THERMAL CONDUCTIVITY OF CNT WATER BASED NANOFLUIDS: EXPERIMENTAL TRENDS AND MODELS OVERVIEW

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ABSTRACT

Thermal conductivity measurement of carbon nanotubes water-based nanofluids is here reported. We have considered in particular the influence of nanoparticle volume fraction, temperature, carbon nanotube aspect ratio and different kind of surfactant (SDBS, Lignin, Sodium polycarboxylate) on thermal conductivity enhancement of nanofluids. The experiments show that TC enhancement of nanofluids produces at very low volume fraction. It is also mainly governed by both volume fraction and temperature increase. However, TC enhancement of nanofluids is weakly affected by carbon nanotubes aspect ratio and surfactant type used in the study.

In addition, various theoretical thermal conductivity models are used to possibly correlate the experimental data and explain the TC enhancement of nanofluids. The selected models do not capture the experimental findings within the range of this parametric study, evidencing the need to develop appropriate model for TC enhancement prediction of CNT nanofluids and measure TC of this kind of nanofluids before performing numerical studies in heat exchangers and cavities.

INTRODUCTION

Since many years, it has been demonstrated that nanofluids (NF) are promising candidates as heat transfer fluids in heat exchangers, cooling devices, and solar collectors [1-3] due to their enhanced thermal properties. Nanofluids are obtained by dispersing a small amount of nanoparticles with high intrinsic conductivity within currently used base fluids, such as water, ethylene glycol, oil, … Among the numerous studied systems, carbon nanotubes (CNT) appear very interesting due to their high thermal conductivity in comparison to Al_2O_3, CuO and TiO_2. In addition, as nanofluid viscosity is related to resistance to flow and pumping power in energy systems, it is better to use low viscosity base fluid. Thermal conductivity is an important

physical properties which can be used to evaluate the efficiency of nanofluids as coolants [4,5], predict heat transfer within exchangers [6,7], natural convection in cavities as well [8,9] and for numerical analyses of these fluids in various systems [10,11].

Some important works dealing with thermal conductivity of CNT nanofluids are briefly reviewed in the following. As for many nanofluids, it is well admitted that TC of CNT nanofluids is enhanced with CNT volume fraction as evidenced by Wan et al. [12] who have compiled a large variety of experimental data. Similar results were also obtained by many authors considering different base fluids such as oil [13], ethylene-glycol [14] and water [15]. The effect of temperature was also previously reported in [4,16,17] showing that TC enhancement of CNT nanofluids is increased with temperature.

The influence of nanotubes aspect ratio was also investigated in some studies. Yang et al. [18] have shown that TC of CNT dispersed in oil increases with the increase of CNT aspect ratio. Similar results were also obtained by Assael et al. [19] considering MWCNT and SWCNT dispersed in water. The structure of CNT can also affect the TC of CNT nanofluids, as shown in [17] who observed that TC decreases when the CNT wall number is increased.

The role of surfactant in dispersing CNT within base fluids, and water in particular, was reviewed in [20]. Actually, in addition to mechanical stirring and sonication, due to the hydrophobic behavior of CNT their dispersion and stability within water is obtained through the use of surfactant. Many types of surfactant can be considered such as sodium dodecylbenzene sulfonate (SDBS) [1,21-23], sodium dodecylsulfate (SDS) [24-26], hemadecyltrimethylammonium bromide (CTAB) [19], gum arabic (GA) [15,27,28], octyl phenol ethoxylate [29] and potentially play a role in TC of CNT nanofluids. Thus, thermal properties of MWCNTs dispersed in water with SDS and SDBS were studied in [23]. The thermal conductivity of SDS and SDBS water mixtures is reported to decrease with the concentration of surfactants. The better thermal performance was obtained with SDBS in comparison with SDS dispersant. It was shown in [28] that GA does not contribute to thermal conductivity enhancement CNT water based nanofluids with low volume content. A first study on comparison of lignin and SDBS as dispersant for CNT water based nanofluids was performed in [30]. The authors demonstrate that lignin can act more a better dispersant than SDBS without affecting TC enhancement of nanofluids.

In addition to experimental results, some theoretical models have also been developed to predict TC enhancement of CNT nanofluids, as nicely reviewed in [31]. Recently, a critical analysis of the TC models for CNT nanofluids was conducted by Lamas et al. [32]. They concluded on the lack of reliability and universality of the models investigated and the need of performing experimental measurement to study the influence of main factors contributing to results, length of nanotubes and volume fraction in particular.

Hence, based on this assessment, the purpose of the present work is to report a comprehensive experimental parametric study on thermal conductivity (TC) of water-based nanofluids containing carbon nanotubes. We report for the first time the coupled effects of volume fraction, temperature, CNT aspect ratio and surfactant type on TC of CNT water-based nanofluids. We have considered here the influence of SDBS (sodium dodecylbenzenesulfonate), lignin, which is a by-product of paper industry and sodium polycarboxylate respectively as stabilizers. The effects of nanoparticle volume fraction within the range 0.005-0.55%, temperature varying between 20 and 40°C, and nanoparticles aspect ratio (90; 160) on the thermal conductivity enhancement is presented and discussed.

