EXPERIMENTAL STUDIES ON THE VISCOSITY OF Fe NANOPARTICLES DISPERSED IN ETHYLENE GLYCOL AND WATER MIXTURE

by

Amir KARIMI [a*]***, Mohamad Amin ABDOLAHI SADATLU*** [b]***, and Mehdi ASHJAEE*** [a]

[a] Department of Mechanical Engineering, College of Engineering, University of Tehran, Tehran, Iran
[b] Department of Engineering and Science, Sharif University of Technology, International Campus, Kish, Iran

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In this paper, experimental studies are conducted in order to measure the viscosity of Fe nanoparticles dispersed in various weight concentrations (25/75%, 45/55% and 55/45%) of ethylene glycol and water (EG-water) mixture. The experimental measurements are performed at various volume concentrations up to 2% and temperatures ranging from 10 °C to 60 °C. The experimental results disclose that the viscosity of nanofluids increases with increase in Fe particle volume fraction, and decreases with increase in temperature. Maximum enhancement in viscosity of nanofluids is 2.14 times for 55/45% EG-water based nanofluid at 2% volume concentration compared to the base fluid. Moreover, some comparisons between experimental results and theoretical models are drawn. It is also observed that the prior theoretical models do not estimate the viscosity of nanofluid accurately. Finally, a new empirical correlation is proposed to predict the viscosity of nanofluids as a function of volume concentration, temperature, and the viscosity of base fluid.

Keywords: *viscosity, Fe nanoparticle, theoretical model, nanofluid*

Introduction

Improving heat transfer capability in the thermal systems such as electronic cooling, heat exchangers, and high energy devices is very essential from the industrial and energy saving perspectives [1]. The poor thermal properties of conventional heat transfer liquids such as water, ethylene glycol, propylene glycol and engine oil makes some impediments for the advances in heat transfer techniques. Therefore, a significant amount of investigations have been performed to enhance the thermal performance. An innovative method for heat transfer process intensification, is adding nano-sized particles in base fluids which has been investigated extensively during the past decade [2, 3]. The nanofluids have attracted great interest because of their potential to increase energy-efficient heat transfer domain, which could have a remarkable impact on energy generation and storage systems [4]. The transport properties of nanofluids such as thermal conductivity and viscosity are essential parameters controlling the heat exchange of the fluid [5]. The nanofluids have some scientific applications in a variety of fields such as cool-

* Corresponding author; e-mail: amir.karimi@ut.ac.ir

ing systems, mechanical engineering and bioengineering [6]. Numerous researches have been conducted concerning transport properties measurement of homogeneous mixtures containing metallic, metallic oxides and carbon nanotubes [7-10].

Ferrofluid, colloidal suspensions of magnetic nanoparticles, such as Fe_3O_4, γ-Fe_2O_3, $CoFe_2O_4$, and FeCo in a base fluid are considered as a functional fluid which are capable to respond under effect of external magnetic field [11]. In recent years, using ferrofluid has received great attention from researchers around the world due to its importance in electronic packing [12], drug delivery [13], and magnetic resonance imaging (MRI) techniques [14].

