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Experimental investigation and proposed correlations for temperature-dependent thermal conductivity enhancement of ethylene glycol based nanofluid containing ZnO nanoparticles

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ABSTRACT

Experimental study of effective thermal conductivity of ZnO/EG nanofluid is presented in this research. The nanofluid was prepared by dispersing Zno nanoparticles in ethylene glycol using a sonicator and adding surfactant. Ethylene glycol based nanofluid containing ZnO nanoparticle with a nominal diameter of 18 nm at different solid volume fractions (very low to high) at various temperatures was examined for the investigation. The thermal conductivity of nanofluids is experimentally measured with THW method and it is found that the thermal conductivity of nanofluids increase with the nanoparticle volume concentration and temperature. Also, based on experimental values of thermal conductivity of nanofluid, three experimental models are proposed to predict thermal conductivity of nanofluids. The proposed models show reasonably excellent agreement with our experimental results.

1. Introduction

Cooling is an essential operation in several industrial applications and engineering designs. In order to enhance the heat transfer rate in a heat exchanger, either the heat transfer area or temperature gradient or the thermal conductivity of fluids exchanging heat has to be increased [1]. Among these techniques, increasing the thermal conductivity of the heat transfer fluids is the most attractive option.

Nanofluid is prepared by dispersing solid particles, fibers, or tubes with lengths of the order of 1–100 nm in traditional heat transfer fluid such as water, oil, and ethylene glycol (EG) which are inherently poor heat transfer fluids. Due to small sizes and very large specific surface areas of the nanoparticles, nanofluids have superior properties like high thermal conductivity, minimal clogging in flow passages, long-term stability, and homogeneity. Hence, nanofluids have a wide range of potential applications like electronic, automotive, and nuclear applications where improved heat transfer or efficient heat dissipation is required [2].

In spite of such potential benefits, the nanofluid technology is still limited for commercial use as there is yet no proven standardized design technique to accurately

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predict important heat transfer characteristics such as the nanofluids' thermal conductivity. Some theories have been proposed over the years by different researchers to explain the thermal conductivity augmentation such as heat transfer due to Brownian motion of particles [3, 4] and the interfacial layer formation at solid–liquid interface [5], but there is a lack of agreement between thermal conductivity values measured at so-called “room temperature”.

The Maxwell [6] model is based on the conduction through a stationary random suspension of spheres. Yu and Choi [7] proposed a modified Maxwell model to account for the effect of nano-layer by replacing the thermal conductivity of solid particles with the modified thermal conductivity of particles which is based on the effective medium theory. The Koo and Kleinstreuer [8] model is based on determining the effective thermal conductivity by incorporating the Brownian motion effect. The Xie [9] model is based on the Fourier's law of heat conduction for low particle loadings including the nanolayer effects. The Burggeman model [10] is based on the differential effective medium theory to estimate the thermal conductivity at high particle concentrations. One of the main objective of this paper is to investigate the accuracy of the existing theoretical models by comparing the predicted value versus the experimental data of thermal conductivity for ZnO/EG nanofluids and to develop new models.

Most experimental observations of nanofluids with just small nanoparticle volume fractions showed that knf will significantly increase when it is compared to the base fluid. As an example, Lee and Choi [11] investigated CuO-water/ethylene glycol nanofluids with particle diameters of 18.6 and 23.6 nm as well as Al_2O_3- water/ ethylene glycol nanofluids with particle diameters of 24.4 and 38.4 nm and discovered a 20% thermalconductivity increase at a volume fraction of 4%.

Li and Peterson [12] provided thermal conductivity expressions in terms of temperature (T) and volume fraction by utilizing curve fitting for CuO-water and Al_2O_3-water nanofluids. Recently, Mintsa [13] provided new thermal conductivity models for Al_2O_3-water and CuO-water nanofluids with particle sizes of 47, 36, and 29 nm by curve fitting of their in-house measured data. Murshed [14] measured a 27% increase in 4% TiO_2-water nanofluids with particle size of 15 nm and 20% increase for Al_2O_3-water nanofluids. However, Duangthongsuk [15] reported a more moderate increase of about 14% for TiO_2-water nanofluids. Quite surprising, Moghadassi [16] observed a 50% increment of thermal conductivity for 5% CuO-monoethylene glycol (MEG) and CuO-paraffin nanofluids.

