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ORIGINAL RESEARCH PAPER

Investigation of Viscosity and Rheological Properties of Copper/Ethylene Glycol Nanofluid

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ABSTRACT: In recent years, significant attention has been devoted to nanofluids to improve the thermal efficiency of conventional cooling fluids. Copper nanoparticles are a proper candidate for this purpose due to their high thermal conductivity. In this study, stable copper nanoparticles with a 34.5 nm average diameter were synthesized via chemical reduction without an inert environment. The synthesized copper nanoparticles and also commercial copper nanoparticles with a 40 nm average size were used in ethylene glycol as the base fluid. Viscosity and rheological behavior of these nanofluids as important factors for assessment of flow behavior in heat exchange equipment were also investigated experimentally. The effects of volume fraction and temperature on nanofluid viscosity were investigated. Viscosity was measured in a 29.5-60 °C temperature range at low weight fractions of 0.0001, 0.0003, and 0.0005. The results were compared with the proposed models for the prediction of nanofluid viscosity, suggesting a correlation. The results show the Newtonian behavior of both nanofluids. Based on the results of a previous study, the heat transfer coefficient and thermal conductivity increased significantly (38.2 % for 0.03 wt. % nanofluid at Re=68 and 39.4% for 0.01 wt. %, respectively). Also, for both cases, nanofluid viscosity was smaller than the base fluid (for nanofluid B, 12.8% reduction at 1.06 vol. %). These results suggest copper nanofluid as an appropriate alternative for application in heat exchange equipment.

KEYWORDS*: Nanofluid Viscosity, Heat Transfer Coefficient, Ethylene Glycol, Copper nanoparticle, Chemical reduction*

INTRODUCTION

Today, improvement of the heat transfer rate of conventional cooling fluids is an essential task for heat exchange equipment. To this end, addition of nanoparticles to the base fluid is considered as a solution. Nanofluid viscosity is altered by the addition of nanoparticles and varies as a function of temperature, nanoparticle size and volume fraction [1]. Also, analysis of viscosity and rheological behavior of nanofluids is important to evaluate the required pumping power [2-6].

The entrance region exhibited a maximum increase in heat transfer, which decreases with an increase in the distance from the inlet. When x/D increased from 63 to 173 at Re=1600, it decreased from 47% to 14%.

Many studies have focused on the viscosity of various nanofluids [7-16]. Yang et al. [17], dispersed 50 nm copper nanoparticles in viscoelastic surfactant solutions and investigated the thermal conductivity and viscosity showing non-Newtonian behavior. Kwak et.al [8], added prolate spheroid shape CuO nanoparticles to EG and investigated the thermal conductivity and viscosity and reported the shear thinning behavior. Ruhani et al. [18], investigated the viscosity of silica nanofluid (20-30 nm) in an EG/water mixture and developed a model for this nanofluid. They reported the enhancement of nanofluid viscosity by increasing the nanoparticle volume fraction. Irani et al. [19], prepared a hybrid nanofluid in water/EG by adding MWCNTs and alumina nanoparticles showing pseudo-plastic behavior. Sahooli and Sabbaghi [20], added CuO nanoparticles to Ethylene Glycol (EG) /water and investigated the thermal properties which showed the enhancement of viscosity by increasing the nanoparticle content and also shear thinning behavior.

EG is used as a cooling fluid in car radiators [21]. Very few studies have been performed on the viscosity of copper-EG nanofluids. For example, Garg et al. [22], prepared Cu-EG nanofluid with a 200 nm size distribution showing an increase in viscosity by nanoparticles addition. Yu et al. [23], added copper nanoparticles with an average size of 10 nm to EG and a significant increase in viscosity was reported. Li et al. [24], investigated the sonication time and temperature effect on the Cu-EG nanofluid with a 50 nm average diameter evaluating the optimal sonication time required for stabilization.

In this study, sodium hypophosphite was used as the reducing agent in the chemical reduction for the synthesis of stable copper nanoparticles. EG-based copper nanofluid was

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prepared using both synthesized and commercial Cu nanoparticles. The viscosity and also shear stress in terms of shear rate were investigated at various nanoparticle concentrations.

While the majority of previous studies on the rheological behavior of nanofluids have shown an increase in the viscosity compared with the base fluid, the results of this study showed a decrease in viscosity at different temperatures by adding copper nanoparticles. Based on the outcomes of this study, a correlation is proposed.

