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Viscosity of alumina nanoparticles dispersed in car engine coolant

Madhusree Kole, T.K. Dey*

Thermophysical Measurements Laboratory, Cryogenic Engineering Centre, Indian Institute of Technology, Kharagpur 721 302, India

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ABSTRACT

The present paper, describes our experimental results on the viscosity of the nanofluid prepared by dispersing alumina nanoparticles (<50 nm) in commercial car coolant. The nanofluid prepared with calculated amount of oleic acid (surfactant) was tested to be stable for more than 80 days. The viscosity of the nanofluids is measured both as a function of alumina volume fraction and temperature between 10 and 50 °C. While the pure base fluid display Newtonian behavior over the measured temperature, it transforms to a non-Newtonian fluid with addition of a small amount of alumina nanoparticles. Our results show that viscosity of the nanofluid increases with increasing nanoparticle concentration and decreases with increase in temperature. Most of the frequently used classical models severely under predict the measured viscosity. Volume fraction dependence of the nanofluid viscosity, however, is predicted fairly well on the basis of a recently reported theoretical model for nanofluids that takes into account the effect of Brownian motion of nanoparticles in the nanofluid. The temperature dependence of the viscosity of engine coolant based alumina nanofluids obeys the empirical correlation of the type: log (μ_{n-f}) = $A \exp(BT)$, proposed earlier by Namburu et al.

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1. Introduction

Nanoparticles have many remarkable properties because of their small sizes and very large specific surface areas [1]. In recent years, the characteristics and applications of nanosized powders have been studied extensively regarding their preparation and processing techniques [2]. Researchers have applied the emerging nanotechnology in the traditional thermal engineering. For example, metallic or nonmetallic nanoparticles are dispersed into conventional heat transfer fluids such as water, glycol, and oil to make a new class of heat transfer fluids, called nanofluids, having superior properties including high thermal conductivity, long-term stability, and homogeneity [3,4]. Along with the enhanced heat transfer properties, the nanofluids also have unique characteristics associated with mass transfer, wetting and spreading and antibacterial activity.

Most published studies on nanofluids deal with the heat transfer behavior including thermal conduction [3–6], phase change (boiling) heat transfer [7–10], and convective heat transfer [11– 14]. Very few studies, however, have been reported on the rheological behavior of nanofluids. Some review articles [15–17] emphasized the significance of investigating the viscosity of nanofluids. It is believed that viscosity is as critical as thermal conductivity in engineering systems that employ fluid flow. This is because; pumping power is proportional to the pressure drop, which in turn

* Corresponding author. *E-mail address:* tapasdey@hijli.iitkgp.ernet.in (T.K. Dey). is related to fluid viscosity. In laminar flow, the pressure drop is directly proportional to the viscosity. Thus from application point of view, ideal nanofluid should not only posses high thermal conductivity but also should have low viscosity.

Masuda et al. [18] measured the viscosity of suspensions of dispersed ultra-fine TiO₂ particles in water. They found that TiO₂ particles of 27 nm average diameter at a volumetric loading of 4.3% increased the viscosity of water by 60%. Wang et al. [19] reported that the effective viscosity of nanofluid containing 5 volume% Al₂O₃ nanoparticles (28 nm) in distilled water, prepared by mechanical blending technique, is enhanced by about 86%. They also found a 40% increase in viscosity of ethylene glycol at a volumetric loading of 3.5% of Al₂O₃ nanoparticles. Their results indicate that the viscosity of nanofluids depends also on dispersion methods. In contrast, Pak et al. [20] found that for 10 volume% concentration of nanoparticles, the viscosities of Al₂O₃ (13 nm)-water and TiO₂ (27 nm)-water based nanofluids are several times higher than that of water. Pak et al. [20] used adjusted pH values and employed an electrostatic repulsion technique for dispersion of nanoparticles. However, viscosity results [20] were significantly larger than the predictions from the classical theory of suspension rheology [21]. Das et al. [7] and Putra et al. [12] measured the viscosity of Al₂O₃-water and CuO-water based nanofluids as a function of shear rate and showed Newtonian behavior of the nanofluids for a range of volume percentage between 1% and 4%. For Al₂O₃-water nanofluids, Das et al. [7] also observed an increase in viscosity with an increase of particle volume fraction. Prasher et al. [22] reported the viscosity of alumina-based nanofluids for various shear rates,

