal conductivity of the SWNTs.

The effects of different weight fraction and aspect ratio of SWNTs in the suspension and of different thermal boundary resistance on the suspension effective properties are quantified in this work. As there has been no experimental or simulation work studying the temperature dependence of the effective thermal conductivity of the SWNT suspensions, the thermal conductivity dependence on temperature of the SWNT-in-water suspension is also studied. Liquid suspension of SWNTs is a basic step in the separation process of nanotube bundles, or of sorting nanotubes by length, diameter or roll-up vector. Understanding the thermal properties of SWNT suspensions can be important in scaling up and optimizing such processes.

2. Simulation methodology

The aspect ratio of SWNTs, as found from experiments, is very large, since the average SWNT diameter, D, is usually not greater than 3.0 nm, while the length of SWNTs, L, can be up to several micrometers¹⁶. Molecular dynamics¹⁷ showed no length dependence of the thermal conductivity of SWNTs, but recent measurements by Wang et al.¹⁸ have shown that the thermal conductivity of SWNTs depends on their length for L<2.5µm. However, the measured change was from ~2600 W/(mK) to ~3500 W/(mK). Both these values are four orders of magnitude higher than the thermal conductivity of the matrix material used herein (see Table 1). We take advantage of this difference in our computational algorithm, by distributing the heat markers uniformly inside a SWNT (see the rules of marker motion below). Using the effective medium theory¹⁹, one can determine¹² that the effects of the aspect ratio on the effective thermal conductivity are negligible for L/D > 20, when there is no percolation through the composite and when the aspect ratio is large, but not infinite. (Note that, according to the analysis of Nan et al.¹⁹ for the case of composites with inclusions that exhibit thermal resistance at the interface between inclusions and matrix material, when L is comparable or equal to the dimensions of the composite, the effective thermal conductivity of the medium for long continuous fibers becomes equal to the result found by the rule of mixtures.¹⁹) In the simulations conducted in this study, an aspect ratio of L/D = 40, 80 and 120 was chosen, and the SWNT diameter was set to be constant with a value of D = 2.40 nm.¹²

The computation of the effective transport coefficients is based on an off-lattice Monte Carlo that has been described at length elsewhere.^{12,15} The computational domain for the numerical simulation is a rectangular cell with SWNTs dispersed randomly and with random orientation. The size of the computational domain for the simulations is presented in Table 2. The computational cell is heated from one surface (the x = 0 plane) with the release of 90000 walkers distributed randomly and uniformly on that surface. The walkers exit at the surface opposite to the heated surface. The cell is periodic in the other two directions. The displacement of the walkers in the matrix is due to Brownian motion and can be described by a normal distribution with a zero mean and a standard deviation that depends on the matrix thermal diffusivity, $D_{\rm m}$. The standard deviation of the distribution in each one of the space dimensions is $\sigma = \sqrt{2D_m\Delta t}$ where Δt is the time increment. The

Brownian motion trajectories of all these random walkers are monitored in time and space for 100 ns with a time increment of 0.02 ns. The temperature distribution is calculated from the number of walkers found in each bin of the computational domain, and it is proportional to the number of walkers in each bin.¹⁵

The details of the algorithm and the physical assumptions of our approach are detailed in ref.12. In summary, the rules of motion of the random walkers are (1) walkers distribute uniformly once inside the SWNTs due to the high SWNT thermal conductivity relative to the thermal conductivity of the surrounding material; (2) the SWNTs are assumed to be dispersed in a way that they do not form bundles and do not bend; (3) the transfer of heat is passive; (4) the thermal boundary resistance is the same for walkers coming in and out the SWNTs, i.e., once a walker in the fluid reaches the interface between the fluid and a SWNT, the walker will move into the SWNT phase with a probability f_{f-CN} , which represents the thermal resistance of the interface and will stay at

the previous position in the fluid with a probability $(1-f_{f-CN})$. Similarly, once a walker is inside a SWNT, the walker will re-distribute randomly within the SWNT with a probability $(1-f_{CN-f})$ at the end of a time step, and will cross into the fluid phase with a probability f_{CN-f} ; (5) the volume fraction of SWNTs in every slice (i.e., every *x* plane) of the computational domain is equal to the volume fraction of the SWNTs in the composite, so that the weighted average of the product of the density times the heat capacity for a slice of the composite is the same throughout the domain; and (6) the boundaries on the *y* and *z* sides are treated as periodic, while those in the *x* (perpendicular to the applied flux) are treated as hard walls.