MATERIALS AND METHODS

The synthesis details through chemical reduction and also XRD characterization of the product are given in a previous study by the authors [25].

Figure1 shows the SEM micrograph (EM3200 of KYKY Co.) of the synthesized copper nanoparticles. The size of particles was obtained from Scherer equation as 34.5 nm. The SEM micrograph shows agglomerated spherical copper nanoparticles. Agglomeration may be enhanced by surface oxidation resulting in the enhancement of electrostatic attraction since despite stability in the detection limits of XRD analysis, a thin surface oxide layer is always expected [26, 27]. Also, spherical copper nanoparticles (40 nm) were purchased from US Research Nanomaterials for comparison of the results. Mass density and purity of the product were reported as 0.21 g/cm^3 and 99.9%, respectively. XRD and TEM analyses of this product can be found elsewhere [28].

Two-step route was also implemented for nanofluid synthesis. Copper nanoparticles were dispersed in the base fluid using sonication (200 Watt for 30 min). Experiments were performed using nanofluids from (a) 34.5 nm synthesized copper nanoparticles (nanofluid A) and (b) 40 nm commercial copper nanoparticles (nanofluid B). The stability of synthesized nanofluid was acceptable; thus during viscosity measurement no surfactant addition was required.

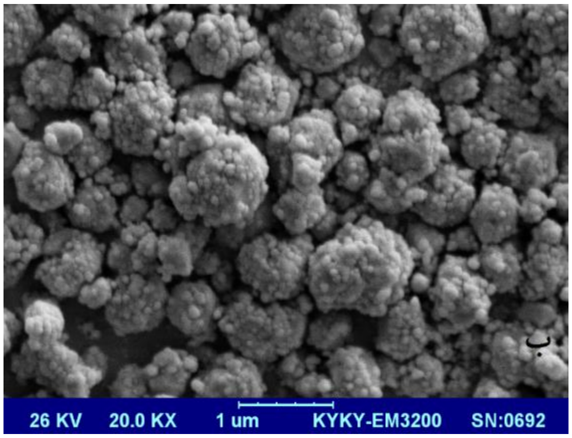


Fig. 1. FE-SEM micrograph of the as-synthesized copper nanoparticles

Figure 2 shows the images taken just after sonication and after 1 and 4 days, respectively. Based on the results, a portion of nanoparticles were settled down after 4 days. Viscosity measurement of the nanofluid was performed using a rotational viscometer (Brookfield model DV-E).

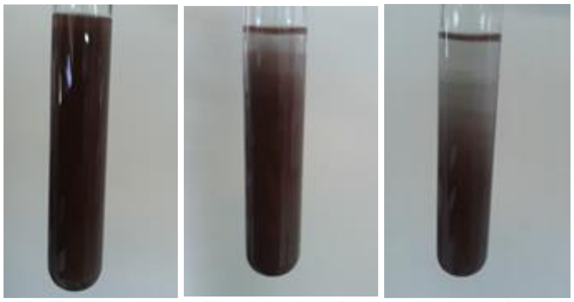


Fig. 2. 0.05 wt. % nanofluid B **a)** just after sonication, **b)** after 24 h and, **c)** after 4 days.

RESULTS AND DISCUSSION

To ensure the accuracy of the viscometer results, viscosity of pure EG was measured at different temperatures and compared with the standard results in the references. As Figure 3 shows the obtained data are consistent with standard viscosities values.

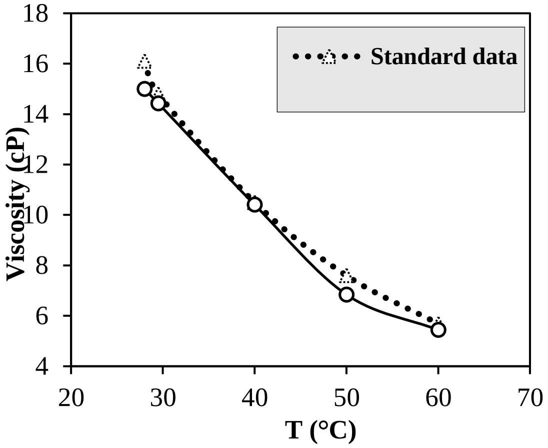


Fig. 3. Comparison of the measured viscosity of pure EG with standard data.

