An experimental study on rheological behavior of hybrid nanofluids made of iron and copper oxide in a binary mixture of water and ethylene glycol: Non-Newtonian behavior

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ABSTRACT
This paper includes an experimental study on rheological behavior of hybrid nanofluids made of iron (Fe) and copper oxide (CuO) in a binary mixture of water and ethylene glycol. Nanofluid samples, with solid volume fractions of 0.05, 0.1, 0.25, 0.5, 1 and 1.5%, were prepared by dispersing an equal volumes of Fe and CuO nanoparticles in a binary mixture of water and ethylene glycol with the proportion of 20–80 vol. %. Viscosity measurements were performed at temperatures ranging from 25 °C to 50 °C and the shear rate range of 3.669–122.3 s⁻¹ for all samples. Experimental findings revealed that the low concentration samples showed Newtonian behavior, while the high concentration samples had non-Newtonian behavior and followed the power law model. Moreover, the power law index and consistency index were obtained using curve-fitting on experimental shear stress-shear rate dependency. The curve-fitting results revealed that the power law index reduced to 0.36, indicating that the high concentration samples possessed shear-thinning behavior at all temperature considered.

1. Introduction
A binary mixture of water and ethylene glycol (EG) is frequently employed as an antifreeze working fluid in very cold regions. Depending on the percentage of water and EG in the mixture, it can withstand temperatures down to −50 °C without any frost. Accordingly, it can be used in many thermal systems such as solar collectors and heat exchangers. Despite the great advantage, the thermal conductivity of the binary mixture is very low [1]. According to many researchers claim, the addition of nano-sized solid materials to this mixture, called nanofluids, can improve its thermal conductivity [2–10]. These studies revealed that the thermal conductivity of nanofluids is a function of size, type and concentration of nanoparticles.

It is obvious that suspending nano-sized solid materials to the liquids varies their rheological behavior. It is a very important issue in calculating the pumping power and estimating the convective heat transfer rate. Therefore, it seems that the consideration of the rheological behavior of nanofluids is essential for the thermal and fluid science and applications. Numerous studies have been conducted on the rheological behavior of nanofluids. The researchers showed that the viscosity of nanofluids increasing with enhancing concentration of solid particles and decreasing temperature [11–15]. In order to evaluate the viscosity of nanofluids, Hojat et al. [16] measured the viscosity of various nanofluids including Al₂O₃, TiO₂ and CuO nanoparticles dispersed in aqueous solution of Carboxy Methyl Cellulose (CMC). Their measurements showed that the base fluid as well as all the suspensions exhibited non-Newtonian (shear-thinning) behavior. Cabaleiro et al. [17] studied the effect of TiO₂ nanoparticles on the rheological behavior of ethylene glycol. They examined the viscosity of the nanofluid in the temperature range of 10–50 °C for nanoparticle mass concentrations up to 2.5%. Their experiments under various shear rates revealed that the nanofluid had non-Newtonian behavior. Phuoc et al. [18] investigated the viscosity of nanofluids containing multi-walled carbon nanotubes (MWNTs). They used MWNTs to enhance or reduce the base fluid viscosity. Their results revealed a reduction up to 20% in the viscosity-reduction case. They also observed a non-Newtonian behavior in the viscosity-enhancement case. Examination of the dynamic viscosity of single-wall carbon nanotubes (SWCNTs) in ethylene glycol was performed by Baratpour et al. [19]. They measured the viscosity of the nanofluid samples with solid volume fractions up to 0.1% at temperatures ranging from 30 °C to 60 °C. Their findings showed that nanofluid samples behaved as Newtonian fluid. Their results
also revealed that the viscosity of the nanofluid increased to 3.18 times that of the base fluid.

