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Curve-fitting on experimental data for predicting the thermal-conductivity of a new generated hybrid nanofluid of Graphene Oxide-titanium oxide/Water

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Abstract

The objective of this experimental study is to study the effects of a hybrid utilization of graphene oxide (GO) and titanium oxide (TiO₂) nano-materials on the water thermal-conductivity. The experiments were carried out for different concentrations of 0.05, 0.1, 0.2, 0.4, 0.6, 0.8 and 1% and in the temperature range of 20 to 50°C. The experiments showed that higher temperature and concentrations of nano-materials result in a higher the thermal-conductivity. The maximum thermal-conductivity enhancement was 32.8%. Moreover, a new accurate correlation for the thermal-conductivity was presented based on the experimental data obtained in the laboratory in terms of temperature and nano-materials volume fraction.

Keywords: Curve-fitting; Hybrid nanofluid; Thermal-conductivity; Graphene and titanium oxides;

1. Introduction

Today, convective heat transfer has a key role in most industrial heat exchangers. This type of heat transfer needs an appropriate fluid. One of the most commonly used fluids in heat exchangers is water. Special properties of water distinguish it from other liquids. It is one of the most abundant compounds on earth. Water is odorless, tasteless, and colorless and it has other properties which make this liquid valuable. Low thermal conductivity of water is one of the weaknesses of using it in heat exchangers. It is clear that a fluid with high thermal conductivity should be used to improve heat transfer in heat exchangers. In order to compensate for this fault and to use other advantages of water, addition of fine metal/metal oxide, carbon materials, etc. to a base fluid is recommended as one of the most effective methods to increase thermal conductivity and consequently convective heat transfer. “*Nanofluid*” is a fluid containing fine metal or non-metal materials (copper oxide, iron oxide, etc.) with dimensions of less than 100 nm in a base fluid (which is mainly water and/or ethylene glycol) as a dispersed stable suspension. The use of nanofluids in the oil, gas, petrochemical, air conditioning and automotive industries improves the performance of heat exchangers. The working fluid has the main role in the heat exchangers. Optimizing the heat transfer properties of these fluids can be an important step in optimizing the performance of the heat exchanger and ultimately increasing productivity in the industry. Today, the issue of reducing or optimizing energy consumption is very important considering the expiration of fossil fuel resources. Consequently, engineers believe that the use of nanofluids can significantly reduce energy consumption in thermal systems, which leads to a reduction in fuel consumption and better protection of the environment [1-6].

Results of heat-transfer experiments on different nanofluid samples showed that they performed better than their expected theoretical performance [7, 8]. In fact, it is possible to reduce the size of heat exchangers using the nanofluids that lead to further heat transfer [9, 10]. Production of smaller and lighter heat exchangers saves costs and energy [11, 12]

Many studies have been reported on the thermophysical properties of nanofluids and it has become specified that it is possible to change the transfer and heat properties of a nanofluid by adding nano-materials. The use of a small amount of nano-materials may even improve heat transfer. Adding a

small percentage of nanoparticles improve nanofluid thermal conductivity significantly as compared with a base fluid. Table 1 shows some studies reported on the thermal conductivity of nanofluids. Studies show that nanofluid thermal conductivity depends on the combination of percentage of nanoparticles and nanofluid chemical, mass percentage of nanoparticles, shape and size of particles, surface active agents (surfactants), temperature, etc..

Table 1. A review on the studies that reported the thermal conductivity of nanofluids

Author	Base fluid	Dispersed particles	Concentration	Maximum enhancement (%)
Abareshi et al. [13]	Water	Fe ₃ O ₄	Up to 3 vol.%	15.1
Afrand et al. [14]	Water	Fe ₃ O ₄	Up to 3 vol.%	90
Jung et al. [15]	Water	Al ₂ O ₃	Up to 1 vol.%	14.4
Mintsas et al. [16]	Water	Al ₂ O ₃	Up to 6 vol.%	34
Das et al. [17]	Water	CuO	Up to 4 vol.%	28.7
Li and Peterson [18]	Water	CuO	Up to 6 vol.%	51
Liu et al. [19]	Water	Cu	Up to 0.1 vol.%	24
Paul et al. [20]	Water	Au	Up to 0.00026 vol.%	48
Li and Xuan [21]	Water	Al	Up to 5 vol.%	27
Kang et al. [22]	Water	Ag	Up to 0.39 vol.%	11
Li et al. [23]	Water	Fe	Up to 5 vol.%	14.9
Wen and Ding [24]	Water	TiO ₂	Up to 0.68 vol.%	6
Duangthongsuk and Wongwises [25]	Water	TiO ₂	Up to 2 vol.%	13.2
Ahmadi Nadooshan [26]	Water-EG	ZnO	Up to 4 vol.%	20
Singh et al. [27]	Water	SiC	Up to 4 vol.%	22.5
Huminic et al. [28]	Water	FeC	Up to 4 vol.%	24.2
Adhami Dehkordi et al. [29]	Water-EG	SWCNTs	Up to 0.65 vol.%	52.7
Shamaeil et al. [30]	EG	DWCNTs	Up to 0.6 vol.%	24.9
Soltanimehr and Afrand [31]	Water-EG	MWCNTs	Up to 1 vol.%	34.7
Yu et al. [32]	EG	Graphene oxide	Up to 0.05 vol.%	61

