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The Rheology of Nanolubricants Based on Fe_2O_3, Al_2O_3, and ZnO Oxide Nanoparticles: A Comparative Study

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This article reports the results of a comparative study of the rheological characteristics of nanolubricants carried out based on Fe_2O_3, Al_2O_3, and ZnO nanoparticles. The base fluid used was 20W40 engine oil. The nanofluid samples were prepared in solid volume fractions (0.25%; 0.50%; 0.75%, and 1%). The measurements were performed at different temperatures, ranging between 20 °C and 60 °C, and at shear rates, ranging from 100 s^{-1} to 300 s^{-1}. The results obtained showed that nanofluids based on Fe_2O_3 and ZnO nanoparticles exhibited a non-Newtonian shear thinning behaviour while the nanofluid based on the Al_2O_3 nanoparticle exhibited a Newtonian behaviour. The Al_2O_3/oil nanofluid had a lower dynamic viscosity than the Fe_2O_3/oil and ZnO/oil nanofluids. The increase in temperature led to a reduction of more than 80% in viscosity for all nanofluids. On the other hand, the increase in the volume concentration of nanoparticles clearly increased the dynamic viscosity.

Keywords: Viscosity, Nanofluid, Rheological behaviour, Nanoparticles

INTRODUCTION

The basic idea of dispersing solid particles in a fluid goes back to Maxwell's work in 1873 [1]. Thirty years later, a new branch of science was born: nanotechnology, which is based on the manipulation of molecules with a nanometric diameter. Since then, nanotechnology has developed rapidly in various areas [2,3], especially nanofluids, which have the potential to contribute to several practical applications, including solar thermal cooling systems, automobile cooling system, electronic cooling, medical fields, detergency, and military systems [4-7]. Nanofluids have thus become a subject of research of immense importance worldwide. Researchers have been mainly interested in studying the thermophysical properties of nanofluid, including thermal conductivity, viscosity, thermal capacity, and density. Among these properties, viscosity is an important property because it indicates the resistance of the fluid to deformation under the effect of shear stress. Indeed, viscosity plays an essential role in the production and use of a product. Many parameters affect the viscosity of nanofluids, including preparation method, type of base fluid, shear rate, temperature, volume concentration, and particle size and shape [8].

The rheological behaviour of liquids is strongly affected by the suspension of nanometric size solid materials, and numerous studies have been conducted in this area. According to the literature, the dispersion of nanoparticles in Newtonian base fluids results in Newtonian behaviour in some nanofluids [9-11] and in non-Newtonian behaviour, mainly shear-thinning, in some other nanofluids [12-16]. The effect of temperature on the viscosity of nanofluids has also been studied, and the results show that the viscosity of nanofluids, similar to that of other fluids, decreases with increasing temperature [17-20]. Viscosity is affected not only by temperature but also by nanoparticle fraction.

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Numerous studies have demonstrated that an increase in the viscosity of nanofluids is linked to the addition of nanoparticles to the base fluid [21,22]. Furthermore, studies have shown that adding even small amounts of nanoparticles, especially at low shear rates and low temperatures, can transform the Newtonian behaviour of the base fluid into non-Newtonian behaviour [23]. To assess the viscosity of nanofluids, Atashrouz *et al.* [24] measured the viscosity of an ethylene glycol-water mixture-based Fe_3O_4 nanofluid, and their results showed that increasing the shear rate led to a reduction in the viscosity (shear-thinning behaviour). Abdul Hamid et al. conducted experimental studies to measure the viscosity of Al_2O_3/water and Al_2O_3/ethylene glycol nanofluids and found an increase greater than 100% compared to the base fluid in relative viscosity with increasing particle concentration [25]. Esfe et al. also measured the viscosity of MWCNT (30%)-Al_2O_3 (70%)/5W50 nanofluid as a function of shear rate, and their results revealed that the viscosity of nanofluid decreased with an increase in temperature and increased with an increase in volume fraction, indicating the non-Newtonian behaviour of nanofluids [26]. Hojjat *et al.* measured the viscosity of various nanofluids, in particular nanoparticles of Al_2O_3, TiO_2, and CuO dispersed in an aqueous solution of Carboxy Methyl Cellulose (CMC). Their results showed that base fluid as well as all suspensions exhibited a non-Newtonian behaviour (shear-thinning) [27]. Also, Das *et al.* measured the viscosity of Al_2O_3/water nanofluids as a function of shear rate and reported that viscosity increased with increasing particle concentration, indicating a non-Newtonian behaviour of nanofluids [28].

