

Contents lists available at ScienceDirect

# Journal of Molecular Liquids



# Experimental investigation on thermal and rheological behaviour of PAG lubricant modified with SiO<sub>2</sub> nanoparticles



S.S. Sanukrishna<sup>a,b,\*</sup>, S. Vishnu<sup>b</sup>, M. Jose Prakash<sup>b</sup>

<sup>a</sup> University of Kerala, Kerala, India

<sup>b</sup> Department of Mechanical Engineering, TKM College of Engineering, Kollam, Kerala 691005, India

#### ARTICLE INFO

Article history: Received 9 December 2017 Received in revised form 9 March 2018 Accepted 10 April 2018 Available online 12 April 2018

Keywords: Nanolubricant Thermal conductivity Rheology Shear thinning Thixotropy

# ABSTRACT

Nanolubricant is prepared by dispersing SiO<sub>2</sub> nanoparticles into synthetic refrigerant compressor oil, polyalkylene glycol (PAG) using two-step method. Thermal conductivity and rheological properties of nanolubricant at various volume fractions (0.07 to 0.6vol %) in the temperature range 20 °C to 90 °C have been investigated. The results show that as the volume fraction increases thermal conductivity and viscosity increase. Conversely, the thermal conductivity and viscosity are found to be decreasing with increase in temperature. The highest thermal conductivity and viscosity of the nanolubricant(volume fraction 0.6%) are 1.31 and 10.34 times greater than that of pure lubricant. The measured thermal conductivity and viscosity are compared with that obtained from classical models and the classical models fail to predict these properties accurately. The non-newtonian shear thinning behaviour of SiO<sub>2</sub>-PAG nanolubricant was confirmed by computing power law and consistency indices. At higher particle concentrations and lower temperatures, nanolubricant shows thixotropic behaviour.

© 2018 Elsevier B.V. All rights reserved.

# 1. Introduction

The enhancement of thermal conductivity of fluids by the dispersion and stabilization of solid particles was established by Maxwell [1] more than a century ago. The nanofluids are proposed as the next generation heat transfer media due to the fact that their thermal transport properties are significantly higher than those of the base liquids [2]. Nanolubricant is a new class of lubricant produced by dispersing nano-sized particles of metals, metal oxides, carbon and its allotropes into conventional lubricating oils. Dispersion of nanoparticles into base oils is a promising approach towards enhancing the thermal, rheological and tribological properties. The viscosity and rheological characteristics are probably more crucial parameters to be measured and analyzed along with thermal conductivity. Few studies are reported in the literature regarding the thermophysical and rheological characteristics of nanolubricants. According to the investigations that have been conducted, the presence of nanoparticles in oils may enhance their lubricating properties compared to the base fluid; this would, in turn, result in an increase in the durability of the components [3-6].Thermal conductivity and viscosity are the thermophysical characteristics of nanofluids that have been studied by many investigators and shown that the thermal conductivity increases with increase in volume fraction and temperature [7,8]. Viscosity plays an important role in the rheological behavior of fluids [9-11]. Temperature, nanoparticles loading and particle size can affect viscosity, thus rheological behaviour of a nanofluid. Viscosity of nanofluids increases with increase in volume fraction and particle size and a reverse trend is observed with increase in temperature [12–14]. Rheological behaviour of nanofluids is quite different from each other and a specific behaviour cannot be established for all nanofluids. Viscosity of Ag-heat transfer oil was studied by Aberoumand et al. [15], and their results indicated that the base fluid which exhibits Newtonian behaviour changes in to non-Newtonian while adding even small amounts of nanoparticles. Recently researchers used hybrid nano-additives to coolants and lubricants with an aim to enhance its thermo physical and heat transfer capabilities and presented optimistic results [16-19]. In refrigeration systems when the refrigerant is circulated through the compressor it carries traces of nanolubricant so that the other parts of the system will have a nanolubricant-refrigerant mixture. Addition of nanoparticles in refrigeration systems results in remarkable improvement in thermophysical, and heat transfer capabilities which in turn enhances the efficiency and reliability. Kedzierski [20-21] shown that lubricant viscosity significantly influences the boiling characteristics of refrigerant/lubricant mixtures. There are also some studies in the literature reporting the performance improvement of refrigeration systems that use nanolubricants [22,23]. Studies show that addition of Al<sub>2</sub>O<sub>3</sub> in compressor oil increases the freezing capacity and COP. Wang et al. [24] showed

<sup>\*</sup> Corresponding author at: TKM College of Engineering, Kerala, India. *E-mail address:* sanukrishna@sctce.ac.in. (S.S. Sanukrishna).

Nomenclature			
English sy m k T Cp	mbols mass of nanoparticles[g] thermal conductivity[W/mK] temperature [ <sup>0</sup> C] specific heat [k]/kgK]		
Greek syn $\beta$ $\varphi$ $\mu$ $\rho$ $\tau$ $\gamma$	nbols ratio of nanolayer thickness to original radius volume fraction [%] dynamic viscosity[cP] density[kg/m <sup>3</sup> ] shear stress(dyne/cm) shear rate(1/s)		
Subscripts bf eff p	base fluid effective particle		
Abbreviat COP PAG TEM SEM EER	ions coefficient of performance polyalkylene glycol transmission electron microscope scanning electron microscope energy efficiency ratio		

that the Energy Efficiency Ratio(EER) of residential air conditioners can be increased by 6% if the polyolester oil is replaced with NiFe2O<sub>4</sub> nanolubricant. In another study, Krishna Sabareesh et al. [25] explored the effect of dispersing low concentration of TiO<sub>2</sub> nanoparticles in the mineral oil based lubricant. They investigated the effect of nanoparticles on viscosity, lubrication qualities, and the performance of refrigeration systems utilizing R12 (Dichlorodifluoromethane) as the working fluid and reported that average heat transfer increases by 3.6%, compressor work reduces y 11% and consequently COP increases in by17%.

