

Thermal Conductivity of Water Based Nanofluids Containing Decorated Multi Walled Carbon Nanotubes with Different Amount of TiO₂ Nanoparticles

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Abstract

In this paper, we report for the first time, thermal conductivity behavior of nanofluids containing decorated MWCNTs with different amount of TiO₂ nanoparticles. TEM image confirmed that the outer surface of MWCNTs successfully decorated with TiO₂ nanoparticles. The results of thermal conductivity behavior of nanofluids revealed that the thermal conductivity and enhancement ratio of thermal conductivity of MWCNTs-TiO₂ at different amount of TiO₂ nanoparticles are higher than those of TiO₂ and MWCNTs nanofluids. Temperature and weight fraction dependence study also shows that the thermal conductivity of all nanofluids increases with temperature and weight fraction. However, the influence of temperature is more significant than that of weight fraction. We also found that decreasing amount of TiO₂ nanoparticles which introduce the outer surface of MWCNTs leads to the augmentation of thermal conductivity of nanofluids containing MWCNTs-TiO₂.

Keywords: MWCNT; TiO₂ Nanoparticles; Decoration; Thermal Conductivity

1. Introduction

Conventional fluids such as water and oil have poor thermal properties that restrict the heat transfer performance compared to most of the solids. Many techniques are available in order to increase heat transfer rates and reduce the size of heat transfer equipment [2]. Thermal conductivity is one of the

thermophysical properties of nanofluid and depends on several parameters such as thermal conductivities of the base fluid and the nanoparticles, the volume fraction, the shape and kind of the nanoparticles, the surface area, and the temperature [1]. The effective thermal conductivity (k_{eff}) of a mixture containing two components is given by [1].

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$$k_{\text{eff}} = \frac{k_p \phi_p (dT/dx)_p + k_b \phi_b (dT/dx)_b}{\phi_p (dT/dx)_p + \phi_b (dT/dx)_b} \quad (1)$$

Where k_p is the thermal conductivity of the particle, k_b is the thermal conductivity of the base fluid and ϕ is the particle volume fraction in the suspension. There are several semi-empirical models for calculation of the thermal conductivity of two-phase mixtures; however, all of them are based on the above definition. So some of the semi-empirical correlations are used for a binary mixture of homogeneous spherical inclusions and some are used for a suspension containing non-spherical particles.

The key idea is to exploit the solid particles with very high thermal conductivity that can be several hundreds of times greater than all of the conventional fluids combined. Various types of particles, such as metallic, non-metallic and polymeric, can be added into fluids to form slurries [2].

A few years ago solid particles of millimeter and micrometer in size were suspended in the conventional fluids to improved thermal behavior but some serious problems emerged by use of these types of fluids. For example, poor stability of the suspension and high erosion and pressure drop in pipelines and equipment.

With the advent of nanotechnology, nanoparticles in size between 1 nm and 100 nm were replaced instead of particles in order of millimeter and micrometer. This type of fluid is called nanofluid [3]. The stability and heat transfer rate of nanofluids are extraordinarily higher than those of suspensions containing particles in the size of millimeter or even micrometer.

Among the various nanoparticles, carbon

nanotubes (CNTs) due to their very excellent thermal conductivity and large aspect ratio are a good choice to prepare nanofluids [4]. The presence of van der Waals attraction between tubes leads to hydrophobic nature of CNTs, so the their dispersibility is very low in water and other organic solvents [5]. Therefore in the preparation of stable suspension, the surfactant addition is an effective way to enhance the dispersibility of CNTs [6]. However, surfactant molecules attaching on the surfaces of CNTs may increase the thermal resistance between the CNTs and the base fluid [7], which limits the enhancement of the effective thermal conductivity.

Modifying the surface of nanotubes with oxygen-containing groups leads to improvement in the interaction of CNTs with the solvent matrix. These functional groups are formed by chemical treatment in mixture of acid such as nitric acid [8]. Zhang *et al.* [9] investigated the heat transfer performance of TiO₂/water nanofluid. They observed that the effective thermal conductivity and thermal diffusivity increase with an increase in the particle concentration. He *et al.* [10] studied static thermal conductivity, heat transfer and flow behavior of stable aqueous TiO₂ nanofluids with different particle sizes and concentrations. They found that the convective heat transfer coefficient increased with increasing nanoparticles concentration.

