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ScienceDirect

Energy Procedia 110 (2017) 101 – 108

Energy Procedia

1st International Conference on Energy and Power, ICEP2016, 14-16 December 2016, RMIT University, Melbourne, Australia

Electrical and thermal conductivities of 50/50 water-ethylene glycol based TiO_2 nanofluids to be used as coolants in PEM fuel cells

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Abstract

Both thermal and electrical conductivities of nanofluids are the key parameters to be optimized for making nanofluids suitable for use as coolants in electrically active thermal applications such as fuel cells. Although nanofluids are considered to be the potential solutions as coolants for Proton Exchange Membrane (PEM) fuel cells, their high electrical conductivity is seen as a challenge in their applications. This study investigates theoretically and experimentally the electrical and thermal conductivities of 50/50 water-ethylene glycol based TiO_2 nanofluids with nanoparticle concentrations in the range of 0.05-0.5 vol%. Though Maxwell model predicts decrease of electrical conductivity with increase of nanoparticle concentrations, an enormous increase of electrical conductivity (~ 900% with 0.5 vol% at 70 ˚C) has been observed compared with that of the base fluid. This experimental result indicates that the Maxwell model is unable to predict the electrical conductivity of nanofluids. On the other hand, the thermal conductivity increases with the increase of concentration of nanoparticles. With 0.5 vol% nanoparticle concentration, the thermal conductivity increased by just over 10% compared to the base fluid.

Peer-review under responsibility of the organizing committee of the 1st International Conference on Energy and Power.

Keywords: Nanofluids; Electrical and thermal conductivities; PEMFCs; coolants.

1. Introduction

The heat transfer characteristics of conventional coolants (e.g. water, ethylene-glycol (EG), mixture of water-EG, etc.) can be improved by enhancing their thermal properties. This can be done by dispersing nano-sized particles (e.g.

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Peer-review under responsibility of the organizing committee of the 1st International Conference on Energy and Power.
doi:10.1016/j.egypro.2017.03.113

metals or metal oxides) into conventional coolants to form what are called nanofluids [1]. Moreover, by increasing the concentration of nanoparticles, the thermal conductivity of fluids increases. Due to the high thermal conductivities of nanofluids, they have attracted substantial attention of researchers in the last couple of decades. However, the electrical conductivity of nanofluids also increases with the increase of nanoparticle concentrations that hinders the applications of nanofluids as coolants in many electrical devices such as Proton Exchange Membrane Fuel Cells (PEMFCs) [2]. The electrical energy efficiency of a PEMFC is usually in the range of 30-50% (depending on its operating point) that means a substantial amount of heat is generated in the fuel cell [3-5]. However, the operating temperature of PEMFCs is relatively low (i.e. 60-80 °C) [6, 7], which limits the opportunity to exchange heat with their surroundings (e.g. at air at 25 °C). That is why the size of the heat exchangers used for cooling PEMFCs are generally relatively large [2]. This relatively large size of radiator can create a challenging situation particularly in applications with packaging limitations, such as automotive applications. Using coolants with better thermal performance (i.e. for heat removal) can address this problem. Previous studies showed that higher thermal conductivities of nanofluids have shown to be effective in reducing the size of the heat exchanger (i.e. by over 25%) used to cool PEMFCs [1, 2]. On the other hand, the suspended nanoparticles also increase the electrical conductivity of the nanofluids which can affect the electrical performance of the PEM fuel cells when they are used as PEMFCs coolants. Hence, it is very important to optimize the thermal and electrical conductivity of nanofluids for PEMFCs coolants.

Kole and Dey [8] investigated ZnO-ethylene glycol (EG) nanofluids prepared by using prolonged sonication (>60 h) that resulted in superior fragmentation and dispersion of ZnO nanoparticles. They measured the thermal conductivity enhancement based on the nanoparticle concentration and temperature. They estimated around 40% thermal conductivity enhancement for 3.75 vol% of ZnO, in ethylene glycol as based fluid, at 30˚C compared with ethylene glycol alone. Sundar et al. [9] also experimentally measured the thermal conductivity of ethylene glycol and water mixture based Al2O3 and CuO nanofluids at different volume concentrations and temperatures. They found that the thermal conductivity enhancement of the Al2O3 nanofluid varies from 9.8% to 17.89% and for CuO nanofluid it varies from 15.6% to 24.56% within the temperature range of 15 ˚C to 50 ˚C at 0.8% volume concentration (i.e. compared with that of the base fluid respectively). Reddy and Rao [10] investigated TiO_2 nanofluids with ethylene glycol-water as base fluid at different volume concentrations and at different temperatures. The thermal conductivity enhancement for water-EG (60/40) based TiO2 nanofluids was measured to be 1.94% and 4.38% respectively compared with that of the base fluid, when the concentration increased from 0.2 vol% to 1.0 vol%. Abdolbaqi et al. [11] investigated the thermal conductivity and viscosity of bioglycol/water based TiO2 nanofluid in the concentration range of 0.5- 2 vol% in temperatures between 30 ˚C and 80 ˚C. They found that the thermal conductivity increases with the increase of concentration while decreases with increasing temperature. They observed a maximum thermal conductivity enhancement of around 12.6% in the volume concentration of 2% and the temperature of 70 ˚C.