Many experimental investigations are performed to study the viscosity of non-magnetic type nanofluids. Pak and Cho [15] studied the viscosity of Al_2O_3 and TiO_2 nanoparticles dispersed in water. At volume concentration of 2.78%, viscosity enhancement of 2.5 times greater than that of pure water has been observed for Al_2O_3 nanofluid. While, it is obtained as 1.3 times for TiO_2 nanofluid at volume concentration of 3.16%. Masuda *et al.* [16] estimated the viscosity of water based TiO_2 nanofluids with a particles size of 27 nm and observed a 60% enhancement of viscosity at 4.3% volume concentration. He *et al.* [17] measured the viscosity of nanofluids containing TiO_2 nanoparticles with average diameters of 95, 45, and 210 nm suspended in water. They observed the enhancement in viscosity ratio of nanofluids with increase in particle size of nanoparticles. Nguyen *et al.* [18] studied the effects of temperature, concentration and particle size on the viscosity of water based nanofluids. They used Al_2O_3 nanoparticles with the size of 36 and 47 nm as well as CuO nanoparticles with the diameter of 29 nm. The experimental measurements are performed at temperature range of 22-75 °C and the particle volume fraction varied between 1% and 12%. They reported that in high concentrations, the greater particles led to higher viscosity enhancement than the smaller ones. Lee *et al.* [19] considered water based Al_2O_3 nanofluids and observed 2.9% viscosity enhancement at 0.3% volume concentration and temperature of 21°C. Murshed *et al.* [20] measured the viscosity of TiO_2 (15 nm) and Al_2O_3 (80 nm) dispersed in water nanofluids at volume concentrations ranging from 1% to 5%. They concluded higher viscosity of nanofluids in comparison with the base fluids. Chandrasekar *et al.* [21] presented some results for the viscosity measurement of water based nanofluids containing Al_2O_3 with particle size of 43 nm and volume concentration up to 5%, at ambient temperature.

In order to decrease the aqueous freezing point of the heat transfer fluid, different ratio of ethylene glycol or propylene glycol are mixed with water. These mixtures are implemented in heat exchangers, automobiles and industrial plants in subzero countries with the long winter climate conditions. Therefore, there is plenty of research about using ethylene glycol/water mixture and propylene glycol/water mixture as a heat transfer fluids. At low temperatures, the ethylene glycol mixture has better heat transfer properties respect to propylene glycol mixture [22]. Kulkarni *et al.* [23] considered nanofluids containing CuO, Al_2O_3, and SiO_2 nanoparticles dispersed in 60/40% EG-water mixture and measured the convective heat transfer coefficient and viscosity of nanofluids. Sundar *et al.* [24] conducted an experimental study on thermophysical properties of 50/50% EG-water mixture based Al_2O_3 and CuO nanofluids. Yiamsawas *et al.* [25] studied the viscosity of Al_2O_3 (120 nm) and TiO_2 (21 nm) nanoparticles dispersed in 20/80% EG-water mixture at volume concentrations of 0-4% and temperature range of 15-60 °C.

Investigations concerning about the viscosity measurement of magnetic nanofluids have been conducted by few researches. The rheological properties of silicon oil based magnetic iron-oxide nanofluid with four volume concentrations up to 12.9% was reported by Yang *et al.* [26]. Odenbach and Stork [27] described the effects of interactions among the surfactant, magnetic particles and base fluid on the viscosity of magnetic nanofluid. Guo *et al.* [28] performed experimental investigations on the viscosity of γ-Fe_2O_3 nanoparticles dispersed in the mixture of water and ethylene glycol, when the volume fractions were varied between from 0.5% to 2%. The adequate sodium oleate was used as surfactant for the dispersion of γ-Fe_2O_3 nanoparticles into the mixture. They concluded that the viscosity of the γ-Fe_2O_3 nanofluids, which showed the Newtonian manner and strongly, depends on the volume concentration and temperature. Abareshi *et al*. [29] measured the viscosity of α-Fe_2O_3 nanoparticles suspended in glycerol as a based fluid. They found that the viscosity of nanofluid decreases with the increment of the temperature. Sundar *et al*. [30] experimentally studied rheological properties of Fe_3O_4 nanoparticles dissolved in different concentrations of EG-water mixture in the volume concentration range from 0 to 1% and temperature ranging from 0 to 40 °C.