First, various theoretical approaches and empirical models for thermal conductivity of CNT nanofluids are presented considering the influence of nanofluid composition and impact of several mechanisms such as volume fraction, temperature, nanoparticle shape and aspect ratio, carbon nanotubes curving and wrapping, interfacial thermal resistance of CNT. All told, seven models were here considered. Finally, experimental results are discussed and compared to theoretical predictions to possibly explain the mechanisms responsible of TC conductivity enhancement of nanofluids.

THERMAL CONDUCTIVITY MODELS FOR CNT NANOFLUIDS

Several theoretical correlations have been developed in the past to predict the thermal conductivity enhancement of CNT nanofluids. The relevance of many correlations have been presented and discussed in recent works [31,32]. Here, we intend to compare some of these correlations with our experimental data, considering different factors that can contribute to the theoretical frame of thermal conductivity enhancement.

At first sight, the thermal conductivity of nanofluids can be estimated by Hamilton and Crosser model [33]. This model, defined by equation (1), can be used when the thermal conductivity of the particles is at least 100 times higher than the one of the liquid phase.

$$k_{nf} = \frac{k_p + (n-1)k_{bf} + (n-1)(k_p - k_{bf})\phi}{k_p + (n-1)k_{bf} - (k_p - k_{bf})\phi} k_{bf} \qquad (1)$$

In the previous equation, k_{nf}, k_p and k_{bf} are the thermal conductivity of nanofluids, nanoparticles and base fluid respectively in W/mK, ϕ is the volume fraction and n is a shape factor linked to nanoparticles sphericity such as $n=3/\psi$. Due to rod shape of nanotubes contained within the nanofluids, the sphericity ψ was here calculated from the average length and diameter of the nanotubes investigated. This leads to n values reported in Table 1 for each tested nanofluids. Table 1 shows that n values vary following CNT aspect ratio (r=l/d, with r the average aspect ratio, l the average length and d the average

diameter respectively) and differs from 6, as generally considered with cylindrical particles.

Xue et al. [34] presented a model for CNT nanofluids which derives from Maxwell model and includes the effect of axial ratio and space distribution of CNT. However, the model does not directly reports on the TC dependence to CNT aspect ratio. This model is defined by equation (2) and depends on the thermal conductivity of nanoparticles and base fluid and the volume fraction.

$$k_{nf} = \frac{1 - \phi + 2\phi \dfrac{k_p}{k_p - k_{bf}} \ln \dfrac{k_p + k_{bf}}{2k_{bf}}}{1 - \phi + 2\phi \dfrac{k_{bf}}{k_p - k_{bf}} \ln \dfrac{k_p + k_{bf}}{2k_{bf}}} k_{bf} \tag{2}$$

Patel et al. [35] developed a model for TC of CNT nanofluids considering combined parallel paths for heat, one through the base fluid, the other one through the CNTs. This model depends on volume fraction and both base fluid and nanoparticle radii, rbf and rp respectively. In absence of measurement, the radius of base fluid is here considered as the molecule radius of water.

$$k_{nf} = k_{bf} \left[1 + \frac{k_p \phi r_{bf}}{k_{bf}(1 - \phi) r_p} \right] \tag{3}$$

The effects of carbon nanotube curving and wrapping was investigated in [36]. Such phenomena depend on base fluids, nanotubes dimensions and presence of surfactants. In this work, several equations were developed to predict TC enhancement of CNT-based nanofluids from a distribution based modelling technology and the use of probability density functions (denoted p(x)). The authors showed that among the models developed, the uniform distribution model, denoted UDM (equation 4, obtained with p(x)=3) and the linear increase distribution model, denoted LIDM, (equation 5 with p(x)=18(1/3-x)) are able to capture the TC enhancement of CNT-based nanofluids investigated in their work. So, the effect of moderate curving and wrapping is captured by the following equation under the consideration of $k_{bf}/k_p \ll 1$.

$$k_{nf} \approx 1 + \phi \ln\left(\frac{27 k_p}{16 k_{bf}}\right) k_{bf} \tag{4}$$

For less curving and wrapping nanotubes, TC of nanofluid can be obtained from ($k_{bf}/k_p \ll 1$)

$$k_{nf} \approx 1 + 6\phi \left[1 + \frac{1}{3} \ln\left(\frac{256 k_p}{19683 k_{bf}}\right) \right] k_{bf} \tag{5}$$

Nan et al. [37] proposed a TC model for carbon-based nanofluids taking into account the effect of interfacial resistance based on multiple scattering theory and the EMT model of Maxwell, considering also the Kapitza resistance at the CNTs medium. This model was expressed as

$$k_{nf} = \frac{3 + \phi(\beta_{11} + \beta_{33})}{3 - \phi\beta_{11}} k_{bf} \tag{6}$$

where

$$\beta_{11} = \frac{2(k^c{}_{11} - k_{bf})}{k^c{}_{11} + k_{bf}} \; ; \; \beta_{33} = \frac{k^c{}_{33}}{k_{bf}} - 1 \tag{7}$$

and

$$k^c{}_{11} = \frac{k_p}{1 + \dfrac{2a_k k_p}{d k_{bf}}} \; ; \; k^c{}_{33} = \frac{k_p}{1 + \dfrac{2a_k k_p}{l k_{bf}}} \tag{8}$$

In these equations, $k_c{}^{11}$ and $k_c{}^{33}$ denote the equivalent TC along transverse and longitudinal axes respectively of a thin interfacial thermal layer and depend on CNT dimensions and Kapitza radius $a_k = R_k k_{bf}$ [21,32], with $R_k = 8 \times 10^{-8}$ m²K/W.

