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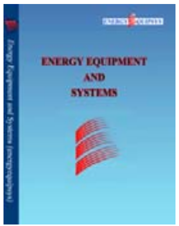
ENERGY EQUIPMENT AND SYSTEMS

Investigation with rheological behavior of liquid paraffin/Al_2O_3 nanofluid: Experimental approach

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ABSTRACT

Liquid paraffin can be used as a coolant fluid in electronic and cutting devices due to its suitable capabilities such as electrical insulating, high heat capacity, chemical, and thermal stability, and high boiling point. In this study, the dynamic viscosity of paraffin containing the alumina nanoparticles has been examined experimentally. The nanofluids with different composition of alumina (0, 1, 2, and 3%) with the diameter of 20 nm were prepared by two-step method and tested by professional Brookfield rheometer in the temperature range of 20 °C to 60 °C and the shear rates of 12 s^{-1} up to 200 s^{-1}. Experimental results indicated that the nano-lubricant behaves as Newtonian fluid in the volume fraction of 0 and 1% only at the temperatures of 50 and 60 °C. While it behaves as non-Newtonian fluid in the volume fraction of 2 and 3% for all measured temperatures. The results showed that the power law model represents the best curve fitting of the experimental data. Therefore, the coefficient values of power-law model including, consistency index and flow index were reported. Finally, an equation of relative viscosity based on the volume fraction and temperature of the combination was proposed by applying the curve fit technique on the experimental data.

Keywords: Experimental Correlation, Al_2O_3, Liquid Paraffin, Nanofluid, Rheological Behavior.

1. Introduction

Liquid paraffin is mainly used as a lubricant in various industrial applications. The most important usage of liquid paraffin is as a coolant because of its high heat capacity (more than 2000 $JKg^{-1}K^{-1}$), especially in electrical devices due to its specifications of electrically insulating and chemical and thermal stability [1-5].

The last decade has experienced the rapid development of nanotechnology, and so the new generation of heat transfer fluids called "Nanofluids" has been developed. The main reason is the higher thermal conductivity of nanofluids in comparison with the base fluid. Kole and Dey[6] reported an enhancement of 10.41% in the thermal conductivity of engine oil/Al_2O_3 nanofluid with only 0.035 volume fraction of nanoparticles. However, the challenging point which must be considered in designing cooling devices is the higher viscosity of nanofluids due to the addition of solid particles. Many types of research postulated the increase of the viscosity of

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nanofluid related to the addition of nanoparticles to the base fluid of engine oil [7-9]. They also reported a non-Newtonian rheological behavior of nanofluid even in small solid fraction of nanoparticles, while the pure base fluid displayed Newtonian behavior. The increase in dynamic viscosity for the 5 wt.% and 10 wt.% of alumina nanoparticles at the temperature of 30 °C, was reported nearly 20% and more than 28%, respectively by Ho and Gao[10]. The viscosity of n-octadecane/TiO_2 dispersions was experimentally investigated by Motahar et al. [11]. The rheological behavior of the samples indicated that nanofluids with low nanoparticle mass fractions demonstrated Newtonian behavior, and for higher mass factions, the shear-thinning behavior was observed.

Hosseini et al. [12] investigated the effect of nanoparticle concentration on the rheological behavior of paraffin-based nanofluid. They showed that the viscosity of the nanofluids increased as the Ni nanoparticle concentration increased. The results showed that this behavior was related to the nanoparticles interactions increase due to the increase of nanoparticles concentration.

The literature showed that predicting the rheological behavior of a Phase Change Material (PCM) plays an important role to design of cooling systems because it affects the free and forced convection heat transfer forces. Researches showed that when the free convection is the dominant heat transfer, the increase solid fraction might dampen the buoyancy-driven flow and heat transfer consequently [13-15]. The higher the viscosity, the higher costs of pumping are demanded [16].

Farsani et al. [17] showed that the addition of nanoparticles to the base fluid can dampen the buoyancy-driven vortices in the liquid phase of the paraffin/alumina melting process.

The viscosity not only changes with solid fraction of nanoparticles but also with temperature [18]. Ferrer et al. [19] reported a polynomial model type equation to predict the paraffin viscosity. Sepyani et al. [20] experimentally examined the effect of ZnO nanoparticles on the rheological behavior of engine oil viscosity to report a new correlation regarding solid volume fraction and temperature.