VISCOSITY MEASUREMENT FOR NANOFLUID A

The previous study on heat transfer coefficient and thermal conductivity of Cu-EG nanofluid approved a significant improvement in comparison with pure EG [25]. Based on this study, thermal conductivity enhancement was 39.4% for the 0.01 wt. % nanofluid. Also, the highest heat transfer coefficient improvemnt was reported as 38.2 % for the 0.03 wt. % nanofluid at Re=68.

Figure 4 shows the viscosity of nanofluid A as a function of nanoparticle content. It seems that spherical copper nanoparticles weaken the intermolecular forces; thus nanofluid viscosity is reduced compared with the base fluid.

Also, the viscosity results from classical models of Einstein [29] and Brinkman [30] are plotted and compared. A significant difference between the experimental data and these classical models can be observed. According to these results, the synthesized copper nanofluid can be considered as an appropriate option in cooling systems because the significant increase in thermal efficiency of EG at low nanoparticle content is accompanied by viscosity reduction.

The results are in contradiction with many nanofluids, where viscosity is reported to be enhanced by adding nanoparticles [8, 9, 31-36].

Figure 5 shows shear stress based on shear rate of this nanofluid at 2.3 and 3.6 vol. %. This nanofluid shows Newtonian behavior with a linear variation. Addition of nanoparticles to a Newtonian base fluid results in a nanofluid with Newtonian behaviour [2].

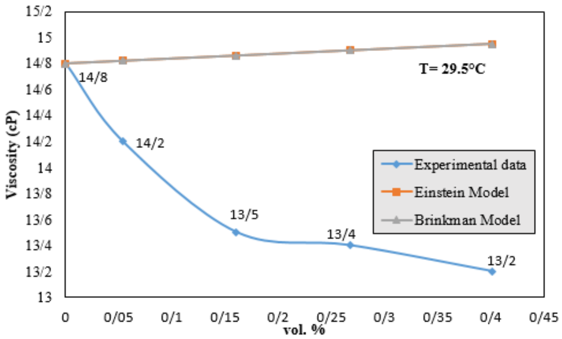


Fig.4. Viscosity vs. nanoparticle vol. % at ambient conditions for nanofluid A.

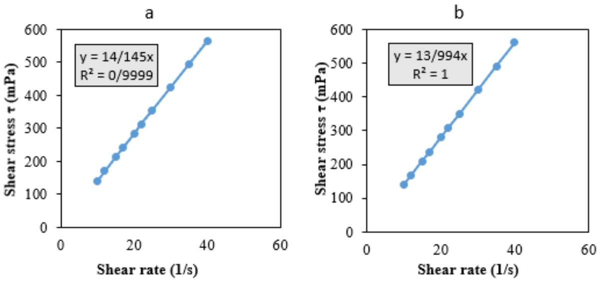


Fig.5. Shear stress based on shear rate of nanofluid A at vol. % of a) 2.3% b) 3.6 %.

VISCOSITY INVESTIGATION FOR NANOFLUID B

Effect of Temperature on the Viscosity of Nanofluid B

Nanofluid B was prepared using 40 nm copper nanoparticles at 0.01, 0.03 and 0.05 weight percent. The viscosity was measured at the room temperature (29.5 ^{0}C), 40, 50 and 60 ^{0}C. Figure 6a shows the variations of EG viscosity with temperature compared with the 0.05 wt. % nanofluid. Figure 6b shows the variations of 0.01 wt. % and 0.03 wt. % samples as a function of temperature. Viscosity is reduced with temperature due to the weakening of intermolecular and inter-particle forces [19, 20]. The nanofluid volume fraction was calculated using nanoparticles weight fraction (w) based on the following equation, where ρ_f and ρ_p are the nanofluid and nanoparticles bulk density, respectively [37]:

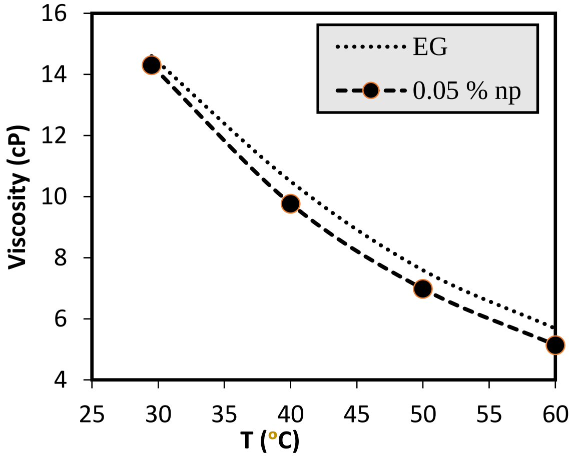
$$\varphi = \frac{w\rho_f}{\rho_p(1-w)+w\rho_f} \quad (1)$$

The equivalent volume fractions based on this equation is given in Table 1:

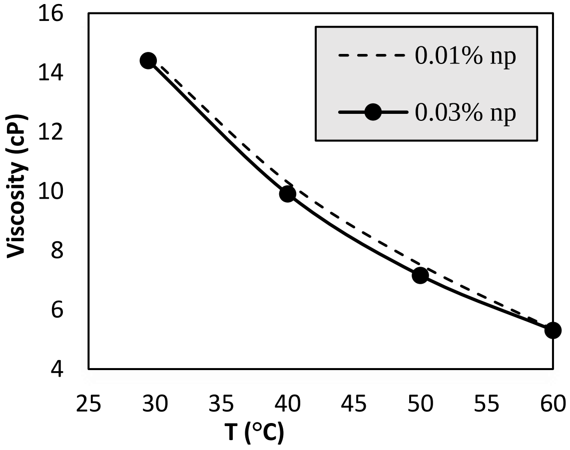
Table 1

Nanoparticle weight and volume percentage and equivalent volume fraction.

Nanoparticle wt. %	Nanoparticle volume fraction	Nanoparticle vol.%
0.01	0.00054	0.054
0.03	0.0016	0.16
0.05	0.0027	0.27



(a)



(b)

Fig.6a) Variation of EG viscosity with temperature compared with the 0.05 wt. % sample of nanofluid B. b) Variations of 0.01 wt. % and 0.03 wt. % samples of nanofluid B with temperature.

For a specific volume fraction, nanofluid viscosity data as well as the viscosity estimations derived by classical models and also a general model provided by Masoumi et al. [38] were plotted as a function of temperature in Fig.7. Based on this figure, for a specific nanoparticle content, the predicted viscosities from existing models are consistent to the experimental data.

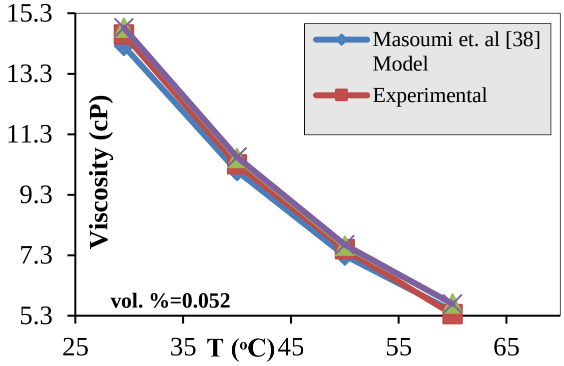


Fig.7. Viscosity variation of nanofluid B with temperature comparing to the existing models.

Effect of nano-Cu content on the viscosity of nanofluid B

In Figure 8, viscosity of nanofluid B and also the predicted viscosity by classical models of Einstein [29], Brinkman [30] and Batchelor [39] and the general model presented by Masoumi et al. [38] are plotted as a function of copper volume fraction. Based on this figure, nano-copper addition to EG (at 29.5 ^{0}C) results in the reduction of viscosity. Increasing nanoparticle concentration leads to 12.8 % reduction at a nanoparticle content of 1.06 vol. %. Similar trends are also obtained at other temperatures (Fig. 9), so that at 0.27 vol. % viscosity reduction is obtained as 6.9, 8.1 and 9.5% at 40, 50 and 60 ^{0}C, respectively. This is opposed to the results predicted by classical models regarding suspensions and microfluids which predict viscosity enhancement at similar conditions.

This is worth mentioning that in classical models, the rheological behavior of suspensions are given at specific conditions including low concentrations and neglecting inter-particle interactions [4, 40].

