New temperature dependent thermal conductivity data for water-based nanofluids

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Abstract

This paper presents effective thermal conductivity measurements of alumina/water and copper oxide/water nanofluids. The effects of particle volume fraction, temperature and particle size were investigated. Readings at ambient temperature as well as over a relatively large temperature range were made for various particle volume fractions up to 9%. Results clearly show the predicted overall effect of an increase in the effective thermal conductivity with an increase in particle volume fraction and with a decrease in particle size. Furthermore, the relative increase in thermal conductivity was found to be more important at higher temperatures. Obtained results compare favorably with certain data sets and theoretical models found in current literature.

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1. Introduction

Since the pioneering work by Masuda et al. [1], suspensions with nanometer scale particles have generated considerable interest in the heat transfer community. The evaluation of the thermophysical properties of such mixtures has since been the focus of several research groups. Results obtained generally show spectacular heat transfer enhancement capabilities when compared to the use of a conventional base fluid alone (for example, water, mineral oils or ethylene glycol). Although suspensions containing larger sized particles have been considered for several decades (see, for example, [2–5]), early work on these so-called “nanofluids” (termed by Choi in 1995 [6]) have shown that various types of oxide nanoparticles have excellent dispersion properties in typical coolants and form surprisingly stable suspensions [7]. These special mixtures also appear to have limited sedimentation, channel clogging and other problems often associated with larger sized particles.

Naturally, the bulk of research efforts so far on this fascinating subject has been on the evaluation of mixture “effective” thermophysical properties. These include properties such as the thermal conductivity and viscosity of various types of nanofluids. Of interest, nanofluid effective viscosities have been evaluated by various groups, [1,8–10]. Results generally show a considerable increase in viscosity with particle volume fraction. In the specific case of nanofluid effective thermal conductivity, a review of pertinent literature reveals an important dispersion in data values obtained by various authors. This lack of agreement can be attributed to a number of factors including particle size and shape, particle clustering, particle sedimentation, etc. Although considerable dispersion does exist, it is clear that the thermal properties of nanofluids are considerably more appealing than those of the base fluid alone. Effective thermal conductivity enhancements of 20% were found for as little as 1 to 5% particle volume fractions for various mixtures (see, for example, [6–8,11]). Other published results on experimental determination of effective thermal conductivities include, amongst others, [1,9,12,13]. Furthermore, only a few results have so far surfaced considering the dependence of the effective thermal conductivity values on temperature for various nanofluids [1,14–16]. Although the dependence of nanofluid thermal conductivity on temperature is shown in these papers, only a few data points are generally presented in each case. The
initial work of the present authors on nanofluid thermal conductivity [17], presented considerable amounts of data points for various particle volume fractions over a temperature range covering 5 °C to 45 °C. As the heating/cooling process covered several days, nanoparticle sedimentation was suspected for cases considering greater particle volume fractions (in particular, 6 and 9%).

For the evaluation of nanofluid effective thermal conductivities, researchers have used different techniques with varying degrees of success. Although the transient hot wire method has been used most extensively (see, for example, [16,18–21], others [14] have used the temperature oscillation technique developed by Roetzel and co-workers [22,23] or steady-state parallel-plate techniques [8]. Considerable discussions are made by the various authors with respect to what technique is best suited for the evaluation of nanofluid effective thermal conductivity.

Undeniably, the available data certainly provides interesting insight into nanofluid properties and heat transfer benefits. However, as stated in Kebinski et al. [24], some of the pitfalls in current research on nanofluids include the lack of agreement between results obtained in different research groups as well as the lack of theoretical understanding of the mechanisms responsible for the spectacular heat transfer enhancement capabilities. Indeed, most researchers seem to agree that although nanofluids are by nature two-phase mixtures, it has become generally accepted that classical theories and correlations developed for two-phase flows cannot, in general, be applied in the case of nanofluids. This view, however, is not shared by some researchers. Indeed, some results found in the recent literature (for example, the work by Zhang et al. [16]) show no anomalous enhancements and are quite well predicted by classical models such as the Hamilton and Crosser model. It is therefore clear that further investigation is required in order to develop more adequate and generally accepted models.