Recently, hybrid nanofluids, made of two different types of nanoparticles in a base fluid, have been attracted the attention of some researchers [20–24]. Regarding the viscosity of hybrid nanofluids, Afrand et al. [25] presented an experimental study on the effects of temperature and concentration of SiO$_2$–MWCNTs hybrid particles on the dynamic viscosity of engine oil (SAE40). Their experiments were performed in the solid volume fraction range of 0–1.0% and temperatures ranging from 25°C to 60°C under different shear rates. Experimental findings showed that the nanofluid samples were Newtonian. Furthermore, their results showed that the maximum enhancement of viscosity of the hybrid nanofluid was 37.4%. Afrand et al. [26] also examined the effects of Fe$_3$O$_4$–Ag hybrid nanoparticles on the rheological behavior of ethylene glycol. They measured the viscosity at different shear rates (12.23–122.3 s$^{-1}$) under temperatures ranging from 25°C to 50°C. Their results demonstrated that the nanofluid samples with solid volume fractions of less than 0.3% had Newtonian behavior, while those with higher solid volume fractions (0.6% and 1.2%) had non-Newtonian behavior. The effect of SiO$_2$–MWCNTs hybrid particles on the viscosity of binary mixture of water and ethylene glycol with proportion of 50–50 vol.% was experimentally investigated by Eshgarf and Afrand [27]. They examined the rheological behavior of the nanofluid samples with solid volume fractions ranging from 0.0625% to 2% in the temperature range of 27.5–50°C. Viscosity measurements under different shear rates showed that the base fluid was Newtonian, while nanofluid samples exhibit a pseudoplastic rheological behavior.

In the recent years, heat transfer and flow of magnetic fluids under various external magnetic fields have been attracted the attention of researchers. They reported the effects of the magnetic field on fluid flow and heat transfer rate [28–34]. However, the literature review showed that few studies focus on the effect of hybrid ferromagnetic nanoparticles on rheological behavior of a binary mixture of water and ethylene glycol. Therefore, as regards Newtonian or non-Newtonian behavior of nanofluids plays an imperative role in thermal and fluid flow applications, for the first time, the evaluation of the rheological behavior of hybrid nanofluids made of iron (Fe) and copper oxide (CuO) in a binary mixture of water and ethylene glycol with the proportion of 20–80 vol.% is presented.

2. Experimentation

2.1. Preparation of nanofluids

Stable samples, with volume fractions of 0.05, 0.1, 0.25, 0.5, 1 and 1.5%, were prepared by suspending an equal volume of Fe and CuO nanoparticles (50:50 vol.% in a specified amount of the binary mixture of EG-water (80:20 vol.%). The characteristics of Fe and CuO nanoparticles are presented in Table 1. Characterizations of both nanoparticles were performed by using XRD as shown in Fig. 1. The average size of nanoparticles is obtained by using the data of XRD image (bruker-D8 Germany) and Debye–Scherrer equation [35]:

\[ d = \frac{0.9 \lambda}{h} \cos \theta \]  

where \( d \) is particle diameter size, \( \lambda \) is wave length of X-ray (0.1541 nm), \( h \) is full width at half maximum and \( \theta \) is the diffraction angle (from Fig. 1).

Based on the known percentage of solid volume fractions, the required masses of Fe and CuO nanoparticles for preparing 600 ml nanofluid samples were calculated from Eq. (2) [36] and presented in Table 2.

\[ \phi = \left( \frac{\rho_{\text{Fe}}}{\rho_{\text{Fe}}} + \frac{\rho_{\text{CuO}}}{\rho_{\text{CuO}}} + \frac{\rho_{\text{Water}}}{\rho_{\text{Water}}} \right) \times 100 \]  

In Eq. (2), \( \phi \) is the percentage of solid volume fraction, \( \rho \) is the density in kg/m$^3$, and \( w \) is the mass in kg.

In the present work, magnetic stirring should not be used, because the iron nanoparticles have a magnetic property. In order to make stable nanofluid, the solutions were exposed to an ultrasonic processor (Hielscher Company, Germany) with the power of 400 W and frequency of 24 kHz for 5–6 h. No particle sedimentation was observed up to 15 days. A summary of the process of preparing nanofluid samples is shown in Fig. 2.

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**Table 1** Characteristics of Fe and CuO nanoparticles.