One possible reason for this reduction can be the very low content of nanoparticles used in this study that seems to facilitate the movement of fluid layers on each other. It seems that at low nanoparticle contents, nanofluid viscosity could be decreased relative to the base fluid, as Chen et al. [41] reported the lower nanofluid viscosity compared with the base fluid by adding MWCNTs to water below 0.4 vol. %. Also, Suganthi et al. [3], investigated the ZnO-PG nanofluid which showed reduced nanofluid viscosity compared to the base fluid at 0-2 vol. % with a 35-40 nm nanoparticle size range. The rheological behavior of 3 nanofluids of CuO, γ-Al_2O_3 and TiO_2 in a carboxymethyl cellulose solution in water were investigated by Hojjat et al. [2]. They reported the relative viscosity to be less than unity for the nanofluid in case of 0.1 and 1 vol. % for CuO nanoparticles and also 0.1 vol. % for TiO_2. The reported results for the effect of volume fraction, size, temperature, etc. on nanofluid viscosity show incomparable trends and contradictions in the results of even similar nanofluids [5, 10].

For example, in case of ZnO-EG nanofluid a significant improvement in viscosity is reported by adding nanoparticles in some studies [14, 43], while reduction is reported in another study [44]. The reasons for these inconsistencies can be attributed to issues such as temperature differences, nanoparticle synthesis techniques and varying nanoparticle contents [45].

Another major reason could be the stabilizer and surfactants present in the nanofluid which are not mentioned in detail in many cases [1, 23, 44, 46]. Also, contradictions can originate from the difference in nanoparticle size, since larger nanoparticles or agglomerates result in higher viscosities [44, 47].

It is also worth mentioning that conventional viscometers are not designed for nanofluids which may lead to inaccurate results. Also, Dynamic flow leads to nanoparticle collision and collision viscosity which is not measurable by conventional viscometers [48].

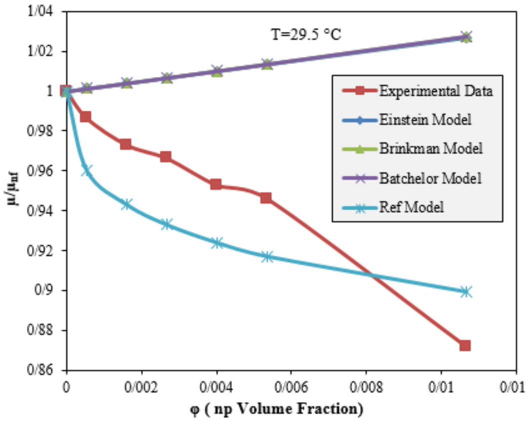


Fig.8. Viscosity variations of nanofluid B vs. nanoparticle content at 29.5^{0}C.

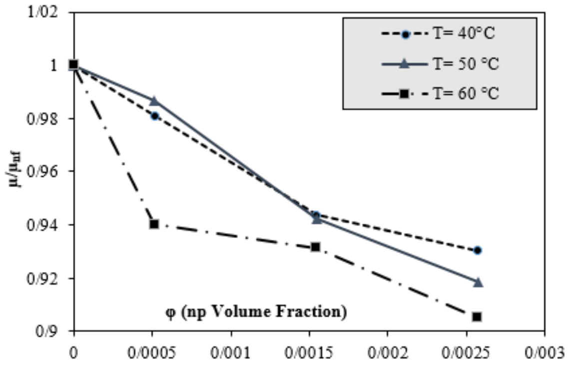


Fig.9. Viscosity variations of nanofluid B vs. nanoparticle content at 40-60 °C

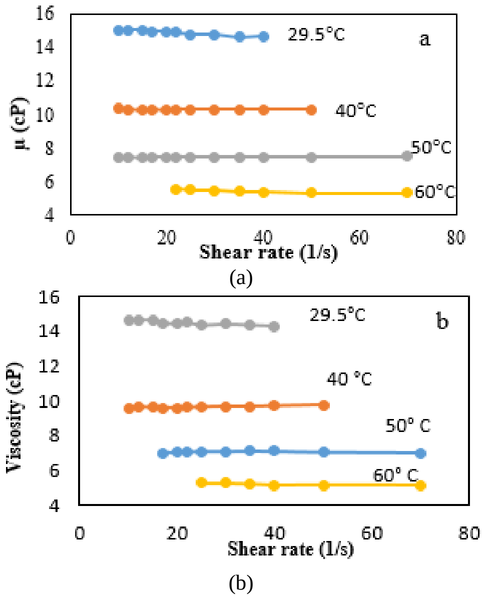
As shown in Figure 8, the general model presented by Masoumi et al. [38], unlike the classical models, has been able to ascertain the trend of viscosity for this nanofluid and also the results match the experimental data well. The important result of Figs. 7 and 8 is that classical models could well predict the temperature effect on nanofluid viscosity with a specific concentration (Fig. 7). However, in case that nanoparticle volume fraction is varied (Fig. 8), there is a difference between the experimental results and models, which suggests that classical models are unable to estimate the effect of volume fraction on nanofluid viscosity.