Various theories have surfaced trying to explain the reasons of the spectacular heat transfer enhancements of nanofluids commonly found in most papers. Several authors state Brownian motion of particles as a prime factor of the thermal enhancement. Kumar et al. [25], for example, elaborate on the strong relationship between this Brownian motion and temperature. Others also consider the effect of the interfacial layer between the fluid and the particle [26,27]. Typically, they have found that the increase of layer thickness increases the thermal conductivity of the nanofluid. New theories and correlations have appeared specifically for nanofluids over the past 3 or 4 years, for example, [25,28–32]. In general, most of these correlations do not agree with each other very well. Although most of these approaches include terms to consider Brownian motion contributions to the effective thermal conductivity of the mixtures (in some form or another), some include other factors such as the impact of interfacial effects (see, for example, [29]). Most of these correlations also consider other important physical parameters such as particle size, particle volume fraction and mixture temperature effects. Although the presented models do seem to agree with certain sets of experimental data, they are most likely to be in considerable disagreement with data and correlations collected by other authors. As a whole, a lack of available experimental data is partially to blame for these discrepancies as most of these models are developed from data sets containing but a few experimental thermal conductivity values. As such, Koo and Kleinstreuer have stated that to improve the accuracy and versatility of their effective thermal conductivity model, more experimental data sets are needed [28].

With the objective to contribute to the expanding nanofluid properties database, thermal conductivity measurements on water based suspensions with alumina and copper oxide nanoparticles are made and results are presented in this paper. We are particularly interested in evaluating the effective thermal conductivities at relatively high volume fractions (up to 18% for room temperature measurements and up to 9% for temperature effects measurements) as well as collecting a considerable number of data points over a wide temperature range for several particle volume fractions.

2. Experimental apparatus and procedures

2.1. Description of experimental apparatus, procedures and validation

Thermal conductivity measurements were made using the Decagon devices KD2 Thermal analyzer. This apparatus has 5% accuracy over the 5 °C to 40 °C temperature range and also meets the standards of both ASTM D5334 and IEEE 442-1981. This portable device uses the transient line heat source approach to determining the fluid thermal properties. It basically comprises a hand-held readout unit and a single-needle sensor that is inserted into the fluid specimen. A single reading generally takes 2 minutes. The first 90 seconds are used to ensure temperature stability, after which the probe is heated for 30 seconds using a known amount of current. The probe also contains a thermistor which measures the changing temperature while the microprocessor stores the data. At the end of reading, the thermal conductivity of the fluid is computed using the temperature difference vs. time data. More information on the theory behind the technique is available in the references [33] and [34]. The KD2 Thermal Analyzer was also used with success by Wen and Ding [35] in their work on nanofluids.

For the thermal conductivity evaluation at room temperature, the fluid specimen is placed inside a test-tube. The apparatus probe is then placed securely in the tube and the thermal conductivity measurement of the fluid is taken. As the determination of temperature-dependent values of thermal conductivity of nanofluids is the primary objective of this paper, the fluid specimen is placed inside a heated, insulated enclosure, Fig. 1. Although no important particle sedimentation is expected in the time frame required to conduct a series of measurements, in order to minimize possible sedimentation, a miniature, mechanical type mixer was used to periodically stir the nanofluid inside the enclosure. The mixer was activated via a switch placed outside the enclosure. The distance between the position of the KD2 probe and that of the mixer shaft is approximately 15 mm. Heating of the enclosure was stopped when the temperature reached approximately 42 °C. The normal temperature...
Fig. 1. Experimental setup.

Fig. 2. Distilled water thermal conductivity data comparison with correlation.