Few studies are available in the literature regarding polyalkylene glycol based nanolubricants. Sharif et al. conducted performance studies on an automotive air-conditioning system with SiO<sub>2</sub>-PAG nanolubricant and reported overall performance improvement [26]. Liu et al. experimentally determined the effect of particle size on the tribological properties of polyalkylene glycol [27]. A comparative study on the thermal conductivity and viscosity of Al<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub> based nanolubricants was conducted by Redhwan et al. [28], but detailed rheological characteristics have not been reported in their studies.

Regardless of the fact, the thermophysical properties of nanolubricants are widely investigated by various researchers; property data related to PAG oil based nanolubricants is scarce. In addition to thermophysical properties, the rheological characterization of nanolubricants can provide valuable insight for practical applications. The present study deals with experimental investigations on the thermal conductivity and rheology of Polyalkylene glycol oil suspended with SiO<sub>2</sub> nanoparticles. The effects of particle concentration and temperature on the thermophysical properties were investigated. The effects of temperature, volume fraction and shear rate were studied to elucidate the rheological behaviour of the nanolubricant. The time dependent viscosity at various temperatures and shear rates has also been examined.

# 2. Experimental method

## 2.1. Materials and characterization

SiO<sub>2</sub> nanoparticles(purity: 99.8%) spherical in shape supplied by Sigma Aldrich Limited, USA with the size of 10-20 nm, molecular weight 60.08 g/mol, boiling point 2230 °C and density 2.6 g/cm<sup>3</sup> were used for the preparation of nanolubricant. The commercially available, fully synthetic, oil based on Polyalkylene Glycol (PAG) with a nominal liquid dynamic viscosity of 30–40 cP at 40 °C, was used as the base fluid. PAG lubricants have better tribological performance than mineral oils when used together with HFCs. Scanning Electron Microscopy (SEM) and Transmission electron Microscopy (TEM) were employed for the morphological characterization of SiO<sub>2</sub> nanoparticles and nanolubricant respectively.

Fig. 1 shows the SEM image of dry  $SiO_2$  nanoparticles as supplied by the manufacturer and it displays the distribution and shape of the nanoparticles. The particles are spherical in shape. It can be seen that the particles are in the form of agglomerates. These agglomerates have to be broken during the preparation of nanolubricant to produce a stable suspension. It can be accomplished through magnetic and ultrasonic agitation.

# 2.2. Preparation of Nanolubricant

Preparation of nanolubricant is the primary step in the experimental studies. Nanofluids are not mere solid to fluid suspensions. In order to achieve even, stable and durable suspension with negligible aggregation of particles, special processes are necessary. Samples examined in these studies were prepared by the two-step method and no surfactants were added. Nanolubricants were prepared at five different particle concentrations (0.07, 0.09, 0.2, 0.4 and 0.6 vol%). The required mass of nanoparticles corresponding to the volume fractions was calculated (Eq. 1) and weighed using a high precision electronic balance.

$$\Phi = \frac{\binom{m}{\rho}_{\text{SiO}_2}}{\binom{m}{\rho}_{\text{SiO}_2} + \binom{m}{\rho}_{PAG}}$$
(1)

The preliminary mixing process of samples was carried out using the magnetic stirrer for 1 h and then agitated using ultrasonic agitator (BRANSON-3800) at a frequency of 40 kHz intermittently for 12 h with sufficient cool-off period in a controlled atmosphere (25-33  $^{\circ}$ C)



Fig. 1. SEM image of dry SiO<sub>2</sub> nanoparticles.



**Fig. 2.** TEM images of SiO<sub>2</sub> suspension (0.6vol %).



Fig. 3. Samples of SiO<sub>2</sub>/PAG nanolubricant at different volume fractions.

to homogenize the samples. No evidence of sedimentation or coagulation was noticed after 120 h of preparation.

Fig. 2 shows the TEM images of the prepared nanolubricant. According to TEM images, the suspension is homogeneous and nanoparticles are well dispersed. The nanoparticles have almost similar characteristics such as physical appearance, shape and size in the suspension also. The average size of nanoparticle was found to be 14.26 nm.

Fig. 3 illustrates the SiO<sub>2</sub>-PAG nanolubricant samples at different volume fractions. in.

### 2.3. Measurement of thermal conductivity

The thermal conductivity of base lubricant and SiO<sub>2</sub>/PAG nanolubricant at different volume fractions were measured with KD2 Pro thermal property analyzer (Decagon devices, Inc., USA). Thermal conductivity in the range of 0.02 to 2.00 W/mK with an accuracy of  $\pm$  0.001 could be measured by this device. The principle of operation of the device is transient hot wire and the maximum deviation is  $\pm$ 5.0%. The probe of this instrument consists of a needle with a heater and temperature sensor inside. Electric current is supplied through the heater and the temperature of the probe is measured over time. Since the transient hot-wire measurement lasts for only a few seconds, the problems related to convection is been eliminated. The apparatus meets the standards of both ASTM D5334-14 [29] and IEEE 442 – 1981 [30]. A refrigerated and heating circulator (F-25, Julabo, Germany) was used to

maintain a constant temperature of the sample with an accuracy of 0.01 °C. The thermal conductivity of SiO<sub>2</sub>/PAG oil nanolubricant at five different particle volume concentrations (0.07 to 0.6vol %) was measured in the temperature range of 20 °C to 50°C. The thermal property analyzer was calibrated with verification standard fluid (Glycerine, CAS 56-81-5) at a controlled temperature of 28 °C and at atmospheric pressure. The measurement of thermal conductivity was performed repeatedly. Based on deviation between thermal conductivity of the standard fluid and the measured thermal conductivity during the calibration, the error in measurement is estimated and the thermal conductivity data reported in the study represents an average of ten measurements with an estimated error of  $\pm 1.6\%$ . The experimental results were compared with that obtained from models given in Table.1.