3. Results and Discussion

3.1. Effects of aspect ratio, weight fraction of SWNTs, and of thermal boundary resistance on the thermal conductivity of SWNT- water and SWNT-oil suspensions

The suspensions used for the simulations were water at 20°C and oil at 40°C. The SWNTs were randomly oriented and the location of the SWNTs was random. The number of SWNTs in the cubic computational cell varied from 214 to 678 for the SWNT-water suspension at 20°C, and from 178 to 587 for the SWNT-oil at 40°C, and depended on the weight fraction of SWNTs in the suspensions. The thermal boundary resistance was chosen based on the experimental and MD simulation studies of Huxtable *et al.*¹¹ and Maruyama *et al.*¹⁰ that have provided estimations of the thermal boundary resistance in conditions similar to the simulations. According to the acoustic theory for the interpretation of thermal resistance⁸, the average probability for transmission of phonons across the interface into the carbon nanotube, f_{f-CN} is related to the thermal boundary resistance, R_{bd} , by

$$f_{f-CN} = \frac{4}{\rho C C_m R_{bd}} \tag{2}$$

where ρ is the fluid density; *C* is the fluid specific heat; and $C_{\rm m}$ is the velocity of sound in the surrounding fluid. The simulations were conducted with different weight fractions of SWNTs (0.5%, 1.0% and 2.0%) and with different thermal boundary resistance ($f_{\rm f-CN} = 1.000$, 0.100, 0.003, 0.008 for water and $f_{\rm f-CN} = 1.000$, 0.100, 0.037 for oil).

Table 2 shows the simulation parameters and the ratios of the effective thermal conductivity and pure suspension thermal conductivity of the SWNT-oil at 40°C and SWNT-water suspensions at 20°C obtained from the simulation runs. The effective thermal conductivity is a function of the thermal boundary resistance, the SWNT weight fraction, and the length scale of SWNTs in the suspensions. For each value of thermal boundary resistance, weight fraction and aspect ratio of SWNTs, the reported thermal conductivity is the average of three separate simulations with different initial SWNT distributions. The standard deviation of the simulation results was within 0.5% of the average value. With the same thermal boundary resistance and the same aspect ratio L/D, the effective thermal conductivities of both SWNT suspensions increase when the weight fraction of SWNTs increases. When the thermal boundary resistance decreases, heat can be transferred easier through the fluid-SWNT interface into the SWNTs, and then transferred through the suspension quite effectively, since the SWNTs have high thermal conductivity. So the effective thermal conductivities of both SWNT suspensions increase, as expected. When the thermal boundary resistance is very large, like the SWNT-water suspension case, the thermal walkers cannot diffuse easily in the suspension because the SWNTs do not allow the thermal walkers to come inside them blocking the diffusion of heat through the suspension. When diffusion is blocked by the SWNTs, the effective thermal conductivity of the SWNT suspension can be decreased, and even become smaller than that of the pure suspension fluid.

Figure 1 shows the simulation results for the ratio of the effective thermal conductivity of the suspensions divided by the thermal conductivity of water and pure oil at 20°C as a function of the thermal boundary resistance and with different aspect ratios of SWNTs (L/D = 40-120) in the suspensions having 1.0 wt% of the SWNTs. Figure 1 shows that the thermal conductivity of the SWNT suspensions increases between the case of L/D = 40 and the case of L/D = 80, but becomes almost constant when the aspect ratio L/D is greater than 80. At the lowest thermal boundary resistance for both suspensions ($f_{f-CN} = 1.0$), the difference of the thermal conductivity enhancement of the SWNT- water and -oil suspensions with the aspect ratios L/D = 80 and 120 is approx. 1%. This means that once the aspect ratio is large enough, the thermal conductivity of the SWNT suspensions is independent of L/D. This trend of the simulation results agrees qualitatively with the analytical solution of Nan *et al*¹⁹ that indicates L/D independence for large L/D.