Validation of procedure was done by comparing thermal conductivity ("k") results obtained for distilled water with available correlations/data in literature [36,37] for the entire considered temperature range, Fig. 2. As one can see, the results obtained for water compare very favorably with the textbook correlation. With the exception of a very few data points, the thermal conductivity values collected, when compared to the textbook correlation, fell within the 5% stated accuracy of the KD2 Thermal analyzer (shown by error bars for each data point). The maximum relative error determined experimentally on the collected data did not exceed 8%. In light of this satisfactory validation, we can conclude with confidence that the consistency and accuracy of the instrument, as well as the experimental procedure, are quite acceptable.

Experiments presented in this paper were conducted on Al$_2$O$_3$–water and CuO–water nanofluids purchased from Nano-phase Technologies. Particle sizes used in these suspensions are 47 and 36 nm for the Al$_2$O$_3$ nanofluid and 29 nm for the CuO nanofluid. The delivered products were in considerably concentrated forms (i.e. approximately 50% wt). For applications as a heat transfer medium, lower volume fractions are preferred, thus the original mixture required dilution to obtain more practical concentrations. In order to determine the volume fraction of the solution, Eq. (1) was used:

\[
\rho_{nf} = \phi \cdot \rho_p + (1 - \phi) \cdot \rho_{bf}
\]

where, \(\phi\) is the particle volume fraction and \(\rho_{nf}\), \(\rho_p\) and \(\rho_{bf}\) are, respectively, the nanofluid, particle material and base fluid (i.e. water) densities. Knowing the density of the base fluid (i.e. water, \(\rho_{bf} = 1000 \text{ kg/m}^3\)) and of the particles (i.e. for the 47 nm Al$_2$O$_3$ nanoparticles, \(\rho_p = 3880 \text{ kg/m}^3\); for the 36 nm Al$_2$O$_3$ nanoparticles, \(\rho_p = 3600 \text{ kg/m}^3\) and for the 29 nm CuO nanoparticles, \(\rho_p = 6500 \text{ kg/m}^3\)) and by measuring the volume and the weight of the mixture (i.e. \(\rho_{nf}\)), one can determine the volume fraction "\(\phi\)" of the mixture. Once the mixture was prepared and the volume fraction determined, thermal conductivity measurements were conducted.

3. Presentation of experimental results and discussion

3.1. Nanofluid thermal conductivity results at ambient temperature

Several measurements at room temperature (in the 21 °C to 23 °C range) were carried out for distilled water and the three types of nanofluids considered with several different particle volume concentrations between 0 and 18%. Figs. 3, 4 show results for, respectively the water/CuO and the Al$_2$O$_3$ nanofluids. In both figures, the data trends ±5% are presented. As can be seen, all data points fall inside the accuracy range of the apparatus. As expected, one can see that the effective thermal conductivity increases with nanoparticle volume fraction. In the particular case of the Al$_2$O$_3$ nanofluids, no clear differences are noticed at room temperature between results obtained with the two different particle sizes, Fig. 4. This would seem to be in accordance with predictions obtained from the model developed by Feng et al. [38]. Indeed, the obtained results at room temperature by these authors seem to indicate that particle size has drastic effects on thermal conductivity only in the case of very small particle sizes (i.e. less than approximately 10 nm). According to their results, no significant dependence of nanoparticle size on thermal conductivity is found for nanoparticles greater than 20 nm in diameter. The model proposed by Feng et al. expresses the effective thermal conductivity as a function of the thickness of the nanolayer, the nanoparticle size, the nanoparticle volume fraction and the thermal conductivities of the suspended nanoparticles and of the base fluid.