Table 1	
Thermal conductivity models.	

Model	Correlation
Maxwell [1]	$\frac{k_{eff}}{k_{bf}} = \left[\frac{k_p + 2k_{bf} + 2(k_p - k_{bf})\varphi}{k_p + 2k_{bf} - (k_p - k_{bf})\varphi}\right]$
Hamilton and Crosser [31]	$\frac{k_{eff}}{k_{bf}} = \frac{k_{p+}(n-1)k_{bf} - (n-1)\varphi(k_{bf} - k_{p})}{k_{p} + (n-1)k_{bf} + \varphi(k_{bf} - k_{p})}$
Timofeeva et al. [32]	$rac{k_{ m eff}}{k_{ m bf}} = [1+3arphi]$
You and Choi [33]	$\frac{k_{eff}}{k_{bf}} = [\frac{k_p + 2k_{bf} + 2(k_p - k_{bf})(1 + \beta)^3 \varphi}{k_p + 2k_{bf} - (k_p - k_{bf})(1 + \beta)^3 \varphi}]$
Redhwanet al [28].	$rac{k_{eff}}{k_{bf}} = 1.2(1 + rac{\varphi}{100})^{0.04}(1 + rac{T - 273.15}{80})^{-0.01}$

Tal	ble	2	

Viscosity models.				
Models	Correlations			
Einstein [35]	$rac{\mu_{eff}}{\mu_{be}} = (1+2.5arphi)$			
Brinkman [36]	$\frac{\mu_{eff}}{\mu_{hf}} = \frac{1}{(1-\omega)^{2.5}}$			
Pak and Cho [37]	$\frac{\mu_{eff}}{\mu_{bf}} = 1 + 39.11\varphi + 533.9\varphi^2$			
Wang et al. [38]	$rac{\mu_{ m eff}}{\mu_{ m bf}}=123arphi^2+7.3arphi+1$			

# 2.4. Measurement of rheological properties

The rheological behaviour of the nanolubricant has been investigated experimentally covering a temperature range of 20-90 °C, shear rates range of 3.75-450/s and particle concentrations range of 0.07–0.6%. A Brookfield LVDV-II + Pro. plate-and-cone rheometer having measurement range between 1.0 and 2000 cP together with a constant temperature circulator (JULABO F-25, Germany) was used for the measurements. The method of measuring follows ASTM D2196-10 [34] which is the standard test methods for rheological properties of materials by the rotational (Brookfield type) viscometer. The torque required to turn the spindle of the rheometer in a fluid is a measure viscosity of the fluid. Torque is applied through a calibrated spring to the spindle immersed in test fluid and the spring deflection measures the viscous drag of the fluid against the spindle. The amount of viscous drag is proportional to the amount of torque required to rotate the spindle. Rheocalc software was used to acquire the data. The spindle (CPA -40Z, cone angle 0.8<sup>°</sup> and cone radius 2.4 cm) used for this study was calibrated with Brookfield viscosity standard fluid. The quantity of sample required for the measurement is 0.5 ml. The maximum uncertainty was found to be 1.9%. All the measurements were performed under steady-state conditions within torque range of 10-100%.

# 2.4.1. The power law model

Ostwald-De Waele model is the most generalized model for non-Newtonian fluids. Ostwald–De Waele power law model (Eq. (2)) was used to quantify the behaviour of nanolubricant, i.e., whether it comes under Newtonian or non-Newtonian fluid.

$$\tau = m\gamma^n \tag{2}$$







**Fig. 5.** Thermal conductivity ratio of SiO<sub>2</sub> –PAG nanolubricant with volume fractions at different temperatures.

The Power Law model is described by two parameters, consistency coefficient (m) and flow behavior index (n). Consistency index is a product's viscosity at one reciprocal second and flow behaviour index indicates the degree with which a material exhibits non-Newtonian flow behaviour. The viscosity of the fluids which follow power law is defined by the following equation:

$$\mu = m\gamma^{n-1} \tag{3}$$

Fluids which obey power-law models are classified into shearthinning and shear-thickening under increasing shear rates. If the magnitude of n < 1, the fluid is known as shear-thinning or pseudoplastic. This means that the apparent viscosity decreases with increase in shear rate. When n > 1, it is shear-thickening or dilatant, i.e. their apparent viscosity increases as shear rate increases. In order to obtain these indices, a logarithmic diagram of shear stress vs shear rate has been drawn and the indices were calculated by the following equation.

$$Ln(\tau) = Ln(m) + nLn(\gamma) \tag{4}$$



Fig. 6. Comparison between experimental thermal conductivity and model predictions at room temperature(30 °C).

The viscosities obtained from experiments were compared with that calculated using correlations. The models considered were depicted in Table.2.

# 2.4.2. Thixotropic effect

A fluid whose viscosity decreases with time under constant shear rate is called thixotropic fluid. The Brookfield rheometer with a

250 3.75/s 20<sup>0</sup>C а · 4.5/s .... 7 5/s 200 11 25/s 15/s18.75/s -22.5/s Viscosity (cP) 150 30/s ···▲··· 37.5/s -⊽- 45/s 100 ····· **\*** 50 0 0.0 0.1 0.2 0.3 0.4 0.5 0.6 0.7 Volume fraction (%) 100 40<sup>0</sup> C b - 3.75/s 4.5/s 7.5/s 80 11.25/s -15/s -18.75/s -22.5/s 0 Viscosity (%) 60 30/s ··· --- 37.5/s -v- 45/s 75/s 40 90/s -0 20 0 0.0 0.1 0.2 0.3 0.4 0.5 0.6 0.7 Volume fraction (%) 50 80°C 3 75/s С 4.5/s7.5/s 40 11 25/s 15/s18 75/s 22.5/s Viscosity (cP) 30 30/s 37.5/s 45/s ..... 75/s 20 -0-90/s 150/s -D- 225/s 10 0 0.0 0.1 0.2 0.3 0.4 0.5 0.6 0.7 Volume fraction (%)

Fig. 7. Variation of viscosity of SiO<sub>2</sub>-PAG nanolubricant with volume fractions at different shear rates at (a) Temperature = 20 °C, (b) Temperature = 40 °C, (c) Temperature = 80 °C.