Nadooshan et al. [21] examined the usage of nanofluid of 10W40 lubricant containing hybrid nano-materials of silica and MWCNTs to find the rheological behavior of viscosity in different temperature and shear stress.

Fazlali et al. al. [22] investigated the rheological properties of paraffin-based CO_3O_4 nanofluid. The results showed these magnetic fluids indicated approximately Newtonian behavior in very low concentrations, whereas they seemed to act as a non-Newtonian-shear thinning fluid by increasing concentration.

To the best of the authors' knowledge, there are not any archival publications considering the rheological behavior of liquid paraffin/Al_2O_3 nanofluids. The objective of this study is to investigate the effect of temperature, nanoparticle concentration, and shear rate on the viscosity of liquid paraffin/Al_2O_3 nanofluid. Furthermore, a new equation is obtained to predict the value of relative viscosity as a function of nanoparticle concentration and temperature of nanofluid.

Nomenclature

K	Consistency (Pa.s^{-n})
n	Power law index
T	Temperature (˚C)

Greeks symbols

γ	Shear rate (s^{-1})
μ	Dynamic viscosity (mpa.s)
ρ	Density (kg/m^3)
τ	Shear stress (N/cm^2)
φ	Nanoparticle volume fraction

Subscripts

nf	Nanofluid
bf	Nanofluid

2. Experimental Methodology

2.1. Materials

- Liquid paraffin (Iran paraffin co., Iran) was used as base fluid to make the composition. Liquid paraffin is a mixture of different hydrocarbons, each containing from 10 up to more than 100 carbon atoms per molecule.
- Commercial spherical-shape Al_2O_3 powders (Alfa Aesar, Ward Hill, MA, USA) with Al_2O_3 content >99.98% and an average diameter of 20nm as nanoparticles.
- Oleic acid (Merck, Germany).
- The specifications of the liquid paraffin and nanoparticles are shown in Tables 1 and 2, respectively [23,24].

2.2. Preparation method

In this research study, the two-step method was used for preparing stable nanofluids. In this method, the purchased nanoparticles are suspended in base fluid with or without the use of surfactants [25]. This process is very suitable to prepare nanofluids containing oxide nanoparticles than those containing metallic nanoparticles [26].

In a typical process, alumina nanoparticles were weighted and mixed with oleic acid as surfactant of 1/3 of its mass fraction using an ultra-balance scale (RADWAG, Poland, see Fig. 1). Kole and Dey [6] reported that the engine oil/alumina nanofluid prepared with calculated amount of oleic acid as surfactant was tested to be stable for more than 80 days. Wang et.al [27] also showed that oleic acid can properly disperse $CaCO_3$ nanoparticles in paraffin.

To combine properly, the compound was put on a magnet stirrer device which was set on 70°C for about 30 minutes. Then, a calculated weight of liquid paraffin was added regularly to the compound. After that, to disperse the nanoparticles in the base fluid and achieve a uniform dispersion and stable suspension, the samples were exposed to an ultrasonic disruptor at 50 °C constant temperature bath and medium frequency for about 3 hours.

Figure 1A shows the prepared samples of nanofluids with the volume fraction of 0%, 1%, 2%, and 3%. Two samples of nanofluids, one with oleic acid and another with propanamine-triethoxysilyl as the surfactants were made and evaluated after 72 hours. Figure 1B demonstrates the samples, once they made and Fig. 1C after the elapsed time of 72 hours, respectively. As it can be observed, the nanofluid with oleic acid shows up a homogenous compound while the other sample has a thick layer of sediment. Figure 1D shows the cylinder spindle used in the rheometer.