Figures 2a and 2b show the effective thermal conductivity using thermal boundary resistance up to 0.4 x 10^{-7} m²K/W (K_{bd} = 25 MW/m²K) at L/D = 40 and at L/D=120, respectively. Since the effective thermal conductivities decrease dramatically with increasing thermal boundary resistance in Figure 2, it is essential to choose the proper thermal boundary resistance within the range of 0.4 -2.0 x 10^{-7} m²K/W (K_{bd} = 5-25 MW/m²K) for validation with experimental data, when they might become available. At the thermal boundary resistance of 0.83 x 10^{-7} m²K/W (K_{bd} = 12 MW/m²K), the effective thermal conductivity of the SWNT-water suspensions can be enhanced only up to 20% at 2.0% wt of SWNTs for short nanotubes (L/D = 40), but up to 167% for long nanotubes. At the lowest thermal boundary resistance simulated for the oil case (R_{bd} = 0.1488 x 10^{-8} m²K/W), the effective thermal conductivity of the SWNT-oil suspension can be enhanced 5.9, 7.2 and 8.8 times with 0.5%, 1.0% and 2.0% wt of SWNTs in the oil, respectively, for long nanotubes (see Fig. 2b). At the lowest thermal boundary resistance simulated for the water case (R_{bd} = 0.064 x 10^{-8} m²K/W), the effective thermal conductivity of the SWNT-water suspension at 20°C can be enhanced 7.3, 9.1 and 11.0 times with 0.5%, 1.0% and 2.0% wt of SWNTs in the water, respectively, for long nanotubes.

Figure 2 also shows that with the same thermal boundary resistance, the SWNT weight fraction is more important in the oil suspension case. Since the specific heat capacity of oil is half of that of water (other physical property values are almost the same), the value of f_{f-CN} for oil calculated by Equation (2) is two times higher than that for SWNT-water. (The specific heat capacities of oil and water are 2.22 and 4.18 J/g K, respectively). This means that phonons can cross the interface of oil and SWNTs more easily than the interface of water and SWNTs. It appears, therefore, that fluids with larger molecules can be more effective in transferring heat to SWNTs suspended in them.

3.2. Temperature effects on the thermal conductivity of the SWNT- water suspensions

The simulations to study the effect of the suspension temperature on the effective thermal conductivity of the SWNT-water suspensions were conducted similarly to the water case at 20° C, described in Section 3.1. The aspect ratio L/D =120 is used. Each of the simulations was conducted assuming that the properties of SWNTs and water were constant for the temperature interval employed in that particular simulation. This is equivalent to applying a very small temperature difference (e.g., less than 1 °C) across the computational domain in each simulation, and repeating this process at different temperatures.

The simulations were conducted at different temperatures (20, 40, 60 and 80° C), with different weight fractions of SWNTs (0.5%, 1.0% and 2.0%). The SWNTs were randomly oriented and the location of the SWNTs was random. The number of SWNTs in the cubic cell varied from 214 to 678.

Since currently available data for the thermal boundary resistance of SWNTs in fluids indicate a range of values between $0.83 \times 10^{-7} \text{ m}^2\text{K/W} (\text{K}_{bd} = 12 \text{ MW/m}^2\text{K})^{11}$ and $2 \times 10^{-7} \text{ m}^2\text{K/W} (\text{K}_{bd} = 5 \text{ MW/m}^2\text{K})^{10}$, simulations were conducted using specifically these two values. It was further assumed that the thermal boundary resistance has a constant value in the temperature range studied here. This assumption is justified because the ratio of the thermal diffusivity of the SWNTs divided by the thermal diffusivity of the water at each one of the temperatures studied differs by less than 5% (the thermal diffusivity of water at different temperatures is listed in Table 1, and the thermal diffusivity of SWNTs remains constant in this temperature interval). In addition, the product of the specific heat capacity times density of water at different temperatures, which is a term appearing in the equation that is used to calculate the thermal boundary resistance (Equation 2), decreases less than 2.5% in the temperature range examined. The velocity of sound in water is also constant. Since the thermal boundary resistance is affected by these factors, one can reasonably assume that it will vary by less than 5%, and one can model the value of the thermal boundary in the simulations as a constant. By applying Equation (1) with constant R_{bd} and using the properties of water from Table 1, the value of f_{w-CN} increases slightly with temperature.