Fig. 8. Viscosity ratio variations with volume fraction at different temperatures at (a) Shear rate = 3.75/s, (b) Shear rate = 37.5/s(c) Shear rate = 90/s.

# 3. Results and discussion

#### 3.1. Thermal conductivity

The thermal conductivity of SiO<sub>2</sub>/PAG nanolubricant was measured covering a temperature range 20-50 °C and volume fractions of 0.07, 0.09, 0.2, 0.4 and 0.6 vol%.

Figs. 4 and 5 show the thermal conductivity and thermal conductivity ratio  $(k_{eff}/k_{bf})$  respectively as a function of volume fraction at different temperatures. The thermal conductivity of nanolubricant increases with increase in particle concentration. The maximum enhancement in thermal conductivity ratio obtained was 1.31 at a volume fraction of 0.6% and temperature 20 °C. There are many potential factors responsible for the anomalous enhancement in thermal conductivity of nanofluids: (i) The Brownian motion effect of nanoparticles in the base fluid is a vital factor for the observed enhancement. The heat is carried by phonons by propagating lattice vibrations in the crystalline solids suspended in fluids. Such phonons are propagating in a random direction. Some ballistic phonon effects could possibly lead to an increase in thermal conductivity. If the ballistic phonons initiated in one particle can reach a nearby particle [39]. The particles may be closer enough due to the increase of particle concentration, and thus enhance coherent phonon heat flow among particles due to Brownian motion; eventually the thermal conductivity becomes a function of nanoparticles concentration.(ii)molecular layering of the liquid: Existence of a



Fig. 9. Temperature dependent viscosity variation at different particle concentrations at (a) Shear rate = 3.75/s, (b) Shear rate = 37.5/s,

nano-layer at the solid-liquid interface and nanoparticle aggregation may constitute major contributing mechanisms for thermal conductivity enhancement in nanofluids. The liquid molecules close to particle surfaces are known to form layered structures and behave much like a solid and which will act as a thermal bridge between the fluid and solid particles. (iii) the increased thermal conductivity of suspended solid particles.

It can also be seen that there is a strong temperature-dependent variation in thermal conductivity at various particle concentrations. At all particle concentrations, thermal conductivity of pure lubricant and SiO<sub>2</sub>-PAG nanolubricant decreases with increase in temperature. This phenomenon can be attributed to the following factors: (i) at elevated temperatures, the liquid molecules move away from each other, and hence the mean free path between them increases. This reduces the probability of collision between the molecules and which eventually decreases the thermal conductivity [26]. (ii)At higher temperatures the separation distance between the particles increases and consequently the near-field radiation (i.e., Coulomb interaction) deteriorates and which in turn reduces thermal conductivity [40].

The experimental results of thermal conductivity were compared with the results obtained from different models and the same is shown in Fig. 6. The model proposed by Redhwan et al. [28] for SiO<sub>2</sub>based nanolubricant over predicts the thermal conductivity, especially at lower particle concentrations. The classical models underpredict the thermal conductivity at all volume fractions. They have been developed for thermal conductivity of nanoparticlesuspended in low viscous fluids like water, ethylene glycol fluids, which considered only thermal conductivities of the base fluid, nanoparticles and particle volume fraction.

# 3.2. Rheological characteristics of Nanolubricant

Viscosity of pure PAG oil and SiO<sub>2</sub>/PAG nanolubricant at different volume fractions (0.07, 0.09, 0.2, 0.4and 0.6 vol%), shear rate (3.75 to 450/s) and temperatures (20 to 90 °C) were measured. In order to evaluate the rheological behavior, Ostwald–de Waele power law model was used.

3.2.1. Effect of particle concentration on viscosity of SiO<sub>2</sub>-PAG Nanolubricant

Fig. 7 shows the variation of viscosity with volume fraction at different temperatures and shear rates.

It is clear that shear rate and volume fraction play a vital role in the viscosity of nanolubricant. As the volume fraction increases, the



Fig. 10. Viscosity variations of Pure-PAG oil with shear rates at different temperatures.

viscosity of nanolubricant also increases and at lower shear rates and temperatures, there is a substantial increase in viscosity with volume fraction. The highest viscosity obtained was 237 cP and it happens at a volume fraction of 0.6% and a shear rate of 3.75/s. Viscosity is a property occurring due to the internal frictional force that develops between different layers of fluids as they move relative to each other. The suspended nanoparticles in the PAG oil would raise its viscosity as a result of the collisions between nanoparticles and the base fluid. By intensifying the amount of solid nanoparticles in a fixed volume of a liquid, greater nano-racemes arise due to Van der Waals forces. In other words, as the concentration of nanoparticle increases, the particles make agglomerations within the suspension. This consequently results



Fig. 11. Variations of viscosity with shear rates at different temperatures, at (a) Volume fraction =0.07%, (b) Volume fraction = 0.09%, (c) Volume fraction = 0.2%, (d) Volume fraction = 0.4%, (e) Volume fraction = 0.6%.

in the increase of internal shear stress in nanolubricant and hence an increase in viscosity.