A new generation of nanofluids has recently been introduced called “hybrid nanofluids”. These nanofluids are potential fluids that provide high heat transfer performance and thermodynamic properties more than heat transfer fluids (oil, water, and ethylene glycol) and nanofluids containing

mono type nanoparticles. In fact, hybrid nanofluid is a new nanofluid which is produced through dispersing two different nano-materials into a common heat transfer fluid. Some researchers studied the thermal conductivity of these types of nanofluids. For example, Sundar et al. [33] examined the thermal conductivity of MWCNTs–Fe₃O₄/water hybrid nanofluid and reported the maximum enhancement of 13.88% at volume fraction of 0.3%. Suresh et al. [34] measured the thermal conductivity of Al₂O₃–Cu/water hybrid nanofluid and presented the maximum enhancement of 12.11% at solid volume fraction of 2%. Sarbolookzadeh Harandi et al. [35] investigated the thermal conductivity of MWCNTs–Fe₃O₄/EG hybrid nanofluid. Their results showed the maximum enhancement of 30 at solid volume fraction of 2.3%. Afrand [36] performed measurement of the thermal conductivity of MgO-FMWCNTs/EG hybrid nanofluid. He increased the solid volume fraction of nano-additives to 0.6% and observed a maximum thermal conductivity enhancement of 21.3%. Nasajpour Esfahani et al. [37] performed an experimental study on thermal conductivity of ZnO–Ag/water hybrid nanofluid. They reported that the maximum thermal conductivity enhancement was 26% at solid volume fraction of 2%. Hemmat Esfe et al. [38] presented an experimental work on ZnO-DWCNTs/EG hybrid nanofluid with 24.9% enhancement in thermal conductivity that occurred at solid volume fraction of 1.9%. Other studies have also done on the thermophysical and heat transfer of the nanofluids, all of which have reported their suitability [39-58].

It is clear that saving energy and improving efficiency of systems are highly important today. The increase of thermal conductivity cools a system quicker, saves energy, and improves efficiency. On the other hand, the unique properties of graphene oxide and appropriate stability of titanium oxide allowed us to produce a new water-based hybrid nanofluid in this study by knowing the construction of the hybrid nanofluids as mentioned above. The use of graphene nanoparticles in water increases thermal conductivity considerably; however, the planar shape of nanofluids increases base fluid viscosity, they increase the pumping power of the nanofluid for a cooling operation, and the nanoparticle is costly. Therefore, this study used titanium nanoparticles combined with graphene oxide for the first time to provide a better viscosity and save costs.

2. Statement of the present research

A graphene oxide–titanium oxide/water hybrid nanofluid is produced in this study. Some methods including ultrasound waves, the addition of surfactants, and base-fluid pH control should be used to produce nanofluid stable samples. After producing a stable sample of the nanofluid, photography, UV-Vis absorption spectroscopy, and DLS (Dynamic Light Scattering) methods are used to determine its stability.

The impact of temperature and concentration of the nanofluid on the thermal-conductivity of graphene oxide–titanium oxide/water hybrid nanofluid is examined after making the nanofluid by measuring the thermal-conductivity of samples. It is then attempted to study the effect of duration of ultrasonic waving on nanofluid thermal-conductivity. Finally, the results of the measurement nanofluid thermal-conductivity are compared with the ones of other nanofluids and a mathematical correlation is developed for the thermal-conductivity prediction. Figure 1 shows the flowchart of the route to achieve the research objectives.