Very recently Zyla and Fal [12] experimentally investigated the thermal and electrical conductivity of ethylene glycol based aluminium nitride nanofluids. They measured the electrical conductivity over the nanoparticle volume concentration range of 1.8 to 7.9% and found 600 times enhancement of electrical conductivity for the highest measured volume concentration of 7.9% for the nanoparticles. They also observed that the electrical conductivity of EG based aluminium nitride nanofluids increases linearly with increasing volume fraction of nanoparticles in the base fluids. Sundar et al. [13] experimentally investigated the electrical conductivity enhancement of water and EG based nano-diamond-nickel (ND-Ni) nano-composite based magnetic nanofluids. They did their experiment with the low particle concentration of 0.02 vol%, 0.05 vol% and 0.1 vol% within the temperature range of 24 65 ˚C. The enhancement values in electrical conductivity for 0.1 vol% of water based ND-Ni nanofluid was measured to be 1339.81% and 853.15% at 24 to 65 ˚C respectively, compared to water as the base fluid. With EG as the base fluid (i.e. with similar nanoparticles), they found ~199% and ~200% enhancement in electrical conductivity at 24 ˚C and 65 ˚C respectively (i.e. compared to the base fluid). Khdher et al. [14] experimentally determined the thermal and electrical conductivity of bio glycol based Al_2O_3 nanofluids in the temperature range of 30-80˚C with the volume concentrations of 0.1%, 0.3%, 0.5%, 0.7% and 1%. They observed that the electrical conductivity of nanofluids increases with the increment of nanoparticle concentration as well as temperature; they also measured the electrical conductivity of 154 μS/cm for 0.5 vol% concentration of particles. Zakaria et al. [15] investigated the thermal and electrical conductivity of water-EG based Al_2O_3 nanofluids with the EG concentration ranging from 0% to 100% and nanoparticle concentration of 0.1, 0.3 and 0.5 vol%. They also found that the electrical conductivity increment as a function of volume concentration. Abdolbaqi et al. [16] experimentally investigated the thermal and electrical conductivities of BioGlycol-water (BGW) mixture based Al_2O_3 nanofluids with the particle concentration of 0.5-2.0 vol%. They found that the electrical conductivity property of BGW dramatically decreased by the addition of nanoparticles (i.e. increasing the concentration). Minea and Luciu did an experimental study on the electrical conductivity of water based Al_2O_3 nanofluids in the concentrations range of 1 vol% to 4 vol%. They observed that the effective electrical conductivity of alumina nanofluids increased linearly with increased volume fractions of the alumina and temperature compared to the base fluid. For 4 vol% of alumina at 70˚C, they recorded the highest electrical conductivity of 4210 μS/cm.

Using nanofluids as PEMFCs coolants, the thermal and electrical conductivities are critical and thus need comprehensive experimental verification. For this paper we selected a 50/50 water-EG as base fluid in order to lower the freezing point of the coolant to allow its use in different sectors including automotive. As a nanoparticle, TiO_2 is selected which is an insulating material and very low electrical conductive compared with that of the metal nanoparticles. Adding EG to water decreases the thermal conductivity of the mixture but adding nanoparticle enhances the thermal properties of the mixture. Hence, the main focus of this paper is to investigate the thermal and electrical conductivities of 50/50 water-EG based TiO_2 nanofluids both theoretically and experimentally in order to determine their applicability as coolants in PEM fuel cells cooling systems.