Murshed et al. [38] developed a model based on three phases concept e.g. nanoparticle, nanolayer and based fluid and taking into account particle size, volume fraction of CNT and interfacial layer. This model writes as follows,

$$k_{nf} = \frac{(k_p - k_{lr})\phi k_{lr}(\gamma_1{}^2 - \gamma^2 + 1) + (k_p + k_{lr})\gamma_1{}^2[\phi\gamma^2(k_{lr} - k_{bf}) + k_{bf}]}{\gamma_1{}^2(k_p + k_{lr}) - (k_p - k_{lr})\phi(\gamma_1{}^2 + \gamma^2 - 1)} \tag{9}$$

Here $\gamma_1 = 1 + t/r_p$ and $\gamma = 1 + t/d_p$ due to cylindrical form of nanoparticles. In addition, k_{lr} represents the thermal conductivity of interfacial layer and t is the thickness of interfacial layer between nanoparticle and base fluid. As reported in [38], t is here taken as 2 nm. This model requires the thermal conductivity of the interfacial layer which is not exactly known and cannot be measured as well. So as suggested in [36], an intermediate TC value between nanoparticles and base fluids was used. So, the TC of interfacial layer was chosen here to be 3 times that of the one of base fluid [38].

It is shown that all these equations require properly determination of both TC and dimensions of nanotubes, and the thermal conductivity of base fluid. It is noted that the equations do not support directly the influence of temperature. It should also be mentioned that the possible effect of particle agglomeration do not appear directly within the previous equations. As reported earlier, the TC of CNT is taken as 3000 W/mK. It is also mentioned that a thermal conductivity model for nanofluids containing nanotubes taking into account the effect of diameter and aspect ratio of nanotubes as well

Brownian effect due to temperature has been developed by Walvekar et al. [28] based on initial model introduced by [39]. This model is here not considered due to its lack of efficiency to correlate our experimental data, as reported earlier [30,40].

MATERIALS AND EXPERIMENTS

MATERIALS

Four types of nanofluids were here selected and investigated. They have been partially studied in our previous works [4,30,41-43]. Nanofluids consists of MWCNT (purity of 90%) dispersed in a mixture of deionized water and ionic surfactant. Three nanofluids, respectively denoted N_1, N_2 and N_3, are composed with the same nanotubes and differ from the surfactant used. The last nanofluid denoted N_4 is obtained from nanotubes with lower aspect ratio and higher density dispersed with the same surfactant that the one used with N_3. Based on manufacturer specification, the composition and the properties of the different nanotubes and nanofluids are summarized in Table 1.

Table 1. Nanotubes and nanofluids properties

	N_1	N_2	N_3	N_4
Nanotube average diameter d (nm)	9.2	9.2	9.2	11.4
Nanotube average length l (μm)	1.5	1.5	1.5	≈1
Average aspect ratio (r=l/d)	160	160	160	90
Density (kg/m³)	1800	1800	1800	2050
Carbon purity (wt.%)	90	90	90	90
Sphericity / n (H-C model)	0.24 /12.5	0.24 /12.5	0.24 /12.5	0.29 /10.23
Surfactant	SDBS	Lignin	Sodium polycar-boxylate	Sodium polycar-boxylate

In all cases, an initial starting suspension with 1% in weight fraction of nanotubes and 2% in weight fraction of surfactant was prepared by Nanocyl. Then, nanofluids with lower volume fraction were obtained from serial dilution of the starting suspension, as reported earlier [41-43], conserving constant