From the data presented in Figs. 3 and 4 two single variable (i.e. particle volume fraction \(\phi\)) correlations can be found by

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simple linear regressions. The relative values of thermal conductivity, \( k_r \), are obtained by dividing the effective thermal conductivity of the nanofluid by the value of the thermal conductivity of the base fluid (i.e. water) at the same temperature (i.e. ambient temperature). Eqs. (2) and (3) are respectively obtained for the water/CuO and water/Al\(_2\)O\(_3\) nanofluids. Both these relationships have a 95\% \( R^2 \) value.

\[
k_r = 1.74\phi + 0.99 \tag{2}
\]

\[
k_r = 1.72\phi + 1.0 \tag{3}
\]

Results obtained in this present work for the 47 nm Al\(_2\)O\(_3\) nanofluid are compared with various experimental results available in current literature, Fig. 5. The linear or spline fits were added by the present authors for clarity and ease of comparison between results. As already stated in available literature, one can see that although the results obtained in this present paper are within the general range of values obtained by other authors, considerable dispersion is found between results from the various sources. As previously mentioned, this dispersion could possibly be attributed to several factors including particle shape and size, particle clustering, particle sedimentation, etc. These factors are influenced by a number of important technical parameters such as nanofluid manufacturing processes, the use of surfactants (see, for example, [13]) or other techniques to aid nanoparticle dispersion (see, for example, [18,39]), as well as different experimental techniques and procedures [40]. Furthermore, as Fig. 5 illustrates quite well, with a few possible exceptions, particle size does seem to have a distinct effect on nanofluid effective thermal conductivity. Generally, the smaller the particle size, the greater the effective thermal conductivity. Although this may contradict the previously mentioned findings of Feng et al. [38], this would seem quite plausible as for a same volume fraction, there are more particles and therefore more contact surface area between the solid and liquid phases (see, for example, [41]). In the specific case of the data collected for this present work, results show that, for example, a nanofluid with a particle volume fraction of say 6% has a 12% increase in thermal conductivity over the base fluid. Furthermore, considering all the data from the various sources, the Hamilton–Crosser equation strikingly seems to represent a good average nanofluid thermal conductivity value at ambient temperature.

3.2. Temperature dependent nanofluid thermal conductivity results

As we are mainly interested in investigating the nanofluid thermal conductivity as a function of temperature, several tests were conducted for various particle loadings. Fig. 6 illustrates a typical nanofluid thermal conductivity behavior in function of temperature. In order to somewhat verify the apparatus accuracy, the trend line with ±5% lines are included with the data. As can be seen, all points fall clearly within the specified accuracy range for the apparatus. Similar accuracy results are found for all other sets of data points.

Figs. 7 and 8 show comparisons of temperature-dependent results with various authors. Results are presented in relative effective thermal conductivity form, representing the ratio of effective thermal conductivities of the nanofluid ("\( k_{nf} \)) with respect to the base fluid ("\( k_{bf} \)) at the same temperature, Eq. (4):

\[
k_r = \frac{k_{nf}(T)}{k_{bf}(T)} \tag{4}
\]

In Fig. 7, results for a particle loading of 4\% (in volume) for a 47 nm particle size Al\(_2\)O\(_3\) nanofluid are compared with those obtained by Das et al. [14], Chon et al. [30] and Masuda et
As can be seen, for this type of nanofluid, thermal conductivity values obtained in this investigation are generally in the vicinity of those obtained by Das et al. [14] and by Chon et al. [30]. Effective thermal conductivity results obtained by Masuda et al. [1] seem to be considerably higher than those obtained in more recent work. They however tested a nanofluid with average particle size of 13 nm, which is significantly smaller than the ones used by the other authors. The same discussion could be used to explain the slightly higher conductivity values obtained by Das et al. However, they do follow the same general behavior. Clearly, temperature has a considerable effect on nanofluid thermal conductivity.

In the case of the CuO based nanofluid, Fig. 8, results indicate the same general behavior as the Al₂O₃ nanofluid. The presented results are compared to those by Li and Peterson [32] and Das et al. [14]. Direct quantitative comparisons are somewhat difficult in this case as particle volume fractions used by the different authors differ somewhat (i.e. 2 and 4% in the case of Li and Peterson, 4% in the case of Das et al. and 3.1% for results presented in this paper). The results obtained by Li and Peterson [32] exhibit effective thermal conductivity values considerably higher than those obtained by Das et al. [14] and those presented in this paper. Obtained results do compare well, at least qualitatively, with those obtained by Das et al. The rates of increase of nanofluid effective thermal conductivity with temperature of both sets of data are essentially the same and the observed differences can, at least in part, be attributed to the fact that the nanofluid specimen tested by Das et al. had a higher particle volume fraction (i.e. 4% vs. 3.1%).