2. Methodology

2.1. Overview

A two-step method has been adopted for preparing nanofluids used in this study. The nanoparticles were purchased from well-known company which is an ISO 9001 certified worldwide manufacturer and supplier of nanoparticles. Ethylene Glycol (EG) also known as Ethanediol, and Milli-Q which is prepared in RMIT chemical lab were used to prepare the 50/50 Water-EG mixture as the base fluid. The thermal and electrical conductivity of TiO_2 nanofluids in the concentration of 0.05 vol% to 0.5 vol% has been investigated both theoretically and experimentally.

2.2. Theoretical approach

Thermal conductivity is the most widely studied property of nanofluids in recent literature [12, 14, 17-31]. However, currently there is no absolutely reliable theory to predict the anomalous thermal conductivity of nanofluids as this property depends on its various parameters [32]. The static model developed by Maxwell is used to determine the effective thermal conductivity of liquid-solid suspensions of mono-disperse, low volume-fraction mixtures of spherical particles which is given by the following Equation [33, 34]:

$$K_{nf} = \frac{K_p + 2K_b + 2(K_p - K_b)\phi}{K_p + 2K_b - (K_p - K_b)\phi} K_b \tag{1}$$

where K_{nf} is the effective thermal conductivity of the nanofluid, K_p is the thermal conductivity of the nanoparticles, K_b is the thermal conductivity of the base fluid, and ϕ is the volume fraction of nanoparticle in the nanofluid.

One of the first models that described the electrical conductivity of suspensions was introduced by Maxwell [35]. This model is applicable only for very low volume concentrations of solid-fluid suspensions with randomly dispersed, uniformly sized and non-interacting spherical particles. The Maxwell model predicts the effective electrical conductivity of suspensions (σ_{nf}). This parameter is a function of the electrical conductivity of particles (σ_p), electrical conductivity of base fluids (σ_b) and the volume fraction (φ) of the particle that can be given as:

$$\sigma_{nf} = \left(1 + \frac{3(\alpha - 1)\phi}{(\alpha + 2) - (\alpha - 1)\phi}\right)\sigma_b \tag{2}$$

where $\alpha = \sigma_p / \sigma_b$ represents the conductivity ratio of the solid and liquid phases.

The Maxwell model is a static model and does not take into account factors such as Brownian motion, aggregation and electrical double layer (EDL). Due to all of these, the Maxwell model does not often predict the experimental results very well [36, 37]. However, for using Maxwell's model, Cruz *et al.* [38] suggested considering three special cases in terms of the electrical conductivities of the particles and the base fluid by applying the DLVO theory. This DLVO theory successfully describes the total interaction energy between particle pairs as a function of the distance between them, stating that it is the balance of the repulsive potential due to the electric charges present in the electrical double layer and the attractive potential due to the ever present long distance van der Waals forces. One of the three cases (i.e. electrical conductivity of nanoparticle is less, equal or more than that of the base fluid) is applicable for insulating nanoparticles (e.g. TiO_2) when $\sigma_p << \sigma_b$, $\alpha \rightarrow 0$ then the equation (2) can be written as:

$$\sigma_{nf} = \left(1 - \frac{3}{2}\phi\right)\sigma_b \tag{3}$$

2.3. Experimental investigation

A KD2 Pro thermal properties analyser manufactured by Decagon Devices, Inc. was used to measure the thermal conductivity of the nanofluids. The KD2 Pro works on the basis of transient hot wire method. The KS-1 sensor is the one used to measure the thermal conductivity of nanofluids. While the default read time for the KS-1 sensor is one minute, when dealing with low viscosity liquid samples, the duration of the read time should be as small as possible to minimise the amount of heat added to the sample. The accuracy of KD2 Pro is +/- 5% for thermal conductivity range of 0.02 to 2.00 W/m.K. IntelliCAL™ CDC401 is a digital, graphite, hand held, 4-pole conductivity probe which was used for measuring electrical conductivity of nanofluids in this study. The accuracy of the electrical conductivity meter is +/- 0.5 % of the reading. A water bath was also used to heat up and stabilize the temperature of nanofluids for measuring electrical and thermal conductivities.

2.4. Uncertainties

The KD2 Pro sensor used in this study was calibrated by introducing the known thermal conductivity fluid such as glycerine provided by the manufacturer with the thermal conductivity of 0.282 W/m.K at 20˚C. The sensor was calibrated each time before taking the measurement of thermal conductivity with the accuracy of less than +/- 2%. For further validation of KD2 Pro thermal analyser, the thermal conductivity of pure ethylene glycol (99.90% minimum) has been measured and compared with available literature data [39] with maximum 0.13% error (Fig. 1).

