Determination of Rheological Behavior of Aluminum Oxide Nanofluid and Development of New Viscosity Correlations

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Abstract

Experimental investigations have been carried out to study the rheological behavior of aluminum oxide nanofluid. Nanoparticles with average particle size of 53 nm were dispersed in a base fluid of 60% (by mass) of ethylene glycol and water. Nanofluids of volumetric concentrations 1 to 10% were tested for determining the viscous properties. It was found that this nanofluid behaved as nonnewtonian at lower temperatures (-35°C to 0°C) and newtonian at higher temperatures (0°C to 90°C). The data showed that the viscosity increases with an increase in concentration and decreases with increase in temperature. Two new correlations were developed expressing viscosity as a function of temperature and concentration.

Keywords: nanofluids, aluminum oxide, viscosity, ethylene glycol, particle concentration, temperature dependency.

1. Introduction

Nanofluids are mixtures of solid nanoparticles with average particle size smaller than 100 nm dispersed in base fluids such as water, ethylene glycol or propylene glycol. Research on nanofluids has received great attention in the last decade due to the prospect of enhanced thermal properties. For example, Eastman et al. (2001) have reported a 40% increase in thermal conductivity of ethylene glycol when copper nanoparticles of 3% volumetric concentration were added to it. Pak and Cho (1998) have shown that at a fixed Reynolds number, convective heat transfer coefficient of an Al₂O₃ nanofluid of
volume concentration 2.78% increases by 75%. Such results have motivated researchers to explore the thermal and rheological properties of nanofluids.

Heating industrial and residential buildings in cold regions requires a great deal of energy. In such severe cold climatic conditions, aqueous mixtures of ethylene or propylene glycol in different volumetric concentrations are typically used to lower the freezing point of the heat transfer medium [McQuiston et al., 2000]. Such heat transfer fluids, which can operate effectively at very low temperatures, are used in building heating systems, automobiles and heat exchangers in industries. It is found that at low temperatures, aqueous mixtures of ethylene glycol have better heat transfer characteristics than propylene glycol [ASHRAE, 2005] and an aqueous mixture of 60% ethylene glycol (by mass), referred to as 60:40 EG/W in this paper provides the freeze protection down to very low temperatures. For this reason, this fluid is most commonly used in the sub-arctic and arctic regions of the world.

Since a very limited amount of data is available on the EG/W based nanofluids, we studied the rheological characteristics of aluminum oxide (Al₂O₃) nanofluids. This research will help understand the viscous behavior of nanofluids, which is crucial for successful application of nanofluids in cold regions. Xuan and Li (2003) have reported conspicuous enhancement of the heat transfer coefficient in nanofluids of low particle concentration without much penalty in pressure loss. Namburu et al. (2008) developed a viscosity correlation for Al₂O₃ nanofluid in EG/W in the temperature range of –35°C to 50°C. However, in building heating applications fluid temperatures as high as 90°C are employed. Therefore, their correlation is inadequate for building heating systems, which motivated the present research for viscosity measurements of Al₂O₃ nanofluids with different volumetric concentrations and at higher temperatures. Rheological characteristics of Al₂O₃ nanofluids of concentrations ranging from 1 to 10% in a 60:40 EG/W base fluid were investigated over a temperature range of –35°C to 90°C for their effective usage in cold regions.

Pumping power requirements and convective heat transfer coefficients of fluids depend strongly on the Reynolds and Prandtl numbers, which in turn, are highly influenced by viscosity. Thus, accurate determination of viscosity of fluids is very important in thermal applications. However, research on
nanofluid viscosity at very low temperatures encountered in sub-arctic and arctic regions are extremely limited. Correlations for nanofluid viscosity attempt to express viscosity as a function of temperature and particle volumetric concentration. Some of the presently available correlations are discussed below.

For suspensions with particle concentrations below 5% Einstein (1956) proposed a viscosity correlation

$$\mu_s = \mu_f \left(1 + \frac{5}{2} \phi \right)$$

(1)

Another correlation for higher concentrations was given by Brinkman (1952)

$$\mu_s = \mu_f \frac{1}{(1-\phi)^{0.5}}$$

(2)

Similar correlation relating viscosity with concentration was proposed by Bicerano et al. (1999)

$$\mu_s = \mu_f (1 + \eta \phi + k_H \phi^2 + \ldots \ldots)$$

(3)

where $\eta$ is the virial coefficient and $k_H$ is Huggins coefficient.