Figs. 9, 10 present the influence of temperature and nanofluid particle volume fraction on effective nanofluid thermal conductivity. The increase in the effective thermal conductivity of nanofluids with temperature is clearly seen. Between 20°C and 40°C, one notes on the average an increase in thermal conductivity of approximately 15% for each type nanofluid. In comparison, the enhancement of thermal conductivity of pure distilled water is approximately 5% between 20°C and 40°C (see Fig. 2). In comparison with distilled water, the addition of nanoparticles gives a better enhancement with temperature for low volume fraction of particles. As previously mentioned, some explain the enhancement of thermal conductivity of nanofluids with the temperature by Brownian motion [25, 28]. Typically, an increase in temperature increases the Brownian motion of particles.

Another seemingly important parameter when considering nanofluids is the particle size. As previously mentioned, at room temperature, no considerable difference was noticed between results obtained with a 47 nm nanoparticle nanofluid and those obtained with a 36 nm particle nanofluid. As can be seen, however, in Figs. 11 and 12, under the influence of increas-
to point out that, when comparing results presented in Fig. 11 (for a 3% particle volume fraction), the difference in thermal conductivity gradually increases with temperature. This would seem to be a possible explanation as to why no discernable difference was noticed for nanofluid effective conductivity at room temperature (see Fig. 4). For a higher particle volume fraction however, i.e. 9%, Fig. 12, thermal conductivities for nanofluids with 36 nm particle diameters are clearly higher than those with 47 nm particles.

3.3. Comparison of experimental results with theoretical predictions

As previously stated, a few theoretical models have so far surfaced attempting to predict nanofluid effective thermal conductivity behavior. It is widely believed that models developed in the past for mixtures containing larger sized particles are not applicable to mixtures containing nano-scale particles. Others have however stated otherwise. Arguably, the most recognized earlier model developed for suspensions is the Hamilton and Crosser model [42] developed in 1962, Eq. (5):

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

(5)

where the subscripts “nf”, “bf” and “p” represent respectively “nanofluid”, “base fluid” and “particle” and “n” is the “shape factor” and defined as Eq. (6):

$$n = \frac{3}{\Psi}$$

(6)

\(\Psi\) is called the “sphericity” and defined as the area-ratio of a sphere over a nanoparticle of given identical volume. If the Al₂O₃ nanoparticles under consideration are considered spherical, \(\Psi = 1\), Eq. (5) is reduced to Eq. (7):

$$k_r = \frac{k_{nf}}{k_{bf}} = \frac{k_p + 2 \cdot k_{bf} - 2 \cdot \phi \cdot (k_{bf} - k_p)}{k_p + 2 \cdot k_{bf} + \phi \cdot (k_{bf} - k_p)}$$

(7)

Visibly, the most notable drawback of the Hamilton Crosser model is that important physical parameters such as temperature and particle size are not considered.

The recent model developed by Koo and Kleinstreuer [28] takes into effect physical parameters such as particle size, temperature and nanoparticle volume fraction as well as the impact of Brownian motion energy transfer by the nanoparticles. They have decomposed the nanofluid effective thermal conductivity into two separate functions, one based on a “static” thermal conductivity, the other a “dynamic” function considering the “Brownian” thermal conductivity, Eq. (8):

$$k_{nf} = k_{static} + k_{Brownian}$$

(8)

In Eq. (8), the dynamic (or Brownian) component is defined as expressed in Eq. (9):

$$k_{Brownian} = 5 \times 10^4 f_1 \phi \rho_{bf} C_{p_{bf}} \sqrt{\kappa T \rho \partial \phi \over \partial T} f_2$$