Garg *et al.* [11] reported the enhancement of thermal conductivity from 3 to 5% for nanofluids containing 1 wt% MWCNTs which were measured at 25°C and the ultrasonication from 20 to 80 min, respectively. Chen *et al.* [12] reported an enhancement of 17.5% of thermal conductivity at volume fraction of 0.01 for an

ethylene glycol based nanofluid containing multi-walled carbon nanotubes.

Meibodi *et al.* [13] investigated the effects of different factors such as nanoparticle size and concentration on thermal conductivity of CNT/water nanofluids. Their results showed that Thermal conductivity of nanofluid is time dependent immediately after ultrasonication and independent of time at longer time. Talaei *et al.* [14] reported the influence of functional group concentration on the thermal conductivity of MWCNT nanofluids. Their results show that increasing the functionalized group causes better stability and higher thermal conductivity if the surface of MWCNT is not damaged in functionalizing process. Xie *et al.* [4] have prepared the stable nanofluids of multi-walled carbon nanotubes into ethylene glycol base fluid and studied the influence of mechanical ball milling on the straight and length distribution of CNTs. Their results demonstrate that the nanotube loading, temperature, straightness ratio, aspect ratio and aggregation play a key role in the thermal conductivity of nanofluids. Raykar *et al.* [15] investigated the effect of temperature and Brownian motion on the enhancement of effective thermal conductivity of carbon nanotube based nanofluids. Their experiments revealed that the Brownian motion has a significant effect on the effective thermal conductivity. Also, they observed that the enhancement of effective thermal conductivity in dilute nanofluids is higher. Hong *et al.* [16] studied the effect of external magnetic field on the thermal conductivity enhancement of nanofluids containing 0.01 wt% nanotube and 0.02 wt% Fe₂O₃ in water under different magnetic strength. Their results showed that

the thermal conductivity of nanofluids under uniform magnetic field is higher than that under non-uniform field. Jha *et al.* [17] reported the effect of decorated carbon nanotubes with various nanoparticles such as Ag, Au and Pd on the thermal conductivity of water and ethylene glycol based nanofluids. Their experiments demonstrated that nanofluids maintain the same sequence of thermal conductivity as that of metal nanoparticles, Ag-MWNTs > Au-MWNTs > Pd-MWNTs. Amiri *et al.* [18] investigated the dispersion stability and thermal conductivity of multiwalled carbon nanotubes nanofluids in the presence of gum arabic (MWCNT-GA) as well as functionalized MWCNT with cysteine (MWCNT-Cys) and silver (MWCNT-Ag). The effect of temperature and weight concentration on the enhancement of thermal conductivity revealed that the covalent functionalization by Ag is more effective than noncovalent functionalization.

Although thermal behavior of CNT nanofluids has been investigated by many researchers, the effect of decorated MWCNTs with different amount of nanoparticles on the thermal behavior has never been reported. Therefore in this study, we want to report for the first time the effect of modified MWCNTs with various amounts of TiO₂ nanoparticles on the thermal conductivity of nanofluids.

2. Materials and experimental

Titanium tetrachloride (TiCl₄, M=189.79, 99%, Merck) without any further purification was used to prepare the TiO₂ nanoparticles using hydrolysis method. The appropriate amount of TiCl₄ was dissolved dropwise in

distilled water under vigorous stirring. This aqueous TiCl_4 solution was stirred for 5 h at ambient temperature then heated for 24 h at 80°C . Finally, TiO_2 nanoparticles were separated from the solution using filtration dried at room temperature and calcined at 370°C for 3 h.

In a typical synthesis of MWCNT- TiO_2 , 100 mg MWCNTs (average diameter of 40-60 nm and lengths ranging from 5 to 15 micrometers) were first dispersed in 50 mL nitric acid (HNO_3 , $M=63$, 65%, Merck), sonicated at room temperature for 2 h in an ultrasound bath and stirred for 2 h at high speed. The acid-treated MWCNTs were rinsed several times with distilled water until the pH value of the solution was close to neutral and then dried at 90°C overnight. Subsequently, a small amount of Hydrochloride Acid (HCl 37 wt%, Merck) and specific amount of TiCl_4 was added dropwise to 100 mL distilled water under rapid stirring, then 75 mg of oxidized MWCNTs were dispersed in this solution using ultrasound bath for 2 h. The mixture was stirred for 22 h at room temperature and then the temperature was increased to 80°C ; the mixture was stirred for 3 h then filtered, dried at 80°C for 1 h and calcinated at 370°C for 3 h.

