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Effective Thermal Conductivity of Aqueous Suspensions of Carbon Nanotubes (Carbon Nanotube Nanofluids)

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This work is concerned with the effective thermal conductivity of aqueous suspensions of multiwalled carbon nanotubes (nanofluids). Stable nanofluids were made using sodium dodecylbenzene sulfonate as the dispersant. The effects of concentration of carbon nanotubes and temperature on effective thermal conductivity were investigated. It was found that effective thermal conductivity increased with increasing concentration of carbon nanotubes, and the dependence was nonlinear even at very low concentrations, which was different from the results for metal/metal oxide nanofluids. The effective thermal conductivity increased with increasing temperature, and the dependence was also nonlinear. At temperatures lower than ∼30°C, approximately linear dependence of the thermal conductivity enhancement on temperature was seen, but the dependence tended to level off above ∼30°C. A comparison between the results of this work and those of published studies showed a large discrepancy in the effective thermal conductivity of carbon nanotube nanofluids. Differences in the interfacial resistances and thermal conductivities of carbon nanotubes used in these studies were proposed to be the main reasons. The experimental results were also compared with some classical macroscopic models for thermal conductivity of homogenous mixtures containing micrometer- or millimeter-sized particles. It was shown that the macroscopic models were inadequate for the prediction of the effective thermal conductivity of nanofluids. Analysis of possible mechanisms for thermal conduction enhancement suggested that networking of carbonnanotubes was likely to be responsible for the observed high effective thermal conductivity of carbon-nanotube nanofluids. Experiments at a temperature above 60–70°C showed that the dispersant failed, which led to destabilization of nanofluids.

I. Introduction

CONVENTIONAL heat-transfer fluids such as air, water, mineral oil, and ethylene glycol play an important role in many industrial sectors including power generation, chemical production, air conditioning, transportation, and microelectronics. These fluids, however, are inadequate for high-heat-flux applications such as superconducting magnets, superfast computing, novel supersonic jet aircraft, and high-power microwave tubes due to their low thermal conductivity. Recent advances in nanoscience and nanotechnology have led to development of a new category of fluids termed nanofluids.[1] Such fluids are liquid suspensions containing a small number of particles with at least one dimension significantly smaller than 100 nm and with a thermal conductivity orders of magnitude higher than that of the base liquid. Nanofluids have been shown to have effective thermal conductivities much higher than that predicted by macroscopic theories.[1–7] Nanofluids have also been shown to give superior performance in convective heat transfer.[8,9]

Nanoparticles of various materials have been used for nanofluid production. These include copper,[7] aluminum,[8] copper oxide,[3] alumina,[4] and titania.[4] More recently, a few studies have been published on the thermal behavior of nanofluids made from carbon nanotubes.[10–12]

Carbon nanotubes (CNTs) are fullerene-related structures that consist of either a grapheme cylinder (the so-called single-wall carbon nanotubes, SWCNTs) or a number of concentric cylinders (the so-called multiwalled carbon nanotubes, MWCNTs). They were first discovered by a Japanese electron microscopist, Sumio Iijima, in 1991 (Ref. 13). Basic research over the past decade has shown that CNTs could have a thermal conductivity an order of magnitude higher than copper, ∼3000 W/m · K for MWCNTs[14] and ∼6000 W/m · K for SWCNTs.[15] This suggests that nanofluids made of CNTs could have a very high thermal conductivity. However, only one of the few published studies shows this picture. Choi et al.[11] measured the effective thermal conductivity of MWCNTs dispersed in synthetic poly(α-olefin) oil. They found a nonlinear dependence of the effective thermal conductivity on the nanotube loading. At a CNT loading of 1% by volume, the enhancement was as high as 160%. This huge enhancement was not observed by Xie et al.[12] for nanofluids made of MWCNTs and water, MWCNTs and ethylene glycol, and MWCNTs and decene, nor by Assael et al.[10] for MWCNT/water nanofluids. The maximum thermal-conduction enhancement observed by Xie et al.[12] was only ∼20% for 1% nanotubes in decene by volume, and that observed by Assael et al.[10] was 20%–40% for 0.6% CNTs in water by volume. As a consequence, one of the objectives of the present work is to investigate possible reasons for the large difference. Another objective is to study the effect of temperature on the effective thermal conductivity of CNT nanofluids, which is more important to engineering applications, and no previous studies have been found in the literature. A third objective is to investigate the mechanisms for thermal-conduction enhancement in CNT nanofluids.