In an industrial cooling application, coolant is used in a closed-loop circulation system where it undergoes both heating and cooling cycles. Hence, a rigorous study on the thermal conductivity of nanofluids is required as a function of temperature.

Das [17] systematically discussed the relationship between the thermal conductivity and temperature for nanofluids, noting a significant increase of k_{nf} (T). More recently, Abareshi et al. [18] experimentally obtained the thermal conductivity of Fe_3O_4-water and asserted that k_{nf} increases with temperature (T). Tavmanet al. [19] measured SiO_2-water, TiO_2-water, and Al_2O_3-water by the 3-ω method and claimed without showing the actual data that there is no anomalous thermal conductivity enhancement with increment of both volume fraction and temperature. Whether anomalous enhancement relationship between knf and temperature (T) exists or not is still an open question [20].

Despite their lower thermal conductivities, metal oxide nanoparticles are preferred over pure metals for preparation of nanofluids because of their resistance to oxidation, lower density, and hence better dispersion [21]. While Al_2O_3 [22,23] and CuO [24] have been widely reported, ZnO remains to be one of the less-explored metal oxides for formulation of nanofluids.

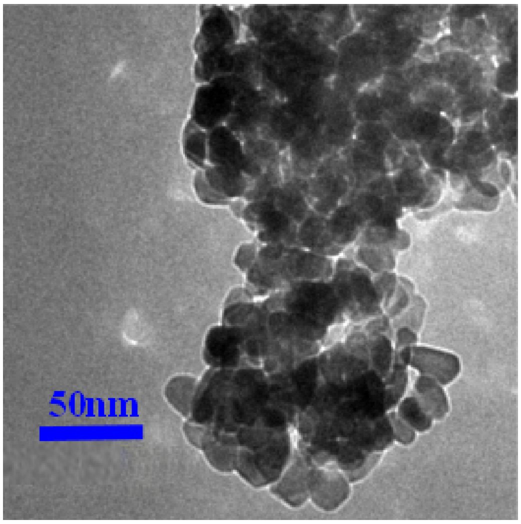
In this experimental study, a two-step method was used to prepare ethylene glycol based nanofluid containing ZnO nanoparticles with different solid volume concentrations. The thermal conductivity of ZnO-EG nanofluid was experimentally measured in terms of temperature and solid-phase concentration. Then, the measured value of thermal conductivity was compared with the theoretical value of existing models such as Yu-Choi, Lu –lin, and Hamilton-Crosser models. Three experimental correlations which are a function of solid volume fraction and temperature are proposed based on the experimental data.

2. Preparation of nanofluid

In the current experiment, ZnO-EG nanofluid was prepared using two-step methods. The preparation of nanofluid must ensure proper dispersion of nanoparticles in the base fluid which includes convenient mechanism such as addition of surfactants or control of pH value to attain the stability of the suspension against sedimentation of nanoparticles. In the current experiment, three effective ways were used to stabilize the suspension. These methods are: use of ultrasonic processor, addition of surfactants and changing the pH value of the nanofluid. Ethylene glycol was used as the base fluid. ZnO nanoparticles with the desired volume concentrations (0.05 (5.0%), 0.04 (4.0%), 0.03 (3.0%), 0.02 (2.0%), 0.015 (1.5%), 0.01 (1.0%), 0.005 (0.5%), 0.0025 (0.25%), 0.00125(0.125%), and 0.000625 (0.0625%)) have been added to pure EG. The mean grain size of ZnO Nanoparticles is 18 nm and was produced by US research nanomaterial, Inc..

After adding nanoparticles, the suspensions were subjected to ultrasonic vibrator for 3-5h in order to get a uniform dispersion and stable suspension. Cetyl Trimethyl Ammonium Bromide (CTAB) surfactants were utilized to ensure better stability without changing nanofluid's thermophysical properties since the surfactant concentrations used in the nanofluid were very low (e.g. volume percentage around 0.01%) [25]. Stability of the prepared nanofluid was studied by measuring the PH values. The PH was measured using a PH meter (HANNA, HI 83141) and the obtained values were far from Iso-Electric Point (I.E.P of Zinc oxide equals to 9.5) of ZnO nanoparticles [26].