<table>
<thead>
<tr>
<th>Characteristic</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Iron (Fe)</td>
</tr>
<tr>
<td>Purity</td>
<td>&gt;99.5%</td>
</tr>
<tr>
<td>Color</td>
<td>Black</td>
</tr>
<tr>
<td>Size</td>
<td>35–45 (nm)</td>
</tr>
<tr>
<td>Morphology</td>
<td>Spherical</td>
</tr>
<tr>
<td>Bulk density</td>
<td>0.45 (g/cm$^3$)</td>
</tr>
<tr>
<td>True density</td>
<td>7.90 (g/cm$^3$)</td>
</tr>
<tr>
<td>Specific surface area (SSA)</td>
<td>8–14 (m$^2$/g)</td>
</tr>
</tbody>
</table>

**Table 2** Required masses of Fe and CuO used for the preparing a volume of 600 ml of hybrid nanofluids.

<table>
<thead>
<tr>
<th>Volume fraction of samples (%)</th>
<th>Mass [±0.001] (g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>IRR</td>
<td>Fe</td>
</tr>
<tr>
<td>0.05</td>
<td>1.185</td>
</tr>
<tr>
<td>0.1</td>
<td>2.370</td>
</tr>
<tr>
<td>0.25</td>
<td>5.925</td>
</tr>
<tr>
<td>0.5</td>
<td>11.850</td>
</tr>
<tr>
<td>1</td>
<td>23.700</td>
</tr>
<tr>
<td>1.5</td>
<td>35.550</td>
</tr>
</tbody>
</table>
2.2 Measurement of viscosity

In this work, in order to measure the nanofluids viscosity a Brookfield DV-I PRIME digital Viscometer was employed. To keep the samples at a constant temperature during viscosity measurements, the Viscometer was equipped with a temperature bath. Measurements were performed at the temperatures of 25, 30, 35, 40, 45 and 50 °C. In order to distinguish the Newtonian or non-Newtonian behavior, conducting tests were repeated at the shear rate range of 3.669–122.3 s⁻¹ for each sample. The accuracy of Brookfield Viscometer is ±1% of full scale range (FSR). FSR could be obtained by Eq. (3),

$$\text{FSR} = \text{TK} \times \text{SMC} \times \frac{10,000}{\text{RPM}}$$

where TK, SMC and RPM are torque constant, spindle multiplier constant and rotational speed respectively.

According to the operating instructions of device, for determining the measurements error 1% of measured viscosity should be added to 1% of FSR.

3. Results and discussion

3.1 Validation of measurements

Before measuring the viscosity of nanofluid samples, we tried to determine the accuracy of the experiments by measuring the viscosity of the base fluid at different temperatures. For this purpose, a comparison of the viscosity of the base fluid was made between the results obtained by the Viscometer and those presented in ASHRAE [1]. As presented in Fig. 3, the experimental findings are in good agreement with the ASHRAE data, and there is a little difference (maximum 5.01%) at all temperatures considered.

3.2 Viscosity of low concentration samples: Newtonian behavior

Fig. 4 displays the viscosity and shear stress versus the shear rate for low concentration samples (0.05% and 0.1%) at different temperatures. The results illustrate a slight decrease in the viscosity of the samples with increasing shear rate. This behavior is due to shear heating considerations occurring in high shear rates. Therefore, the viscosity of these samples can be considered independent of shear rate. It can also be found from Fig. 4 that there is a linear relationship between shear stress and shear rate. This means that low concentration samples have Newtonian behavior. However, it can be observed that by increasing the concentration of nanoparticles, the viscosity of nanofluid is increased, while it decreases with increasing temperature. In fact, the more nanoparticles into the base fluid would increase its viscosity as a result of the interactions between the nanoparticles and the base fluid. By increasing the amount of nanoparticles, nano-clusters may be formed due to van der Waals forces between the nanoparticles. The nano-clusters barricade the movement of layers of the fluid on each other, leading to a higher augmentation in viscosity. Moreover, with augmenting temperature, the intermolecular interactions between the molecules become weak and consequently the viscosity reduces.

3.3 Viscosity of high concentration samples: Non-Newtonian behavior

Fig. 5 shows the viscosity and shear stress versus the shear rate for high concentration samples (0.25, 0.5, 1 and 1.5%) at different temperatures. The results demonstrate a considerable decrease in the viscosity of the samples with increasing shear rate. As an example, at the solid volume fraction of 1.5%, for an increase in shear rate from 3.669 s⁻¹ to 24.46 s⁻¹, the viscosity decrease...
approximately 57, 59, and 70%, at the temperatures of 30, 40 and 50°C, respectively. These variations are very important for engineering applications such as pumping power and convective heat transfer. In fact, this behavior may be caused by the arrangement of nanoparticles at high shear stresses. With increasing shear stress the nanoparticles arrangement becomes more disciplined as a result of fluid on each layer becomes easier and viscosity decreases. According to this figure, it can be claimed that the viscosity of high concentration samples is dependent on shear rate, consequently non-Newtonian behavior. Since the viscosity decreases with increasing shear rate, it can be found that the samples possess shear-thinning behavior. Therefore, the high concentration samples follow the power law model expressed in Eq. (4) with a power law index of less than unity ($n < 1$).