surfactant/carbon nanotubes weight ratio of 2. Consequently, it is assumed that preparation of nanofluids does not affect the TC measurement performed in the following. Finally, the whole volume fraction range investigated varies between 0.005% and 0.55% at ambient temperature. It should be mentioned that the volume fraction of N_4 is slightly lower due higher nanotubes density. As evidenced by Table 1, impact of average CNT aspect ratio and surfactant type are presently studied.
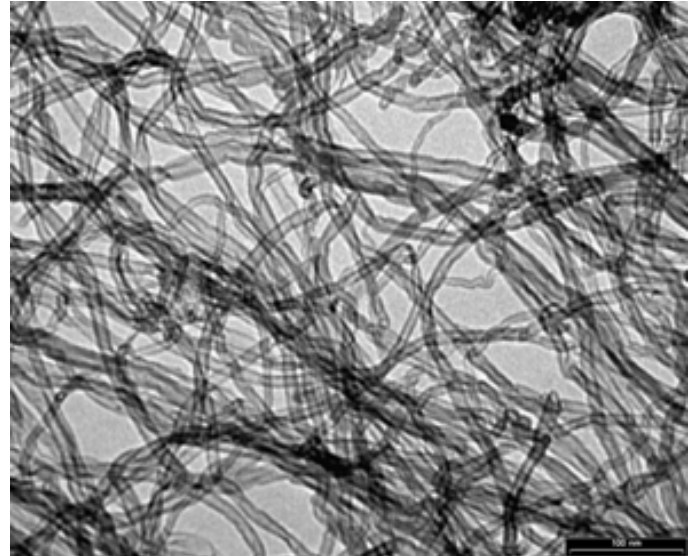
(a)
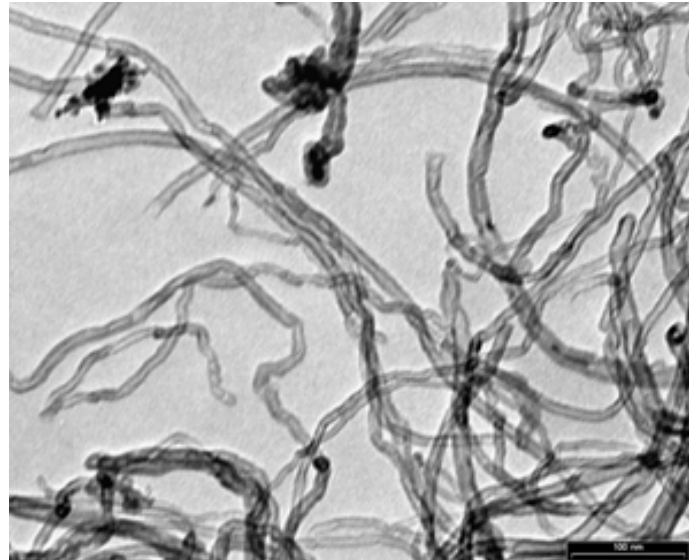
(b)

Figure 1. TEM picture of nanotubes – (a): nanotubes with aspect ratio of 160; (b): nanotubes with aspect ratio of 90.

The size of nanotubes and the structure of nanofluids at 1% in weight fraction were also evaluated from TEM analysis. Figures 1 shows that the size of nanotubes used are well in agreement with manufacturer specifications, and that the nanotubes are not necessarily straight. Similar conclusions are

obtained from Figure 2. Figure 2 also shows that nanotubes at the mass fraction of 1%, when dispersed in base fluid, are mainly entangled. This corresponds to a volume fraction of 0.55% and 0.49% for N_3 and N_4 respectively. The CNT appear randomly oriented with no apparent preferential direction, and form a connected network of conducting nanotubes. A similar analysis was previously reported for N_1 [44] and N_2 [30] showing that N_1 can form aggregates at this concentration.
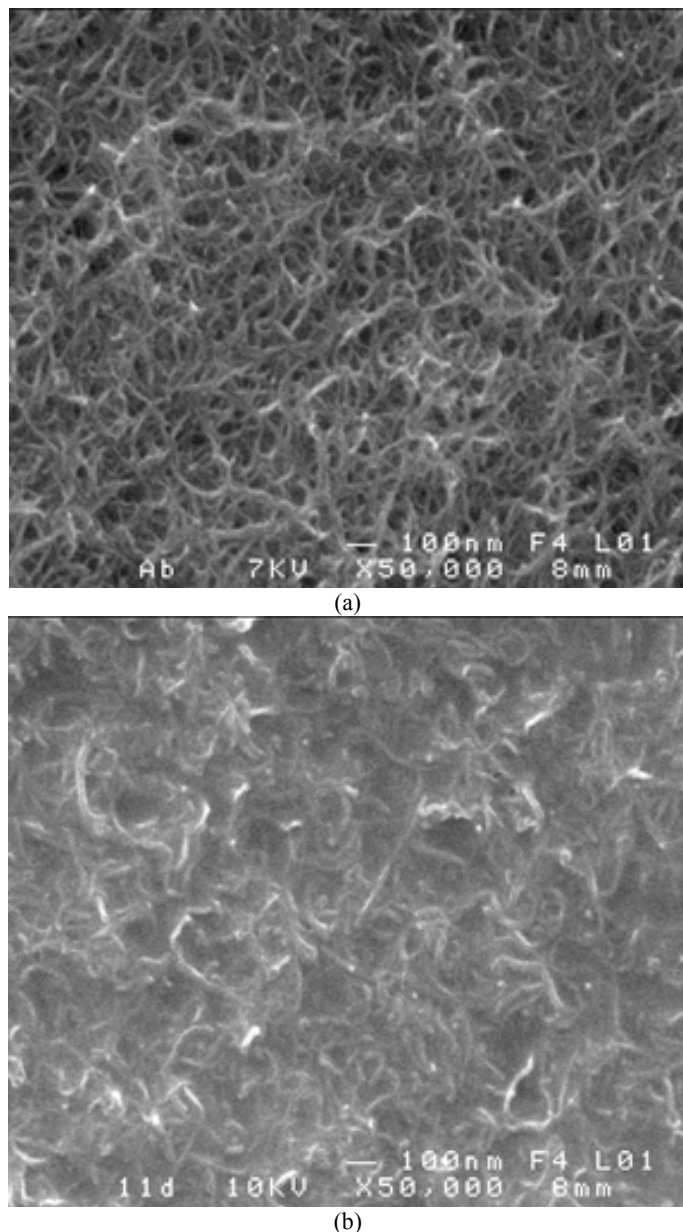

(a)

(b)

Figure 2. TEM picture of dried nanofluids with 1% in weight fraction − (a): N_3; (b): N_4.