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temperature, and particle volume fraction. Their data demonstrated that viscosity is independent of shear rate proving that the nanofluids are Newtonian in nature. Temperature and volume fraction dependence of viscosity of SiO₂ based nanofluids were investigated by Namburu et al. [23]. Non-Newtonian behavior of the nanofluids has been observed at sub-zero temperatures. Based on the experimental data an empirical correlation relating the viscosity with particle volume fraction and nanofluid temperature was suggested. Viscosity of ethylene glycol based nanofluids containing titania nanoparticles (8 wt.%) was measured by Chen et al. [24] and Newtonian behavior was seen over a wide shear rate range at temperatures between 293 and 333 K. The observed rheological behavior was explained by aggregation mechanism. A new equation [24], based on Krieger–Dougherty equation, was found to predict the volume fraction dependent viscosity of the nanofluids. Theoretical understanding of viscosity of nanofluids is very limited and hence most of the rheological data on nanofluid is usually compared with some of the well known classical models [21,24,25-27]. Only theoretical model for nanofluid viscosity so far reported is by Masoumi et al. [28], which includes the effect Brownian motion of nanoparticles in the base fluid and the model was shown to predict the viscosity of alumina-water nanofluid. However, the validation of Masoumi's viscosity expression [28] has not been tested yet on other nanofluids.

Engine coolant is one of the most commonly used commercial fluids adopted in automobiles. In the present communication, we report on the synthesis of car engine coolant based nanofluids containing Al₂O₃ nanoparticles and a detailed investigation on their effective viscosity as a function of Al₂O₃ volume concentrations, shear rate and temperature. Results have been discussed both in terms of several classical models and the theoretical prediction for viscosity of nanofluids proposed recently [28].

2. Models for the viscosity of nanofluids

From the theoretical point of view, understanding various properties of nanofluid represents a new challenge to the researchers in fluid dynamics and heat transfer. There exist very few established theoretical formulas that may be used to predict the effective viscosity of nanofluids and most of such models are derived from well known Einstein model [21]. As nanofluid is a two-phase fluid, one may expect that it would have common features with solid–liquid mixtures. However, the question regarding the applicability of these classical models for use in nanofluids still remains doubtful. Some of the widely used models for nanofluids are mentioned below.

Einstein's model [21] can be used for relatively low volume fractions, ($\phi \leq 0.02$), which is given as:

$$\mu_{nf} = \mu_{bf}(1+2.5\phi) \tag{1}$$

where μ_{bf} is the viscosity of the base fluid.

Brinkman [25] extended Einstein formula for use with moderate particle concentration as:

$$\frac{\mu_{nf}}{\mu_{bf}} = (1 - \phi)^{-2.5} \tag{2}$$

Batchelor [26] considered the effect of Brownian motion of particles on the bulk stress of an isotropic suspension of spherical particles and derived the viscosity expression as:

$$\frac{\mu_{nf}}{\mu_{bf}} = 1 + 2.5\phi + 6.5\phi^2 \tag{3}$$

Various recent studies [29–31] suggest that the high viscosity of nanofluids is likely to be associated with the aggregation of nanoparticles for which the Krieger–Dougherty equation [27] has been used by many authors. The Krieger–Dougherty model has a form:

$$\frac{\mu_{nf}}{\mu_{bf}} = \left(1 - \frac{\phi_a}{\phi_m}\right)^{-[\eta]\phi_m} \tag{4}$$

where ϕ_m is the maximum concentration at which flow can occur, ϕ_a the effective volume fraction of aggregates and $[\eta]$ is the intrinsic viscosity, which for monodisperse systems has a typical value of 2.5.

Chen et al. [24] argued that as aggregates do not have constant packing throughout the structure, the packing density may be assumed to change with radial position. In such a situation, ϕ_a may be taken as: $\phi_a = \phi \left(\frac{a_a}{a}\right)^{3-D}$, where, a_a and a are the radii of aggregates and primary nanoparticles, respectively. The term D is defined as fractal index, which for nanoparticles has a typical value of 1.8 [29–31]. Incorporating ϕ_a , Chen et al. [24] modified the Krieger–Dougherty equation as:

$$\frac{\mu_{nf}}{\mu_{bf}} = \left(1 - \frac{\phi}{\phi_m} \left(\frac{a_a}{a}\right)^{1.2}\right)^{-[\eta]\phi_m} \tag{5}$$

A simple expression was proposed by Kitano et al. [32] involving ϕ_m was also used to predict the viscosity of two phase mixture:

$$\frac{\mu_{nf}}{\mu_{bf}} = \left[1 - \left(\frac{\phi}{\phi_m}\right)\right]^{-2} \tag{6}$$

Till recently, there has been no report on the exact theoretical estimation for nanofluid viscosity. Masoumi et al. [28] in 2009 presented a new analytical model to predict nanofluid viscosity considering the Brownian motion of nanoparticles and showed its applicability for Al₂0₃-water system. The expression for the effective viscosity of nanofluid is given as:

$$\mu_{nf} = \mu_{bf} + \frac{\rho_N V_B d_N^2}{72C\delta} \tag{7}$$

In the above expression, V_B , ρ_N and d_N are Brownian velocity, density and diameter of the nanoparticles respectively. $\delta \left(=\sqrt[3]{\frac{\pi}{6\phi}}d_N\right)$ represents the distance between the centers of the particles and *C* is a "correction factor".