In addition stability is achieved because of very large repulsive force among the nanoparticles when PH is far



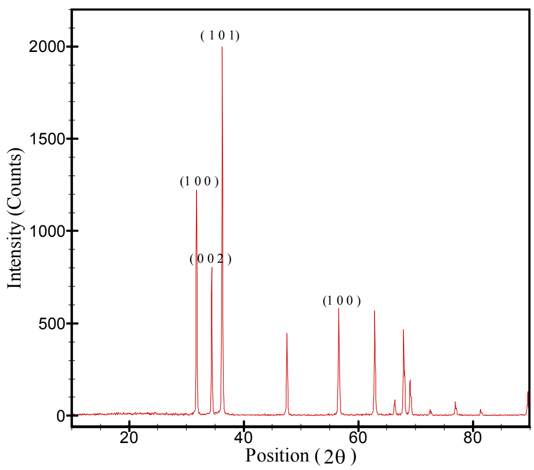


Fig. 1. TEM image and XRD patterns of ZnO nanoparticles

from isoelectric point. When the PH value is far greater or smaller than I.E.P, the particles can hardly agglomerate because the nanoparticle suspension has a higher surface potential [27].

In this article, a transmission electron microscope (TEM) and XRD pattern were used to approximate the size and shape of the ZnO nanoparticles. Fig. 1 illustrates that the shape and size of nanoparticles are spherical and about 18 nanometer, respectively. This method is commonly used by a wide range of researchers [28, 29].

3. Thermal conductivity of nanofluid

3.1 *Thermal conductivity measurement*

The measurement of thermal conductivity is one of the significant aspects to analyze the thermal properties of ZnO- EG nanofluid.

Hamilton–Crosser (H–C) [30] model was one of the basic models to predict the thermal conductivity of solid–liquid mixtures, K_{eff}. This model is applied to estimate thermal conductivity of the mixtures for which the ratio of solid phase thermal conductivity to that of liquid phase is greater than 100. The H-C equation is:

$$\frac{k_{eff}}{k_f} = \frac{k_p + (n-1)k_f + (n-1)\varphi(k_p - k_f)}{k_p + (n-1)k_f - \varphi(k_p - k_f)} \quad (1)$$

where k_p and φ are thermal conductivity and volume fraction of nanoparticles, respectively and k_f is the thermal conductivity of base fluid. *n* is the empirical shape factor given by:

$$n = \frac{3}{\psi} \quad (2)$$

where ψ is the particle sphericity defined by the ratio of the surface area of a sphere with a volume equal to that of the particle, to the surface area of the particles. For spherical particles the value of *n* is 3.

Another model applied for calculating the thermal conductivity of the nanofluids was proposed by Yu and Choi [31] which can be expressed as:

$$\frac{k_{eff}}{k_f} = \frac{k_p + 2k_f + 2\varphi(k_p - k_f)(1+\beta)^3}{k_p + 2k_f - \varphi(k_p - k_f)(1+\beta)^3} \quad (3)$$

where, β is the ratio of the nano-layer thickness to the original particle radius.

Lu and Lin also proposed another model to predict the thermal conductivity of nanofluids as below for spherical particles [32]:

$$\frac{k_{eff}}{k_f} = 1 + 2.25\varphi + 2.27\varphi^2 \quad (4)$$

Among measuring different thermophysical properties, thermal conductivity is generally regarded as the most difficult property to be measured due to some errors associated during the measuring operation. In the current study, the thermal conductivity of ZnO-EG nanofluids with different solid volume fraction was measured by using a KD2 Pro (decagon Inc.) thermal property analyzer with a maximum error of about 5%. The "thermal conductivity ratio" is defined as the ratio of nanofluid thermal conductivity to the water thermal conductivity.

The measured thermal conductivity ratio of ZnO dispersed ethylene glycol based nanofluid is shown in Fig. 2 as a function of the solid volume fraction of nanoparticles.

In order to compare, the enhanced thermal conductivity ratio calculated using Hamilton–Crosser model (Eq. 1), Yu–Choi model (Eq. 3), and Lu-Lin model (Eq. 4) is also shown in Fig. (2).

The thermal conductivity ratio increases nonlinearly with an increase in solid concentration. The maximum value of thermal conductivity belongs to the maximum solid volume fraction.