Most research works on the viscosity of nanofluids presented until now focus on nanofluids containing nonmagnetic nanoparticles suspended in various base fluids. There are few reports on the viscosity of magnetic nanofluid in the literature and to the best of the authors' knowledge, the investigations on viscosity of Fe magnetic nanoparticles dispersed in various weight concentration of ethylene glycol and water mixture are scarce. The main aim of this paper is to investigate the viscosity of Fe magnetic nanoparticles in different base fluids of 25/75%, 45/55%, and 55/45% EG-water mixture with the influence of volume concentration and temperature. The EG-water mixture has better properties compared to self water and self ethylene glycol due to the mixture has low freezing point and lower viscosity. Finally, the experimental results are compared with some creditable theoretical models and a new empirical correlation is presented to predict the viscosity of nanofluids.

Sample preparation and characterization

The preparation of nanofluid is carried out by dispersing the nanocrystalline powder samples in the base fluid, which is called two-step procedure. In this work, spherical-shaped Fe nanoparticles (Nanostructured and Amorphous Materials, Inc., USA) are used. Ethylene glycol and water mixture with three various concentrations such as 25/75%, 45/55%, and 55/45% of EG-water mixture are utilized as base fluids. In order to obtain a uniform suspension of particles in the base fluid, the dispersion of nanofluid are subjected to ultrasonic vibration (Hielscher UP400S, Germany) for 50 minutes which generates ultrasonic waves of 400 W at 24 kHz. In order to avoid the agglomeration of nanoparticles, adequate sodium dodecyl benzene sulfonate (SDBS) is used as a surfactant. Nanofluids are prepared at volume concentrations of 0.25%, 0.5%, 1%, 1.5%, and 2% to measure the viscosity of nanofluids at different volume concentrations and temperatures. The pH of nanofluid is adjusted by using sodium hydroxide (NaOH) solution and is measured by a pH meter (AZ8686). Zeta potential and average particle diameter (Z-average) are determined by dynamic light scattering (DLS) with a Zetasizer Nano ZS (Malvern Instruments Ltd., UK). The DLS cumulants analysis provides the characterization of a sample through the mean hydrodynamic diameter (Z-average) for the size, calculated from the intensity of scattered light. The physical stability of colloidal dispersions can be evaluated through the zeta potential. If the zeta potential is high, the particles are stable due to the high electrostatic repulsion force between particles. In present study, the zeta potential of nanofluid ($\varphi = 2\%$) is –36.78 mV at pH = 9.7 which indicates that the nanofluids are stable (absolute value more than 30 mV). The zeta potential measurements are repeated after 5 days. Comparing the measurements with primary data show that changes in zeta potential are insignificant. The size and shape of the nanoparticles are determined by TEM, ZEISS EM10-C at 100 kV. The TEM image of nanoparticles is shown in fig. 1. The average particle diameters are obtained to be around of 25 nm, using the Dynamic Light Scattering (DLS) method (fig. 1).

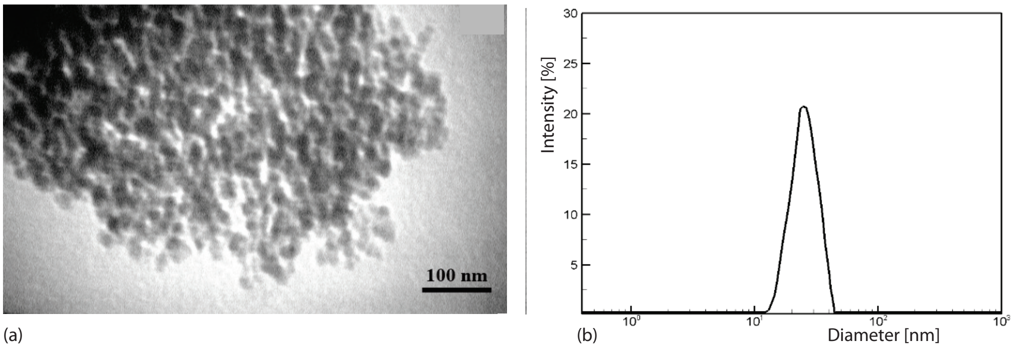

Figure 1. (a) TEM image of nanoparticles, (b) diameter distribution of nanoparticles obtained by dynamic light scattering