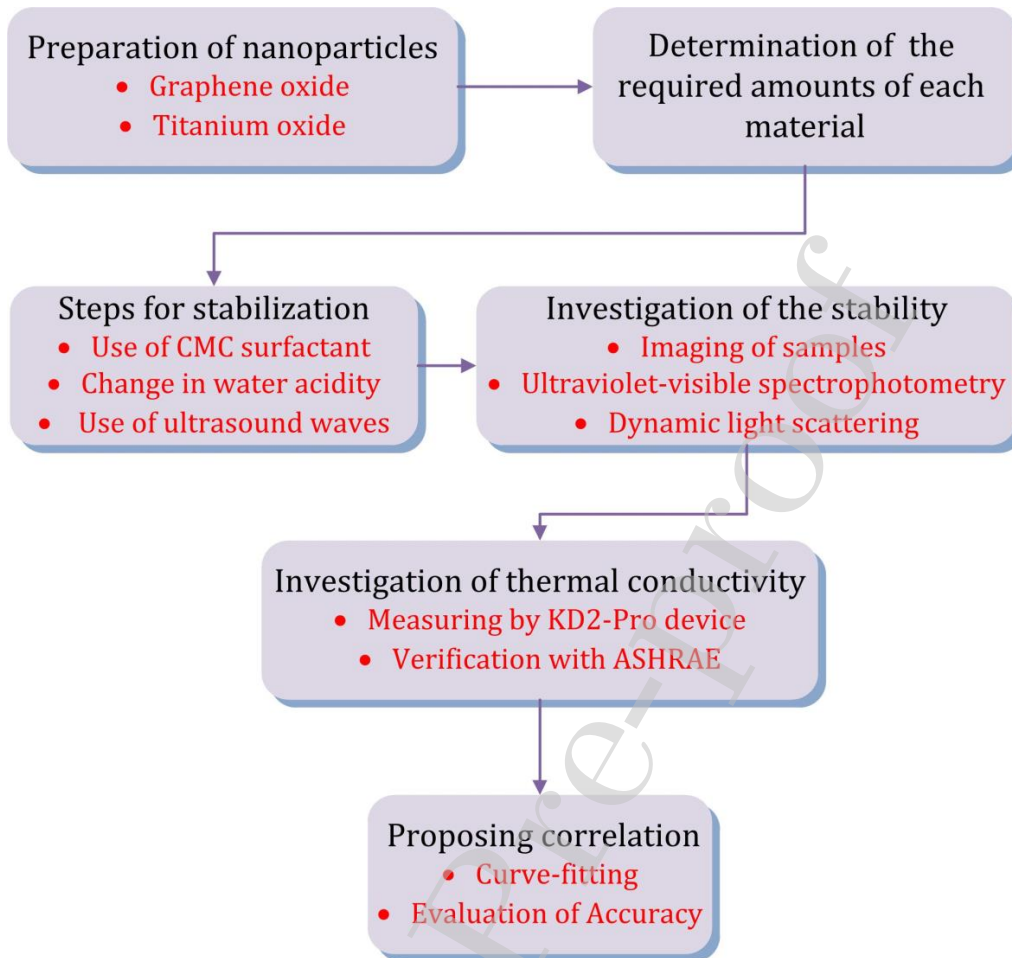


Fig. 1. The process of doing this research

3. Used materials

This study used distilled water as the base-fluid and graphene oxide and titanium oxide as the nanoparticles. The ratio of using nano-materials in all volume fractions of the test was considered 50% of each. The nanoparticles were produced using a chemical analysis method. The nanoparticles were made by *US Research Nanomaterials, Inc.*

3.1. Graphene Oxide

Studies show that graphene oxide nanoparticle, as compared with other common metal nanoparticles such as aluminum oxide, copper oxide, etc., has an appropriate specific surface area, high thermal-conductivity, a very low average of particle size, and very low density. However, the only major problem of this group of nanoparticles is their high cost.

One of the advantages of graphene oxide is its ease of diffusion in water and other organic solvents and various matrices, which is due to the presence of oxygenated agents. Graphene nano-sheets have some features including high thermal-conductivity, large specific surface area, hydrophilicity, insolubility, and low density. Table 2 shows the nanoparticle specifications. The XRD of graphene oxide is shown in Fig. 2.

Table 2. Specifications of graphene oxide

Material	Purity (%)	Appearance	Color	Thickness (nm)	Number of layers	Specific surface area (m ² /gr)
Graphene oxide	99	Sheet	Black	3.4-7	7-11	100-300

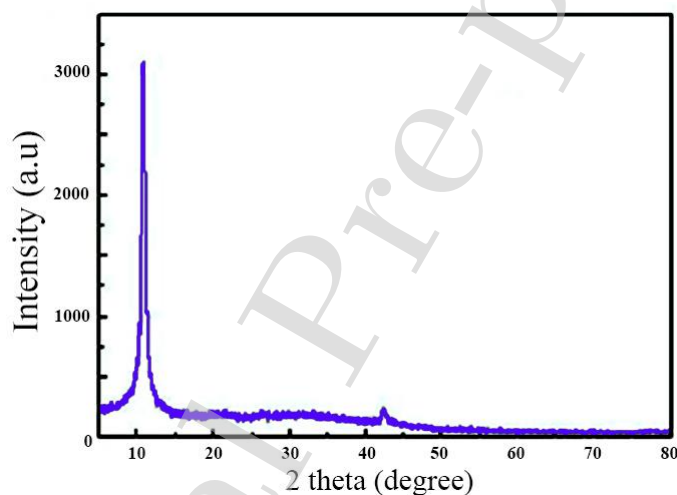


Fig. 2. XRD of graphene oxide nano-sheets

3.2. Titanium Oxide

Titanium oxide nanoparticles have high purity, limited range of particle size, spherical structure, and relatively high specific surface area. Moreover, titanium oxide nano-materials have chemical stability, high thermal-conductivity, and lower thermal expansion coefficient.