Also, it can be seen that the rate of thermal conductivity increase at low concentration is much greater than that at high concentration. The reason may be that the increase in nanofluid viscosity is much higher than the enhancement in thermal conductivity at higher solid concentrations. The values of thermal conductivity ratio recorded from the current experimental study are higher than that predicted by applying the Hamilton–Crosser (H–C) and Lu-Lin models but Yu-Choi model predicts values higher than the measured data.

It shows that the aforementioned models are unable to calculate the thermal conductivity of ZnO-Water nanofluid. This is because of the fact that the effects of significant factors such as the particle size, temperature, and interfacial layer on the thermal conductivity of nanofluids have not been considered in these models. It is important to note that the thermal conductivity of nanofluids depends on parameters like the thermal conductivity of solid particles and base fluid, particle concentration, shape of particles, thickness and thermal conductivity of nanolayer [33], stability of nanofluid, and temperature.

3.2 *Effect of temperature on thermal conductivity*

The effect of temperature on enhancement of thermal conductivity of nanofluids was also studied by measuring the thermal conductivity of nanofluids with a wide range of solid volume fractions from 0.0625% to 5% at different temperatures ranging from 24.7 to 50°C in Fig. 3. Thermal conductivity is measured after placing the nanofluid inside a temperature bath with sufficient isolation to prevent heat dissipation during the experiment.

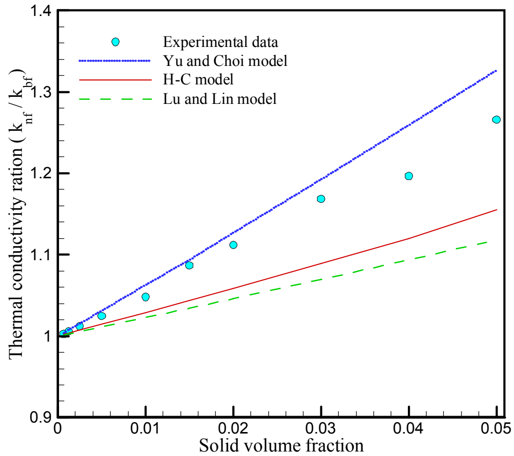


Fig. 2. Comparison between theoretical models and experimental data

As the temperature increases, serious increases in thermal conductivity are evident for all solid volume fractions, especially for high concentrations. Significant increase of relative thermal conductivity with respect to increasing temperature indicates that thermal conductivity depends strongly on temperature. This behavior is consistent with the previously reported results for nanofluids [34, 35, 17].

This is due to higher temperatures of the fluid that the nanoparticles agglomeration would break more easily and the nano-size particles will disperse more uniformly inside the water. Indeed, from a theoretical (i.e. kinetics) view-point, with the increment of the nanofluid's bulk temperature (T), molecules and nanoparticles are moreactive and are able to transfer more energy from one location to another per unit of time.

3.3 *Proposed model:*

In this study, three experimental correlations have been proposed for thermal conductivity of ZnO-EG nanofluid (18nm) based on the experimental data as follow:

Experimental model 1:

$$\frac{k_{nf}}{k_{bf}} = 0.24859\ T^{\,2.504\varphi^{0.7974}} + 0.7492 \qquad (5)$$

$$R^2 = 0.99$$

Experimental model 2:

$$\frac{k_{nf}}{k_{bf}} = \frac{(46.59 + T)}{(46.098 - 135.23\varphi)} - 0.02187\ T \qquad (6)$$
$$R^2 = 0.99$$

Experimental model 3:

$$\frac{k_{nf}}{k_{bf}} = 1.1164\ T^{0.10467 + 0.9802\varphi} - 0.1743 Ln(T) \qquad (7)$$

Fig. 4 shows the comparison between experimental data and proposed correlations at different solid volume fractions:

As can be seen from Fig. 4, these models predict thermal conductivity of ZnO-EG nanofluid perfectly.

3.4 *Deviation analysis of thermal conductivity:*

As to the margin of deviation between thermal conductivity calculated by our proposed correlations and experimental data, this study used experimental data as a benchmark for results of proposed correlations. The deviation between experimental results and calculated data of empirical equations can be computed as below:

$$Dev = \left[\left(Nu_{Exp} - Nu_{pred}\right) / Nu_{pred}\right] \times 100\% \qquad (8)$$

Fig. 5 Shows the measured margin of deviation (for three proposed models) according to Eq. (8) with respect to the solid volume fraction of nanoparticles. As it can be seen, these correlations predict the thermal conductivity of nanofluids well. It can be observed that the maximum values of deviation is about 1.5% at T=25 (°C) while this value is about 3% at T=50 (°C).