$$\tau = \frac{m \dot{\gamma}^n}{\rho}$$  \hspace{1cm} (4)

where $\tau$, $\dot{\gamma}$ denote the shear stress (mPa) and the shear rate (1/s), respectively. Moreover, $m$ is consistency index (mPa·s$^n$) and $n$ is the power law index.

By inspection of Fig. 5, it can also be observed that by increasing the solid volume fraction, the viscosity becomes more dependent on shear rate because of the complex interactions between the base fluid and hybrid nanoparticles. Moreover, by comparing Figs. 4 and 5, it can be found that for an increase in the solid volume fraction from 0.05% to 1.5%, the viscosity increases extremely (approximately 2200%).

3.4. Power law index and consistency index

Based on Eq. (4) and using curve-fitting, the power law index ($n$) and consistency index ($m$) have been obtained for each sample at different temperatures. Fig. 6 shows the power law index as a function of the temperature for all samples. It can be seen that, for low concentration samples (0.05 and 0.1%), the power law index is close to 1, which confirms the Newtonian behavior of low concentration samples as shown in Fig. 4. It can be found from Fig. 6 that the power law index decreases with increasing the solid volume fraction, which means that non-Newtonian behavior becomes important for high concentration samples as shown in Fig. 5. Fig. 6 also reveals that power law index is almost constant with temperature changes. In fact, non-Newtonian behavior of samples is not dependent on temperature.

Fig. 7 displays the consistency index of all hybrid nanofluid samples as a function of the temperature. Results illustrate that the consistency index of nanofluids increases with increasing the solid volume fraction, which means that non-Newtonian behavior becomes important for high concentration samples as shown in Fig. 5. Fig. 6 also reveals that power law index is almost constant with temperature changes. In fact, non-Newtonian behavior of samples is not dependent on temperature.

Fig. 7 displays the consistency index of all hybrid nanofluid samples as a function of the temperature. Results illustrate that the consistency index of nanofluids increases with increasing the solid volume fraction, which means that non-Newtonian behavior becomes important for high concentration samples as shown in Fig. 5. Fig. 6 also reveals that power law index is almost constant with temperature changes. In fact, non-Newtonian behavior of samples is not dependent on temperature.
Fig. 5. Viscosity and shear stress versus shear rate for high concentration samples at different temperatures.
In order to evaluate the accuracy of the curve-fitting, the comparison of shear stress obtained by predicted m and n with that obtained by experiments for all samples is illustrated in Fig. 8. It can be observed that most points are near the equality line or on it. This figure shows that there is an excellent agreement between experimental data and the results obtained by m and n.

4. Conclusion

In this work, the rheological behavior of Fe-CuO/EG-water hybrid nanofluid at temperatures ranging from 25°C to 50°C for different samples with the solid volume fractions of 0.05, 0.1, 0.25, 0.5, 1% and 1.5% has been experimentally investigated. The results illustrated that with increasing the concentration of nanoparticles, the viscosity of all nanofluid samples increased, while it decreased with increasing temperature. Viscosity measurements at various shear rates revealed that the low concentration samples had Newtonian behavior, while the high concentration samples possess shear-thinning behavior. Moreover, a comparison between the viscosities of various nanofluid samples showed that for an increase in the solid volume fraction from 0.05% to 1.5%, the viscosity increases approximately 2200%. The power law index and consistency index were obtained by curve-fitting on experimental findings. The curve-fitting results for high concentration samples demonstrated that non-Newtonian behavior became important for high concentration samples (n = 0.36). Results also revealed that non-Newtonian behavior of samples was not dependent on temperature. Furthermore, results of consistency index illustrated that it increased with increasing the solid volume fraction and decreased with increasing temperature.

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References


