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Exploiting the thermal and rheological potentials of graphene-PAG nanolubricant for the development of energy efficient refrigeration systems

S.S. Sanukrishna [a,*], M. Jose Prakash [b]

[a] Department of Mechanical Engineering, Sree Chitra Thirunal College of Engineering, Thiruvananthapuram 695018, India
[b] Department of Mechanical Engineering, TKMIT, Kollam 691505, India

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ABSTRACT

The addition of nanoparticles is one of state of the art methods to enhance the thermophysical and heat transfer characteristics of cooling and lubricating fluids. Exploring the energy-saving potentials of novel material graphene as a lubricant additive is the primary focus of this study. The thermal and rheological properties of Poly Alkylene glycol(PAG)-graphene nanolubricant at different volume fractions are investigated to pose as an energy-efficient alternative lubricant in refrigeration systems. Moreover, genetic algorithm based regression correlations are proposed to predict the thermal conductivity and viscosity of the graphene-PAG nanolubricant. The results show that the addition of graphene nanoplatelets to the oil has the potential to improve the thermophysical characteristics of the lubricant. The presence of platelet shaped nano graphene particles enhance the thermal and rheological characteristics of the colloidal suspension. The proposed regression models exhibit excellent agreement with the experimental data. Thermal and rheological studies revealed that the application of graphene-PAG nanolubricant is an option to improve the energy efficiency and overall performance of HVAC systems.

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1. Introduction

The world energy utilization is expanding exponentially per year. The growth in numbers of refrigeration and air-conditioning systems both in commercial, industrial, and residential sectors are one of the major reasons for the expanding pattern of energy consumption across the globe. In view of energy security and environmental concern, the energy efficiency of such systems needs to be enhanced. The emergence of nanotechnology leads to the development of superior materials and heat transfer media having better thermal, mechanical, and morphological characteristics which will benefit in energy, communication, biomedical, and other diverse sectors[1,2]. Increasing demands in energy, miniaturization of systems, and precarious economic crisis mandate energy-efficient and high performing cooling media and lubricating fluids in diverse thermal applications and systems.

One of the fortunate approaches to increase the thermal transport phenomena in conventional heat transfer fluids is the use of nanoparticles as colloidal material [3]. Low thermal conductivity and heat transport capabilities are the primary hurdles to overcome for the development of sustainable and efficient systems in the arena of heat transfer. Thus it is imperative to increase the thermo physical and heat transport characteristics of these fluids from an energy-conserving perspective. Present and future HVAC systems would necessitate energy efficient primary and secondary working fluids such as refrigerants and lubricants. Consequently, investigations on energy saving through heat transfer enhancement and better tribological performance have gained great attention nowadays [4]. The addition of nanoparticles to the conventional heat transfer fluids as a heat transfer modifier have been gained consideration for the past few years [5]. Recently carbon and its novel allotropes such as graphene, fullerene, and nanotubes have emerged as an excellent additive to the coolant, lubricants, and heat transfer fluids including refrigerants, due to its remarkable, unique thermal, mechanical, electrical, and optical properties[67]. However, graphene, among them is not effectively

* Corresponding author.
E-mail address: sanukrishna@sctce.ac.in (S.S. Sanukrishna).

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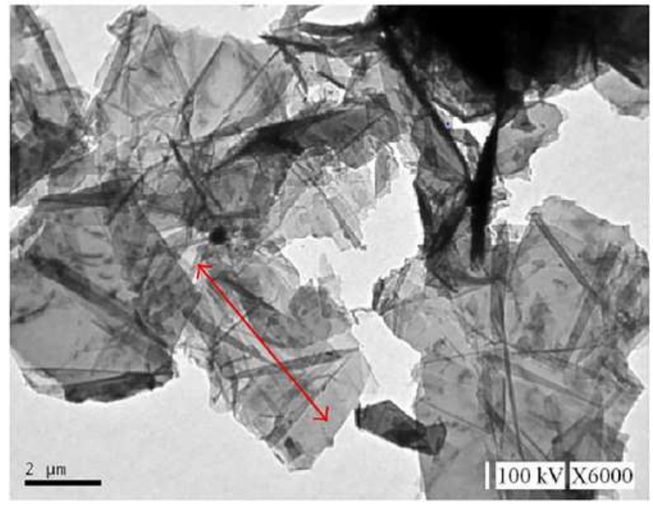

Fig. 1 (*continued*)

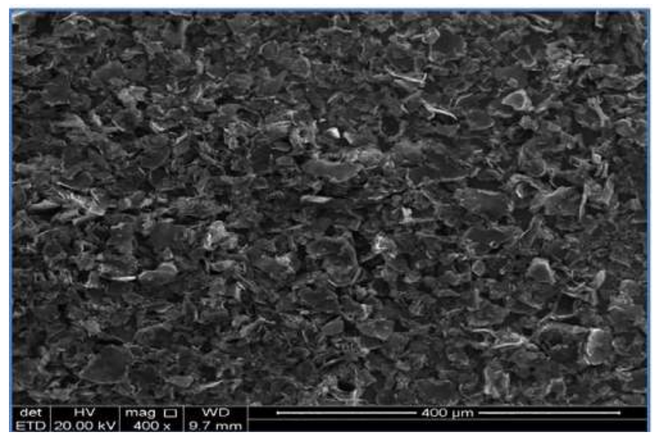

Fig. 1. (a) SEM image of graphene nanoparticles (b) TEM image of graphene suspension.