II. Experimental

A. Materials and Nanofluids Preparation

Deionized water and MWCNTs were used in this work as the continuous and dispersed phases, respective. The carbon nanotubes were supplied by the Tsinghua Nafine Nano-powder Commercialization Engineering Center (China). The nanotubes had an average diameter of 20–60 nm and a length of a few tens of micrometers, and were produced catalytically from hydrocarbon materials on nanocatalysts under high pressure. Figure 1 shows a scanning electron microscopy (SEM) image of the sample. It can be seen that the nanotubes are entangled, which makes it difficult to disperse and to stabilize them in water. Various measures were therefore used in preparing nanofluids, which included disentangling nanotubes and dispersing them with an appropriate dispersant.[16–18]

It is known that CNTs have a hydrophobic surface, which is prone to aggregation and precipitation in water in absence of a dispersant. Some efforts were therefore made in the initial stage of this work

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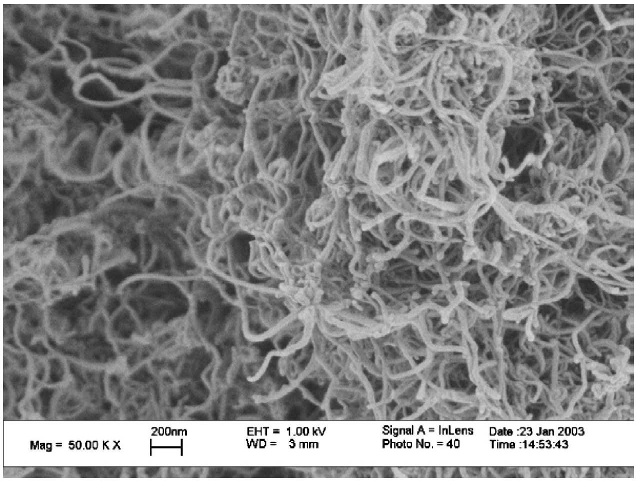


Fig. 1 SEM image of multiwalled carbon nanotubes as received.

to search for an appropriate dispersant and its minimum amount. After many trial and error tests, sodium dodecyl benzene sulfonate (SDBS) purchased from Sigma–Aldrich (U.K.) was found to able to stabilize carbon nanotubes very well. The minimum amount of SDBS was identified to be ∼17% by weight with respect to CNTs. To give some allowance, all nanofluids were prepared by using 20% by weight of SDBS with respect of CNTs.

A typical process for nanofluid preparation involves in series a) sonicating a CNT sample with known weight in an ultrasonic bath for more than 36 h, b) dispersing the disentangled CNTs into a preset amount of water containing SDBS, c) subjecting the mixture to ultrasonication for 24 h, and d) treating the resulting suspension with a high-speed magnetic stirrer for 1 h. CNT nanofluids made in this way were found to be very stable for months without sedimentation. Figure 2a shows a SEM image of the dispersed sample (0.46% by volume). It can be seen that CNT aggregates have largely been loosed/broken. This can also be seen from a transmission electron microscopy (TEM) image of the dispersed sample shown in Fig. 2b.