Mixing nanoparticles with the base liquid alters the viscosity of the nanofluid, thus affecting its rheology. Hence, understanding the rheological properties of nanofluids is of critical importance for the further development of their practical applications. For instance, the enhancement of the rheological properties of nanolubricants can improve the performance of engine lubrication system. As a result, various methods have been developed to improve the capability of lubricants for various applications. One of the methods suggested in this regard is the use of nanoparticles in lubricants. However, few comparative studies have been conducted to investigate the effects of adding nanoparticles on the rheological behaviour of engine oil. The objective of this paper is to propose a comparative study of the rheological behaviour of nanofluids based on iron oxide (Fe_2O_3), aluminum oxide (Al_2O_3), and Zinc oxide (ZnO) nanoparticles. To this end, after nanofluids were prepared, rheological measurements were carried out. Then, the variation in the viscosity of nanofluids was examined as a function of shear rate, temperature, and volume concentration, and the rheological behaviours of nanofluids were compared to each other.

EXPERIMENTATION

In this study, nanofluids were prepared using a two-step method. Nanoparticles used in this method were first prepared as dry powders using chemical or physical methods and were then dispersed into the base liquid for the desired nanofluid. The base fluid used was 20W40 engine oil. In total, three nanofluids, namely, Fe_2O_3/oil, Al_2O_3/oil, and ZnO/oil, were prepared with solid volume fractions of 0.25%, 0.5%, 0.75%, and 1%. For this, Equation (1) of the volume fraction was used to determine the weight of various nanoparticles corresponding to each volume fraction.

$$\varphi = \frac{V_{nanopartides}}{V_{nanopartides} + V_{20W40}} = \frac{\left(\frac{W}{\rho}\right)_{nanopartides}}{\left(\frac{W}{\rho}\right)_{nanopartides} + \left(\frac{W}{\rho}\right)_{20W40}} \times 100 \quad (1)$$

In this equation, ρ is density, φ is the solid volume fraction of nanoparticles, and w is the weight of nanoparticles. The amount of nanoparticle calculated was weighed using a precision digital balance and was then added to the base fluid. The mixture was stirred using a magnetic stirrer. Subsequently, to obtain a homogeneous suspension, the mixture was subjected for three hours to ultrasonic homogenization using an ultrasonic homogenizer (1200 w, 20 kHz; Hielscher Ultrasonics GmbH, Teltow, Germany). The preparation of nanofluids by this method made it possible to suppress agglomeration and aggregation of nanoparticles and obtain stable suspensions for up to 72 h. The specifications of nanoparticles provided by Sigma-

Aldrich (USA) are listed in Table 1. The nanoparticles and different samples of nanolubricants are illustrated in Fig. 1. The rheological measurements were carried out using an MCR 302 rotational rheometer (Anton Paar, Graz, Austria). The geometry used was double-gap concentric cylinder geometry. The accuracy and repeatability of the apparatus were ±1.0% and ±0.2%, respectively. The tests were carried out with different shear rates, ranging from 100 s^{-1} to 300 s^{-1}, and at different temperatures, ranging from 20 °C to 60 °C. To minimize repeatability, the experiments were repeated at different temperatures, concentrations, and shear rates, and the average data were calculated.

Table 1. Characteristics of Nanoparticles

	Fe_2O_3	Al_2O_3	ZnO
Purity	≥99%	≥98%	≥99%
Molecular weight (g mol^{-1})	159.69	101.96	81.39
Average particle size (nm)	58	≤58	60
Appearance	Brown	White	White