(9)

where \(f_1 = f(\phi)\), \(f_2 = f(\phi, T)\), \(C_{p_{bf}}\) is the specific heat of the base fluid, \(\kappa\) is the Boltzmann constant, \(d_p\) is the average
particle diameter. Details and values for functions $f_1 = f(\phi)$, $f_2 = f(\phi, T)$ are found in the referenced work. The authors however also stipulate that to improve the accuracy and versatility of their $k_{eff}$ model, more experimental data sets are required. The model developed by Chon et al. [30] also includes temperature and particle size effects, Eq. (10):

$$k_r = 1 + 64.7\phi^a \left( \frac{d_{ef}}{d_p} \right)^b \left( \frac{k_p}{k_{lff}} \right)^c Pr^d Re^e$$

(10)

where $Pr$ and $Re$ are, respectively, the Prandtl and Reynolds numbers and $a = 0.746, b = 0.3690, c = 0.7476, d = 0.9955$ and $e = 1.2321$. The Reynolds number is based on the Brownian motion velocity. The developed model was based on measurements on water–Al$_2$O$_3$ nanofluids.

The two variable linear regression relationships proposed by Li and Peterson [32] (one for water/alumina nanofluids, the other for water/copper oxide nanofluids, respectively, Eqs. (11) and (12)) relate nanofluid effective thermal conductivity to temperature and nanoparticle volume fraction. The developed relationships cover a relatively small temperature range, say 27°C to 36°C for three particle volume fractions (2%, 6% and 10%).

$$k_r = 1 + 3.761088\phi + 0.0179247 - 0.30734$$

(11)

$$k_r = 1 + 0.7644815\phi + 0.018689T - 0.46215$$

(12)

The results obtained in this present study are compared to these recent aforementioned correlations developed by Chon et al. [30], Koo and Kleinstreuer [28] and Li and Peterson [32] as well as with the classic Hamilton–Crosser relationship [42]. Figs. 13–15 illustrate comparisons in results considering the influence of particle volume fraction on effective thermal conductivity obtained in this present work with various proposed correlations in available literature. The data presented was measured at ambient temperature. As can be seen, the data from this current work compares well with the correlation developed by Chon et al. [30]. Furthermore, one can see that the results obtained from the Li and Peterson [32] relationship, Eq. (12), show $k_r < 1.0$ for the entire $\phi$ range. This can be explained by the fact the correlation was developed, as previously mentioned, for a temperature range between 27°C to 36°C. The ambient temperature when the data was collected for this present work was in the vicinity of 20°C. Although the correlations developed by Li and Peterson [32] yield excellent results in the prescribed temperature range, more data points will need to be considered in order to cover a wider range of temperatures. Also of interest, as can be seen in all three cases, i.e. Figs. 13–15, the Hamilton–Crosser relationship yields good results for small values of particle loadings.

The general influence of temperature on nanofluid thermal conductivity with the comparisons can be appreciated from the results presented in Figs. 16 and 17. As can clearly be seen, the results presented in this paper are, as in the case of the influence of particle volume fraction, quite well described by the correlation developed by Chon et al. It is also interesting to note that, as found by [16], the Hamilton–Crosser model remains a good, at least at ambient temperature, representation of nanofluid effective thermal conductivity. As we can however see, both relationships developed by Li and Peterson strongly over estimate the conductivity values for the nanofluids considered in this present paper.

4. Conclusion

New experimental data of the temperature dependence of the thermal conductivity of alumina and copper oxide based nanofluids have been presented in this paper. Results clearly show the predicted overall effect of an increase in the effective thermal conductivity with an increase in particle volume frac-

References

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Fig. 16. Temperature dependent nanofluid effective thermal conductivity for water/Al₂O₃ nanofluid.

Fig. 17. Temperature dependent nanofluid effective thermal conductivity for water/CuO nanofluid.

Fig. 17. Temperature dependent nanofluid effective thermal conductivity for water/CuO nanofluid.

Fig. 16. Temperature dependent nanofluid effective thermal conductivity for water/Al₂O₃ nanofluid.

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