The measurement of thermal conductivities was done using KD2 Pro thermal property analyzer purchased from Decagon Devices Inc. The mentioned instrument has three different probes and measures the thermal conductivity according to the transient hot wire technique. In this study we used the single-needle (KS-1) 60 mm long and 1.3 mm diameter and accuracy ± 0.01 W/(mK) from 0.02-0.2 W/(m K).

Measurements were carried out in the range of temperature and weight fraction varying from 25 to 70°C and 0.25 wt% to 1.5 wt%, respectively. A certain amount of each nanoparticle was dispersed in water and the mixture was put in an ultra-sound bath for 2 h. In order to change and control the temperature, the KD2 Pro device was connected to a constant temperature bath (Thermo Haake K10 TT4310) which has a circulator and was able to maintain temperature uniformity. For better accuracy, we maintained the sample and probe in double walled cylindrical container having liquid circulating facility at constant temperature and waited for about fifteen minutes between readings. Fig. 1 illustrated the use of KD2 Pro device. A number of measurements were taken for each sample and only measurements resulted with the mean correlation coefficient $r^2 > 0.9998$ were considered. Transmission electron microscopy (TEM) micrographs of acid-treated and modified MWCNTs with TiO_2 nanoparticles were obtained using a LEO 912AB system operating at 120 kV.



Figure 1. Experimental set up (KD2 Pro device) for thermal conductivity measurements.

3. Results and discussion

3-1. Acid treatment of MWCNTs

During the formation of multiwalled carbon nanotubes the open ends of them are often closed by catalyst particles. Treatment of MWCNTs in nitric acid mainly removed the catalyst particles. Therefore, the tube ends are opened and largely decorated with oxygen containing groups such as acid carboxylic (-COOH) and hydroxyl (-OH) which could change the sp² hybridization of MWCNTs to sp³ hybridization [19]. The introduction of these oxygen containing groups on the outer surface of MWCNTs leads to high dispersion of CNTs in polar solution such as water [20]. Fig. 2 shows the TEM image of acid treatment of MWCNTs in nitric acid. As can be seen, acid treatment leads to the opening and cutting of MWCNTs to the short length.

3-2. Decoration of MWCNTs

As a result of oxidation of MWCNTs with HNO₃, oxygen containing groups act as active sites and adsorb titanium ions which are produced by hydrolysis of TiCl₄. Adsorption of titanium ions on the outer surface of MWCNTs due to the electrostatic attraction led to the nucleation of TiO₂ nanoparticles. Then calcination of MWCNT-TiO₂ results in the nanocrystalline TiO₂ on the outer surface of MWCNTs which is consistent with the results reported by Abbasi *et al.* [21]. Fig. 3 illustrates the TEM image of decorated MWCNTs. It is observed that the outer surface of MWCNTs was decorated with TiO₂ nanoparticles. The formation mechanism of TiO₂ nanoparticles is as shown below:

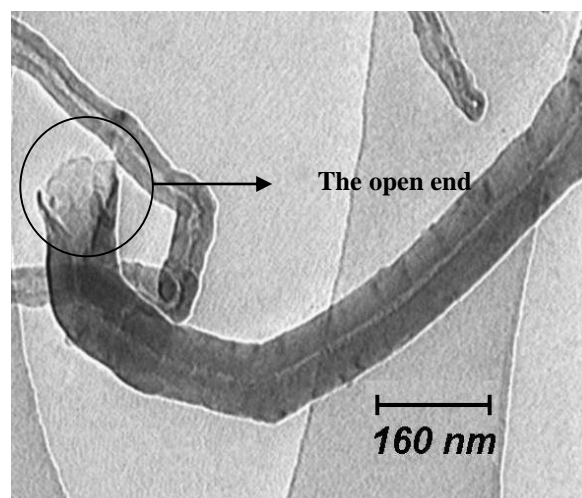
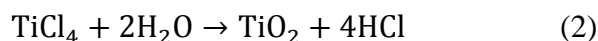


Figure 2. TEM image of acid treated MWCNTs.