While the model presented by Masoumi et al. [38], is considered to be much precise due to taking into account the Brownian motion and the relative velocity between the nanoparticles and the base fluid. The proposed relationship for the prediction of viscosity for this nanofluid with 40 nm copper nanoparticle, obtained using curve fitting is suggested in Eq. 2:

$$\mu/\mu_{\mathrm{nf}} = 1 - 8.8\varphi - 234\varphi^2 \tag{2}$$

Rheological behavior of nanofluid B

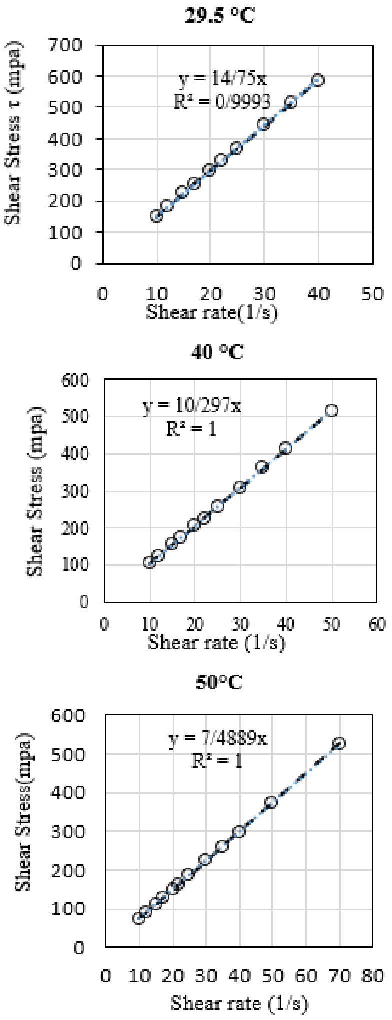
The viscosities of Newtonian fluids like diluted oils are constant with the variation of shear rate. Also, for Newtonian fluids, shear stress has a linear trend vs. shear rate [42]. Fig. 10 shows viscosity variations vs. shear rate for 0.01 and 0.05 wt. % nanofluids, respectively. Based on the results, viscosity is not dependent on shear rate for all temperatures indicating Newtonian behavior in both cases. Also, Figure 11 shows curves of shear stress based on shear rate for the 0.01 wt. % nanofluid which also approves the Newtonian behavior.



(a)

(b)

Fig.10. Rheological behavior of a) 0.01 wt. % b) 0.05 wt. % nanofluid B.