Finally, to examine the characteristics and stability behavior of nanofluid, the Laser Particle Analyzer-VASCO Flex TM (ORDUAN TECHNOLOGIES, France) was

Table 1. Properties of liquid paraffin[25]

Results	**Items**
25-80 (12.5-16.5) mPa.s	Dynamic viscosity at 20 (40) °C
-12°C	Melting point
200-190 °C	Flashpoint
0.800-0.860	Specific gravity
0.001-0.005	Carbon residue wt %
28-30	Color saybolt
0.1-0.5	Aromatic mass %
390 °C	Initial boiling point
470 °C	Final boiling point

Table 2. Physicochemical property of Al_2O_3 nanoparticle

Parameter	**Value**
Color	White
Purity	99.9%
True density	3.6 gr/cm^3

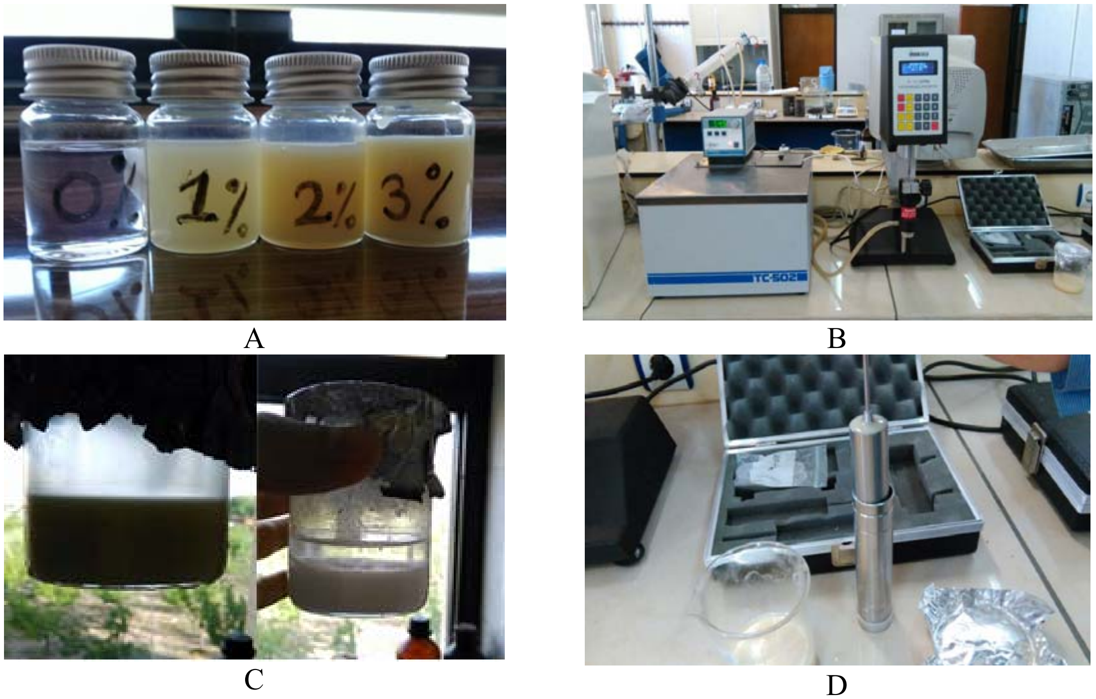

Fig.1 A: The prepared nanofluids with the volume fraction of 0%, 1%, 2%, and 3% , and B: the Brookfield digital rheometer-DV-III set up. C: Two samples of nanofluids, with oleic acid (left) and with propanamine-triethoxysilyl (Right) , and D: the L1- cylinder spindle of the rheometer

used to test the average dispersed size of nanoparticles in the base fluid which integrated by the Dynamic Light Scattering (DLS) approach. The results are shown in Fig. 2. As it was shown, the frequency distribution of nanoparticles, around the original diameter of 20nm, demonstrates that it should be few aggregation or accumulation within the solution to ensure that it acts stable.

2.3. Viscosity measurement

Viscosity data of the prepared composition were measured for volume fractions of 0, 1, 2, and 3% and the temperatures between 20-60 oC in the atmospheric pressure. The digital Brookfield rheometer (Brookfield Engineering Laboratories, Inc.) equipped with a temperature bath with the accuracy of ±1.0%