THERMAL CONDUCTIVITY MEASUREMENT

A KD2 Pro thermal property analyzer (Decagon Devices Inc.), which is based on the transient hot wire method, was equipped of a KS-1 sensor to evaluate the thermal conductivity of both nanofluids and base fluids (deionized water and surfactant). The experimental set-up for thermal conductivity measurement was previously used and described in [4,42,43]. In short, each sample is placed in a vessel, itself located in a temperature control bath. The sensor probe and a platinum probe (with an accuracy of 0.1°C) are inserted within the sample. Before starting TC measurement, both the sample and the probes were first maintained 30 min at the required temperature. Once temperature stability is achieved, ten measurements were recorded with 5 mins of interval between each measurement. This was done to reduce experimental uncertainty. So, the thermal conductivity values reported in the following consist of an average of the ten measures performed at 20, 30 and 40°C respectively for each tested volume fraction of nanofluids and base fluids.

It is important to note that accuracy and reliability of thermal conductivity measurement system was also previously evaluated with distilled water within the temperature range of 20-50°C, leading to a maximum relative standard deviation of 3.5% [4,42,43].

RESULTS AND DISCUSSION

As indicated in Materials, due to constant surfactant/carbon nanotubes weight ratio, the quantity of surfactant increase with CNT volume fraction. Moreover, three kinds of surfactant were used here. So, the effect of surfactant content and nature is first investigated reporting the TC of base fluid in function of surfactant quantity. This is shown by Figure 3 at 30°C. As reported earlier [30], it is observed that TC of base fluid decreases when the amount of surfactant is increased, and that the TC is lower than the one of deionized water at the same temperature. This means that surfactants penalize the TC of NF. It is also shown that this effect is independent of surfactant type presently investigated, within the experimental uncertainty. Similar tendencies were obtained at lower and higher temperatures (20 and 40°C), TC being decreased and increased when the temperature is decreased and increased respectively.

Figure 4 shows the effect of temperature and volume fraction on the thermal conductivity enhancement of NF for two nanofluids N_3 and N_4. A similar behavior was also noticed for N_1 and N_2. As often reported in literature, TC of NF increases when both the CNT nanotube volume fraction increases and temperature as well. The TC quickly enhances for lower CNT volume fraction, in particular at 30 and 40°C. In addition, at these temperatures, TC do not follow a linear trend with volume fraction.

The thermal conductivity of NF is reported in Figure 5 (left) for N_1, N_2 and N_3 at 20°C evidencing the effect of surfactant on TC. While TC of base fluid decreases with the amount of surfactant, figure 5 (a) shows that TC of nanofluids increases with CNT volume fraction. When temperature is increased, similar trend is also noticed. Figure 5 also shows that surfactant nature do not influence TC enhancement except at higher volume fraction for SDBS. This can be explained by the