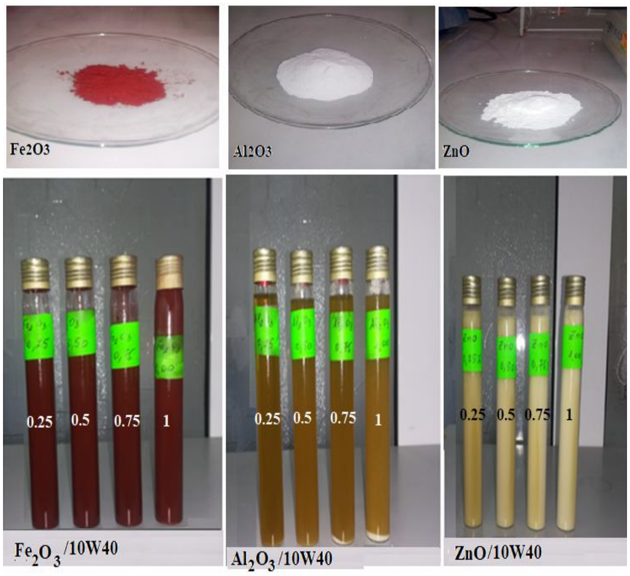


Fig. 1. Fe_2O_3, Al_2O_3, and ZnO nanoparticles and their corresponding nanofluid samples.

RESULTS AND DISCUSSION

XRD Analysis

Figure 2 shows the XRD patterns of Fe_2O_3, Al_2O_3, and ZnO nanoparticles displayed in the range of 0-80°. The average particle size of the nanoparticles was found to be 58 nm.

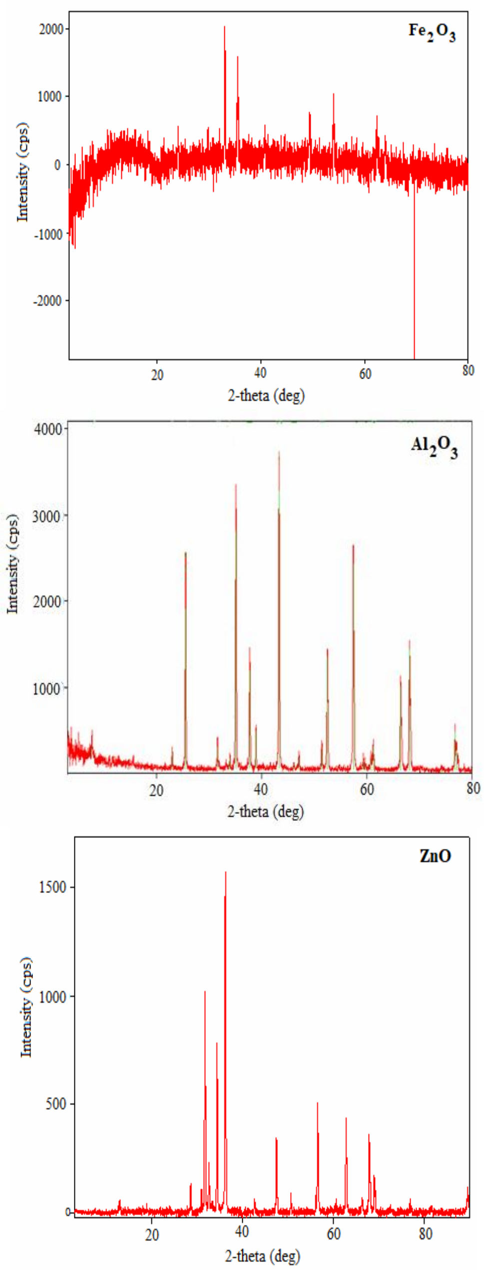


Fig. 2. The XRD patterns of Fe_2O_3, Al_2O_3, and ZnO nanoparticles.

The Effect of Shear Rate on Viscosity

The dynamic viscosities of different samples, as a function of shear rate at different temperatures, are plotted together in Fig. 3. According to this figure, the variation of viscosity was within 1.5% at 20 °C, suggesting that the behaviour of nanofluids was identical and that they can be approximately treated as Newtonian fluids. However, at 40 °C and 60 °C, the nanofluids exhibited a non-Newtonian behaviour.

Figure 4 illustrates the viscosity of different nanofluids with respect to the shear rate at various temperatures. In addition, Fig. 4 shows the purely Newtonian behaviour of the base fluid within the measured shear rates. By adding a number of solid nanoparticles to the base fluid, the rheological behaviour of the fluids changed and viscosity became a function of the shear rate. As can be seen in Fig. 4, the viscosity of Al_2O_3/oil had only a minimal effect on the shear rate whereas the Fe_2O_3/oil and ZnO/oil nanofluids exhibited a clear shear-thinning behaviour. Such shear-thinning behaviour possibly arose from the alignment of nanoparticle clusters in the flow direction caused by the increasing shear rate, thereby resulting in an initial decrease in viscosity. Other researchers have also observed similar behaviour [29,30]. The study of this behaviour is detailed by determining the power index *n*.