Fig. 8 shows the variation of viscosity ratio ( $\mu_{eff}/\mu_{bf}$ ) with volume fraction and temperature at three different shear rates. The viscosity ratio increases with increase in temperature and this is because of the fact that, the rate of decrease of viscosity of the pure lubricant is higher than that of nanolubricant, in other words, the nanolubricant sustains adequate viscosity than pure lubricant at elevated temperatures. An abrupt increase in relative viscosity at higher volume fractions and at lower shear rates also been manifested. The maximum viscosity ratio is found as 10.34, which is obtained at a lower shear rate of 3.75/s at volume fraction of 0.6%. The reason for this anomalous increase in relative viscosity may be related to the fact that at higher volume fraction the nanoparticle clustering is more and the applied lowers hear rate (3.75/s) is not sufficient enough to break the nano-clusters. As the shear rate increases, the relative viscosity is found to be decreasing. The maximum viscosity ratio observed at a higher shear rate of 90/s is 2.32. From this it is clear that SiO<sub>2</sub>-PAG nanolubricant is more appropriate to use at moderate temperature range and shear rates.

# 3.2.2. Effect of temperature on viscosity

For the understanding of viscosity variation with temperature, the dynamic viscosity versus temperature at different volume fractions is plotted in Fig. 9. The results reveal that irrespective of the shear rate, the viscosities of the pure lubricant and nanolubricant diminish with increase in temperature. The reasons behind this phenomenon can be explained as follows: At lower temperatures, the nano-racemes hinder the motion of lubricating oil layers on each other.

Increasing the temperature helps the particles to overcome Van der Waals attractive forces and which may disintegrate the clusters of nanoparticles suspended in base fluid and hence the intermolecular interactions between the molecules become weak and this phenomenon leads to decrease in viscosity. Specifically, due to temperature rise, the Brownian motion will be enhanced which may cause increase in chaos and this will decrease the viscosity of nanolubricant.

### 3.2.3. Rheological behaviour of base lubricant and nanolubricant

In order to study the rheological behaviour of pure lubricant and nanolubricant, the relation between viscosity and shear rate at different temperatures are plotted. The rheological studies were performed over the range of shear rate from 3.75 to 450/s covering the temperature range from 20 °C–90 °C. Fig.10 shows the variation of viscosity with shear rate for pure lubricant. As expected, pure lubricant exhibits Newtonian behaviour within the shear rate range considered.

The viscosity of the nanolubricant samples was measured over the same range of shear rate. Fig. 11 shows the effects of shear rate and temperature on apparent viscosity. The apparent viscosity of nanolubricant decreases with shear rate increment regardless of temperature and solid volume fraction. The results illustrate that at low concentrations (0.07% and 0.09%) change in viscosity with increase in shear rate is small. However, at higher concentrations, a considerable decrease in the viscosity of the samples with shear rate is noticed. That is viscosity of high concentration samples is more dependent on shear rate, consequently displays non-Newtonian behaviour.

The alterations in the structure and arrangement of intermingling particles can be connected to shear thinning behaviour of welldispersed suspensions. Shearing may cause the particles to orient in the direction of flow and its gradient. This can disrupt the agglomerates in the suspension and hence reduce the amount of fluid immobilized by the nanoparticles. The interaction forces may then decrease and which in turn lowers the flow resistance and the apparent viscosity. The viscosity decreases exponentially at lower shear rates. i.e. the nanolubricant exhibit Non-Newtonian behaviour with significant shear thinning characteristics. The shear viscosity of the nanolubricant predominantly decreases for a shear rate lower than 100/s. Over this



Fig. 12. Logarithmic diagram of shear stress-shear rate of pure PAG oil at different temperatures.

shear rate value, the apparent shear viscosity tends to a Newtonian plateau.

Figs. 12 and 13 show the logarithmic diagram of shear stress vs shear rate of pure lubricant and nanolubricant respectively. The volume fraction is varied from 0.07 to 0.6%. The curves are fitted with R-squared value higher than 0.99. Here, n and m are the power law and consistency indices respectively. From figures, it is clear that, regardless of temperature and volume fraction,SiO<sub>2</sub>-PAG nanolubricant behaves as a non-Newtonian fluid. However, the high concentration samples follow the power law model expressed in Eq. (2) with a power law index much less than unity (n < 1). Meanwhile, the pure PAG oil behaves like Newtonian fluid with power-law index n  $\approx$  1 at all temperatures.

Fig. 14 depicts the variation of power-law index with particle dosing level and temperature. According to power law index, shear thinning is confirmed for nanolubricant at all temperatures and particle concentrations and significant shear thinning behavior is manifested at volume fractions beyond 0.2%.

Maximum shear thinning (n < 1) is recorded for a volume fraction 0.6%. That is the addition of more nanoparticles results in non-Newtonian behaviour. The possible reasons for the this change can be attributed to; the addition of nanoparticles to the host lubricant alters the structure of the base oil. The nanoparticles act as the interfaces between the oil layers and interconnect them. Since there is difference in Brownian nature of nanoparticles and based fluid particles, the molecular links were broken and tends to initiate new molecular links [41]. If the particle concentration increases the phenomenon of breaking and formation of new links become vigorous and consequently fluid exhibits substantial shear thinning.

The balance of acting forces (Brownian motion, viscous resistance, intermolecular Van-der-Waals interaction, and electrostatic interactions) between ions and dipoles of the nanoparticles and the fluid determines whether the particles to move independently (well-dispersed suspensions) or to move together (agglomerated suspensions) [42]. At higher concentrations, increased nanoparticle agglomerates restrict the fluid flow and as the shear rate increases, de-agglomeration happens and consequently, a dramatic viscosity reduction and non-Newtonian shear-dependent behavior occurs.