3. Experimental

Commercially available engine coolant for automobiles is a halfand-half mixture of propylene glycol and water. The density of engine coolant is determined to be 1.047 g/cc. Engine coolant based nanofluids are prepared with Al₂O₃ nanoparticles (M/S, Sigma–Aldrich, nominal diameter <50 nm, density 4 g/cc) with volume fraction between 0.001 and 0.015. It may be noted that Alumina – Engine coolant based nanofluid could be stabilized only with addition of appropriate amount of oleic acid (surfactant). The solid particles in the coolant are deagglomerated by intensive ultrasonication (Ms Hielscher model UP200S) for 3 h. Finally, the suspension is homogenized for 1 h by magnetic force agitation. The suspension stability of the prepared nanofluid has been tested for 80 days without any trace of visible particle sedimentations (Fig. 1a–e).

Viscosity of the nanofluids is measured by Brookfield programmable viscometer (model: LVDV-II-Pro) connected to a PC controlled Julabo temperature controlled bath which can vary the fluid temperature between -20 and $100 \,^{\circ}$ C and control to $\pm 0.1 \,^{\circ}$ C. The viscometer drives a spindle immersed in nanofluids. Due to rotation of the spindle, a viscous drag of the fluid against the spindle is developed, which is measured by the deflection of the calibrated spring. Model LVDV-II-Pro viscometer has a viscosity measurement range between 1.5 and 30,000 mPa s. For the present study, the Brookfield ultra low (UL) adapter with recommended spindle (model ULA-49EAY, spindle code 00) has been used. UL adapter has provision for cooling/heating fluid circulation. Total



Fig. 1. (a–d) Photographs of nanofluids showing no sedimentation after 80 days. (e) Nanofluid prepared without surfactant. Sedimentation of Al_2O_3 nanoparticles is seen within two hours of preparation of nanofluid.

volume of nanofluid required in UL adapter is ~16 ml. Temperature of the fluid is measured by a calibrated Pt-100 temperature sensor. For the UL adapter, the spindle (code: 00) and speed combinations, the viscometer gives very satisfactory results when the applied torque is between 10% and 100%. The operation of the viscometer and data collection (viz, viscosity, shear stress, shear rate, RPM, torque and temperature) is performed using Wingather[®] software. All the measurements are performed under steady state conditions and the measured value is within 2.5% of the true viscosity. The viscometer has been tested and calibrated with the calibration fluid provided by Brookfield Engineering Laboratories. The schematic of the viscosity measurement set up is shown in Fig. 2.

4. Results and discussion

The viscosity (μ_{bf}) of the engine coolant (base fluid) as a function of shear strain rate $(\dot{\gamma})$ between 10 and 50 °C is shown in

Fig. 3a. It may be seen that over the entire measured temperature range the viscosity of the engine coolant is independent of the shear strain rate, indicating a Newtonian behavior. As car engine coolant is a 50:50 mixture of propylene glycol and water, the present result may be compared with that reported by Namburu et al. [23] for 60:40 ethylene glycol and water, which also behaves as a Newtonian fluid. Similar plots for coolant-alumina nanofluids with varying nanoparticles concentration is given in Fig. 3b-f. It is clear from the Figs. 3b-f that addition of even 0.001 volume fraction of alumina nanoparticles in the coolant, the resultant nanofluid transforms to non-Newtonian fluids. For nanofluids with low loading of Al₂O₃ (viz., 0.001 and 0.004) the fluids display Newtonian behavior only at temperatures higher that \sim 40 °C. However, for nanofluids with higher volume fraction (>0.004) of Al₂O₃, non-Newtonian behavior is evident in the entire measured temperature range of 10-50 °C. Non-Newtonian nature of the present nanofluids at room temperature (30 °C) are further ascertained by plotting the shear stress (τ) vs. shear strain rate ($\dot{\gamma}$) (Fig. 4a). Shear stress (τ) vs. shear strain rate ($\dot{\gamma}$) data of all the present nanofluids fits well with the characteristic equation of a Bingham plastic:

$$\tau = \tau_{\rm v} + \mu \dot{\gamma} \tag{8}$$

where μ is the coefficient of viscosity and τ_y is the yield stress necessary before the fluid starts deforming. The yield shear stress (τ_y) is determined from the intercept of the fitted straight line (Fig. 4a). The yield stress (τ_y) estimated at 30 °C increases gradually from 0.05416 dyne/cm to 0.87898 dynes/cm as Al₂O₃ nanoparticles loading in coolant increases from 0.001 to 0.015 volume fraction. It may be noted that the yield stress (τ_y) follows a power–law relationship with the volume fraction of the added alumina nanoparticles (Fig. 4b). The dependence of τ_y on ϕ is expressed as: $\tau_y = (0.50063)\phi^{1.3694}$ with a correlation factor (R^2) of 0.99678. Similar, power–law dependence of (τ_y vs. ϕ) was also reported by Tseng et al. [33] for BaTiO₃ powders in ethanol–isopropanol mixtures.

Fig. 5a shows the viscosity of the Al_2O_3 nanofluids at various temperatures as a function of Al_2O_3 volume fraction. In general, nanofluid viscosity (μ_{nf}) increases appreciably with increasing nanoparticle loading in the base fluid. This is consistent with the



Fig. 2. Experimental schematic for measurement of nanofluid viscosity between -20 and +100 °C.



Fig. 3. (a) Viscosity vs. shear strain rate ($\dot{\gamma}$) for pure engine coolant. (b)–(f) show plots of viscosity (μ_{nf}) vs. shear strain rate ($\dot{\gamma}$) for engine coolant–Al₂O₃ nanofluids with Al₂O₃ volume fractions between 0.001, 0.004, 0.007, 0.010 and 0.015 at various temperatures between 10 and 50 °C.

fact that increase in nanoparticle concentration in nanofluid increases the fluid internal shear stress, hence the viscosity. Viscosity of the present nanofluid at room temperature as a function of Al₂O₃ nanoparticle volume fraction has been estimated on the basis of the expressions proposed by Einstein (1), Brinkman (2), Batchelor (3), Krieger–Dougherty (4), Chen et al. (5) and Kitano et al. (6). The maximum solid concentration (ϕ_m) is determined from $(1 - \mu_{rel}^{-1/n})$ vs. ϕ plot [34], which generally follows a linear relationship over a wide range of particle concentration. Taking the flow index n (=3), the extrapolation of $(1 - \mu_{rel}^{-1/3})$ to unity gives ϕ_m ,

which for the present nanofluid is estimated to be 0.595. Fig. 5b shows the measured viscosity of the nanofluid as a function of Al_2O_3 nanoparticle volume fraction at 30 °C, along with the viscosity calculated using the above classical models [Eqs. (1)–(6)]. It may be seen that all the above models, including those by Krieger–Dougherty [27] and Chen et al. [24] largely under predict the measured viscosity of the nanofluid. Failure of Krieger–Dougherty equation and its modified version by Chen et al. [24] indicates the absence of aggregation of Al_2O_3 nanoparticle in engine coolant based nanofluid.





Fig. 4. (a) Shear stress (τ) vs. Shear strain rate ($\dot{\gamma}$) for various nanofluids with different volume concentration of Al₂O₃ nanoparticles. Solid lines are the fitted ones according to Eq. (8). (b) Yield stress (τ_y) as a function of volume% of Al₂O₃ nanoparticles in the nanofluid. Line is the fitted power-law equation: $\tau_y = (0.50063)\phi^{1.3694}$.

In a recently published report, Masoumi et al. [28] derived the effective viscosity of nanofluid as: $\mu_{nf} = \mu_{bf} + \mu_{app}$, where μ_{app} is the apparent viscosity arising from the effects of nanoparticles in the fluid. As Reynolds number (R_e) is very small ($\ll 1$) for nanoparticles, Masoumi et al. [28] considered the flow around a nanoparticle in the Stokes regime and used creeping flow solution [35] with a correction factor (*C*). Equating the total shear stress (τ_{total}) over the nanoparticle surface with that derived from "work-energy principle", the apparent viscosity (μ_{app}) was calculated and finally the nanofluid viscosity (μ_{nf}) is expressed as given in Eq. (7). The Correction factor (C) in Eq. (7) is calculated from: $C = \mu_{bb}^{-1}(a\phi + b)$, where *a* and *b* are experimental parameters, which for the engine coolant-Al₂O₃ nanofluid are estimated to be -0.00004 and 7.1274×10^{-7} , respectively. Using Eq. (7), relative viscosity $(\mu_{nf}|\mu_{bf})$ of the nanofluids at room temperature is calculated and is shown in Fig. 6a. Similar estimation of $(\mu_{nf}|\mu_{bf})$ at 10 °C and 45 °C are also shown in Fig. 6b and c, respectively. It may be seen that the viscosity of the nanofluid calculated from Eq. (7) predicts the measured viscosity fairly well. It may also be