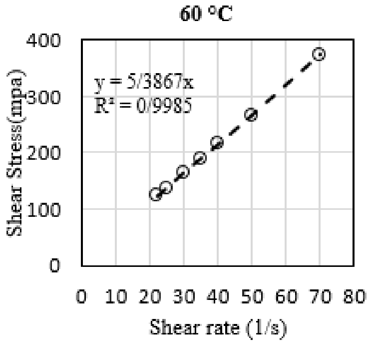


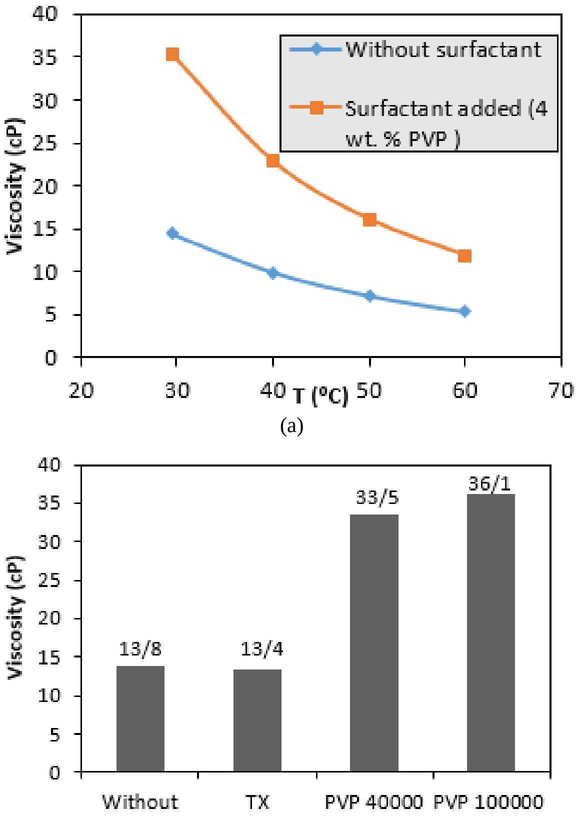
Fig.11. The curves of shear stress based on shear rate for 0.01 wt. % nanofluid B.

Effect of stabilizer on nanofluid viscosity

In order to investigate the stability, TX100 and PVP with two molecular weights of 10^5, 4×10^4 were added to nanofluid B to enhance its stability. The optimal concentration for PVP 10^5 and TX100 are determined as 4 wt. % and 0.12 wt. %, respectively. Figure 12a shows the variations of 0.03 wt. % samples containing 4 wt. % PVP 10^5 as a function of temperature. Based on the results, this surfactant has caused a significant increase in nanofluid viscosity. Increasing temperature from 29.5 to 60^{0}C resulted in the reduction of viscosity from 35.4 to 11.9 cP. Also, temperature increase results in the fading of viscosity difference between the samples with and without stabilizer. Based on Figure12b, PVP (4×10^4 and 10^5) results in the enhancement of viscosity 2.4 and 2.6 times compared with the original nanofluid, respectively due to the long hydrocarbon chain; which is uneconomical from power supply point of view. However, TX100 as a nonionic surfactant soluble in EG has reduced the viscosity by 2.9%. Also, stability is obtained at much lower contents which is favorable.

One reason for the discrepancy between some of the reported results for nanofluid viscosity is the stabilizer type and content. Based on the results, the stabilizer added to nanofluid can reduce or enhance the nanofluid viscosity; which can be a major reason for the difference in the results of similar nanofluids in different studies.

Studying the viscosity of nanofluids suggest that, due to the fact that the mechanisms and theories are still not fully understood, there is still no comprehensive model for predicting the viscosity and rheological behavior of nanofluids.



(a)

(b)

Fig.12.a) Viscosity of 0.03 wt. % samples of nanofluid B based on temperature with and without stabilizer. b) Effect of stabilizer on nanofluid viscosity at 30^{0}C.

CONCLUSION

The special thermal properties of copper nanoparticles suggest copper nanofluid as a potential candidate for application in areas such as cooling equipment in electronics, radiators and heat exchangers. Stable copper nanoparticles with 34.5 nm size were synthesized by chemical reduction in a non-inert environment and used to prepare nanofluid A. Also, nanofluid B was prepared using commercial 40 nm copper nanoparticles. The previous investigation of thermal properties of nanofluid A reported 38.3 % increase for heat transfer coefficient at 0.03 wt. % and also 39.4% enhancement for thermal conductivity at 0.01 wt. % nanofluid. In both cases, viscosity measurements indicated reduction compared to the base fluid and for 1.06 vol. % of nanofluid B 12.8% reduction was reported. This reduced viscosity of copper nanofluid in EG, beside the thermal properties reported in a previous investigation, suggests copper nanoparticles as a convenient option for the preparation of cooling nanofluids especially for EG as the base fluid.

In both cases, the rheological investigations of the synthesized nanofluids showed Newtonian behavior. The experimental data were compared to the models and the results show that classical models cannot predict the effect of nanoparticle content on nanofluid viscosity. A viscosity correlation in terms of the nanoparticles volume fraction for this nanofluid was suggested. Based on the results, surfactants can affect the nanofluid viscosity which can greatly increase or even in some cases reduce the viscosity. It can be concluded that the addition and type of surfactants has a major effect on the reported viscosity of nanofluids and can be largely responsible for the contradictions reported in various papers.

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