Consistency index is the viscosity of a fluid per shear rate. It gives a clear picture of the viscosity of a fluid quantitatively. As the consistency factor increases, the fluid becomes more viscous. It can be seen from Fig. 15, the consistency index of nanolubricant is influenced by both temperature and the particle concentration. The consistency index of



Fig. 13. Logarithmic diagram of shear stress-shear rate at different temperatures, at (a) volume fraction = 0.07%, (b) volume fraction = 0.09%, (c) volume fraction = 0.2%, (d) volume fraction = 0.4%, (e) volume fraction = 0.6%.

the nanolubricant increases with particle volume fraction and decreases with reduction in temperature, which means substantial variation in viscosity. The variation in consistency index at low volume fractions is marginal. For instance, increasing of particle volume fraction from 0 to 0.6% at a temperature of 90 °C the difference in consistency index noted is 0.46, which is an indication of higher consistency of apparent viscosity, on the other hand, the difference between the maximum and minimum values



Fig. 14. Power law index of SiO<sub>2</sub>-PAG nanolubricant with volume fractions at different temperatures.

at a lower temperature of 20 °C is 2.45. ie. consistency index is drastically increased as a result of temperature decrement, which is the clear indication of an increase in apparent viscosity. The basis of this phenomenon is the formation of nano-clusters in the base fluid which is due to Van der Waals force between the fluid's molecules.

Fig. 16 shows the time evolution of shear viscosity for volume fractions 0.07 and 0.6 at different temperatures. The viscosity decreases and shows thixotropic behavior. It is evident that the thixotropic effect is more prominent at higher particle concentrations and lower temperatures. However, at higher shear rates, the influence of shearing time on viscosity is marginal. This can be explained on the basis of nanofluid microstructure and the attractive force between the particles. Nanofluids are structured materials with particles or aggregates, even at low concentrations. So, during the increasing of shear rates, the structure and the flocculants of the nanolubricant break down and this will cause a decrease in viscosity. Increasing the shearing time allow the material to rebuild and retrieve the structure and maintains its viscosity in a constant manner. According to Aladag et al. [43], during the reverse phase,



Fig. 15. Consistency index of SiO2-PAG nanolubricant with volume fractions at different temperatures.

reducing the stress rate may cause a growth of the flocculants and/or allow the particulate network to rebuild. The rebuild of initial structure is influenced by shearing time rather than shear rate.

#### 3.2.4. Modelling of viscosity

Fig. 17 illustrates a comparison between experimental data of viscosities and that obtained from different models. It is observed that classical models underpredict the viscosity of SiO<sub>2</sub>/PAG nanolubricants at higher concentrations. Prasher et al. [44] reported that the Einstein model could not adequately predict the viscosity of propylene glycol and ethylene glycol-based nanofluids. The classical models utilize the parameters like volume concentration and viscosity of the base fluid as the depending factors of effective viscosity. While the experimental results show that the temperature, type of nanoparticle, and shear rate have a noteworthy influence on the effective viscosity of nanofluids. In addition to the aforesaid parameters, there are other critical factors to be considered while modelling of the viscosity of nanofluid/nanolubricant. Anoop et al. reported that there are specific effects of pressure [45] and near-wall velocity on viscosity [46]. According to them, "fluid velocities of nanofluids are slightly higher than those of the base fluid in the wall region and this variation in near-wall shear rate could be used for correction of viscosity measurements while using nanofluids".

# 4. Conclusions

In the present experimental study, thermophysical and rheological properties of SiO<sub>2</sub>/PAG nanolubricant were investigated. The effect of particle concentration and temperature on thermal conductivity and viscosity were elucidated. The experimental results were compared with that obtained from existing models. The rheological properties are investigated at various shear rates, particle concentrations, temperatures. The following conclusions have been drawn from the present experimental investigation.

- (i) The thermal conductivity of the nanolubricant increases with increase in volume concentration and decreases with the intensification of temperature. The maximum thermal conductivity ratio obtained was 1.31 at 0.6 vol% and 20 °C.
- (ii) The classical models underpredict the thermal conductivity and viscosity of SiO<sub>2</sub>-PAG nanolubricant. The model proposed by Redhwan et al. for SiO<sub>2</sub> based nanolubricant overpredicts the thermal conductivity.
- (iii) The viscosity of nanolubricants increases with increase in volume fraction and decreases with increase in temperature. Moreover, the viscosity augmentation was found to be higher compared to that of thermal conductivity.
- (iv) The maximum enhancement in viscosity is observed as 10.84, which is found at a volume fraction 0.6% and shear rate 3.75/s.
- (v) Unlike pure lubricant, shear rate plays a vital role on the behavior of nanolubricant.
- (vi) Nanolubricant shows significant thixotropic behavior at higher particle concentrations and lower temperatures
- (vii) Non-Newtonian shear thinning of nanolubricant was evidenced. However at higher shear rates, shear thinning is insignificant and nanolubricant behaves almost like a Newtonian fluid.
- (viii) At elevated temperatures and shear rates SiO<sub>2</sub>-PAG nanolubricant sustains adequate viscosity than pure lubricant.
- (ix) This novel type of nanolubricant shows outstanding potential for replacements as an advanced lubricant.

#### Acknowledgment

The authors are grateful to Sophisticated Test and Instrumentation Centre (STIC), Cochin University of Science and Technology, India for the assistance in material characterization.



Fig. 16. Influence of shearing time on viscosity (a) at 0.07 vol%, 20 °C, (b) at 0.07 vol%, 40 °C, (c) 0.07 vol%, 60 °C, (d) 0.6 vol%, 20 °C (e) 0.6 vol%, 40 °C (f) 0.6 vol%, 60 °C.

# References

- [1] M.L. Levin, M.A. Miller, Maxwell's treatise on electricity and magnetism, Clarendon Pressvol, Second ed, 135, 1981, p. 425.
- Ja. Eastman, S.U.S. Choi, S. Li, LJ. Thompson, S. Lee, Enhanced thermal conductivity through the development of nanofluids, MRS Proc. 457 (1997) 3–11, https://doi.org/ [2] 10.1557/PROC-457-3.
- [3] K. Sepyani, M. Afrand, M. Hemmat Esfe, An experimental evaluation of the effect of ZnO
- nanoparticles on the rheological behavior of engine oil, J. Mol. Liq. 236 (2017) 198–204. [4] M. Kole, T.K. Dey, Investigation of thermal conductivity, viscosity, and electrical conductivity of graphene based nanofluids, J. Appl. Phys. 113 (2013) 1–8 084307 https://doi.org/10.1063/1.4793581.
- [5] M.A. Akhavan-Behabadi, F. Hekmatipour, S.M. Mirhabibi, B. Sajadi, Experimental investigation of thermal-rheological properties and heat transfer behavior of the heat



Fig. 17. Comparison between experimental viscosity ratio and model predictions at room temperature (30 °C).

transfer oil-copper oxide (HTO-CuO) nanofluid in smooth tubes, Exp. Thermal Fluid Sci. 68( (2015) 681–688, https://doi.org/10.1016/j.expthermflusci.2015.07.008.