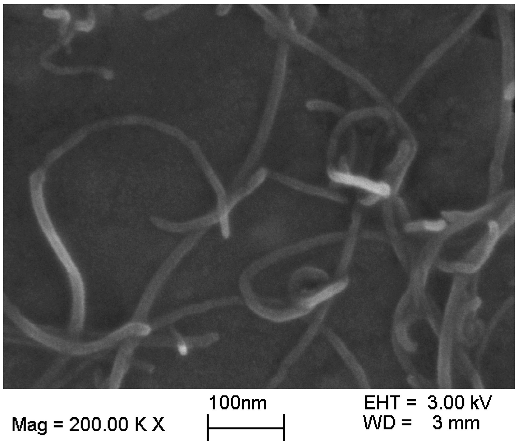
B. Measurement of the Effective Thermal Conductivity

The effective thermal conductivity of nanofluids was measured using a KD2 thermal property meter (Labcell, Ltd, U.K.), which is based on the transient-hot-wire method. The KD2 meter has a probe with length 60 mm and diameter 0.9 mm, which integrates in its interior a heating element and a thermoresistor. The probe is connected to a microprocessor for controlling and conducting the measurements. The KD2 meter was calibrated by using deionized water before any measurement. In order to study the effect of temperature on the effective thermal conductivity of nanofluids, a thermostat bath (GD120-S12, Grant, U.K.) was used, which was able to maintain temperature uniformity within ±0.02°C. Five measurements were taken for each CNT concentration at a given temperature. This ensured that the uncertainty of thermal conductivity measurements was within 3%.

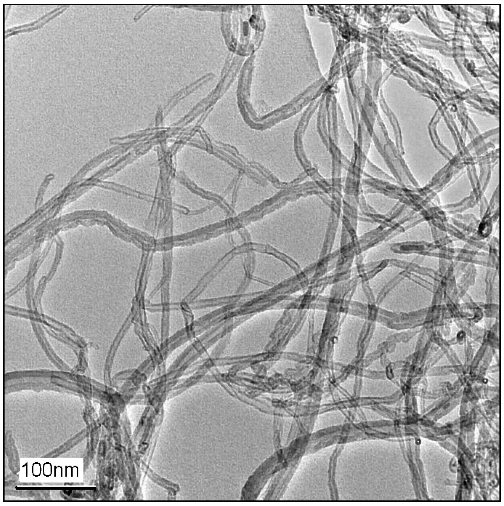
Nanofluid samples were put in cylindrical containers with diameter ∼40 mm and height ∼70 mm. To avoid evaporation of water in nanofluids, the containers were sealed using rubber stoppers. The rubber stoppers had small holes in the middle through which the thermal meter probes were inserted vertically into the nanofluids.

III. Experimental Results and Discussion

Nanofluids with CNT concentration of 0–0.84% by volume were tested. The tests were conducted over a temperature range of ∼10–70°C. In the following, experimental results will be presented and discussed in Sec. III.A. Comparison of the results with macroscopic models will be made in Sec. III.B. Further discussion of the mechanisms of thermal-conduction enhancement will be given in Sec. III.C.



a) SEM image



b) TEM image

Fig. 2 SEM and TEM images of dispersed CNTs in deionized water.

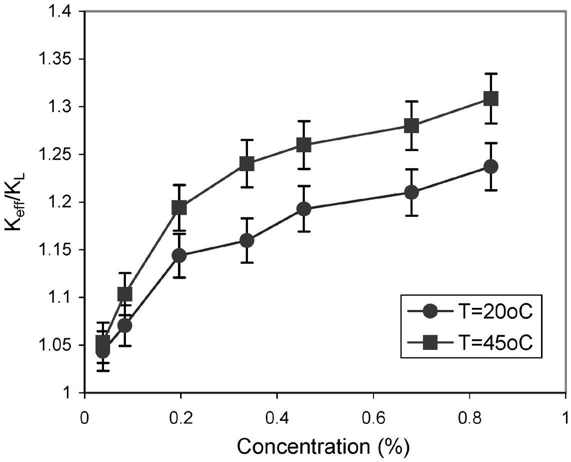


Fig. 3 Effect of CNT concentration on effective thermal conductivity of nanofluids at two temperatures.