The use of titanium oxide nanoparticles in the hybrid nanofluid modifies the physical structure of graphene oxide nano-sheets, which helps to reduce the nanofluid viscosity as compared with graphene oxide/water nanofluid and reduces the production cost of the hybrid nanofluid with the modified

feature as compared with graphene oxide/water nanofluid. Table (3) shows the nanofluid specifications and its XRD is respectively shown in Fig. 3.

Table 3. Specifications of titanium oxide

Material	Purity (%)	Appearance	Color	Diameter (nm)	Density (gr/m ³)	Specific surface area (m ² /gr)
Titanium oxide	99.9	Spherical	White	20	3.9	200

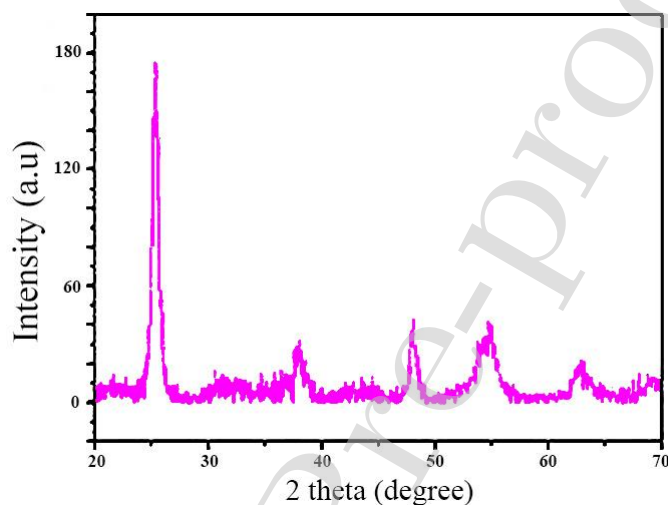


Fig. 3. XRD of titanium oxide nanoparticles

3.3. Base-fluid

The PH of distilled water is neutral (about 7). The specific conductivity of distilled water is very low, as the electrical conductivity of water enhances with the dissolution of salts. The boiling point of distilled water, due to its purity, is lower than the one in ordinary water. It never corrodes due to having no soluble materials. Distilled water is not a desirable and pleasant drink and it has lost its taste due to the removal of ions. Distilled water has commonly been used in industry because of the change in some of its physical properties as compared with ordinary water.

4. Experimental work

4.1. Production Samples

The nanofluid samples used in this study were made of graphene oxide and titanium oxide nano-materials in a hybrid manner in a water-based fluid with the concentrations of 0, 0.05, 0.1, 0.2, 0.4, 0.6, 0.8, and 1%. The required amount of each substance for producing 100 ml of nanofluid in a certain volume fraction is given as:

$$m_{TiO_2} = 5 \times 10^{-6} \rho_{TiO_2} \varphi \quad (1-a)$$

$$m_{GO} = 5 \times 10^{-6} \rho_{GO} \varphi \quad (1-b)$$

$$m_{H_2O} = \rho_{H_2O} \left[0.100 - \left(\frac{m_{GO}}{\rho_{GO}} + \frac{m_{TiO_2}}{\rho_{TiO_2}} \right) \right] \quad (1-c)$$

where φ is the nanofluids volume fraction in percent. m_{GO} , m_{TiO_2} , and m_{H_2O} are respectively the mass of graphene oxide nano-sheets, titanium oxide nanoparticles, and base-fluid in grams. ρ_{GO} , ρ_{TiO_2} , and ρ_{H_2O} are respectively the symbols of the density of graphene oxide and titanium oxide nanoparticles, and the mass density of base-fluid in kg/m^3 .

As it is impossible to measure the accurate volume of nanoparticles using the above relation, the values of density and mass of nanoparticles are used to determine different concentrations. A 0.001g digital scale was used for measuring the weight of nanoparticles.

pH is one of the major parameters for maintaining stability in nanofluids. Studies show that water-based nanofluids containing graphene oxide and titanium oxide particles have the highest surface charge of particles for long-term stability at a pH higher than the neutral state [47,48]. This study uses carboxymethyl cellulose (CMC) surfactants to create an alkaline environment and to increase the surface charge of particles. For each specimen of nanofluid, $\frac{1}{4}$ of nanoparticles mass was used. The surfactants increased pure water pH from a neutral state and pH was approximately 9 for all different test samples. The surfactants were manufactured by Sigma Corporation and it had a considerable effect on the stability of GO-TiO₂/water hybrid nanofluid as compared with the state without surfactants.

A magnetic mixer was employed to scatter nano-materials in the base-fluid and each sample was exposed to the magnetic mixer for an hour. To establish stability, nanofluid samples were exposed to ultrasound waves. A nanofluid sample was exposed to ultrasound waves in an Ultrasonic device for 30, 40, 50, and 60 minutes. It was observed that the sample maintained uniform stability after 50-60 minutes. All the samples were then exposed to ultrasound waves for 45 minutes in different volume fractions. Figure 4 shows an image of a sample during the ultrasound process.