- [6] M.H. Esfe, S. Saedodin, O. Mahian, S. Wongwises, Thermal conductivity of Al2O3/ water nanofluids, J. Therm. Anal. Calorim. 119 (2014) 1817–1824, https://doi.org/ 10.1007/s10973-014-3771-x.
- [7] W. Duangthongsuk, S. Wongwises, Measurement of temperature-dependent thermal conductivity and viscosity of TiO<sub>2</sub> -water nanofluids, Exp. Thermal Fluid Sci. 33 (2009) 706–714, https://doi.org/10.1016/j.expthermflusci.2009.01.005.
   [8] B. Buonomo, O. Manca, L. Marinelli, S. Nardini, Effect of temperature and sonication
- [8] B. Buonomo, O. Manca, L. Marinelli, S. Nardini, Effect of temperature and sonication time on nanofluid thermal conductivity measurements by nano-flash method, Appl. Therm. Eng. 91( (2015) 181–190, https://doi.org/10.1016/j.applthermaleng.2015. 07.077.
- ??? M. Afrand, D. Toghraie, B. Ruhani, Effects of temperature and nanoparticles concentration on rheological behavior of Fe<sub>3</sub>O<sub>4</sub> - Ag/EG hybrid nanofluid: an experimental study, Exp. Thermal Fluid Sci. 77 (2016) 38–44, https://doi.org/10.1016/j. expthermflusci.2016.04.007.
- [10] O.A. Akbari, D. Toghraie, A. Marzban, G. Ahmadi, The effect of velocity and dimension of solid nanoparticles on heat transfer in non-Newtonian nanofluid, Phys. E Low-dimensional Syst. Nanostructures, 2016 Accepted manuscript https://doi.org/ 10.1016/j.physe.2016.10.013.
- [11] E. Shahsavani, M. Afrand, R. Kalbasi, Experimental study on rheological behavior of water–ethylene glycol mixture in the presence of functionalized multi-walled carbon nanotubes: a novel correlation for the non-Newtonian nanofluid, J. Therm. Anal. Calorim. (2017) 1–9.
- [12] M. Hemmat Esfe, S. Saedodin, An experimental investigation and new correlation of viscosity of ZnO-EG nanofluid at various temperatures and different solid volume fractions, Exp. Thermal Fluid Sci. 55 (2014) 1–5 doi: 01110.1016/j.expthermflusci. 2014.02.011.
- [13] M.A. Serebryakova, S.V. Dimov, S.P. Bardakhanov, S.A. Novopashin, International journal of heat and mass transfer thermal conductivity, viscosity and rheology of a suspension based on Al<sub>2</sub>O<sub>3</sub> nanoparticles and mixture of 90% ethylene glycol and 10% water, Heat Mass Transf. 83 (2015) 187–191, https://doi.org/10.1016/j. ijheatmasstransfer.2014.12.002.
- [14] S. Bobbo, L. Fedele, A. Benetti, L. Colla, M. Fabrizio, C. Pagura, S. Barison, Viscosity of water based SWCNH and TiO<sub>2</sub> nanofluids, Exp. Thermal Fluid Sci. 36( (2012) 65–71, https://doi.org/10.1016/j.expthermflusci.2011.08.004.
- [15] S. Aberoumand, A. Jafarimoghaddam, M. Moravej, H. Aberoumand, K. Javaherdeh, Experimental study on the rheological behavior of silver-heat transfer oil nanofluid and suggesting two empirical based correlations for thermal conductivity and viscosity of oil based nanofluids, Appl. Therm. Eng. 101( (2016) 362–372, https://doi. org/10.1016/j.applthermaleng.2016.01.148.
- [16] M. Afrand, Experimental study on thermal conductivity of ethylene glycol containing hybrid nano-additives and development of a new correlation, Appl. Therm. Eng. 110 (2017) 1111–1119.
- [17] E. Dardan, M. Afrand, A.H. Meghdadi Isfahani, Effect of suspending hybrid nanoadditives on rheological behavior of engine oil and pumping power, Appl. Therm. Eng. 109 (2016) 524–534.
- [18] M. Afrand, K. Nazari Najafabadi, N. Sina, M.R. Safaei, A.S. Kherbeet, S. Wongwises, M. Dahari, Prediction of dynamic viscosity of a hybrid nano-lubricant by an optimal artificial neural network, Int. Commun. Heat Mass Transf. 76 (2016) 209–214.