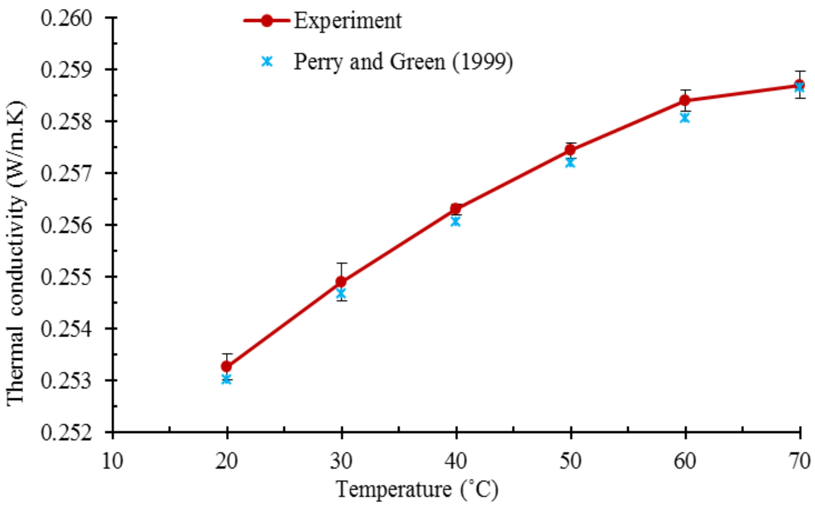

Fig. 1. Comparing the measured thermal conductivities of ethylene glycol with available literature data in Perry's chemical engineer's hand book [39]

The electrical conductivity meter was also calibrated by introducing a fluid of known electrical conductivity, i.e. pure ethylene glycol (99.90 %), and 50/50 water-EG mixture. The measured data closely matched with the available literature [40], with errors of 4.8% and 3.3% for ethylene glycol and 50/50 water-EG mixture respectively Table 1.

Table 1. Comparison of measured electrical conductivity of ethylene glycol and 50/50 water-ethylene glycol mixture (by volume) with the industrial data provided by MEGlobal [40] at 20 ˚C

Liquid	Measured (μS/cm)	MEGlobal (μS/cm)	Difference (%)
Ethylene glycol	3.14	3.3	4.8
50/50 water-EG mixture	5.03	5.20	3.3

3. Results and discussions

3.1. The effect of temperature and concentration of nanoparticles on the thermal conductivity of nanofluids

Fig. 2 and Fig. 3 show that the enhancement of thermal conductivity of nanofluids increased with an increase of nanoparticle volume concentration as well as the temperature compared with that of the base fluids and the increment is almost linear. The same enhancement trend in nanofluids thermal conductivity has been observed previously [12, 20, 21, 30, 41-44] with different types of nanoparticles and base fluids. By increasing the temperature from 20 ˚C to 70 ˚C, the maximum enhancement of thermal conductivity was found to be ~6% for 0.05 vol% TiO_2 nanofluids compared with the thermal conductivity of the base fluid (i.e. 50/50 water-EG mixture). The thermal conductivity of nanofluids increase mainly due to the facts that the nanoparticles alter the fluid composition that affects the energy transport process; specifically the random Brownian motion of nanoparticles and interfacial interactions between the nanoparticles and the liquid molecules enhance energy transport inside the liquid [45].

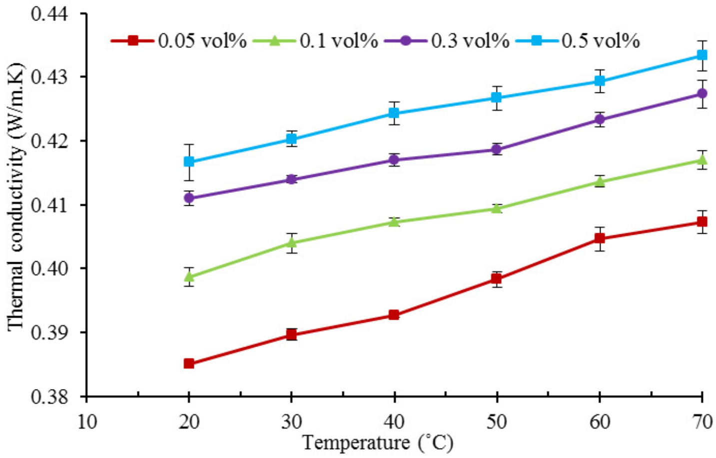

Fig. 2. Variation of effective thermal conductivity of base fluid (50/50 water-EG) and various volume concentration of 50/50 water-EG based TiO_2 nanofluids with temperature