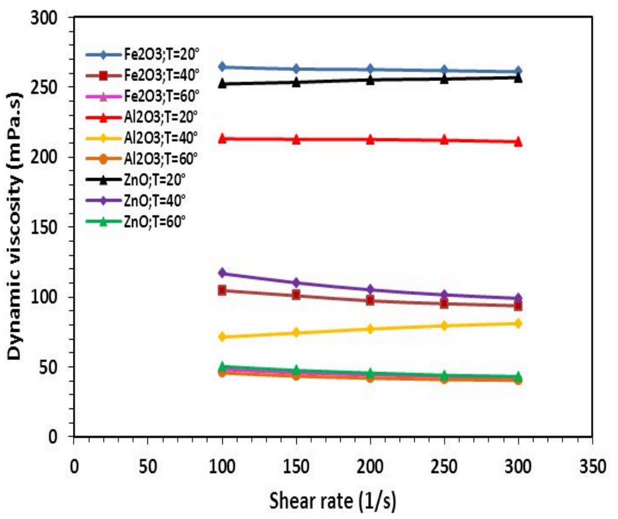


Fig. 3. Dynamic viscosity *versus* shear rate at different temperatures; φ = 1%.

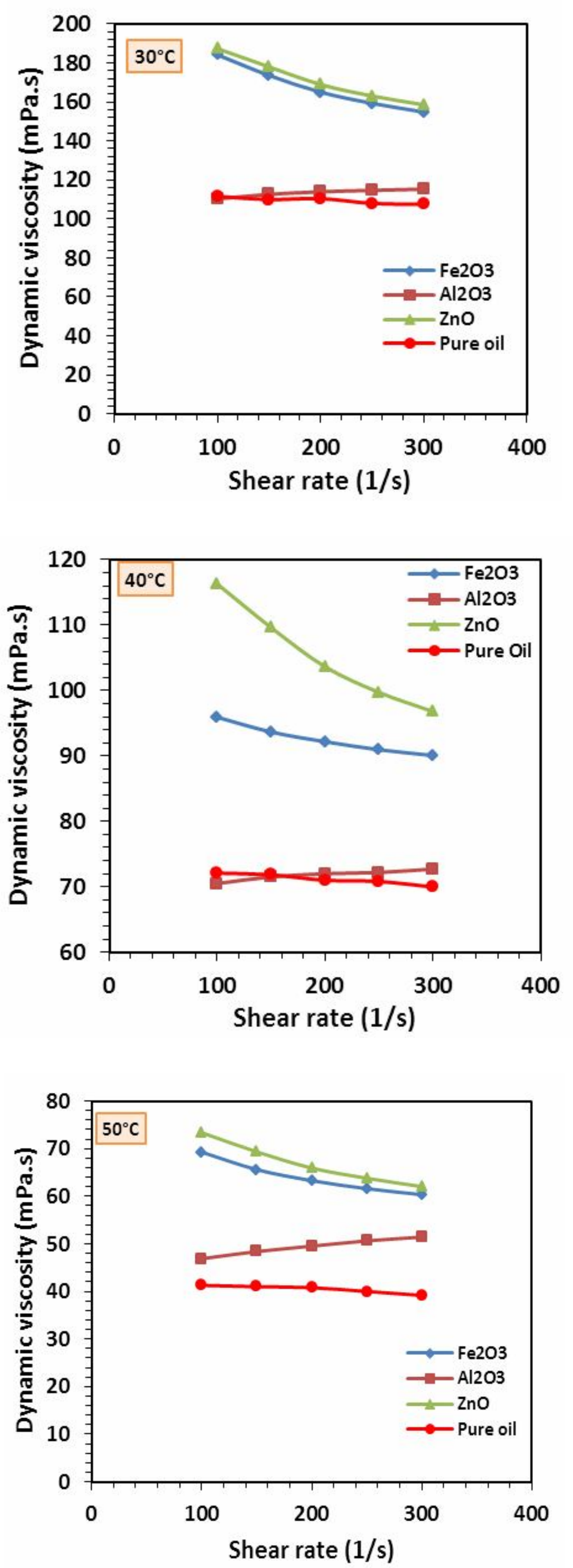


Fig. 4. Dynamic viscosity *versus* shear rate; φ = 0.5%.