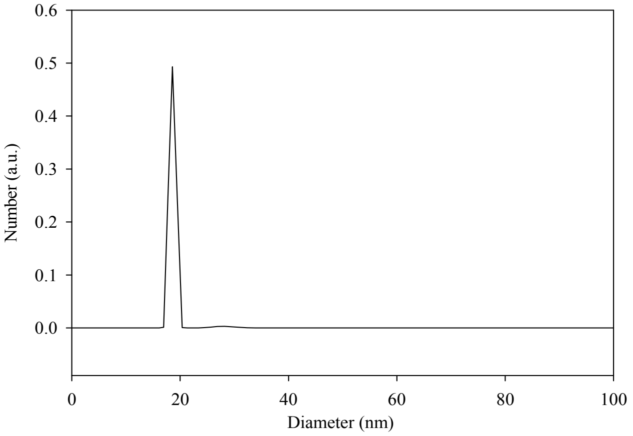

Fig. 2. Particle size distribution of Al_2O_3 nanoparticles in liquid paraffin

was employed to measure the viscosity of the samples. The measurements were carried out for shear rate ranges of 15 s^{-1} up to 200 s^{-1}. Cylinder spindle has been employed to perform the present experiments (See Fig. 1). The accuracy of the rheometer is verified using the viscosity of the standard fluids at 25 ^{o}C before the measurements according to the instruction manual.

3. Results and discussion

In this section first, the rheological behavior (Newtonian or non-Newtonian) of the nanofluids is investigated accurately. Then, a new correlation will be proposed to predict the viscosity trend based on the volume fraction and temperature changes.

3.1. Rheological behavior

Figure 3 shows the dynamic viscosity and shear stress versus shear rate (i.e., flow curves) for the base fluid and nanofluids with different concentrations.

The range of shear rate for the base fluid and nanofluid of 1% volume fraction is 15 s^{-1} to 200 s^{-1}, while it is 15 s^{-1} to 154 s^{-1} for nanofluids of 2% and 3% volume fraction.

The point is that the reported ranges were configured base on the torque capability of the instrument through the nanofluids. Due to higher resistance to flow, the range of shear rate was more limited for the concentrated nanofluids. It can be inferred from the results that the considered nanofluids behave as shear-thinning fluids since there is a sharp reduction in the viscosity of nanofluids with an increase of shear rate at lower shear rates, and the viscosity becomes gradually constant at higher shear rates.

This means that the studied nanofluids show the Newtonian pattern at the higher shear rates. The shear-thinning region is up to 90 s^{-1}. In other words, the nonlinear change of shear stress versus shear rate remains up to the higher values of the shear rate in concentrated nanofluids. Before this value of shear rate, the viscosity decreases as the shear rate increases, nonlinearly.

According to the obtained results, increasing the concentration of the nanofluids enhances their shear thinning behavior. The shear thinning properties is related to the probable deagglomeration of nanoparticles or rearrangement in the direction of the shearing force, which decreases viscous drag [28].

The results also depict that the relation between shear stress and shear rate is non-linear at lower shear rates but becomes linear at higher shear rates even at low nanoparticle concentrations. However, for the concentrated nanofluids the nonlinearity is more considerable. This trend associated with the interactions between nanoparticles and the base fluid at lower values of shear rates. The higher solid fraction of nanoparticles in the base fluid, the greater yield stress are reported [29]. In concentrated nanofluids, the interactions between nanoparticles and the base fluid are more pronounced which leads to stronger initial resistance to fluid shearing. As the shear rate increases, the resistance is eventually overcome and the fluid more easily flows.

The variation of viscosity with shear rate for many of the non-Newtonian nanofluids can be described by several models; the most commonly used being the Ostwald-de Waele or power law model. The power law model is demonstrated in Eqs. (1) and (2), where τ is the shear stress, K is the consistency factor, $\dot{\gamma}$ is the shear rate, n is the power law index and, μ is the effective or apparent viscosity.

$$\tau = K\dot{\gamma}^{n} \tag{1}$$

$$\mu = K\dot{\gamma}^{n-1} \tag{2}$$

when $n = 1$, Eq. (1) represents a Newtonian fluid, while a power law index of $0 < n < 1$ and $n > 1$ respectively represents a shear thinning and a shear thickening fluid. The shear thinning behavior reported for nanofluids by many researchers [30].

In addition to measuring viscosity, Brookfield digital rheometer is able to obtain the power law index and the consistency factor by fitting the measured data with the power law model. Table 3 displays the reported power law index and the consistency factor for the studied nanofluid samples.