Fig. 5. (a) Nanofluid viscosity (μ_{nf}) plotted as a function of Al₂O₃ volume fraction at different temperatures. (b) Points are the measured relative viscosity $\left(\frac{\eta_{nf}}{\eta_{bf}}\right)$ shown as a function of Al₂O₃ volume fraction. The solid lines are the relative viscosity predicted by various classical models.

noted that compared to the classical models, agreement of Eq. (7) to the measured viscosity of the present nanofluids is in general much superior.

Temperature dependence of viscosity of coolant-alumina nanofluid having various volume fractions of alumina nanoparticles is plotted in Fig 7. Viscosity of the nanofluids decreases exponentially with increase in temperature of the nanofluid, presumably due the weakening of inter-particle and inter-molecular adhesion forces. Similar trends have also been observed in other varieties of nanofluids [36,37]. In absence of any proper theoretical formulations to predict the temperature dependence of viscosity, different empirical correlations [23,38-40] have been proposed by several authors (Table 1) depending on the nature and type of nanofluids. Present set of data on temperature dependence of viscosity of coolant-alumina nanofluids, were tested to fit the above correlations. However, except for the correlation proposed by Namburu et al. [23], no other correlations (Table 1) give an acceptable agreement to the temperature dependence of viscosity of the present nanofluids and the curve-fit parameters (A and B) of Eq. (13) is given in Table 2. As shown in Fig. 7, the temperature dependence of the viscosity of the present nanofluids computed using Namburu correlation (13)



Fig. 6. (a – c). Symbols (\bullet) are the measured relative viscosity $\begin{pmatrix} \eta_n \\ \eta_n \end{pmatrix}$ of the nanofluid plotted as a function of Al₂O₃ nanoparticles volume fraction at 10, 30 and 45 °C. Solid lines and symbols (\blacktriangle) are the values predicted by Mausomi et al. [28].

agrees extremely well (maximum deviation: <2%) with the measured viscosity of engine coolant– Al_2O_3 nanofluids. It may be mentioned that the expression derived by Masoumi et al. [28], which explains fairly well concentration dependence of the viscosity of



Fig. 7. Temperature dependence of the nanofluid viscosity (μ_{nf}). Solid lines are the fitted values calculated from equation proposed by Namburu et al. [23].

Table 1

Various correlations proposed for temperature dependence of nanofluid viscosity.

Authors & ref	Correlations	Eq. nos.
White [35]	$\ln rac{\mu_{nf}}{\mu_{bf}} = a + b \Big(rac{T_0}{T} \Big) + c \Big(rac{T_0}{T} \Big)^2$	(9)
Reid et al. [38]	$\mu_{nf} = A \exp(B/T)$	(10)
Yaws [39]	$\log(\mu_{nf}) = A + BT^{-1} + CT + DT^2$	(11)
Kulkarni et al. [40]	$\ln(\mu_{nf}) = AT^{-1} - B$	(12)
Namburu et al. [23]	$\log(\mu_{nf}) = A \exp(BT)$	(13)

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Curve-fit values of the parameters *A* and *B* with correlation factor.

Vol fraction	Α	В	R^2
0.001	1.83442	-0.01345	0.9993
0.004	1.88642	-0.01244	0.9994
0.007	1.98529	-0.01226	0.9995
0.010	1.98752	-0.01128	0.9954
0.015	2.1355	-0.00999	0.9974

the present nanofluid, does not give acceptable agreement to its temperature dependence.

5. Conclusions

Car engine coolant based alumina nanofluids of excellent stability has been prepared. The volume concentration and temperature dependences of their viscosity are investigated. Addition of small amount of alumina nanoparticles transforms the Newtonian behavior of the pure engine coolant to a non-Newtonian fluid and it behaves as a Bingham plastic with small yield stress. Yield stress (τ_y) calculated from the measured shear stress (τ) vs. shear strain rate ($\dot{\gamma}$) data display a power–law dependence on the particle volume fraction (ϕ). An empirical correlation of the type, log (μ_{nf}) = $A \exp(BT)$, accurately explains the observed viscosity temperature dependence. We confirm that the expression derived recently by Masoumi et al. [28], considering the influence of Brownian motion of nanoparticles in the base fluid, predicts fairly well the particle concentration dependence of nanofluid viscosity.

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