In Eqs. (1)-(3) $\mu_s$ = suspension viscosity, $\mu_f$ = viscosity of base fluid and $\phi$ = particle volumetric concentration.

It should be noted that in all the three correlations mentioned above, the suspension viscosity is expressed as a function of particle concentration and temperature does not appear exclusively as a variable. However, it is well known that viscosity of liquids is a strong function of temperature. White (1991) proposed a correlation including temperature dependence of viscosity for pure fluids ($\mu_f$)

$$\ln \frac{\mu_f}{\mu_0} \approx a + b \left( \frac{T_0}{T} \right) + c \left( \frac{T_0}{T} \right)^2$$

(4)

In Eq. (4) $\mu_0$, $T_0$ are reference viscosity and temperature (absolute), respectively. The parameters $a$, $b$ and $c$ are dimensionless curve-fit constants, which depend on the type of fluid considered. For example, in the case of pure water $a = -2.10$, $b = -4.45$ and $c = 6.55$. 
The Andrade’s equation cited by Reid et al. (1987) is an exponential correlation between the viscosity of fluids and their temperature

$$\mu_f = Ae^{B/T}$$ (5)

where $A$ and $B$ are curve-fit parameters.

Yaws presented a viscosity correlation valid for many industrially important chemical liquids

$$\log(\mu_f) = A + \frac{B}{T} + CT + DT^2$$ (6)

where, $A$, $B$, $C$ and $D$ are curve-fit parameters unique to a liquid.

In a study of copper oxide (CuO) nanoparticles suspended in water and for a temperature range of 5–50°C, Kulkarni et al. (2006) proposed a correlation

$$\ln(\mu_{nf}) = A\left(\frac{1}{T}\right) - B$$ (7)

In Eq. (7), $\mu_{nf}$ is the nanofluid viscosity and curve-fit parameters $A$ and $B$ are expressed as functions of the nanoparticle concentration ($\phi$). Since the above correlation was developed for an aqueous base fluid, it is not applicable at sub-zero temperatures.

Namburu et al. (2008) developed a viscosity correlation for various concentrations (1-10%) of Al$_2$O$_3$ nanofluids in a temperature range of -35°C to 50°C for a 60:40 EG/W base fluid

$$\log(\mu_{nf}) = Ae^{BT}$$ (8)

where $A$ and $B$ are expressed as polynomial functions in nanoparticle concentration ($\phi$). However, this correlation is not applicable for temperatures above 50°C. Upon careful inspection it was found that this correlation may have deviations in the range of ±30% from the measured viscosity values. Therefore, the present study aimed at developing new correlations for a broader temperature range extending up to 90°C, which is typical of fluids used in building heating. This will help develop the next generation of heat transfer fluids applicable in cold regions.

2. Experimental setup and procedure
The original nanofluid procured from Alfa Aesar (2007) is a 50% (by mass) of Al₂O₃ nanoparticle dispersion in water. The average particle size is 53nm and the particle density is 3.6 gm/cc. Nanofluid samples of different particle volumetric concentrations (1, 2, 4, 6, 8 and 10%) were prepared by adding exact amount of ethylene glycol and water to the original nanofluid with a precision mass balance of 0.1mg accuracy. In the newly prepared samples, 60:40 EG/W was the base fluid. Subsequently, the sample was placed in an ultrasonic agitator for a minimum of 90 minutes to ensure uniform dispersion of the nanoparticles.

The experimental setup for measuring the rheological property of Al₂O₃ nanofluids is shown in Figure 1. The setup consists of an LV DV-II+ Brookfield programmable viscometer (Brookfield, 1999) and a Julabo temperature-controlled bath. For different spindle combinations the viscometer has the ability to measure viscosities in the range of 1.5–30,000 cP [1 cP = 1 mPa.s]. The test fluid, whose viscosity is to be measured, is placed inside the sample chamber. The motor of the viscometer rotates a spindle immersed in the test fluid. Viscous drag of the fluid against the spindle due to rotation is measured by a sensitive calibrated spring attached to the spindle. Temperature of the test fluid is controlled between –35°C to 90°C by a programmable computer connected to the Julabo temperature bath. During viscosity measurements, sample temperature is recorded by a Resistance Temperature Detector (RTD) sensor attached to the sample chamber. For a specified spindle type, the viscosity measurements are more accurate when rotational speed combinations produce a torque above 10%. Computer-1 connected to the viscometer, records the data through the WINGATHER® software (Brookfield, 1999), which includes rotational speed of the spindle (RPM), torque (%), viscosity (cP), shear stress (dyne/cm²), shear strain rate (1/s), temperature (°C) and time duration for which the readings are taken.
Figure 1. Experimental setup for viscosity measurement of nanofluids