A. Experimental Results and Discussion

Figure 3 shows the effective thermal conductivity, K_{eff}, normalized by the thermal conductivity of the base liquid, K_L, as a function of CNT concentration at two temperatures, 20 and 45°C. Addition of CNTs to water significantly enhances the thermal conductivity and the enhancement increases with increasing CNT concentration and temperature. At a CNT concentration of 0.84%, ∼23.7% enhancement has been achieved at 20°C; the enhancement increases to ∼31% at 45°C. Figure 3 also shows a nonlinear dependence of

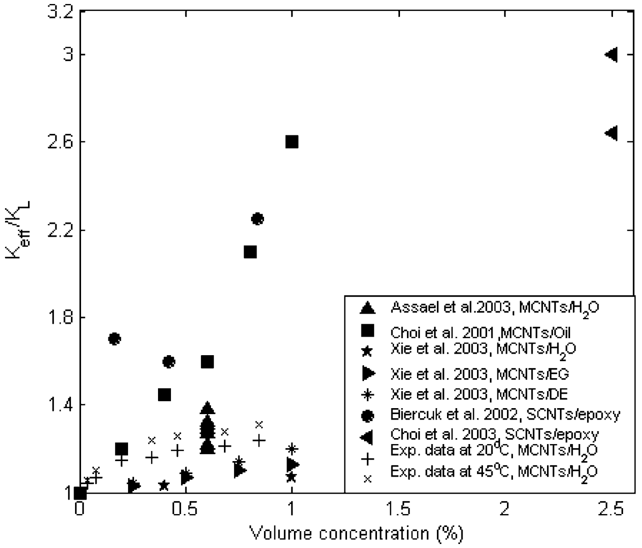


Fig. 4 Comparison of published data on thermal conductivity for CNT nanofluids and CNT/polymer composites.[10–12,19,20]

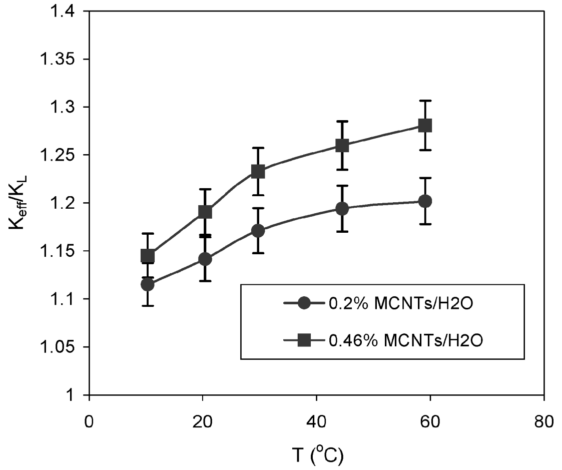
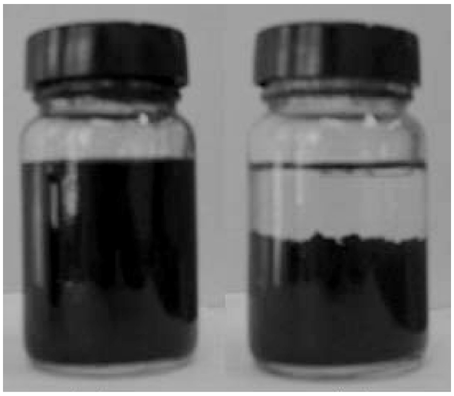


Fig. 5 Effect of temperature on the effective thermal conductivity of nanofluids of two concentrations.



a) b)

Fig. 6 Pictures of 0.2% CNT nanofluids a) before and b) after bems heated up to 69.4°C.

the normalized thermal conductivity on CNT concentration even at very low concentrations. This differs from the results for nanofluids using metal and metal oxide particles, for which approximately linear relations are observed; see, for example, Choi,[1] Lee et al.,[3] and Xuan and Li.[7]

Figure 4 compares reported experimental data on nanofluids obtained at approximately room temperature. Also included are data for carbon nanotube/polymer composites.[19,20] As some data are reported as weight percentages, conversion is needed. In these cases, the densities of CNTs and epoxy are assumed to be 1200 and 1000 kg/m^3, respectively. A huge difference is seen in the extent of thermal conductivity enhancement between the reported data, ranging from less than 10% to more than 160% for 1% by volume of CNTs.