Experimental procedure

It is important to study whether the nanofluid behaves like a Newtonian or a non-Newtonian fluid. In the case of nanofluids, some researchers have been reported that nanofluids exhibit Newtonian characteristics [4, 28, 30]. Namburu *et al.* [4] studied the viscosity of copper oxide nanoparticles dispersed in EG-water mixture. They reported that EG-water mixture based copper oxide nanofluids exhibit Newtonian behavior at volume concentration varying up to 6.12%. The results of Guo *et al.* [28] exhibited that the γ-Fe_2O_3 nanofluids show the Newtonian behavior at various volume concentrations between 0 to 2%. Sundar *et al.* [30] have been observed Newtonian behavior for EG-water mixture based Fe_3O_4 magnetic nanofluids at different volume concentrations between 0% and 1% and in the temperature range of 0-50 °C. The nanofluids containing spherical nanoparticles at relatively modest volume fractions (~3 vol.%) show Newtonian behavior [31, 32]. In this study, according to the TEM image of fig. 1, the nanoparticles are on a spherical shape. The experimental measurements are also performed to study whether the fluid shows Newtonian or non-Newtonian behavior after addition of nanoparticles by cone and plate rheometer. It is observed that the shear stress for nanofluids at different volume concentrations up to 2% depends linearly on the shear rate. This indicates that the nanofluids behave in Newtonian manner. The viscosity of Fe nanoparticles suspended in different base fluids like 25/75%, 45/55%, and 55/45% (by weight) EG-water mixture is measured by using falling ball viscometer (Lovis 2000M microviscometer, Anton Paar Company) in the temperature range of 10-60 °C with the accuracy of 0.5%. In Newton's law of motion for a falling ball, the net forces including buoyancy, gravity and viscous is equal to zero at equilibrium state. The method applies Newton's law of motion under force equilibrium on a falling sphere ball when it reaches a terminal velocity. Moreover, the drag force can be estimated from Stokes' law. A measuring system includes a capillary tube and stainless steel ball with a diameter of 1.59 mm and 1.5 mm, respectively. This system is used to measure the viscosity of nanofluids in the range of 0.3 to15 mPa·s. Capillary tubes require at least 0.8 mL sample volume quantity. The ball is located into fall tube after pouring nanofluids. To ensure no air bubbles are entrapped in capillary tube, the screw which is positioned on the top of the tube is tightened. In order to set the temperature measurement, this viscometer has a temperature control system to tune the temperature in range of 5-100 °C with 0.02 °C accuracy. Knowing the fall time of the ball to drop a specific distance through the sample test in an inclined cylindrical tube, the dynamic viscosity is determined. Each experiment is performed three times in order to assure the experiment's repeatability then the results are averaged.

Results and discussion

Before starting the measurements, the calibration procedure with base fluids like 25/75%, 45/55%, and 55/45% EG-water mixture at temperatures ranging from 10 °C to 60 °C are conducted. The comparison between the experimental data with the data from ASHRAE handbook [22] are made. Maximum deviation between experimental measurements and results of ASHRAE handbook [22] is 2.4%. The variation of viscosity of Fe nanoparticles dispersed in different base fluids versus the temperature at different volume concentrations is shown in fig. 2. The results disclose that the viscosity of nanofluid increases with increase in volume concentration and decreases with increase in temperature. The absolute viscosity of EG-water mixture (25/75 wt%) based nanofluids is 1.21 times and 1.72 times higher compared to base fluid at volume concentration of 0.25% and 2%, respectively (T = 10 °C). The absolute viscosity of EG–water mixture (45/55 wt% and 55/45 wt%) based nanofluid at 2% volume concentration is 1.75 and 2.14 times compared to the base fluid. The results show that the viscosity of Fe nanofluids decreases with increasing temperature. This is due to the decrease in the viscosity of the base fluid at higher temperatures. The similar trend of viscosity enhancement with the effect of temperature for EG-water mixture based nanofluids is observed by other researchers [4, 28, 30].