**Calibration:** Relationship between the shear strain rate (\( \dot{\gamma} \)) and shear stress (\( \tau \)) for a newtonian fluid is given by the equation

\[
\tau = \mu \dot{\gamma} \tag{9}
\]

where \( \mu \) is the coefficient of viscosity. To verify the accuracy of our experimental setup and procedure, viscosity of the Brookfield Calibration Fluid 10, which is newtonian, was measured. The shear stress and shear strain rates at 13 different rotational speeds of the spindle within the torque limit of 10% - 90% (as recommended by the Brookfield LV-II viscometer manual, 1999) were recorded and the results are shown in Figure 2. A straight line passing through the origin fits the stress-strain rate data very well and therefore, clearly exhibits newtonian behavior. From the slope of the stress-strain rate curve a viscosity value of 9.44cP was recorded using the WINGATHER® software. This measured value is within a very acceptable error limit of 2.6% of the true viscosity provided by Brookfield Engineering Laboratories.
3. Results and discussion

Namburu et al. (2008) had collected viscosity data of Al₂O₃ nanofluid of various concentrations in the temperature range of -35°C to 50°C. In the present research, viscosity measurements were extended to 90°C and new correlations were developed, which gave improved accuracy and applicability over a broader range of temperatures.

It is crucial to determine whether the nanofluid displays newtonian or nonnewtonian behavior with variation of particle concentration and temperature. Figure 3 displays the viscosity versus shear strain rate for Al₂O₃ nanofluid of 10% volumetric concentration over a temperature 238K (-35°C) to 363K (90°C). Because the viscosity changes with shear strain rate from 238K to 273K, the nanofluid behaves as a nonnewtonian fluid in this temperature range. However, beyond 273K, the viscosity remains constant for all values of shear strain rate indicating a newtonian behavior.
Figure 3. Viscosity variation with shear strain rate of Al₂O₃ nanofluid of 10% concentration at various temperatures (238K to 363K).

Plots such as Figure 3 were generated for all concentrations and non-Newtonian behavior was observed in the lower temperature range (238K to 273°C). However, in the higher temperature range (273K to 363K) the fluid behaved as Newtonian.

In order to characterize the non-Newtonian behavior, the stress-strain data in the lower temperature range were plotted to ascertain the rheological behavior of the nanofluid. The shear stress ($\tau$) versus shear rate ($\dot{\gamma}$) plot (Figure 4) of the nanofluid with 10% concentration at 243K shows that the data fits quite well with the characteristic of a Bingham plastic. It is observed that a yield stress ($\tau_y$) is necessary before the fluid starts deforming. The equation for the straight line shown in Figure 4 is

$$\tau = \tau_y + \mu \dot{\gamma}$$

(10)

A yield stress $\tau_y$ of 2.0161 dyne/cm² and a viscosity $\mu$ of 238.08 cP were observed in this case.
Therefore, in the lower temperature range the Al₂O₃ nanofluid behaves as a Bingham plastic. This observation is in agreement with the conclusions made by Macosko and Mendes (1996), where they specify that concentrated suspension of solid particles in newtonian liquids show a yield stress followed by nearly newtonian flow. Similar plots as Figure 4 were created for concentrations of 1, 2, 4, 6 and 8% and the yield stress (the intercept in Figure 4) values increased with concentration and decreased with temperature.

The results of viscosity measurements in the higher temperature range, where the nanofluid is newtonian, are discussed next. The stress-strain behavior of a 10% concentration Al₂O₃ nanofluid at 323K shows a newtonian behavior (Figure 5).
Following this, the stress-strain curves were plotted (Figure 6) for a nanofluid with 4% concentration tested in the higher temperature range. It is confirmed that in this temperature regime, the nanofluid exhibits newtonian behavior. Similar behavior was observed in case of the other concentrations tested.
The influence of particle concentration on viscosity in both the temperature regimes is displayed in Figure 7. It shows that the Al₂O₃ nanofluid viscosity increases with an increase in concentration and it decreases with an increase in temperature. The upper plot shows viscosity variation in the lower temperature regime and the lower plot shows the same for the higher temperature regime.