The huge enhancement in thermal conduction of CNT nanofluids (Fig. 4, ~160% for 1% CNTs at the room temperature) obtained by Choi et al. was not observed in this work. Additionally, Choi et al.[11] observed an upward-concave dependence of the effective thermal conductivity on CNT concentration, whereas our results show a downward-concave dependence. The exact reason for the discrepancies is unclear, but differences in the properties of the base liquid, the CNTs, and the dispersant are believed to be responsible. The results obtained in this work seem close to those reported by Xie et al.[6] and Assael et al.[10]

It is interesting to note that interaction may occur between the effects of temperature and CNT concentration. At CNT concentrations lower than ~0.3%, the difference between the two temperatures in thermal-conduction enhancement increases with CNT concentration. The difference seems to disappear at CNT concentrations larger than ~0.3%, beyond which the slopes of the two curves are approximately the same.

Figure 5 shows the effective thermal conductivity as a function of temperature for two CNT concentrations, 0.2% and 0.46%. At temperatures lower than ~30°C, an approximately linear dependence of the thermal conductivity enhancement on temperature is seen, but the dependence tends to level off above ~30°C. These observations are similar to the observations of Biercuk et al.[19] on CNT/epoxy composites, but different from that of Das et al.[21] on alumina/water nanofluids; they found that the thermal-conductivity ratio increased with temperature in a linear fashion.

Figures 3 and 5 suggest that higher thermal conductivities occur at higher temperatures. However, caution must be exercised due to the presence of dispersant, which may destabilize suspensions under high-temperature conditions. Figure 6 shows the pictures of nanofluids with 0.2% CNTs by volume before and after being heated up to 69.4°C. Clearly, strong aggregates formed during the process of heating. Further investigation showed that the phenomenon occurred at a temperature between 60 and 70°C, and the process was irreversible even after a prolonged period of ultrasonication at the room temperature.

B. Comparison with Macroscopic Models

Many macroscopic models have been proposed to calculate mixture properties. Most of them are based on the so-called effective medium theory (EMT), developed about a century ago for calculating the dielectric properties of composite materials. Examples of EMT-based models include the Maxwell–Garnett self-consistent model and the Bruggeman model,[22] the Hamilton and Crosser (HC) model,[23] and the Jeffrey[24] and Davis[25] models. These models have been applied to liquid–solid mixtures, gas–solid mixtures, and solid–solid composite materials. Good agreement has generally been obtained for micro- or millimeter-sized particles dispersed homogenously in continuum media. A comparison of these models shows that, for spherical particles (shape factor 3 for the HC model), little difference between these models occurs at particle volume fractions lower than ~0.03, whereas the Bruggeman model gives a higher prediction than other models at concentrations higher than ~0.05 (Ref. 26). It should be noted that the shape factor in the HC model can be used as a fitting parameter to match the experimental results. In the following, comparison is only made between the Bruggeman and HC models and the experimental results. Figure 7 shows the comparison, where the shape factor in the HC model is taken as a fitting parameter and three arbitrary values, 30, 60, and 100, are used due to unavailability of the exact length of carbon nanotubes. It can be seen that the Bruggeman model fails to predict the measured results. The HC model with the shape factor as a fitting parameter