The amount of TiO₂ nanoparticles introduced to the outer surface of MWCNTs was affected by the used volume of precursor (TiCl₄). Increasing the amount of used TiCl₄ leads to the augmentation of the titanium ions which were produced during the hydrolysis. Therefore, the attachment probability of titanium ions to the functional groups increases. Hence, these attached ions grow and change to the TiO₂ nanoparticles that covalently attach on the outer surface of MWCNTs. This is in agreement with results of previous investigation [22]. In this study, we synthesized two kinds of MWCNTs-TiO₂ with 34% and 61% of TiO₂ nanoparticles.

3-3. Measurement of thermal conductivity of nanofluids

Before taking the main measurements the experimental apparatus was calibrated with glycerol and allowed 15 minutes of equilibration time. The standard thermal conductivity of the glycerol is 0.285 W/(m·K) at 20°C. To minimize the error during the

measurements, the needle should be approximately centered in the vial and must not be touching the side of the vial, to ensure that the system is not undergoing rapid temperature drift. Even the heat from holding the vial in one's hand for a few seconds, or the cooling from direct air conditioning flow can decrease the accuracy of the measurement. Therefore, for more accuracy the needle and nanofluid should be placed at the same temperature and avoid free convection (e.g. insulated the vial and needle). The thermal conductivity of water as a base fluid was measured at the mentioned temperatures and was compared with the reference data presented in the standard data [23,24]. As can be seen in Fig. 4, the measured data are a little smaller than those of the reference data.

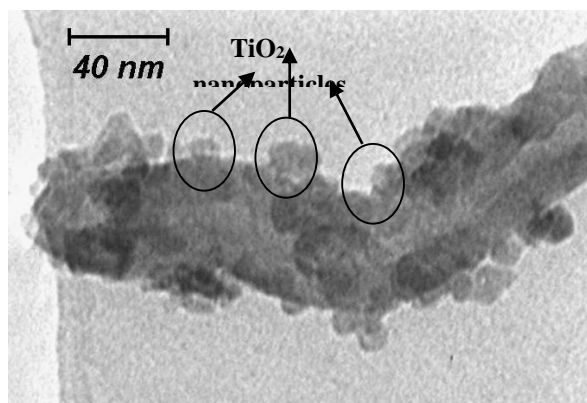


Figure 3. TEM image of MWCNTs-TiO₂.

Fig. 5 depicts the measured thermal conductivity of TiO₂/water nanofluids as a function of temperature at four different mass fractions. The average size of prepared nanoparticles is in the range of 21 nm to 45 nm. However, a high proportion of them is about 21 nm to 24 nm. Due to the influence of temperature on the thermal conductivity of nanofluids, nanofluids may be used under different temperatures. Therefore, we want to

investigate the temperature effect on the thermal conductivity and thermal conductivity enhancement ratio of nanofluids. In Fig. 5, it is clear that the thermal conductivities of TiO₂ nanofluids are higher than those of the base fluids at all the tested temperatures. Also, it can be observed that the thermal conductivity increases with increasing temperature and TiO₂ concentration. Meanwhile it can be deduced that the temperature has a more significant effect than the mass concentration. For the nanofluid containing sphere metal or metaloxide nanoparticles the strong temperature dependence of thermal conductivity was due to the Brownian motion of nanoparticles [25].

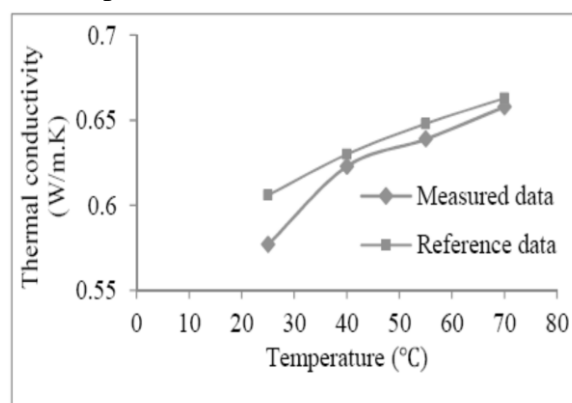


Figure 4. Comparison of the thermal conductivity between measured data and reference data.