Figure 7. Variation of viscosity with volumetric concentrations at two distinct temperature regimes

Analysis of the rheological data compiled in Figure 3 through 7 confirmed distinct behaviors in two regimes: one in the lower temperature range and the other in the higher temperature range. Therefore, two corresponding correlations must be developed to correctly describe the viscous behavior of the Al₂O₃ nanofluid.

Correlations developed by Namburu et al. (2008), Kulkarni et al. (2006), Yaws (1997), White (1991) and Andrade’s equation in Reid et al. (1987), were tested to fit the experimental data. However, it was found that none of these correlations fit the data properly. Therefore, by careful statistical analyses an exponential model was derived using the LABFIT® (2008) software to arrive at the best-fit correlations.
for the experimental data. While fitting the data, temperature and concentration were taken as the independent (predictor) variables and viscosity was taken as the dependent (response) variable. This equation fits the data with $R^2 > 0.99$ in both the low and high temperature regimes

$$\mu_{nf} = A e^{(B/T+C\phi)}$$ (11)

where $\mu_{nf}$ is viscosity of the Al$_2$O$_3$ nanofluid in centipoise (cP), $T$ is the absolute temperature in K. The curve-fit parameters $A$, $B$ and $C$ are characteristics of the nanofluid for the given temperature regimes (238K-273K) and (273K-363K). In Eq. (11), particle concentration ($\phi$) is expressed in percent volume of particles in the base fluid and varies from 0 to 10. Table-1 summarizes the curve-fit parameters for the lower and higher temperature regimes. Viscosity of the nanofluids for different particle concentrations and temperatures were computed from the correlation given by Eq (11). The experimental and curve-fitted viscosity values are plotted against temperature for both the low (Figure 8) and high (Figure 9) temperature regimes.

Table 1. Viscosity curve-fit parameters for the low and high temperature regimes

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<tbody>
<tr>
<td>$A$</td>
<td>1.2200 x 10^{-6}</td>
<td>2.3920 x 10^{-4}</td>
</tr>
<tr>
<td>$B$</td>
<td>4285</td>
<td>2903</td>
</tr>
<tr>
<td>$C$</td>
<td>0.1448</td>
<td>0.1265</td>
</tr>
<tr>
<td>$R^2$</td>
<td>0.9984</td>
<td>0.9958</td>
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Unlike the correlations proposed by Kulkarni et al. (2006) and Namburu et al. (2008), where the curve-fit coefficients are dependent on the particle concentration \( \phi \), the new correlations [Eq. (11)] contains constants \( A \), \( B \) and \( C \), which are independent of \( \phi \). In Eq. (11), both \( T \) and \( \phi \) appear in a form similar to the well-known Andrade’s equation for viscosity. Except for high temperatures (353K and 363K), the experimental and curve-fit values agree within a deviation of \( \pm 10\% \). Therefore, the correlations are much simpler and predict viscosity better over a wider range of temperatures. At 363K, the deviation is higher and is attributed to the operational limitation of the viscometer, whose lower limit is 1.5 cP. This error can be reduced by using a more precise viscometer (e.g. the cone-plate viscometer) to obtain very low viscosity values at higher temperatures.

4. Conclusions

1. Aluminum oxide nanoparticles in a 60:40 EG/W base fluid exhibit a nonnewtonian behavior at a lower temperature range of 238K to 273K for all particle concentrations. It behaves as a Bingham
plastic with small yield stress, which decreases with decrease in volumetric concentration and increase in fluid temperature.

2. In the higher temperature range (273K to 363K) the nanofluid behaves as a newtonian fluid.

3. The viscosity of nanofluids increases with an increase in particle concentration. For example, viscosity of 10% concentration aluminum oxide nanofluid is about 4 times the value of that of the base fluid at 238K.

4. Two empirical correlations have been developed to accurately determine the viscosity for two temperature regimes. In both regimes, as the temperature increases, nanofluid viscosity decreases exponentially.

5. The new empirical correlations expressed by Eq. (11) for the Al₂O₃ nanoparticles in a 60:40 EG/W base fluid exhibit an exponential relationship between the viscosity and volume concentration.

References