It should be noted that these values are reported by Brookfield digital rheometer with the R^2 value about 1.0. It is clear that the based fluid and nanofluid of 1% volume fraction have Newtonian nature ($n \approx 1$) only in the temperatures of 50 and 60°C, whereas they show non-Newtonian in the temperatures of 20, 30, and 40°C. The nanofluids with 2% and 3% volume fraction show the non-Newtonian shear thinning behavior ($n < 1$) in all measured temperatures.

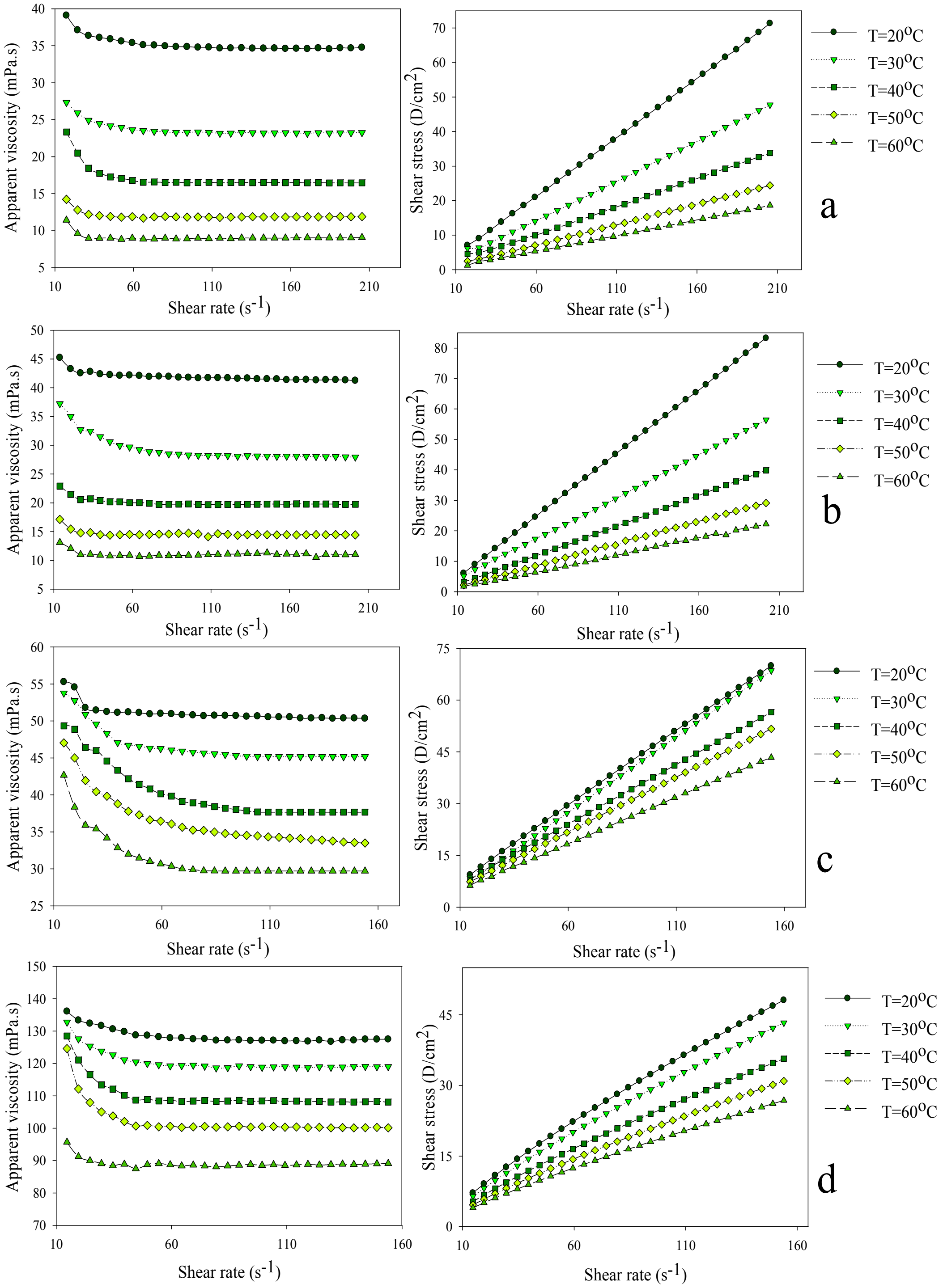

Fig.3. Viscosity and shear stress versus shear rate for (a) base fluid and nanofluid with (b) 1%, (c) 2%, and (d) 3% volume fraction with the effect of temperature