dispersion state of CNT with this surfactant at this volume fraction, as NF appear mainly in the form of aggregates [41]. The effect of CNT aspect ratio is shown in Figure 5 (b) at 30°C. It should be mentioned that this effect is negligible at 20°C; it is increased at 40°C. Higher the aspect ratio, higher is the TC enhancement at high volume fraction. So, this agrees well with previous published results [18,19]. However, the maximum relative deviation, which is around 0.5% at 40°C, is only about 5%. This means that the effect of CNT aspect ratio on TC is here rather weak.
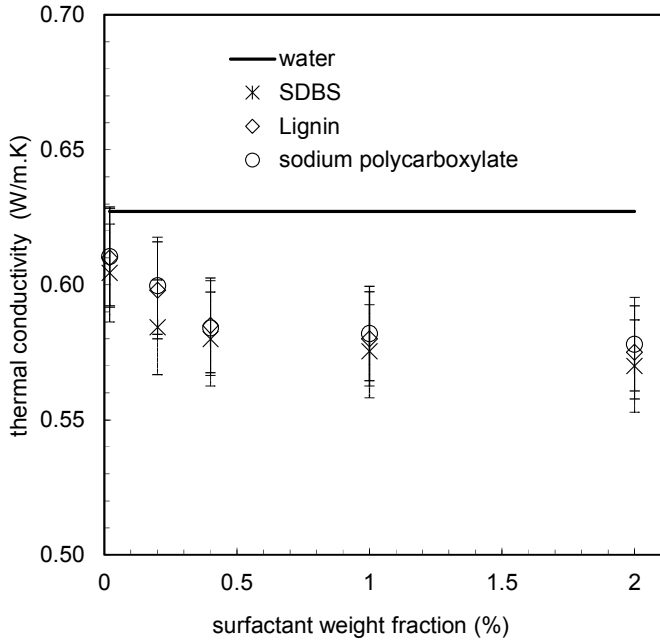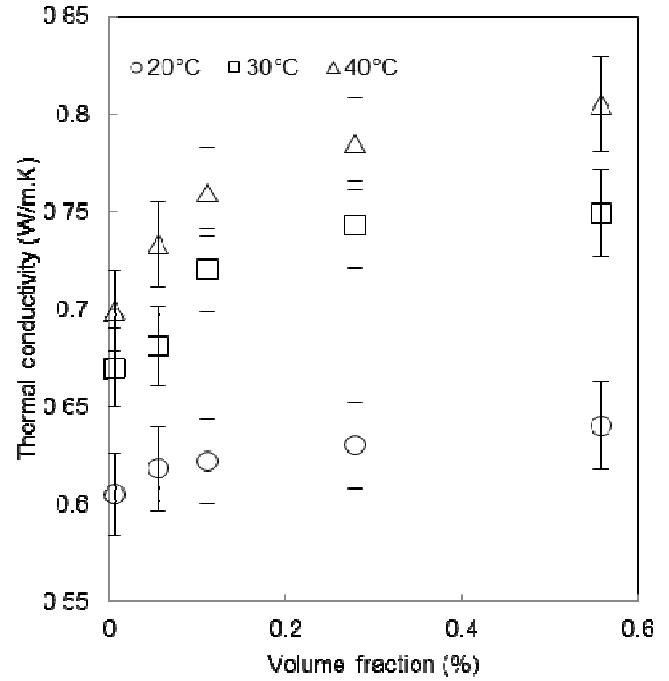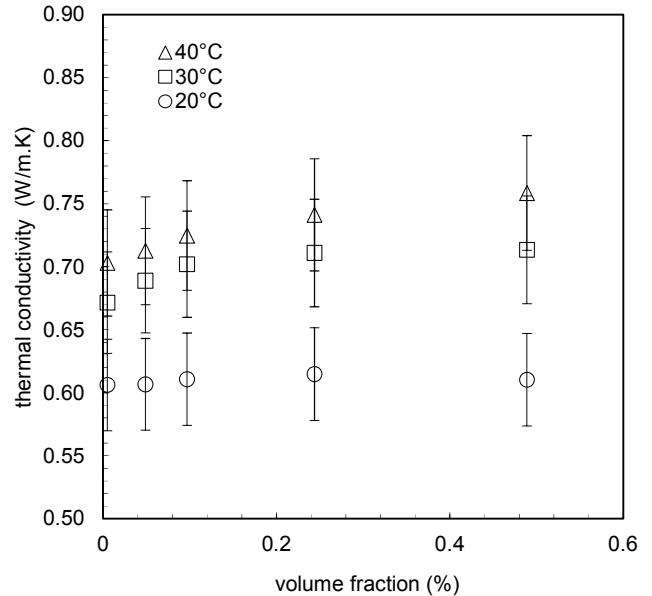

Figure 3. Thermal conductivity of base fluids at 30°C – Impact of nature and quantity of surfactant.

Based on these experimental findings, we focus now on the comparison between experimental data and theoretical correlations presented above considering mainly the influence of volume fraction, temperature and CNT aspect ratio. So, N_3 and N_4 nanofluids are only discussed in the following.

As for TC, Figures 6 and 7 show that relative TC (RTC) of nanofluids which is defined as the ratio of TC of NF to the TC of base fluids. It is shown from these figures that RTC increases quite linearly at 20°C. When the temperature is increased, RTC of nanofluids sharply increases at low volume fraction up to 0.111%. Then, the increasing goes up more slowly. This also evidences that the penalizing effect of surfactant on TC of water reported before is not predominant in comparison with TC of nanotubes. As expected also, the better RTC is achieved for both the higher volume fraction and temperature.
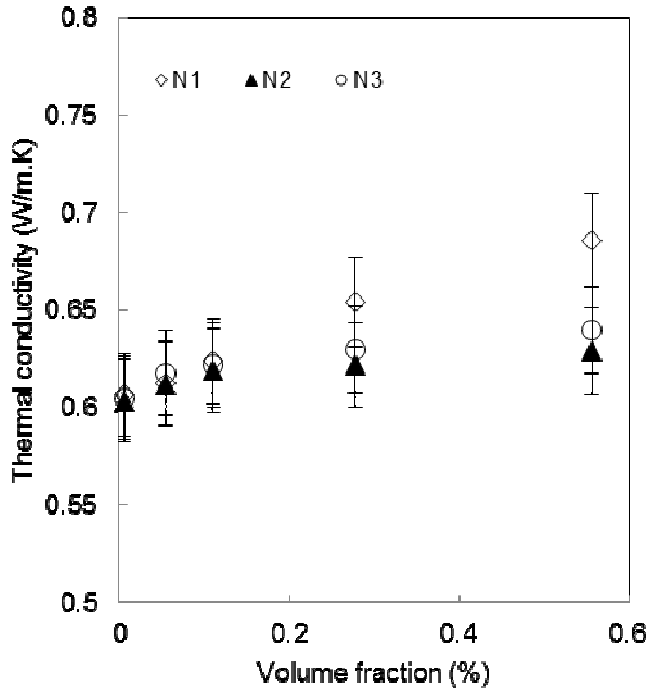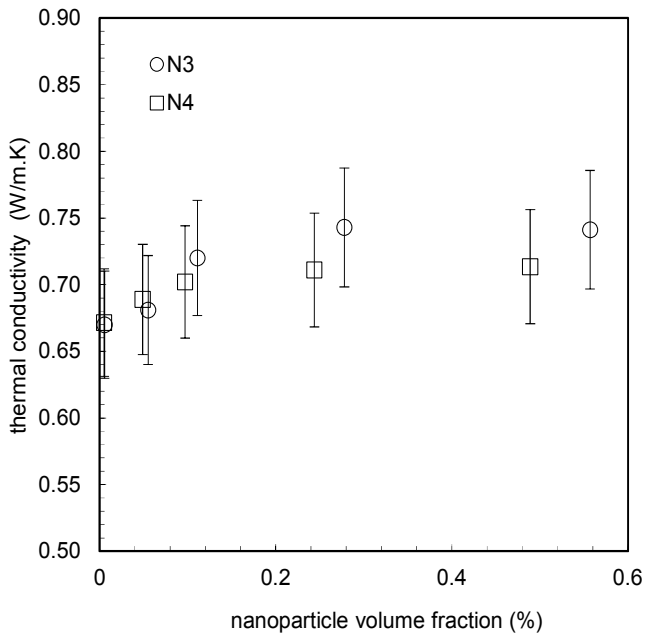