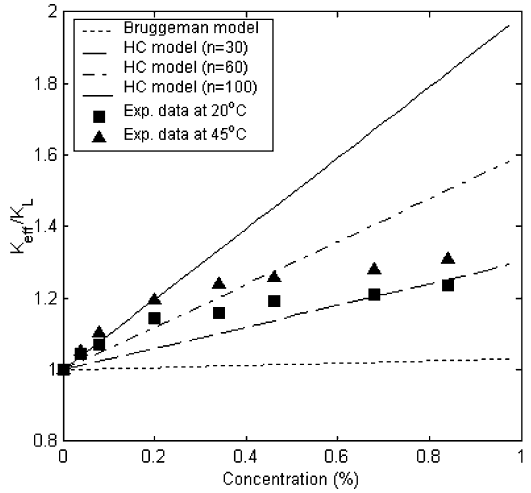


Fig. 7 Comparison of experimental data with macroscopic models.

is also seen to be inadequate. In particularly, the HC model (and the Bruggeman model) does not reflect the nonlinear dependence of the measured effective thermal conductivity on the concentration of carbon nanotubes. More fundamental work is clearly needed on this aspect.

C. Mechanisms of Heat Transfer Enhancement

A number of mechanisms have been proposed to explain the observed increase in thermal conductivity due to the addition of metal/metal oxide nanoparticles. These include Brownian motion,[2] interfacial ordering,[2,27] ballistic transport of energy carriers,[2] and nanoparticle clustering.[3] In the following these mechanisms are discussed with reference to CNT nanofluids.

First of all, the Brownian motion is unlikely to play a direct role in enhancing heat conduction. This has been discussed in detail by Keblinski et al.[2] through comparing the characteristic times for the Brownian motion with those for thermal diffusion in the base liquid, and by Ding et al.[26] from the viewpoint of microconvection.

Interfacial layering refers to a phenomenon at the liquid–particle interface where liquid molecules are more ordered than those in the bulk liquid. The ordered structure could have a higher thermal conductivity than that of the bulk hence enhancement of the effective thermal conductivity.[2,27] However, doubts may be raised when there is a surfactant/dispersant; see below and also Ding et al.[26]

Nanoparticle clustering or, more appropriately for CNT nanofluids, nanotube networking can be a likely mechanism. Nanotube networking also provides an avenue for ballistic transport of energy carriers. Biercuk et al.[19] showed that a topologically connected CNT network could be formed in CNT/epoxy composites at a CNT volume fraction as low as 0.1–0.2%. Consider cylindrical particles dispersed in a lower-thermal-conductivity medium, if particles are topologically connected, and the thermal resistance at the connection points is negligible, geometrical considerations gives[28]

$$K_{\mathrm{eff}}/K_L = \langle\cos^2\theta\rangle(K_s/K_L)\phi_s \tag{1}$$

where K_s is the thermal conductivity of carbon nanotubes, ϕ_s is the volume fraction of the dispersed phase, and $\langle\cos^2\theta\rangle$ is a geometrical factor equal to $\frac{1}{3}$ for randomly dispersed particles. Nan et al.[29] studied the effective thermal conduction of a mixture with randomly dispersed cylindrical particles. For cylindrical particles with aspect ratios greater than 100, they arrived at the following expression by using effective medium theory:

$$\frac{K_{\mathrm{eff}}}{K_L} = \frac{3 + 2\phi_s(K_s/K_L)}{3 - 2\phi_s} \tag{2}$$

If K_s and K_L are taken as 3000 W/m · K for MWCNTs[14] and 0.6 W/m · K for deionized water, and we assume 1 vol% CNT randomly dispersed in water, Eqs. (1) and (2) give thermal conductivity ratios (K_{eff}/K_L) of 17 and 35, respectively. These values are about an order of magnitude higher than the measured data shown in Figs. 3–5. Two possible reasons are believed to be principally responsible for the huge disparity: 1) the actual thermal conductivity of carbon nanotubes may be much lower than 3000W/m · K, and 2) thermal resistance exists at the interface of CNTs and the base liquid and at the interface between connected CNTs. They are discussed in the following:

1. *Thermal Conductivity of CNTs*

The value of K_s of 3000 W/m · K is the thermal conductivity in the axial direction. Thermal conductivity in the radial direction of CNTs could be much lower.[30] This practically implies that the overall thermal conductivity could be much lower than 3000W/m · K. Furthermore, the value of 3000W/m · K is taken from the high end of the reported data, and a much lower value of 200W/m · K has been reported by Yang et al.[31] It should also be noted that the thermal conductivity of CNTs also depends on their diameter (a larger diameter gives a lower thermal conductivity[14]) and vacancy and defect concentrations (vacancy and defect have a negative effect on thermal conduction[30]). Additionally, the thermal conductivity of the bulk of CNTs is lower than that of single CNTs (both SWCNTs and MWCNTs).[14,30,31]

2. *Thermal Resistance at the Interface*

The thermal resistance at the interface can be expressed by the Kapitza conductance, G, which is defined as

$$q = -G\Delta T \tag{3}$$

where q is the heat flux and T is the temperature. Huxtable et al.[28] investigated interfacial heat flow in SWCNT suspensions in the presence of sodium dodecyl sulfate (SDS) surfactant using the picosecond transient absorption method. It was found that the interfacial thermal conductance was exceptionally small, $G = \sim 12$ MW/m^2 · K. Further molecular modeling indicated that the interfacial conductance between nanotubes and molecular fluids was controlled by the phonon–phonon coupling within the nanotubes. The nanotubes and the surrounding surfactant molecules were coupled by a small number of low-frequency vibrational phonon modes. Energy contained in high-frequency phonon modes within the carbon nanotubes must first be transferred to the low-frequency modes through phonon–phonon coupling before being-exchanged with the surrounding medium. As a consequence, most of the temperature drop occurred at the nanotube–polymer interface. If one takes the measured conductance as an example and assumes that the polymer has a thermal conductivity of 0.3 W/m · K, then thermal resistance at this interface is equivalent to a polymer layer with thickness 25 nm. The presence of surfactant could therefore impose a large thermal resistance to heat transfer and result in a lower effective thermal conductivity.

The interfacial conductance is influenced by the properties of dispersed system and surfactant. For example, Wilson et al.[32] found $G = 130$ MW/m^2 · K for citrate-stabilized platinum nanoparticles in water, and $G = 5$ MW/m^2 · K for nanoparticles of alkanethiol-terminated AuPd alloy in toluene. As a consequence, prediction of the effective thermal conductivity requires the knowledge of the interfacial resistance, which is likely to be responsible for the observed huge difference in the measured results as shown in Fig. 4. More detailed work is needed in this regard.

IV. Conclusions

Experimental work has been carried out on the effective thermal conductivity of carbon nanotube/water nanofluids with sodium dodecylbenzene sulfonate (SDBS) as the dispersant. The effects of CNT concentration and temperature on the effective thermal conductivity have been examined, and possible mechanisms for the thermal conduction enhancement discussed. The following conclusions could be drawn:

1) The effective thermal conductivity increases with increasing CNT concentration, and the dependence is nonlinear even at very

low concentrations. This differs from the results for metal/metal oxide nanofluids.

2) The effective thermal conductivity increases with increasing temperature, and the dependence is also nonlinear. At temperatures lower than ~30°C, an approximately linear dependence of the thermal-conductivity enhancement on temperature is seen, but the dependence tends to level off above ~30°C.

3) The enhancement of the thermal conductivity could not be adequately predicted by the macroscopic models.

4) High temperatures may result in failure of the dispersant and hence destabilization of nanofluids. For nanofluids formulated with SDBS, temperature should not be higher than ~60–70°C.

5) There exists a large difference in the reported thermal conductivity of CNT nanofluids. The interfacial resistance and uncertainty in the thermal conductivity of CNTs are proposed to be the main reasons.

6) The observed thermal-conduction enhancement is likely to be associated with networking of carbon nanotubes.

Acknowledgments

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