It is evident that for lower solid volume fraction, correlations can predict the thermal conductivity of nanofluid well.

a)

b)

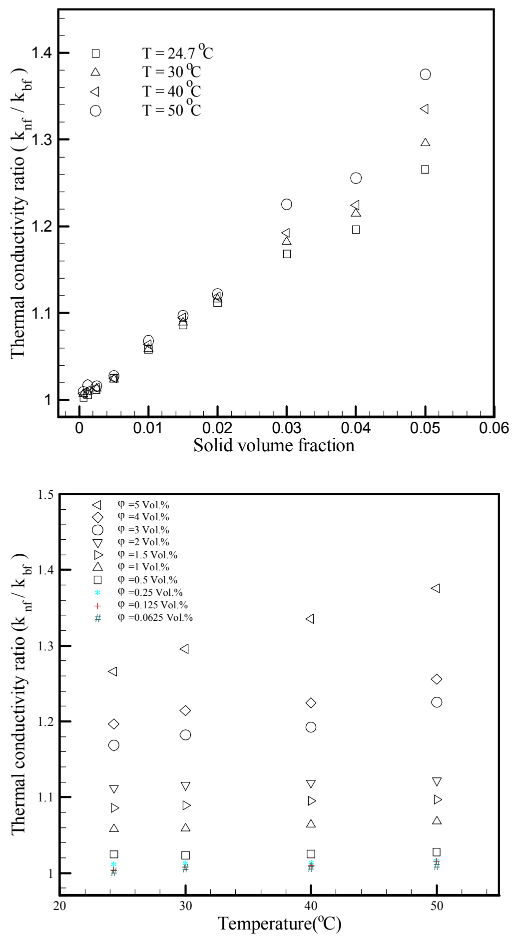


Fig. 3. Thermal conductivity ratio a) at different temperature versus solid volume fraction b) for various concentration with respect to temperature

a)

b)

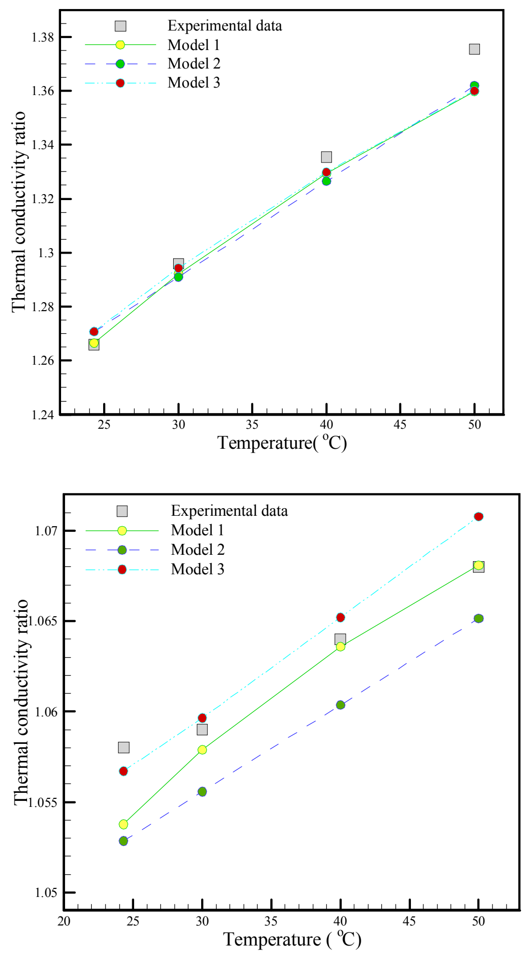


Fig. 4. comparison between experimental data and proposed correlationsat a) $\varphi = 5\%$, b) $\varphi = 1\%$