From Fig. 6 it can be inferred that thermal conductivity enhancement ratio is affected by both temperature and concentration. Whereas k_{nf} and k_w represent the thermal conductivities of the nanofluid and the base fluid, respectively and k_{nf}/k_w is the thermal conductivity enhancement ratio. By increasing the temperature and mass fraction from 25 to 70°C and 0.25 to 1.5 wt%, the thermal conductivity of TiO₂ nanofluid was increased between 1.38% and 5.16%. The

maximum enhancement of thermal conductivity of TiO₂/water nanofluids occurs at 70°C for nanofluids containing 1.5 wt%.

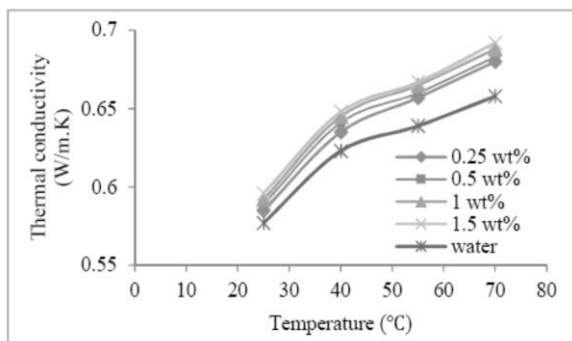


Figure 5. Dependence of the thermal conductivity of TiO₂/water nanofluids on temperature at different mass fractions.

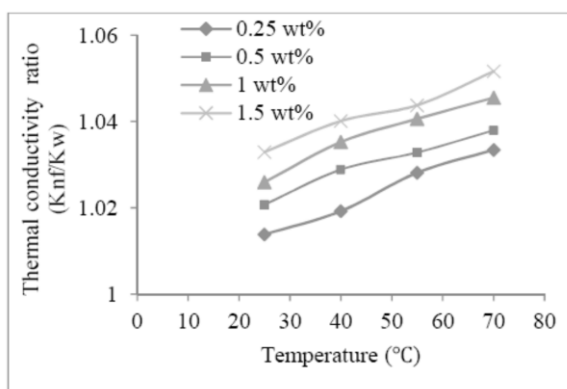


Figure 6. Variation of thermal conductivity enhancement ratio of TiO₂ nanofluids with temperature.

Fig. 7 and 8 depict the thermal conductivity and thermal conductivity enhancement ratio of the MWCNTs nanofluids as a function of temperatures respectively. The mass fraction of the MWCNTs in water is 0.25, 0.5, 1 and 1.5 wt%. According to the Fig. 7, it is seen that the thermal conductivity of water-based nanofluids containing MWCNTs show augmentation with respect to temperature and weight fraction of MWCNTs. It should be mentioned that in the tested weight fraction range, the influence of weight fraction is not

significant and can be negligible. But as can be observed, the effect of temperature is more important. According to the FTIR analysis, the outer surface of MWCNTs is functionalized with oxygen containing groups such as –OH which are introduced during the purification of as prepared MWCNTs (data not shown). Therefore, in the water-based nanofluids containing MWCNTs, in addition to the Brownian motion of MWCNTs, the chemical functionalized groups have a key effect on the amount of energy which transfers into the nanofluids by changing the temperature [26]. As we know, by increasing the temperature, the hydrogen bond of water was weakened. Therefore the structure of

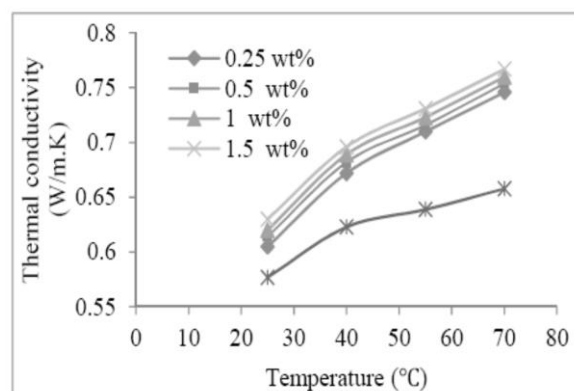


Figure 7. Dependence of the thermal conductivity of MWCNTs/water nanofluids on temperature at different mass fractions.