The Ostwald-de Waele equation presented below was used to identify the rheological behaviour of fluids:

$$\tau = k\gamma^{n} \qquad (2)$$

where n is the power-law index, k is the consistency index, and τ is the shear stress. The fluid viscosity, as a function of the power-law, was defined by the following equation:

$$\eta = k\gamma^{(n-1)} \qquad (3)$$

In Equation (3), η represents the apparent viscosity and $\dot{\gamma}$ is the shear rate. In addition, n determines the rheological behaviour of fluids as follows: If n is equal to 1, the fluid is Newtonian, if $n < 1$, the fluid is shear-thinning, and if $n > 1$, the fluid is shear-thickening.

In order to obtain the indices n and k, logarithmic curves of shear stress versus shear rate were plotted. Then, the indices were calculated by the following equation:

$$\ln(\tau) = \ln(k) + n\ln(\gamma) \qquad (4)$$

Figure 5 shows the shear stress *vs*. shear rate curves of nanofluids for a solid volume fraction of 0.5%. The values were obtained from the linear regression of the curves in the logarithmic diagram ln(τ), as a function of ln(γ). The indices n and k were determined for all nanofluids at different temperatures and solid volume fractions. The results are listed in Table 2.

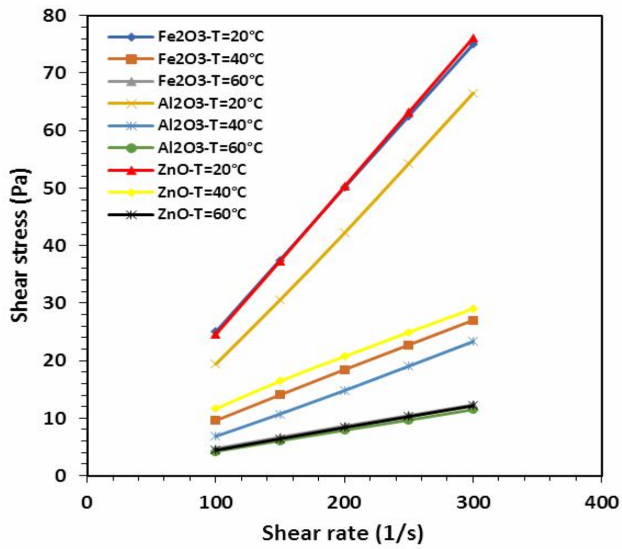


Fig. 5. Shear stress *versus* shear rate; φ = 0.5%.

The variations in the n and k indices of each nanofluid at different temperatures with a 0.5% volume fraction of nanoparticles are shown in Fig. 6. As can be seen in Fig. 6, at the same temperature, the value of the k index of the Al_2O_3/oil nanofluid was lower than that of the other two nanofluids, indicating that the viscosity of Al_2O_3/oil nanofluid was lower than that of Fe_2O_3/oil and ZnO/oil nanofluids. However, the k index decreased with increasing temperature for all nanofluids. Regarding the variations in the power-law index, the value of n for nanofluids based on

Table 2. Consistency and Power-Law Indices

Concentration	Temperature	Fe_2O_3		Al_2O_3		ZnO	
(% vol.)	(°C)	k	n	k	n	k	n
0.5	20	0.25	0.99	0.11	1.12	0.21	0.95
0.5	40	0.12	0.94	0.07	1.11	0.18	0.89
0.5	60	0.08	0.87	0.06	0.91	0.08	0.87
0.75	20	0.14	1.10	0.11	0.99	0.14	1.11
0.75	40	0.12	1.09	0.10	0.95	0.12	1.11
0.75	60	0.04	0.97	0.04	0.98	0.07	0.9
1	20	0.27	0.98	0.14	1.08	0.23	0.93
1	40	0.15	0.90	0.07	1.11	0.21	0.84
1	60	0.08	0.87	0.04	0.98	0.09	0.85

Fe_2O_3 and ZnO was less than 1, which confirms the non-Newtonian shear-thinning behaviour of these nanofluids compared to the nanofluid based on Al_2O_3, which exhibited a Newtonian behaviour. However, at a high temperature (60 °C), the *n* index was almost the same for all nanofluids, which reflects the same rheological behaviour.