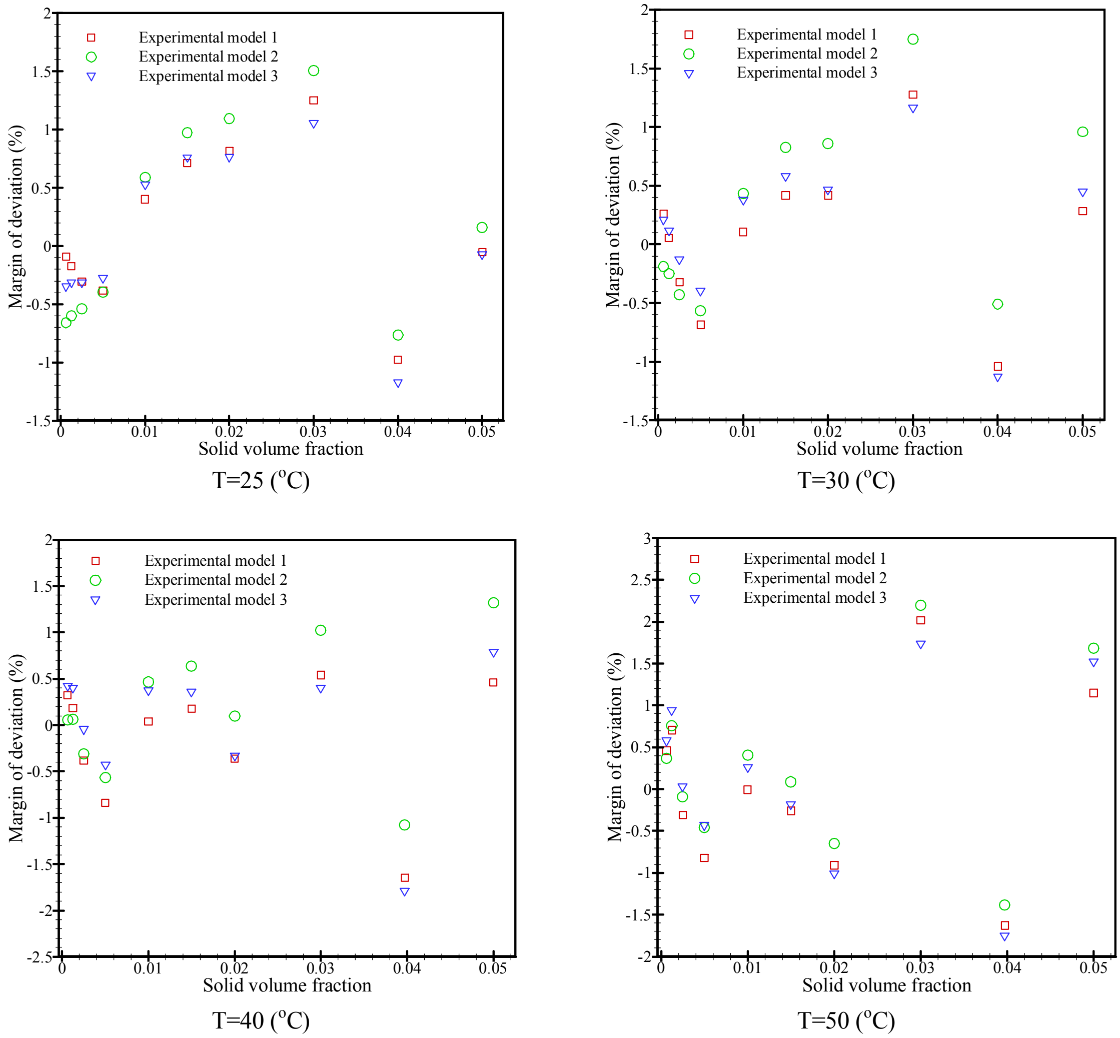


Fig. 5. Margin of deviation for different temperatures

3.5 Sensitivity analysis:

A sensitivity analysis is conducted based on Ref. [36] for three proposed models. The sensitivity analysis shows how much the thermal conductivity is sensitive to the changes in particle loading at a given temperature. In the current study, the sensitivity analysis is performed by considering 10% change in particle loading. For example, consider the volume fraction of 2% and temperature of 30 ˚C. The sensitivity in this temperature can be calculated as following:

$$\text{Sensitivity}(\%) = \frac{k(\phi = 2.2\%) - k(\phi = 2\%)}{k(\phi = 2\%)} \times 100 \quad (8)$$

Figures 6-8 provide the results of the sensitivity analysis for different proposed models. It is evident that with an increase in the temperature, the sensitivity of thermal conductivity increases. This means that at a moderate volume fraction, adding a specified amount of nanoparticles to the nanofluid at a high temperature is more effective than at a low temperatures.