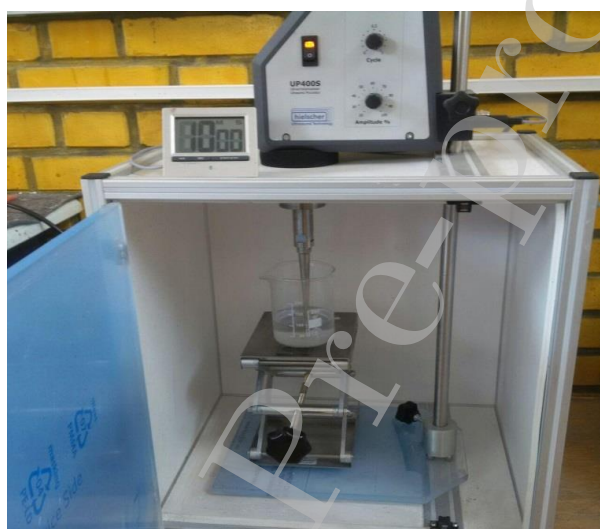


Fig. 4. Image of a sample during ultrasound process

4.2. Stability

Ultraviolet-visible spectrophotometry was used to examine the stability of samples. The results are used for the quantitative description of suspension colloidal stability. One of the features of the device is that it can be used for all base-fluids. The law used in this method is that light intensity is altered by its absorption or diffusion through the fluid. Suspension stability is often investigated by measuring the deposited volume in terms of deposition time. The disadvantage of this method is that it is not suitable for the nanofluids with high concentration, especially the solution of carbon nanotubes. A *Spectrophotometer-Biochrom WPA Lightwave II* was used to examine nanofluid stability.

An absorption test was carried out to measure the absorption intensity using pure water for calibration of the device and examining the control sample. The graphene oxide-titanium oxide/water hybrid nanofluid was then tested in different periods. Figure 5 shows the absorption test results in 60, 150, and 240 minutes after nanofluid production.

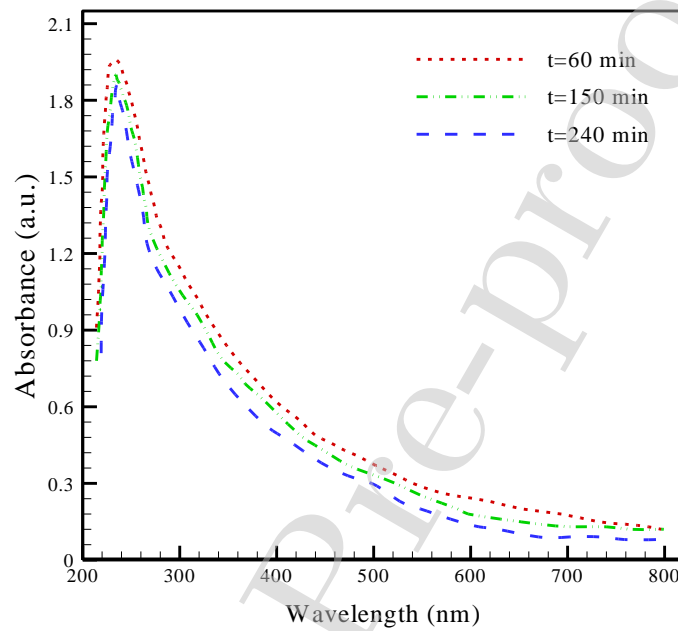


Fig. 5. Absorption test results in 60, 150, and 240 minutes after nanofluid production

The test results show that maximum absorption intensity is at the wavelength of 234 nm. To examine the stability, the test was repeated without any change in nanofluid status at different periods. The intensity of nanofluid absorption reduced from 1.96 to 1.86 within 240 minutes after the first test, which indicates the nanofluid stability is acceptable.

Figure 6 shows the images taken from the samples three hours after producing nanofluid samples.