The physical properties of water at different temperatures are summarized on Table 1. Figure 3 shows the temperature effect on the effective thermal conductivity of SWNT-water suspension. The reported thermal conductivity is the average of three simulation runs with different initial SWNT random distributions. With the same weight fraction of SWNTs and the same thermal boundary resistance, the effective thermal conductivities of the SWNT-water suspension increases when the temperature increases, due to the better diffusion of the heat walkers. This result is consistent with experimental measurements of previous researchers for multi-walled carbon nanotube nanofluids^{20,21} (as shown in Figure 3) and for carbon nanotube/epoxy³. Once the weight fraction of SWNTs increases, the effective thermal conductivity increases under the same suspension temperature and the same thermal boundary resistance. When the temperature of water increases from 20 to 80°C, the thermal conductivity of the suspension can be enhanced by up to 8%.

A temperature enhancement factor f_T can be defined as the ratio of (K_{eff}/K_{water}) divided by (K_{eff}/K_{water}) at 20°C. This factor is shown in Figure 4 as a function of temperature. Under the same water temperature, the factor f_T is almost constant and independent of the weight fraction of SWNTs

in the water and of the thermal boundary resistance. By fitting the calculated f_T data, we can obtain the following equation:

$$f_T = 0.95713 + 0.00230T - 0.00001T^2 \tag{3}$$

where *T* is the water temperature (in $^{\circ}$ C) and the coefficient R² for the fit is 0.98. Figure 4 also shows data points obtained from the experiments of multi-walled carbon nanotubes (MWNTs) in suspension reported by Wen and Yulong.²⁰ (Note that the surfactant used in the work of Wen and Yulong to suspend the MWNTs was similar to the surfactant used in the work of Huxtable *et al.*¹¹). The experimental data fall above the empirical fit suggested by Equation (3) for 46% vol MWNT and closer to the fit for 20% vol. As Equation (3) is independent of the weight fraction of the SWNTs and the thermal boundary resistance, this equation can predict the increase of the ratio K_{eff}/K_{water} with the water temperature. Since the equation has been obtained empirically, one should be careful not to expend its range of applicability for cases that are far beyond the range of SWNT weight fractions used here.

4. Conclusions

A computational model for systematically studying the effects of the thermal boundary resistance, weight fraction and the aspect ratio of SWNTs in oil and in water, as well as the effect of different water temperature on the thermal conductivity of SWNT-water suspensions, using a random walk algorithm has been successfully developed. This model can be applied to any suspension with a very wide range of weight fraction of SWNTs in the suspension, given that the inclusions are not in contact with each other.

In both SWNT-oil and SWNT-water suspensions, the effective thermal conductivity increases once the weight fraction of the SWNTs increases and the thermal boundary resistance is kept constant. With the same SWNT weight fraction, the effective thermal conductivity increases when the thermal boundary resistance decreases. The simulation results appear to show that fluids with larger molecules can be more effective in transferring heat to SWNTs suspended in them. Increasing the suspension temperature can enhance the thermal conductivity of the SWNT-water systems, and this enhancement can be described as a function of temperature only, independent of weight fraction and thermal boundary resistance.

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TABLES

Suspensions	Oil at $40^{\circ}C^{a}$	Water at $20^{\circ}C^{b}$	Water at $40^{\circ}C^{b}$	Water at $60^{\circ}C^{b}$	Water at $80^{\circ}C^{b}$
3	40 C	20 C	40 C	00 C	80 C
Density, g/cm ³	0.819	0.998	0.992	0.983	0.972
Specific heat capacity, J/g K	2.222	4.182	4.178	4.184	4.196
Thermal conductivity, W/mK	0.145	0.598	0.630	0.654	0.670
Thermal diffusivity, nm ² /ns	79.67	143.35	152.07	159.04	164.29
Velocity of sound, m/s ^c	1477.00	1496.70	1496.70	1496.70	1496.70

Table 1. Properties of oil and water used in the simulations

^aGlavatskih *et al.*²²

^bHandbook of chemistry and physics, 6-9 and 6-10²³

^cVelocity of sound is obtained from handbook of chemistry and physics, *velocity of sound in various media*, 14-35. Water and oil are considered as distilled water and castor oil respectively²⁴.