4. Conclusion

In this paper, we have experimentally investigated the effective thermal conductivity of ethylene glycol based nanofluid containing ZnO nanoparticles. For this purpose, ZnO nanoparticle with an average diameter of 18 nm is provided and is dispersed in different volume of pure ethylene glycol to obtain desirable solid volume fraction. Then, ZnO/EG nanofluid with various concentrations from

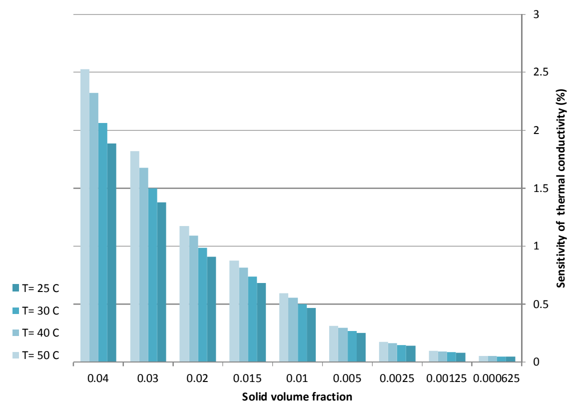


Fig. 6. Sensitivity analysis of thermal conductivity predicted by the experimental model "1" at different temperatures and concentrations

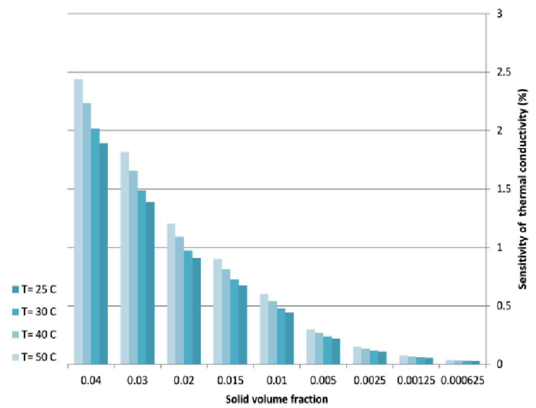


Fig. 7. Sensitivity analysis of thermal conductivity predicted by the experimental model "2" at different temperatures and concentrations

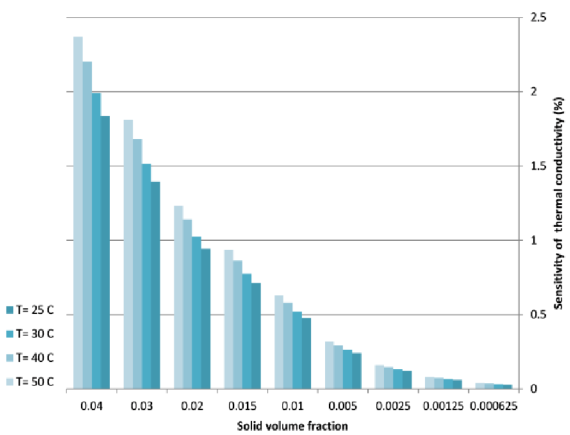


Fig. 8. Sensitivity analysis of thermal conductivity predicted by the experimental model "3" at different temperatures and concentrations

0.0625% to 5% was prepared using ultrasonication andadding surfactant. The thermal conductivity of nanofluid was measured using KD2-Pro thermal property analyzer in various temperatures for any concentration. Results have shown that the thermal conductivity ratio increases nonlinearly with an increase in the solid concentration and the maximum value of thermal conductivity belongs to the maximum solid volume fraction.

Further, as the temperature increases, serious increases in thermal conductivity are evident for all solid volume fractions, especially at high concentrations. Significant increase of relative thermal conductivity with respect to increasing temperature indicates that thermal conductivity is strongly dependent on temperature.

Thermal conductivity increases with both increasing temperature and increasing solid volume fraction. Furthermore, three correlations for estimating the thermal conductivity were proposed based on the experiment data. Data obtained from the theoretical and proposed correlations have been compared with measured data.

The results show that experimental data are in an excellent agreement with correlations which have been presented in this investigation.

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