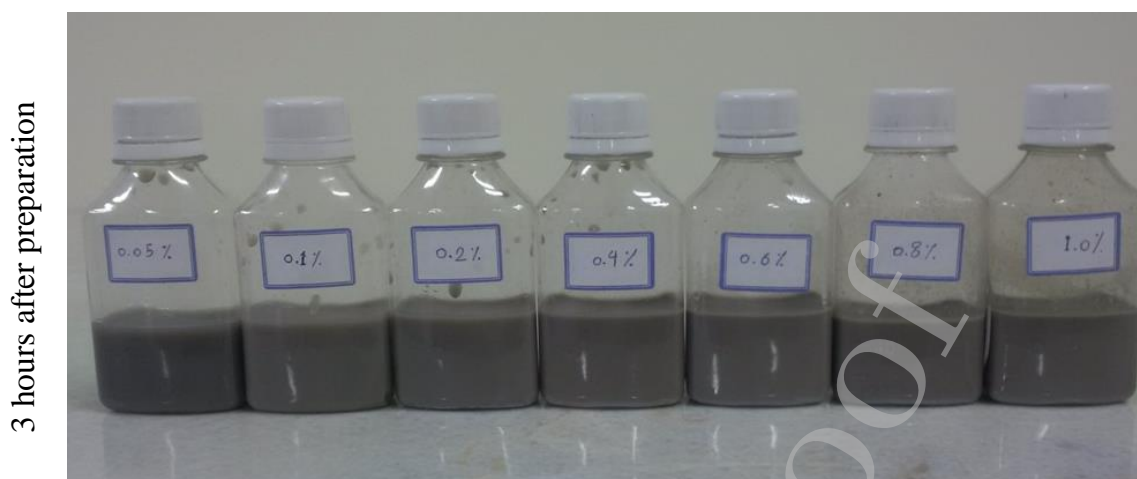


Fig. 6. Images of nanofluid samples at different time periods

4.3. Measurement of Thermal-conductivity

This research used a KD2-Pro device to determine the thermal-conductivity of the hybrid nanofluid. The device mainly operates employing the transient hot-wire method as one of the most precise methods. KD2-Pro is a battery-powered portable device employed for thermal properties measurement of different materials as per EN55022:1987 reference standard. It is used for the thermal and conductivity properties of liquids, solids, and soil. The operating temperature range of the device is 0-50°C and the operation range of the device probes is 50-150. The KS-1 sensor of the device with a maximum precision of 5% was used in this test.

As one of the objectives was to assess the effect of temperature on nanofluid thermal-conductivity, the temperature of the samples should have been maintained at a fixed value during each measurement. The temperature range in the test was 20-50°C. A 7-liter-capacity WNB-7 Memmert bath was employed for maintaining the temperature stability of the samples. Heat transfer in this bath is carried out by natural convection. The temperature range of the device is 25-95°C with a precision of 0.1°C. Chamber temperature sensitivity was adjusted to 0.1°C. The samples were placed in the temperature bath for measurement to reach the desired temperature and the thermal-conductivity of the nanofluid samples were measured by a KD2-PRO device. Figure 7 shows how to measure thermal-conductivity while the sample is in the temperature bath.



Fig. 7. Setup of thermal-conductivity measurement in temperature bath

5. Results and discussion

5.1. Measurements Verification

To verify the measurements, the thermal-conductivity of pure water measured in this study was compared with the values reported for water in the ASHRAE Handbook [59] as shown by Figure 8. The figure shows that the maximum difference is lower than 1%, which indicates an appropriate accuracy for all the measurements.

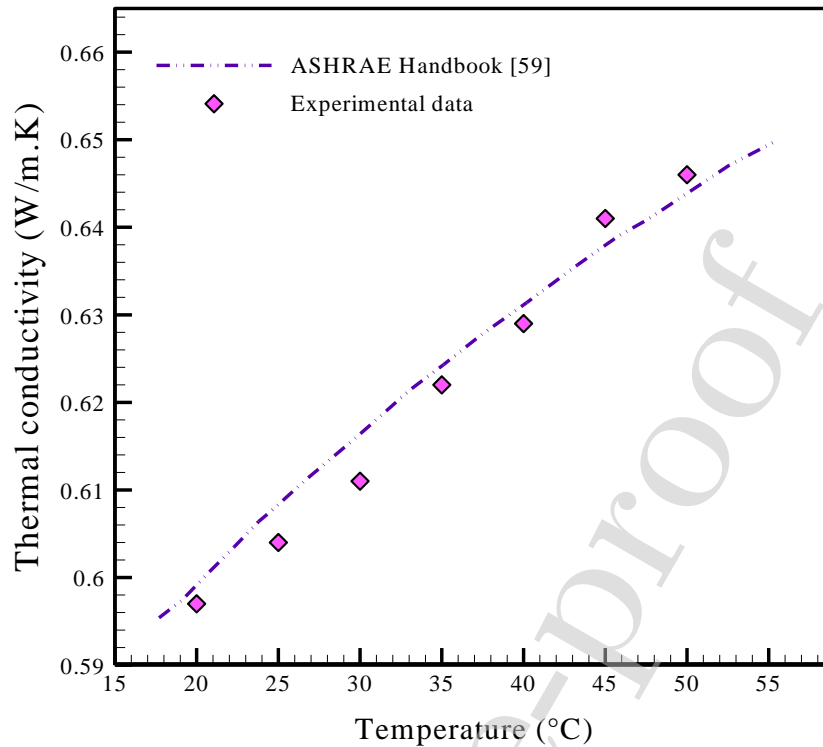


Fig. 8. Measured thermal-conductivity in comparison with ASHRAE data [59]

5.2. Effects of nano-materials volume fraction and temperature

Figure 9 shows the absolute and relative thermal-conductivities of the nanofluid against nano-materials volume fraction at different temperatures. The thermal-conductivity of nanofluid over the base-fluid at an identical temperature is called “relative thermal-conductivity”. It shows that the absolute and relative thermal-conductivities enhances with increasing the concentration at all temperatures. The increase is due to the fact that nanoparticles move among base-fluid molecules and sometimes collide with one another through Brownian motion. In a special case, when two particles impact with each other, the heat transfer created between the solid nanoparticles may increase the nanofluid thermal-conductivity. Figure 9 illustrates that the gradient of thermal-conductivity increase is higher in lower concentrations. This phenomenon may be attributed to the fact that nano-materials have a large specific surface area due to the low amount of agglomeration of nanoparticles in lower volume fractions. Larger specific surface area leads to higher thermal-conductivity.