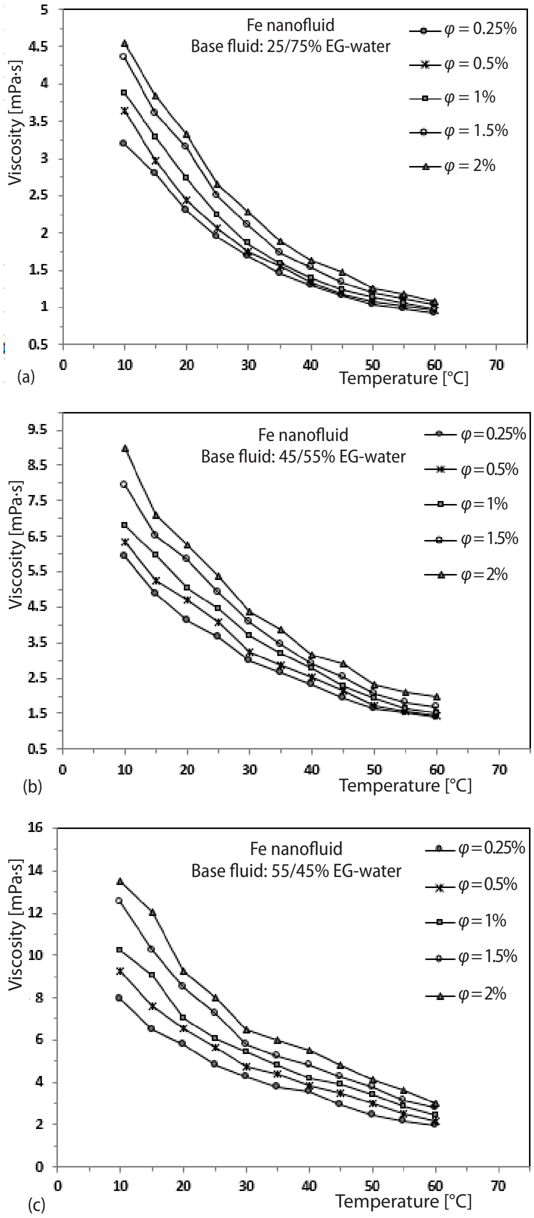

Figure 2. Viscosity of Fe nanoparticles in (a) 25/75%, (b) 45/55%, and (c) 55/45% EG-water *vs.* temperature at different volume concentrations

Figure 3 shows that the viscosity ratio of EG–water mixture (25/75 wt.%) based nanofluid *vs*. temperature at different volume concentrations. It can be seen from this figure that the viscosity ratio of nanofluid decreases with an increase in temperature. At higher concentrations, the change in viscosity ratio as temperature increases from 10 °C to 60 °C is maximal. This could be due to the agglomeration or clinging of the nanoparticles with each other at higher concentrations. The measured viscosity value of Fe nanoparticles in EG-water nanofluids versus volume concentration at two different temperatures of 30 °C and 40 °C are shown in fig. 4. It is observed

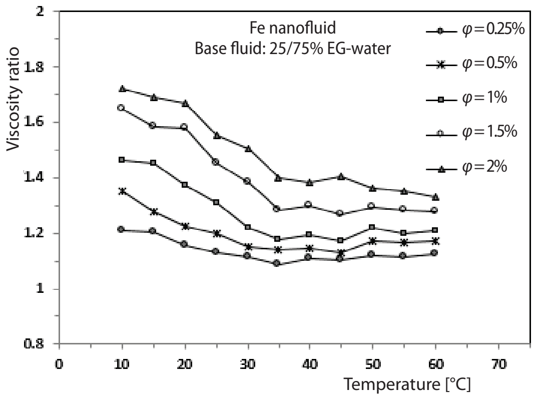

Figure 3. The viscosity ratio of Fe nanoparticles in (25/75 wt.%) EG-water mixture *vs*. temperature at different volume concentrations