Table 2. Summary of the simulation parameters and simulation results of the SWNT dispersed randomly in the suspensions

Computational cell: 100 x 100 nm ³ (L/D=40)												
			$200 \times 100 \times 100 \text{ nm}^3 \text{ (L/D=80)}$									
				$300 \times 100 \times 100 \text{ nm}^3$ (L/D=120)								
Simulation parameters			Num	Number of walkers: 90000 Time increment: 0.002 ns						02 ns		
		SWN	SWNT diameter: 2.4 nm			Rat	Ratios of L/D: 40-120					
		Ther	Thermal equilibrium value $C_{\rm f}$: 0.25									
	R _{bd}		Weight fractions of SWNTs, wt %									
$f_{fluid-CN}$	$[10^{-8} \text{m}^2 \text{K/W}]$	0.5	1.0	2.0	0.5	1.0	2.0	0.5	1.0	2.0		
	$(K_{bd},[MW/m^2K])$	L/D = 40			L/D = 80			L/D = 120				
K _{eff} /K _{water} , SWNTs dispersed randomly in water at 20°C												
Volume fraction, vol %		9	17	29	9	17	29	9	17	29		
(number of SWNTs)		(214)	(393)	(678)	(214)	(393)	(678)	(214)	(393)	(678)		
0.003^{a}	20.000 (5)	0.95	0.95	0.96	1.40	1.59	1.77	1.56	1.79	2.01		
0.008^{b}	8.310 (12)	1.06	1.13	1.21	1.82	2.12	2.41	2.03	2.36	2.67		
0.100	0.640 (156)	2.20	2.73	3.22	4.24	5.07	5.91	4.38	5.26	6.14		
1.000	0.064 (1563)	4.01	5.25	6.44	7.20	8.98	10.08	7.31	9.09	11.04		
			K _{eff} /K	_{oil} , SWN	Ts dispe	rsed ran	domly i	n oil at 4	0°C			
Volum	e fraction, vol %	7	14	25	7	14	25	7	14	25		
(num	ber of SWNTs)	(178)	(333)	(587)	(178)	(333)	(587)	(178)	(333)	(587)		
0.037^{c}	4.000 (25)	1.59	2.14	3.70	2.65	3.19	3.73	2.89	3.45	4.01		
0.100	1.488 (67)	1.88	2.64	4.78	3.51	4.26	5.03	3.73	4.48	5.30		
1.000	0.149 (671)	2.24	3.21	6.02	5.67	7.09	8.68	5.68	7.15	8.78		

^aCalculated from the thermal boundary resistance obtained from the MD work of Maruyama *et* al.¹⁰

^bCalculated from the thermal boundary resistance obtained from experimental work of Huxtable *et al.*¹¹

^cCalculated from the thermal boundary resistance obtained from the MD simulation of Octane of Huxtable *et al.*¹¹.



Aspect ratios L/D

Figure 1. Length scale effect on the effective thermal conductivity of SWNT-water and -oil suspensions with 1.0 wt% of the SWNTs. For each value of thermal boundary resistance and aspect ratio of SWNTs, the thermal conductivity is the average of three simulations with different initial SWNT distributions.



(a)





(b)

Figure 2. Effective thermal conductivity of randomly dispersed SWNT-oil and SWNT-water suspension at (a) L/D = 40 and (b) L/D = 120. For each value of thermal boundary resistance and weight fraction of SWNTs, the thermal conductivity is the average of three simulations with different initial SWNT distributions.



Figure 3. Temperature effect on the effective thermal conductivity of SWNT-water suspension at L/D =120. For each value of thermal boundary resistance and weight fraction of SWNTs, the thermal conductivity is the average of three simulations with different initial SWNT distributions. The experimental data are for MWNT suspensions.²⁰



Figure 4. Temperature effect on the ratio of (K_{eff}/K_{water}) divided by (K_{eff}/K_{water}) at 20°C. The experimental data are for MWNT suspensions.²⁰