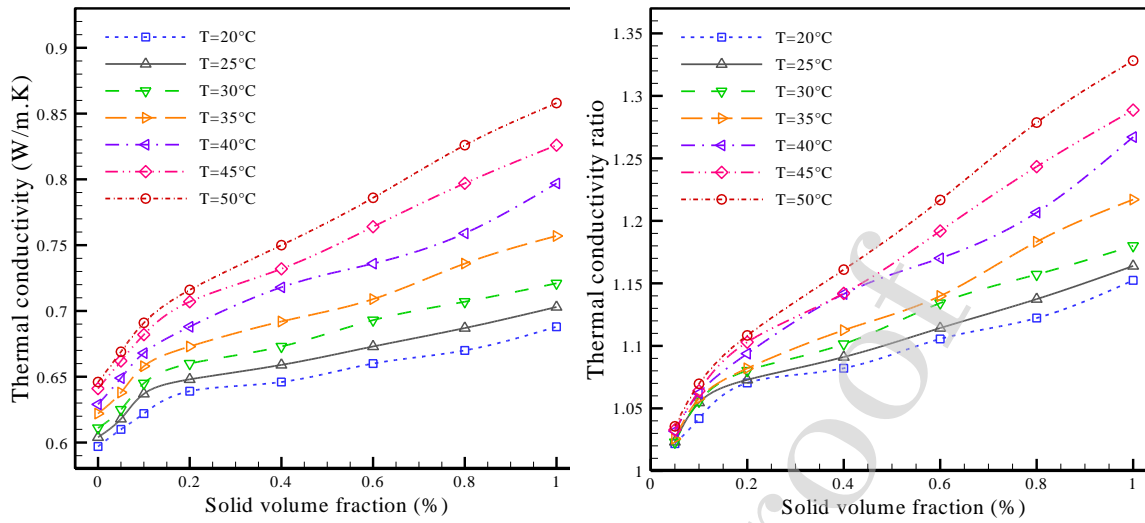


Fig. 9. Absolute and relative thermal-conductivities of the nanofluid against volume fraction at different temperatures

Figure 10 shows the absolute and relative thermal-conductivities of the nanofluid against temperature for all nanofluid samples. As the figure shows, a higher thermal-conductivity is gained from all the samples with increasing the temperature due to the increase of Brownian diffusion and further impacts of particles. The increase is higher in higher volume fractions because the number of nanoparticles exceeds in richer nanofluids and the effect of temperature on Brownian diffusion and random collision of nanoparticles is more tangible. For instance, the relative conductivity increases from 1.023 to 1.180 in the volume fraction of 0.05-1% at 30°C and the maximum volume fraction has the maximum relative heat conductivity. As the figure indicates, the maximum relative heat conductivity, i.e. 1.328%, is related to 50°C in 1 Vol.%.

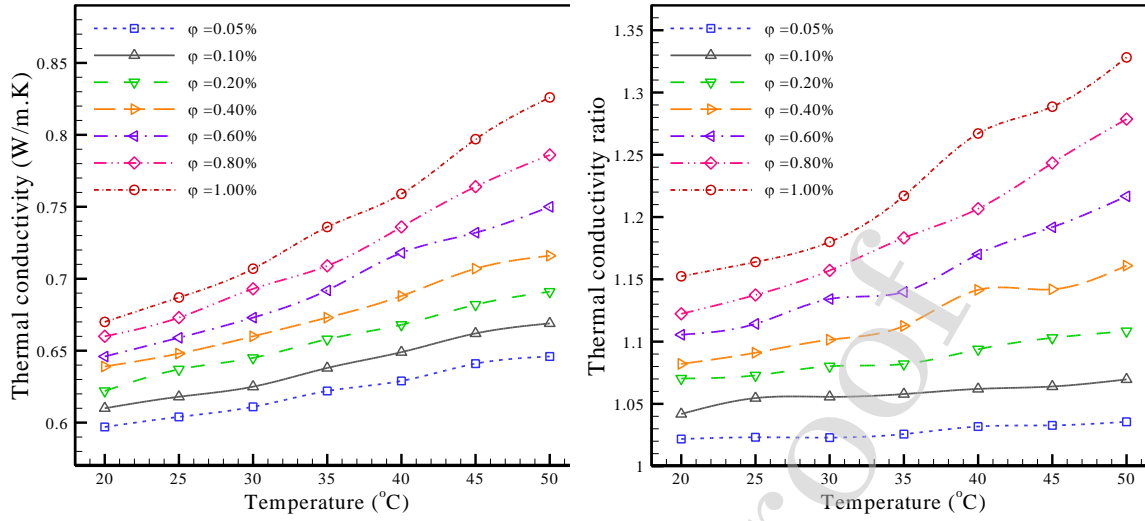


Fig. 10. Absolute and relative thermal-conductivities of the nanofluid in terms of temperatures at for all nanofluid samples