(a)

(b)

Figure 4. Thermal conductivity of N3 (a) and N4 (b) – Impact of CNT volume fraction and temperature.
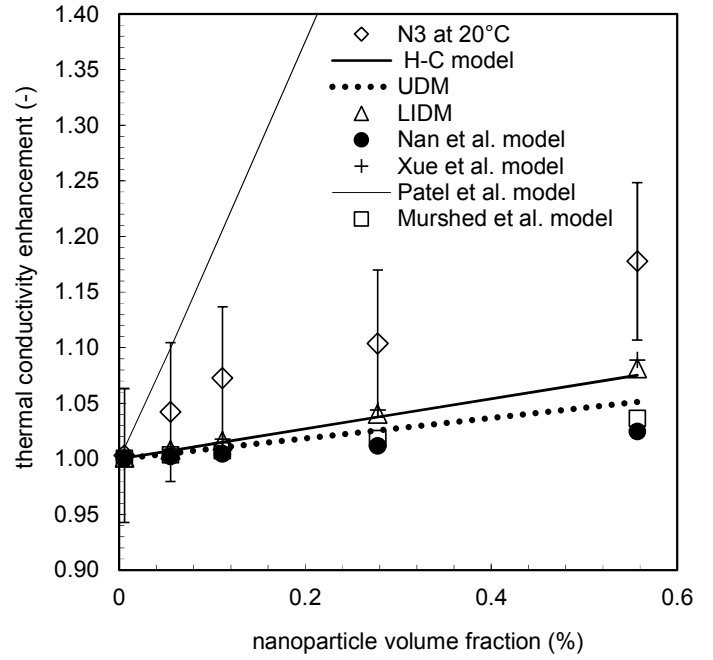
(a)

(a)

(b)

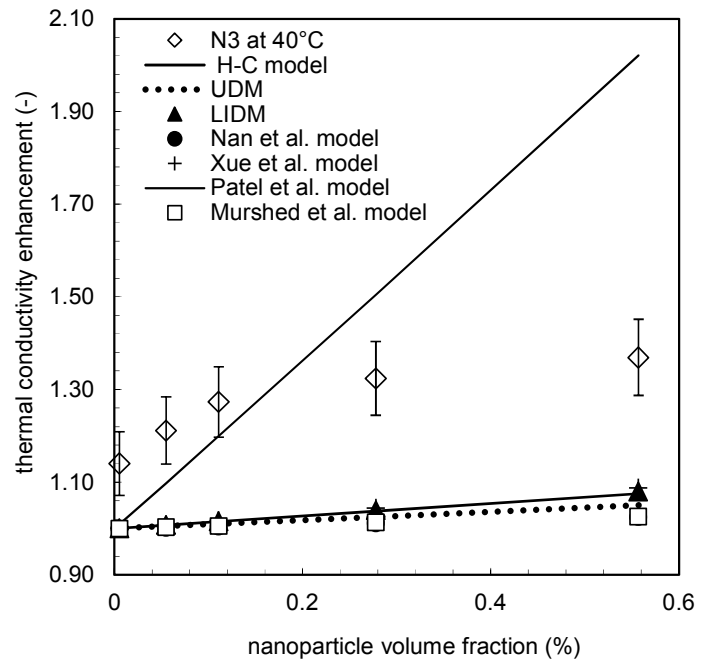(b)

Figure 5. Effect of surfactant (a) and CNT aspect ratio (b) on thermal conductivity of nanofluids.