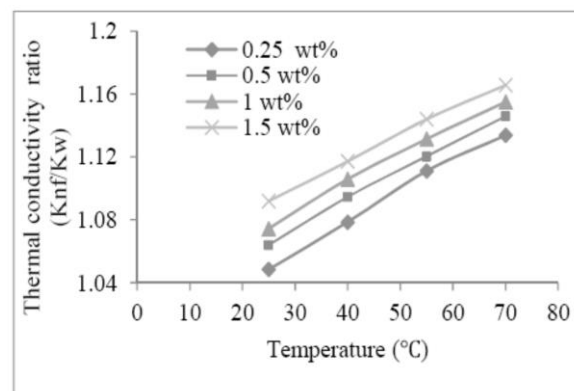


Figure 8. Variation of thermal conductivity enhancement ratio of MWCNTs/water nanofluids with temperature.

enhancement ratio of MWCNTs nanofluids with temperature.

water molecules was destroyed and the number of free water molecules increased. These free water molecules can be arranged around the MWCNTs surface. This liquid layer which was produced due to the chemical surfaces of MWCNTs and van der Waals force between the water molecules has a higher thermal conductivity than the bulk liquid [27]. As can be seen in Fig. 8 the thermal conductivity enhancement ratio of the MWCNTs nanofluids increases with respect to the temperature and weight fraction. In the weight fraction ranging from 0.25 wt% to 1.5 wt% and testing temperature it was observed that the enhancement ratio varied between 5% and 16.5%. The comparison between the maximum thermal conductivity enhancement ratio of TiO₂ nanofluids and that of MWCNTs nanofluids shows that the MWCNTs increase the thermal conductivity much higher than TiO₂ nanoparticles. This can be due to the excellent intrinsic thermal conductivity of MWCNTs.

Thermal conductivity of nanofluids containing MWCNTs-TiO₂ with 61% and 34% of TiO₂ nanoparticles are presented in Fig. 9a and b, respectively. It is clear that the application of MWCNTs-TiO₂ at different amount of TiO₂ nanoparticles increases the thermal conductivity of nanofluids with respect to the temperature and weight fraction. From Fig. 9 it can be inferred that the thermal conductivity of nanofluid containing MWCNTs-TiO₂ with 61% and 34% of TiO₂ nanoparticles is equal to 0.784 and 0.805 at temperature of 70°C and weight fraction of 1.5 wt% respectively. Therefore, it can be deduced that the thermal conductivity of

MWCNT-TiO₂ nanofluids increases as TiO₂ nanoparticles content decreases. Because decreasing amount of TiO₂ nanoparticles leads to the augmentation of a portion of MWCNTs in the hybrid which has higher intrinsic thermal conductivity.

Fig. 10a and b depict the thermal conductivity enhancement ratio of the MWCNTs-TiO₂ nanofluids with 61% and MWCNTs-TiO₂ with 34% of TiO₂ nanoparticles respectively. It is clear that the maximum thermal conductivity enhancement of nanofluid containing MWCNTs-TiO₂ with 61% and MWCNTs-TiO₂ with 34% of TiO₂ nanoparticles is equal to 19.4% and 22.34% respectively.

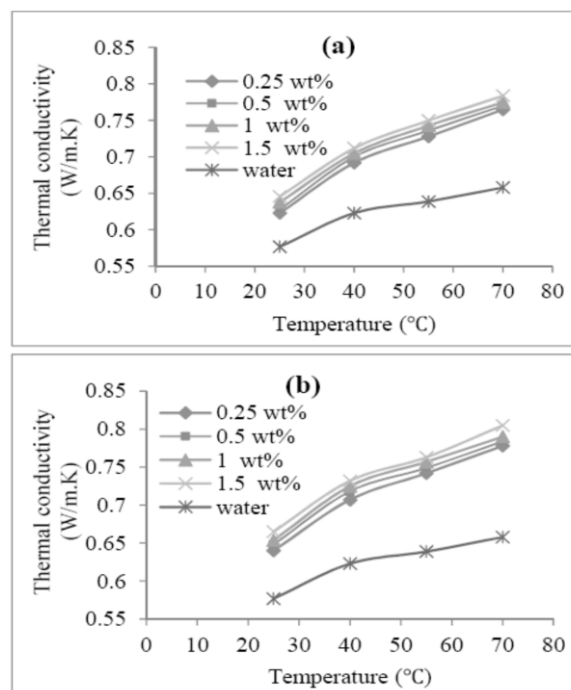


Figure 9. Dependence of the thermal conductivity of MWCNTs-TiO₂/water nanofluids on temperature at different mass fractions, (a): MWCNT-TiO₂ (61%), (b): MWCNT-TiO₂ (34%).