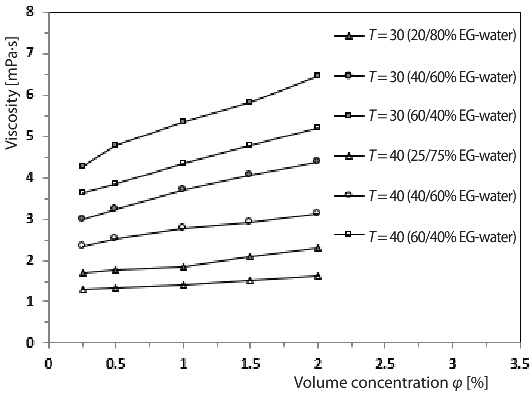

Figure 4. The viscosity of Fe nanoparticles in different base fluids *vs*. volume concentration at T = 30 and 40 °C

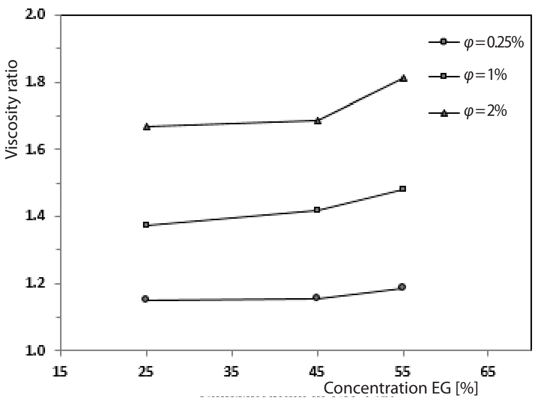

Figure 5. The viscosity ratio of Fe nanofluids *vs*. concentration of EG mixed with water at φ = 0.25, 1, and 2%

that with an increase in concentration level, the viscosity of nanofluids increases. More number of the nanoparticles comes into contact with the base fluid by increasing volume concentration of nanofluid. The increment of total surface area in contact with the base fluid leads to more resistance to the movement of the base fluid molecules, thereby increasing the viscosity of the nanofluid. Figure 5 displays the viscosity ratio of nanofluids versus percentage of ethylene glycol (EG%) mixed with water. It can be clearly seen that the viscosity ratio of nanofluids increases with an increase in percentage of ethylene glycol (EG%) mixed with water. The viscosity of nanofluid is dependent on the viscosity of base fluid. By increasing the viscosity of base fluid, the stability of nanofluid is increased [33]. This leads to an enhancement in the viscosity of nanofluids. The change in the viscosity of nanofluids can cause the increase of pressure drops and affect the performance of nanofluid-engineered systems. Therefore, the mixture of EG-water (25/75 wt.%) is more helpful to use as a base fluid.

Theoretical models for the effective viscosity

Several empirical correlations have been introduced in order to predict the effective viscosity of nanofluids. As demonstrated in previous works, the classical models such as Einstein [34], Brinkman [35], and Batchelor [36] failed to estimate the effective viscosity of nanofluids [25, 30].

Corcione [37] proposed a model to calculate the viscosity of nanofluids in wide regions of validity (the ranges of the nanoparticle diameter, volume concentration and temperature are 25-200 nm, 0.01-7.1% and 20-50 °C) by curve fitting many experimental data points. This formula is defined as:

$$\frac{\mu_{nf}}{\mu_{bf}} = \frac{1}{1 - 34.87\left(\frac{d_p}{d_f}\right)^{-0.3} \varphi^{1.03}} \tag{1}$$

where d_f is the equivalent diameter of a base fluid molecule, given by:

$$d_f = 0.1\left(\frac{6M}{N\pi\rho_{fo}}\right)^{1/3} \tag{2}$$

in which M is the molecular weight of the base fluid, N – the Avogadro number, and ρ_{fo} – the mass density of the base fluid determined at temperature of 20 °C (T_0= 20 °C).