Table 3. The power law model values of liquid paraffin-alumina

	Temperature (℃)									
	20		**30**		**40**		**50**		**60**	
φ (%)	K	n	K	n	K	n	K	n	K	n
0	0.394	0.97	0.277	0.96	0.219	0.94	0.114	1.01	0.909	1.00
1	0.454	0.98	0.353	0.95	0.224	0.97	0.155	0.99	0.112	1.00
2	0.549	0.96	0.536	0.96	0.659	0.88	0.564	0.89	0.503	0.88
3	0.816	0.81	0.731	0.80	0.603	0.81	0.523	0.82	0.453	0.81

3.2. Effects of concentration and temperature on viscosity

Figure 4 illustrates the effect of temperature and concentration on viscosity of the studied nanofluids.

The results are reported for the range of shear rate which the viscosity is constant ($\dot{\gamma}$ >200 s^{-1}). In other words, the considered nanofluids behave as a Newtonian fluid at shear rates above 200 s^{-1}. Generally, it can be observed that the increase of temperature resulted in the reduction in viscosity. The higher tthe emperature, the lower viscosity of the base fluid happens due to the weakness of adhesion forces among the fluid molecules. Furthermore, in higher temperatures, the connection between the particles, which prevents the movement of the fluid layers on each other, weakens. In other words, the increase in temperature reduces the van der Waals forces between the particles [6, 21].

It can also be observed that, with increasing the particles volume fraction, the viscosity of nanofluid enhances. This observation can be described by the fact that internal shear stress of fluids is related to loading of particles, and it increases by increments of particle loading in the base fluid. This is caused by rising in internal friction of the fluid that enables it to resist more against flowing, which is along with the viscosity enhancement.

3.3 The Curve fitting equation to the relative viscosity

To determine the increased viscosity of nanofluids compared to that of the base fluid, the relative viscosity is defined as the ratio of the nanofluid viscosity (μ_{nf}) to base fluid viscosity (μ_{bf}). The variation of relative viscosity with volume the fraction of nanoparticles for various temperatures is shown in Fig. 5. These data are also obtained from the shear rate range in which the

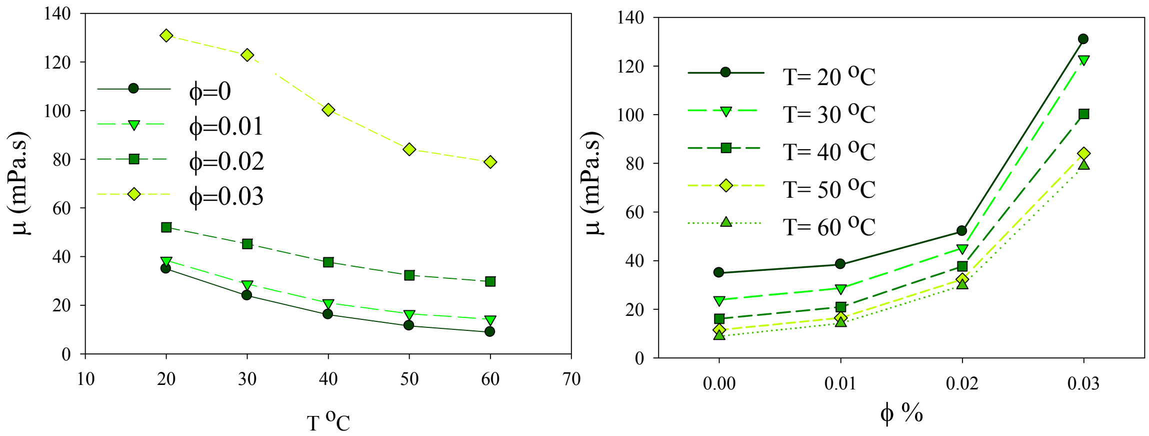

Fig. 4. The dynamic viscosity versus solid volume fraction and temperature at shear rates above 200 s^{-1}.