Effect of Temperature on Viscosity

Figure 7 shows the dynamic viscosity, as a function of temperature, for nanofluids at different volume fractions and a shear rate $\dot{\gamma}$ = 200 s^{-1}. The results showed a general trend, according to which the dynamic viscosity decreased when the temperature increased at a constant volume fraction for all nanofluids. More specifically, when the temperature increased from 20 °C to 60 °C, the dynamic viscosity decreased 85%, from 260 mPa s to 40 mPa s. While maximum viscosity was observed in the nanofluid Fe_2O_3/oil with a 1% volume fraction, minimum viscosity was found in the base fluid. An increase in temperature resulted in an increase in the intermolecular distance of the nanoparticles and the base fluid, as a consequence of which their resistance against the flow and viscosity of nanofluids were reduced. The viscosity curves converged almost to the same value T = 60 °C, suggesting that the effect of solid volume fraction was negligible compared to that of temperature.

Effect of Concentration on Viscosity

The solid volume fraction is a very important parameter that affects the rheological behaviour of nanofluids. The dynamic viscosity of the studied nanolubricants with respect to solid concentration at different temperatures and different shear rates is displayed in Fig. 8. As can be seen in Fig. 8, the viscosity of nanofluids based on Fe_2O_3 and ZnO nanoparticles was significantly higher than that of the nanofluid based on Al_2O_3 nanoparticles. The rheological behaviour of the Al_2O_3/oil nanofluid was different from that of the other two nanofluids. Also, Fig. 8 illustrates that as the solid volume fraction increased, the viscosity increased too, except for the Al_2O_3 nanofluid, in which the dynamic viscosity decreased at low volume fractions (between 0.25% and 0.5%) but increased at volume fractions higher than 0.5%. Several experimental studies have concluded that the

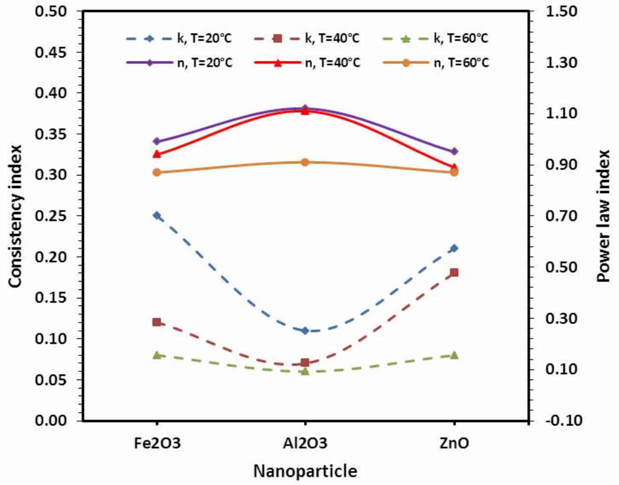


Fig. 6. Consistency and power-law indices for φ = 0.5%.

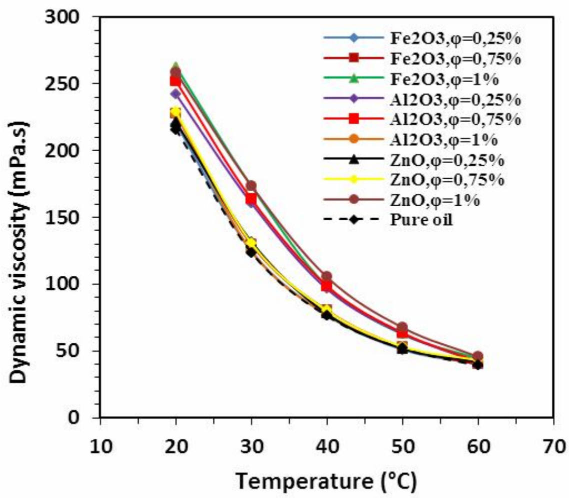
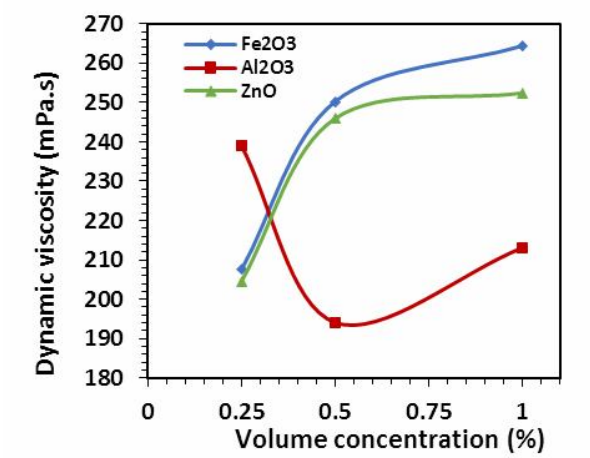
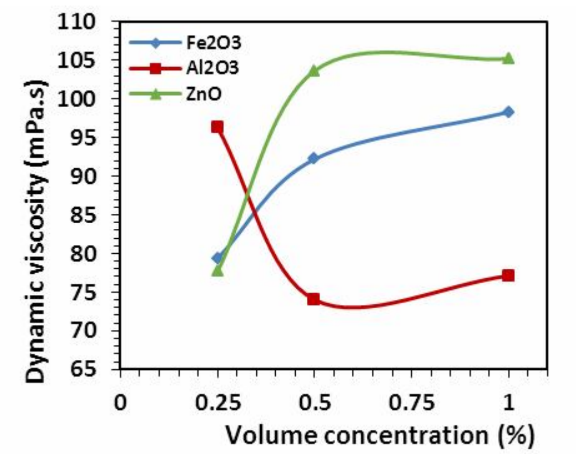