Sundar *et al.* [30] proposed a correlation by suspending Fe_3O_4 nanoparticles in ethylene glycol and water mixture at the volume concentration range of 0% to 1%, as follows:

$$\frac{\mu_{nf}}{\mu_{bf}} = (1+\varphi)^{0.68} \tag{3}$$

The viscosity ratio of the theoretical models and present correlation versus volume concentration at temperature of 30 °C is shown in fig. 6. The classical models consider only volume concentration and the viscosity of base fluid as a variable. The Corcione [37] and Sundar *et al.* [30] models give better prediction of the viscosity of nanofluids compared with the classical models. This is because these models consider the effects of more parameters. The classical models do not estimate the viscosity of nanofluid precisely with the effect of temperature [30]. So, an empirical correlation containing two dimensionless parameters are derived for the viscosity ratio () by using the multiple regression method based on the experimental results, as follows:

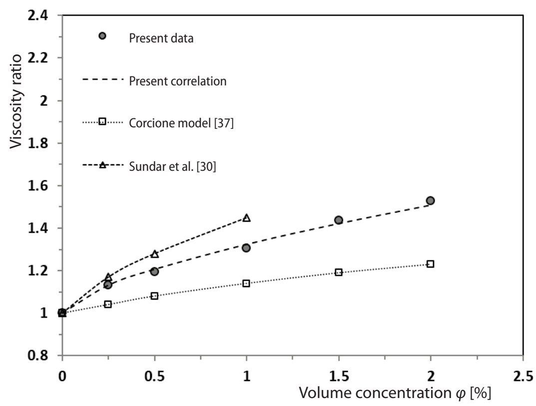

Figure 6. Comparison between the viscosity ratios of Fe nanoparticles in a mixture of EG-water (25/75 wt.%) and the predicted values from current correlation and previous models (*T* = 30°C)

$$\frac{\mu_{nf}}{\mu_{bf}} = 1 + a\varphi^b\left(\frac{T}{T_0}\right)^c \tag{4}$$

where φ is the particle volume fraction ($0 < \varphi < 2\%$), T – the temperature of nanofluid ($10 < T <$ 60 °C, T_0= 10 °C) and μ_{bf} is the viscosity of EG-water mixture (25/75%, 45/55%, and 55/45%). Coefficients a *a*, *b,* and, *c* are calculated from experimental data, which are listed in tab. 1.

Figure 7 shows a comparison between experimental viscosity of nanofluids with those obtained from presented correlation. A good agreement is observed between experimental data and presented equation at various volume concentrations and temperatures. The present model is helpful to determine the

Table 1. Coefficients of correlation for different base fluids

Base fluid	Coefficient			Average error
	a	*b*	*c*	
25/75% EG-water	10.44	0.6523	–0.5124	1.96 %
45/55% EG-water	11.49	0.7002	–0.0719	2.79 %
55/45% EG-water	20.14	0.7639	–0.0921	2.92 %