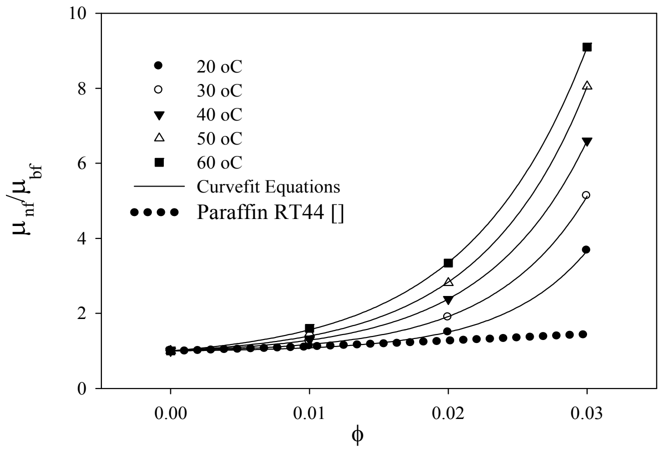

Fig. 5. Relative viscosity versus solid volume fraction for various temperatures and data for paraffin RT44[31]

nanofluid behaves as Newton a ian fluid and the viscosity is constant. Generally, the results show that the relative viscosity increases with the increase of solid volume fraction and temperature. The figure also illustrates the trend of relative viscosity associated with the equation employed by Arasu and Mujumdar [31] for paraffin RT44/alumina nanofluid. As it can be observed this equation predicts the relative viscosity only for dilute concentration of nanofluid in low temperatures.

A comparison between this equation and the experimental data show that the rheological behavior of paraffin/alumina needs more

To predict the relative viscosity of the considered nanofluids, a new model is proposed in the present investigation. This model captures the effect of concentration and temperature:

$$\frac{\mu_{nf}}{\mu_{bf}} = 1 + \frac{a\left(e^{b\varphi} - 1\right)}{T} \quad (3)$$

$$a(T) = -0.7792 + 0.3363e^{0.0645T} \quad (4)$$

$$b(T) = 235.5581 - 5.1449T + 0.0913T^2 - 0.0006T^3 \quad (5)$$

The coefficient a and b can be calculated dependent on the temperature in degree centigrade.

Figure 5 shows the model predictions for the relative viscosity of the considered nanofluids at different temperatures. It is clear that there is an agreement of model predictions and experimental data.

3.4. Deviation measurement

Table 4 indicates the deviation from the target calculated by Eq. (6) in various temperatures and solid volume fractions. As can be observed, the deviations are slight, meaning that the suggested correlation can sufficiently predict the liquid paraffin/Al_2o_3 relative viscosity.

$$\text{Margin of deviation} = \left|\left(\frac{\mu_{\text{measured}}}{\mu_{\text{proposed}}} - 1\right) \times 100\right| \quad (6)$$

4.Conclusion

The rheological behavior of paraffin/alumina nanofluids in the various volume fractions (0-3%), temperatures (20-60°C), and shear rates (0-200 s^{-1}) have been investigated experimentally. The base fluid and the dilute samples (0 and 1%) show the Newtonian behavior only in the high temperature (50 and 60°C), while the non-Newtonian shear thinning characteristic (n <1) is observed in the concentrated compounds (2 and 3%) in all measured temperatures. The shear stress changes non-linearly at lower shear rates but becomes linear at higher shear rates even at low nanoparticle concentrations. However, all

Table 4. Measured, calculated, and the margin deviation % values.

	Measured values					Proposed model					Margin of deviation (%)				
	Temperature (℃)														
φ (%)	20	30	40	50	60	20	30	40	50	60	20	30	40	50	60
0	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	0.00	0.00	0.00	0.00	0.00
1	1.1	1.2	1.3	1.43	1.6	1.08	1.17	1.26	1.38	1.56	1.57	2.46	3.12	3.33	2.40
2	1.49	1.89	2.34	2.81	3.34	1.51	1.90	2.26	2.73	3.36	1.29	0.75	3.59	2.87	0.51
3	3.687	4.92	6.23	7.31	9.24	3.71	5.06	6.08	7.48	9.10	0.58	2.90	2.35	2.26	1.54

of the studied samples behave as a Newtonian fluid at shear rates above 200 s^{-1}. Additionally, the curve fit equation of relative viscosity is proposed to predict its value based on the volume fraction and temperature.

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