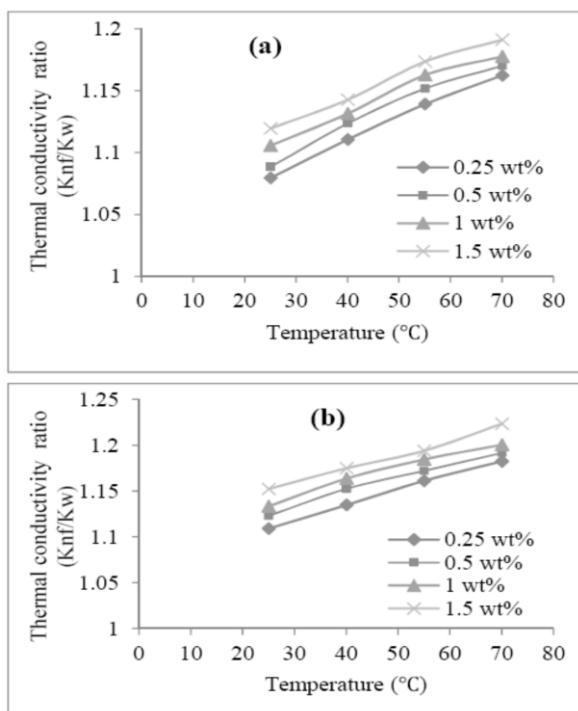


Figure 10. Variation of thermal conductivity enhancement ratio of MWCNTs-TiO₂ nanofluids with temperature (a): MWCNT-TiO₂ (61%), (b): MWCNT-TiO₂ (34%).

The comparison between thermal conductivity of nanofluids containing the TiO₂ nanoparticles, pristine MWCNTs and decorated MWCNTs with different amount of TiO₂ nanoparticles is illustrated in Fig. 11. As can be deduced from this figure, the sequence of thermal conductivity of nanofluids containing different amount of nanoparticles are the intrinsic thermal conductivity of nanoparticles (the thermal conductivities of the order given by MWCNT-TiO₂ (34%) > MWCNT-TiO₂ (61%) > MWCNTs > TiO₂). This is in agreement with results of previous investigation [17].

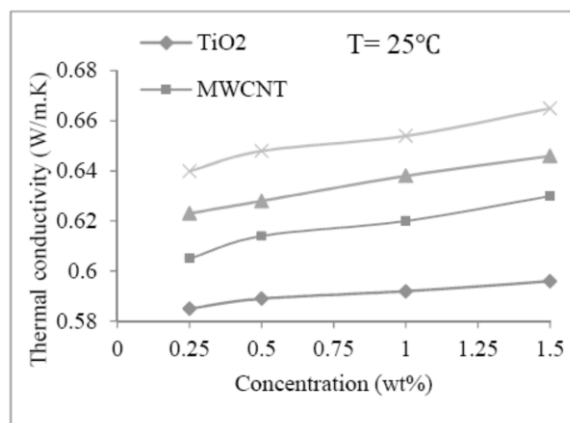


Figure 11. Comparison the thermal conductivity of MWCNT-TiO₂ (34%), MWCNT-TiO₂ (61%), MWCNTs and TiO₂ at 25°C.

4. Conclusions

The present study measured the thermal conductivity of nanofluids containing TiO₂ nanoparticles, pristine MWCNTs and decorated MWCNTs with 34% and 61% of TiO₂ nanoparticles and investigated the effects of weight concentrations and temperature. The results show that the thermal conductivities of all studied nanofluids are higher than those of the base fluids at all the tested temperatures and concentrations and the influence of temperature on the thermal conductivity and enhancement ratio of the thermal conductivity is more significant than the weight fraction. Also, it has been observed that thermal conductivity of MWCNTs nanofluid is higher than that of TiO₂ nanofluid. In addition, the results revealed that the nanofluids containing MWCNTs-TiO₂ have higher thermal conductivity than that of the TiO₂ and MWCNTs nanofluids and thermal conductivity increases as TiO₂ nanoparticles content of hybrid decreases.

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