With the various level of nanoparticles concentrations used in this experimental study (i.e. 0.05-0.5 vol%), the enhancement of thermal conductivity was found to be ~10%, when temperature varied between 20 ˚C and 70 ˚C. It is noteworthy that the rate of thermal conductivity enhancement is more sensitive to concentration rather than temperature. It is clear from Fig. 3 that the TiO_2 nanofluid exhibits higher thermal conductivities compared with the base fluid (i.e. at different concentrations). It was also observed that the Maxwell's model under-predict the thermal conductivity of the nanofluids. This shortfall was also observed by few other researchers [20, 29, 46-58]. The experimentally-measured thermal conductivities of 50/50 water-EG based TiO_2 nanofluids (i.e. measured during this study), showed a close agreement with the experimental thermal conductivities (i.e. at different concentrations) reported by Reddy *et al.* [59] (i.e. with the maximum difference of less than 1%) .

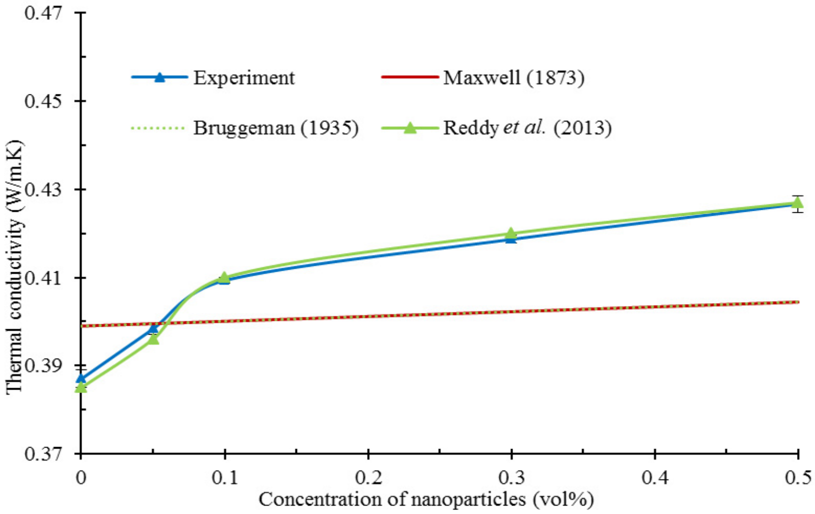

Fig. 3. Comparisons between the experimental results of this study, experimental results of Reddy et al. [60], and the values predicted by the Maxwell and Bruggeman theories at 50 ˚C for 50/50 water-EG based TiO_2 nanofluids

3.2. The effect of temperature and nanoparticle concentration on the electrical conductivity of TiO_2 nanofluid

It was observed that the electrical conductivities of the 50/50 water-EG based TiO_2 nanofluids increased almost linearly with the temperature (Fig. 4). With 0.05 vol% concentration, almost 91% enhancement of electrical conductivity was observed compared to the base fluid when the temperature was increased from 20˚C to 70˚C. From similar increase in the temperature, the enhancement in the electrical conductivity was found to be 52% for 0.5 vol% concentration. This is mainly due to the fact that nanoparticles are less sensitive to temperature compared with the liquids, and the increased temperature causes a reduction in the viscosity of the base fluids and an increase in the Brownian motion of nanoparticles, that leads to a further increase in the electrical conductivity [61-63].

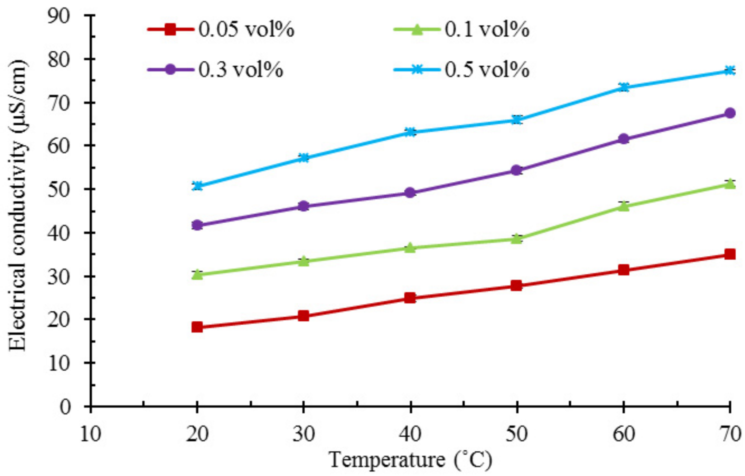

Fig. 4. The effect of temperature on the electrical conductivity of base fluid (50/50 water-EG) and various volume concentration of 50/50 WEG based TiO2 nanofluids