5.4. Introducing a new correlation

Literature has shown that conventional models would not be able to predict the nanofluids thermal-conductivity. Soltanimehr and Afrand [31] proved that maximum thermal-conductivity of nanofluid in the volume fraction of 1% was only 3% Based on the Maxwell model; while their proposed nanofluid in the same volume fraction showed an approximate increase of 35%. This research showed that thermal-conduction increased by about 34% in the volume fraction of 1%. Therefore, it can be argued that the Maxwell Model is useless to predict the nanofluid thermal-conductivity. Consequently, this section introduces a new model to determine the thermal-conductivity of graphene oxide-titanium oxide/water hybrid nanofluid based on the results and measurements in terms of concentration and temperature. Curve fitting is used first to present Equation 2 for water thermal-conductivity based on temperature. Then, Equation 3 is presented for predicting the thermal-conductivity of nanofluid using the values in proportion to thermal-conductivity.

$$k_{bf} = 0.5642e^{0.002748T} \quad (2)$$

$$\frac{k_{nf}}{k_{bf}} = (1.017 + 0.072 \times 1.029^T \times \varphi^{0.773}) \quad (3)$$

In Equations 2 and 3, k is the thermal-conductivity in W/m.K, T is the temperature in °C, and ϕ is the volume fraction in %. Indices bf and nf refer to the base-fluid and the nanofluid, respectively.

Figure 11 compares the values of the thermal-conductivity calculated by Equation 3 and the experimental values to assess the accuracy of the proposed relation. The figure shows that most experimental data are in agreement with the predicted line showing the accuracy of the proposed relation.

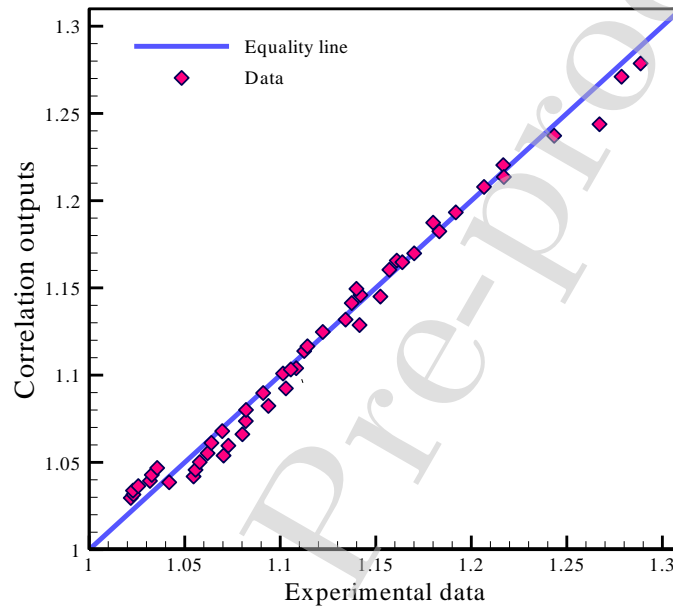


Fig. 11. Thermal-conductivity ratio obtained by proposed equation in compared with the experimental data

6. Conclusion

In this research, the thermal-conductivity of GO-TiO₂/water hybrid nanofluid in the temperature range of 20 to 50°C for samples with volume fractions of 0.05, 0.1, 0.2, 0.4, 0.6, 0.8 and 1% was examined and the following results are obtained:

- The thermal-conductivity enhanced for a higher concentration and temperature. The highest thermal-conductivity enhancement was 32.8%, which was related to the temperature of 50°C and a volume fraction of 1%.
- The slope of variation of thermal-conductivity ratio in the concentrations range of 0.4 to 1% was higher than that in range of 0.05 to 0.4%.

- To achieve proper stability, samples were exposed to ultrasonic waves for 45 minutes. 45 minutes was an optimal time. In this way, two samples with volume fractions of 0.1 and 0.6% have been subjected to ultrasound over different times.
- In the UV-Vis absorption spectroscopy test, the absorption rate of nanofluid reduced from 1.96 to 1.86 after 240 minutes of the first experiment, which showed acceptable nanofluid stability.
- An accurate equation was presented in terms of volume fraction and temperature to determine GO-TiO₂/water thermal-conductivity.
- At the end, the results of experimental data and the proposed model were compared showing the accuracy of the proposed equation as well as the comparison of these results with the ASHRAE Handbook of the appropriate validation.

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[68] **Highlights**

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- Using curve-fitting to propose a correlation for predicting thermal conductivity
- Preparing homogeneous and stable samples of GO-TiO₂/water hybrid nanofluid
- Presenting effects of temperature and GO-TiO₂ concentration on thermal conductivity
- 32.8% improvement in thermal conductivity of the hybrid nanofluid compared to water

[70]

Journal Pre-proof