Fig. 7. Dynamic viscosity *versus* temperature at different volume concentrations, γ = 200 s^{-1}.

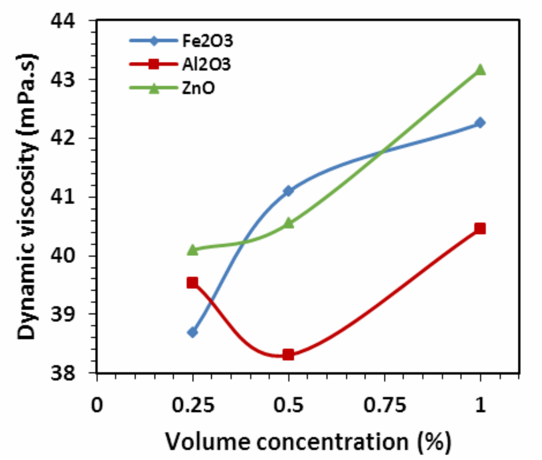
addition of nanoparticles increases fluid viscosity. The presence of nanoparticles affects the fluid shear stress due to the increase in the internal friction of the fluid, which, in turn, allows the fluid to resist the flow more, which is synonymous with increased viscosity. However, increasing the number of solid particles in a specific amount of fluid leads to larger nanoclusters due to van der Waals forces between the particles. These nanoclusters prevent the movement of oil layers on each other, leading to a higher viscosity [31].



T = 20 °C, $\dot{\gamma}$ = 100 s^{-1}



T = 40 °C, $\dot{\gamma}$ = 200 s^{-1}



T = 60 °C, $\dot{\gamma}$ = 300 s^{-1}

Fig. 8. Dynamic viscosity *versus* solid volume fraction.

CONCLUSIONS

In the present paper, a comparative study of the rheological behaviour of nanofluids was carried out. Nanofluids based on Fe_2O_3, Al_2O_3, and ZnO were prepared at different concentrations. The viscosity of the nanofluids was examined in terms of different variables, including shear rate, temperature, and volume fraction. Based on the results, the following conclusions can be drawn:

- The viscosity of Al_2O_3/oil nanofluid was significantly lower than that of Fe_2O_3/oil and ZnO/oil nanofluids;
- Ostwald-de Waele equation allowed the researchers to identify the non-Newtonian shear-thinning behaviour of Fe_2O_3/oil and ZnO/oil nanofluids and the Newtonian behaviour of Al_2O_3/oil nanofluid;
- All studied nanofluids had a viscosity that decreased with increasing temperature. The viscosity decreased around 85% at temperatures ranging from 20 °C to 60 °C;
- The viscosity of Fe_2O_3/oil and ZnO/oil nanofluids increased by the increase in the concentration of solid nanoparticles. For nanofluid Al_2O_3, the dynamic viscosity decreased at low volume fractions (below 0.5%) but increased at volume fractions beyond 0.5%. At a high temperature, the effect of solid volume fraction became negligible compared to that of the temperature.

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