Figure 6. Relative TC enhancement of N3 in function of nanoparticle volume fraction at 20°C (a) and 40°C (b) – Comparison with theoretical correlations.
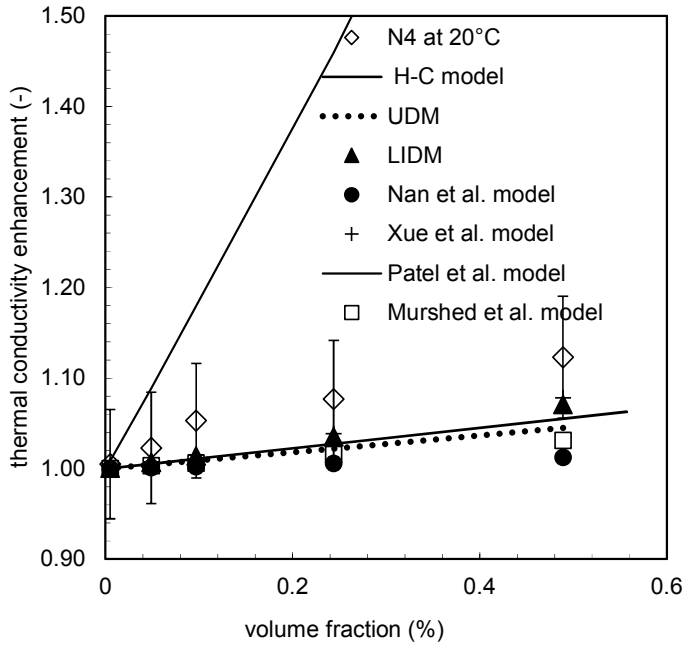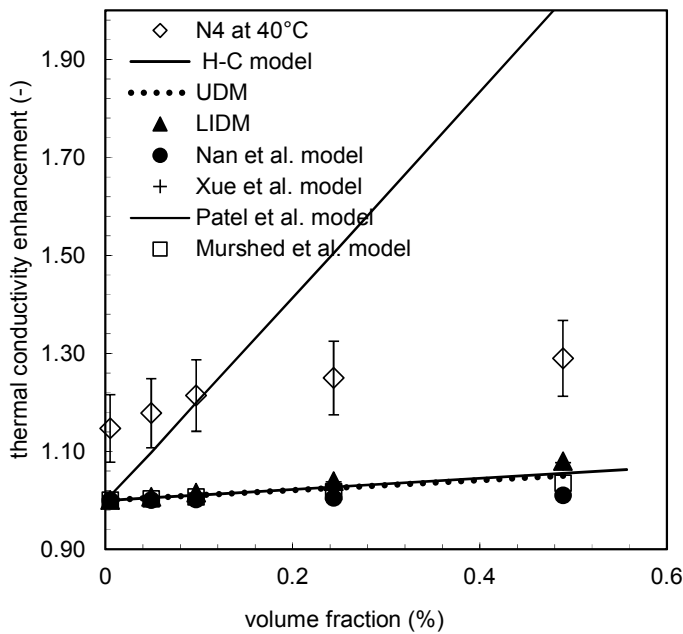
(a)

(b)

Figure 7. Relative TC enhancement of N4 in function of nanoparticle volume fraction at 20°C (a) and 40°C (b) – Comparison with theoretical correlations

It is also observed from both figures that the theoretical predictions are much lower than the experimental data except for the model of Patel et al. [35] which largely overpredicts the experimental data. However, some of the proposed models are able to correlate the experimental data at 20°C (within the experimental uncertainty), the best correlation being obtained with the LIDM model [34] and the model of Xue et al. [36]. An insight in these models indicates that at this temperature, TC is affected by curving/wrapping and special distribution of CNT. It should be mentioned that the accuracy of the model of Murshed et al. [38] can be improved increasing the TC of interfacial layer. When the temperature is increased, all the models greatly deviate in comparison with experimental data and do not report in particular the great enhancement in TC at low volume fraction in nanotubes showing the potential of Brownian motion of CNT even at low temperature. So, the conclusions are not so far easy and one factor cannot be only used to predict RTC enhancement from theoretical point of view.

Finally, this experimental study evidences the potential of carbon nanotubes water-based nanofluids as heat transfer media and coolants for thermal applications due to the great enhancement of TC even at low volume fraction. In addition, none of the selected TC models properly predict TC enhancement of carbon nanotubes water-based nanofluids within the entire range of volume fraction, temperature and nanotubes aspect ratio. While several factors affecting TC have been considered, there is little difference between the studied TC models apart with the model of Patel et al. [35]. So, the results suggest that none of the mechanisms presently investigated is predominant.

CONCLUSION

A parametric experimental study of thermal conductivity of water-based carbon nanotubes was presented. In particular, we have considered the influence of surfactant used to stabilize the nanotubes, carbon nanotubes volume fraction and aspect ratio and temperature as well. It was observed that TC enhancement of CNT nanofluids increase with volume fraction and temperature. The thermal conductivity enhancement is really significant at very low volume fraction, in particular at 30 and 40°C. The effect of the used surfactants on TC enhancement of nanofluids is weak, TC is also weakly affected by the influence of CNT aspect ratio considered here.

The comparison between experiments and models show that theoretical predictions presented above cannot clearly capture the TC enhancement of CNT water based nanofluids presently investigated within the entire range of volume fraction considered and temperature. This evidences a need both to develop appropriate model for TC enhancement prediction of CNT nanofluids and measure TC of this kind of nanofluids before performing numerical studies in heat exchangers and cavities.

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