Fig. 5 shows the enhancement of electrical conductivity of 50/50 water-EG based TiO_2 nanofluids for the concentration range of 0.05 vol% to 0.5vol%, at 50 ˚C. It is clearly seen that the electrical conductivity increases almost linearly with the increase of nanoparticles volume concentrations. This increment trend has been observed by other researchers as well [8, 62-64] for different nanofluids. At 50 ˚C, the electrical conductivity of the nanofluid at

0.5 vol% concentration was observed to be almost ten times (~900%) larger than that for the base fluid (i.e. 50/50 water-EG). This is while the Maxwell's modified model, as described by equation (3) even suggests a slight reduction in electrical conductivity by increasing the concentration. It was discussed by earlier studies that the Maxwell's model is applicable for the dilute suspensions $(\phi << 1)$ with the particles size larger than tens of micrometres [65, 66]. The size difference offers a possible explanation for the above-mentioned discrepancy. The enhancement in electrical conductivity is because of the fact that an increase in volume fraction increases the charge transport due to increase in the number of charge carriers. Along with the increase of charge carriers, some factors such as Brownian motion, Electrical Double Layer (EDL) interactions, agglomeration or even electrochemical properties of nanoparticles cause the increase of electrical conductivity of the nanofluids [67, 68] that the Maxwell model does not take into account.

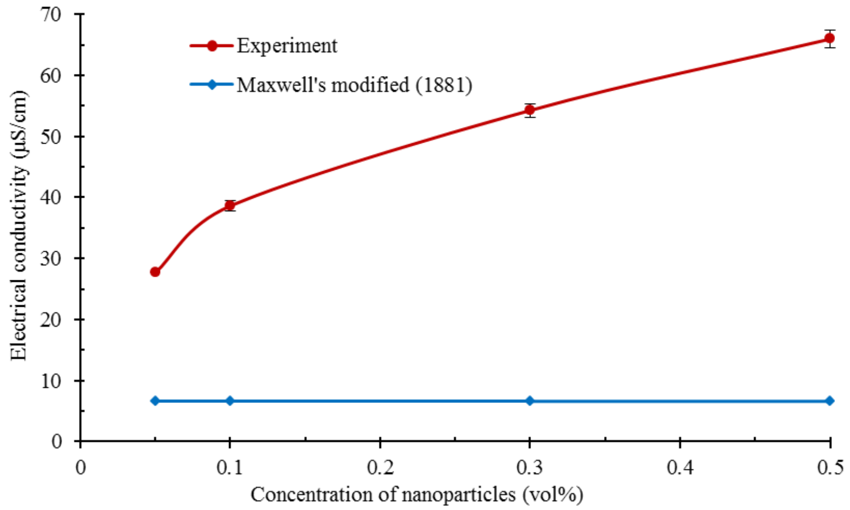

Fig. 5. The electrical conductivity of TiO2 nanofluids at 50˚C: a comparison between the experimentally measured electrical conductivity in this study and those suggested by the Maxwell (1881) electrical conductivity model

4. Conclusion

The thermal and electrical conductivities of 50/50 water-EG based TiO_2 nanofluids have been investigated both theoretically and experimentally. The thermal conductivity increased with the increase of both temperature and concentration of nanoparticles. The enhancement of thermal conductivity was found to be ~10%, when temperature varied between 20 ˚C and 70 ˚C. Around 91% enhancement of electrical conductivity for TiO_2 nanofluids relative to the base fluid was measured for 0.05 vol% concentration for increasing the temperature from 20˚C to 70˚C while for 0.5 vol% concentration the enhancements was found to be 52%. The enhancement of thermal and electrical conductivity of 50/50 water-EG based TiO_2 nanofluids were found to be almost linear with the increase of temperature and concentration of nanoparticles. The classical Maxwell's thermal conductivity model and Cruz *et al.* [38]'s modified Maxwell's electrical conductivity model under-predict the enhancement of thermal and electrical conductivities for a TiO_2 nanofluid.

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