- [19] M. Vafaei, M. Afrand, N. Sina, R. Kalbasi, F. Sourani, H. Teimouri, Evaluation of thermal conductivity of MgO-MWCNTs/EG hybrid nanofluids based on experimental data by selecting optimal artificial neural networks, Phys. E Low-Dimensional Syst. Nanostructures, 85, 2017, pp. 90–96.
- [20] M.A. Kedzierski, The effect of lubricant concentration, miscibility, and viscosity on R134a pool boiling, Int. J. Refrig. 24 (2001) 348–366.
- [21] M.A. Kedzierski, Effect of concentration on R134a/Al<sub>2</sub>O<sub>3</sub> nanolubricant mixture boiling on a reentrant cavity surface, Int. J. Ref. 49 (2014) 36–48, https://doi.org/10. 1016/j.ijrefrig.2014.09.012.
- [22] N. Subramani, M.J. Prakash, Experimental studies on a vapour compression system using nanorefrigerants, Int. J. Eng. Sci. Technol. 3 (2011) 95–102.
- [23] D.S. Kumar, R. Elansezhian, Experimental study on Al<sub>2</sub>O<sub>3</sub> -R134a nano refrigerant in refrigeration system, Int. J. Mod. Eng. Res. 2 (5) (2012) 3927–3929.
- [24] R. Wang, Q. Wu, Y. Wu, Use of nanoparticles to make mineral oil lubricants feasible for use in a residential air conditioner employing hydro-fluorocarbons refrigerants, Energ, Build. 42 (2010) 2111–2117.
- [25] R. Krishna Sabareesh, N. Gobinath, V. Sajith, S. Das, C.B. Sobhan, Application of TiO2 nanoparticles as a lubricant-additive for vapor compression refrigeration systems – an experimental investigation, Int. J. Refrig. 35 (2012) 1989–1996, https://doi.org/ 10.1016/j.ijrefrig.2012.07.002.
- [26] M.Z. Sharif, W.H. Azmi, A.A.M. Redhwan, R. Mamat, T.M. Yusof, Performance analysis of SiO2/PAG nanolubricant in automotive air conditioning system, Int. J. Refrig. 75 (2017) 204–216.
- [27] X. Liu, N. Xu, W. Li, M. Zhang, L. Chen, W. Lou, X. Wang, Exploring the effect of nanoparticle size on the tribological properties of SiO2/polyalkylene glycol nanofluid under different lubrication conditions, Tribol. Int. 109 (2017) 467–472.
- [28] A.A.M. Redhwan, W.H. Azmi, M.Z. Sharif, R. Mamat, N.N.M. Zawawi, Comparative study of thermo-physical properties of SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> nanoparticles dispersed in PAG lubricant, Appl. Therm. Eng. 116 (2017) 823–832, https://doi.org/10.1016/j. applthermaleng.2017.01.108.
- [29] ASTM D5334-14, Standard Test Method for Determination of Thermal Conductivity of Soil and Soft Rock by Thermal Needle Probe Procedure, ASTM International, West Conshohocken, PA, 2014www.astm.org.
- [30] IEEE STD 442, IEEE Guide for Thermal Resistivity Measurements, The Institute of Electrical and Electronics Engineers, Inc., New York, NY, 1981.
- [31] R. Hamilton, O.K. Crosser, Thermal conductivity of heterogeneous two component systems, Ind. Eng. Chem. Fundam. 1 (3) (1962) 187–191.
- [32] E.V. Timofeeva, A.N. Gavrilov, J.M. Mccloskey, Y.V. Tolmachev, S. Sprunt, L.M. Lopatina, J.V. Selinger, Thermal conductivity and particle agglomeration in alumina nanofluids: experiment and theory, Phys. Rev. 76 (2007) 28–39, https://doi.org/10. 1103/PhysRevE.76.061203.
- [33] W. Yu, S.U.S. Choi, The role of interfacial layers in the enhanced thermal conductivity of nanofluids: a renovated Maxwell model, J. Nano Part. Res. 5 (2003) 167–171.
- [34] ASTM D2196-10, Standard Test Methods for Rheological Properties of Non-Newtonian Materials by Rotational (Brookfield Type) Viscometer, ASTM International, West Conshohocken, PA, 2010www.astm.org.
- [35] H. Einstein, C. Journals, A. Contact, M. Iopscience, Investigations on the Theory of the Brownian Movement, Dover, New York, 1956.
- [36] H.C. Brinkman, The viscosity of concentrated suspensions and solutions, J. Chem. Phys. 571 (1952) 1–2.
- [37] B. Pak, Y. Cho, Hydrodynamic and heat transfer study of dispersed fluids with submicron metallic oxide, Exp. Heat Transfer 11 (1998) 37-41.
- [38] X. Wang, X. Xu, Thermal conductivity of nanoparticle fluid mixture, J. Thermo. Phy. Heat Transfer. 13 (1999) 474–480.
- [39] S. Shen, A. Narayanaswamy, G. Chen, Surface phonon polaritons mediated energy transfer between nanoscale gaps, Nano Lett. 3 (2009) 2909–2913.
- [40] G. Domingues, S. Volz, K. Joulain, J. Greffet, Heat transfer between two nanoparticles through near field interaction, Phys. Rev. Lett. 85901 (2005) 2–5, https://doi.org/10. 1103/PhysRevLett.94.085901.
- [41] M.H. Esfe, Experimental investigation, model development and sensitivity analysis of rheological behavior of ZnO/10W40 nano-lubricants for automotive applications, Phys. E Low-dimensional Syst. Nanostructures, 2017.
- [42] E.V. Timofeeva, M.R. Moravek, D. Singh, Improving the Heat Transfer Efficiency of Synthetic Oil with Silica Nanoparticles Improving the Heat Transfer Efficiency of Synthetic Oil with Silica Nanoparticles, 2011 1–25.
- [43] B. Aladag, H. Salma, N. Doner, T. Mar, D. Steven, To Cite This Version: HAL Id: hal-00707410 Experimental Investigations of the Viscosity of Nanofluids at Low Temperatures, 2012 876–880.
- [44] R. Prasher, D. Song, J. Wang, P. Phelan, Parametric experimental study of viscosity of Nanofluids, ASME, Int. Mech. Eng. Congr. Expo. 2006 (2006) 21–24.
- [45] K. Anoop, R. Sadr, M. Al-Jubouri, M. Amani, Rheology of mineral oil-SiO2nanofluids at high pressure and high temperatures, Int. J. Therm. Sci. 77 (2014) 108–115.
- [46] A. Kanjirakat, R. Sadr, Near-wall velocity profile measurement for nanofluids, AIP Adv. 6 (2016)https://doi.org/10.1063/1.4939986.