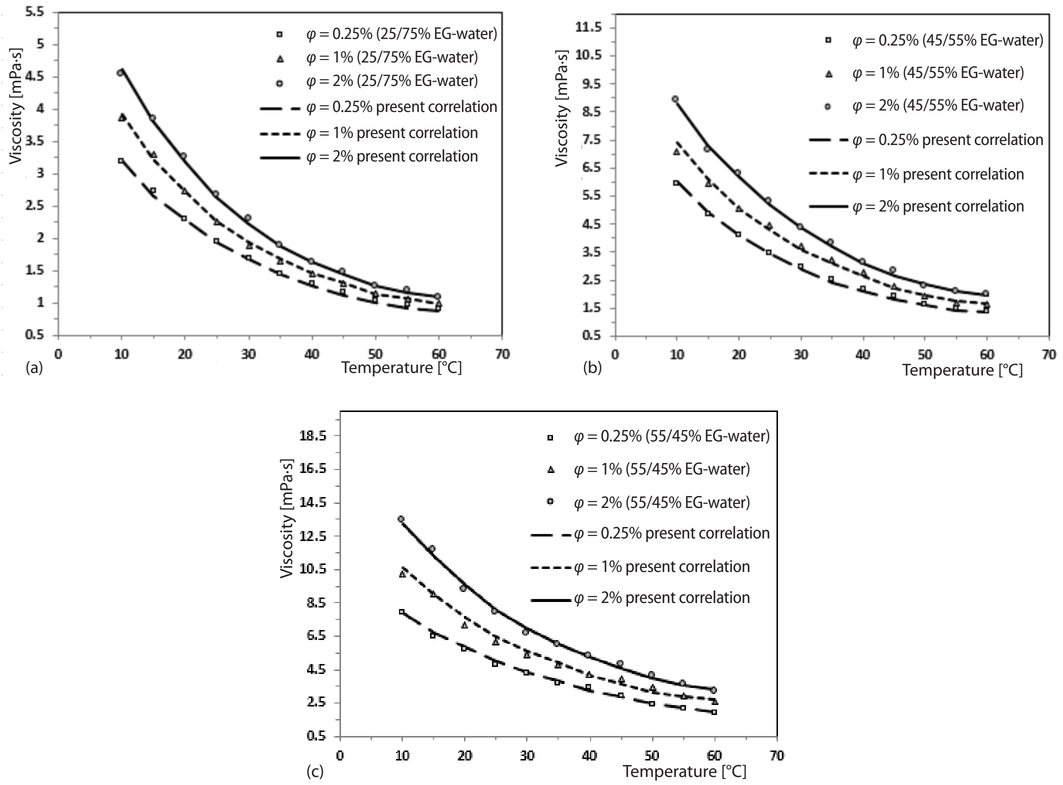

Figure 7. Comparison of the experimental viscosity of Fe nanoparticles in a mixture of EG-water with the presented correlation at three different volume concentrations of 0.25, 1, and 2%

viscosity of Fe nanofluid at specific range of volume concentration and temperature within the range of the present study.

Conclusions

In present study, the viscosity of Fe nanoparticles dispersed in ethylene glycol and water mixture is measured in the volume concentration range of 0% to 2% and temperature range of 10-60 °C. The experimental results show that the viscosity of nanofluids increases with increase in volume concentration and decreases with increase in temperature. The influence of concentration of ethylene glycol mixed with water on the viscosity of nanofluid is discussed. The viscosity of 25/75% EG-water based nanofluid is 1.72 times, 45/55% EG-water based nanofluid is 1.75 times and 55/45% EG-water based nanofluid is 2.14 times compared to the base fluid at 2% volume concentration. It is observed that the viscosity ratio of nanofluids increases with an increase in percentage of ethylene glycol (EG%) mixed with water. The mixture of EG–water (25/75 wt.%) based nanofluid exhibits lower viscosity enhancement compared to others and more helpful to use as a base fluid. The classical models fail to determine the viscosity of nanofluids. Finally, a new empirical correlation is proposed to predict the viscosity of nanofluids.

Nomenclature

T – temperature, [°C]
wt. – weight, [%]
DLS – dynamic light scattering, [–]
K – thermal conductivity, [$Wm^{-1}K^{-1}$]

D – diameter, [mm]
d_p – particle diameter, [nm]

Greek symbols

μ – viscosity, [mPa·s]
φ – volume concentration, [%]

Acronyms

EG – ethylene glycol
TEM – transmission electron miscroscopy

Subscripts

bf – base fluid
nf – nanofluid
eff – effective
max – maximum

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