explored as an additive to the lubricants and refrigerants to enhance the thermo physical and tribological performance. The innovative lubricants appended with novel one-dimensional graphene nano-platelets are expected to have outstanding potentials in energy conservation outlook, especially in refrigeration and air conditioning systems. The application of ultra-fine metal, ceramic, oxides, composite particles has been employed as an additive to lubricating oils for improving their thermo physical, heat transfer, friction reduction, and anti-wear properties[8,9,10,11]. The studies about the thermal, rheological and tribological properties of coolants, lubricants, thermal fluids, etc. suspended with nano-sized particles in the literature emphasize the heat transfer augmenting potentials [12,13,14]. Many researchers have established the potentials of nano-sized metallic, ceramic and oxide particles as an additive to improve thermo physical properties thermo fluids [1516].The rheological behavior of nano lubricants is least characterized researchers [1718,14], few of them established the temperature and particle concentration dependence on these properties. The nanolubricants display reduced apparent viscosity at elevated temperature, more over with smaller particle size, significant increase in viscosity has not been observed. However reverses trends were reported with increased volume fraction [19]. As far as the rheological properties of nanofluids are concerned, we cannot forecast a specific behaviour. Different nanoparticles having different morphological characteristics, which may exhibit unusual behaviour within the host fluids. Aberoumand et al. [16] reported that Ag based nano-oil behaved as a non-Newtonian fluid at very low particle mass fraction. The comprehensive experimental studies concerning the thermal, rheological and tribological features of graphene nano-palatelets appended with poly alkylene glycol based refrigerant compressor oil is not reported in literature. Unfortunately the studies pertaining to the shear dependent viscosity and flow behaviour of graphene-PAG nanolubricants and correlations to predict the behaviour are unavailable in literature to the best of author's knowledge and belief. In the present investigation, comprehensive studies on the thermophysical and rheological behaviours of graphene-PAG (Refrigerant compressor oil) nanolubricant at different volume fractions, temperature and shear rates have been performed to ascertain its effects according to ASTM standards. Moreover, first time, a genetic algorithm based regression correlations are suggested to predict the thermal conductivity and viscosity of graphene-PAG nanolubricant by considering the effect of particle concentration, temperature, density and shear rate. These studies eventually lead to the development of energy-efficient refrigeration systems.

2. Materials and characterization

Graphene nano-platelets procured from SIGMA ALDRICH (USA) are employed for the experimental investigation. The commercially available, fully synthetic, Polyalkylene Glycol (PAG) lubricant is used as the base lubricant. It is well known that Glycol based lubricants show excellent tribological performance compared to mineral oils when used together with HFCs. The morphological characteristics such as shape, size etc., of the nanomaterials are performed with the help of Scanning Electron Microscopy (SEM) and Transmission Electron Microscopy (TEM). The distribution and shape of dry graphene nanoparticles and nanoparticle suspension are shown in the SEM and TEM images Fig. 1 (a) and 1(b) respectively. SEM images of the particles show that they are platelets in shape and are seems to be flocculated in the dry state. These agglomerates can be segregated using a magnetic stirrer and ultrasonic agitator during the preparation of nanolubricants. According to TEM images the suspension is found to be homogeneous and nano-platelets are well dispersed and distributed within the host fluid. It is also observed that the nanoparticles exhibit almost similar morphological characteristics such as physical appearance, shape, and size in the suspension too.

The two-step method, is employed to prepare the nanolubricants at various volume fractions. Five different particle concentrations are considered for the studies (0.07 to 0.6 vol%). The primary disintegration process of platelets within the base lubricant was carried out with the aid of magnetic stirrer for 1 h, and the samples are ultrasonically agitated using an ultrasonic agitator at a frequency of 40 kHz to homogenize the colloid, for 6 h. Visual inspections are performed first to identify any indication of sedimentation or coagulation. No evidence was noticed after five days of preparation according to visual inspection. In order to quantify the stability of nanolubricant samples, U-V visible spectroscopy analysis is conducted.

2.1. Measurement of stability

The ultraviolet–visible spectroscopy is widely used to analyse fluids and solids with the help of ultraviolet radiant energy and visible and near-infrared energy of the electromagnetic spectrum. The transmitted light from the sample provides the absorbance spectra of the sample fluid. The analytical values can be represented in terms of the absorbance of light energy. The basic principle of analysis is Bouguer-Beer law or Beer-Lambert rule, which states that the absorbance of a solution is directly proportional to the concentration of the species in the solution and the path length. In the present study, the stability of nanolubricants is measured with UV–vis spectroscopy by determining the absorbance after the first and fifth days of preparation.

2.2. Measurement of thermal conductivity

The thermal conductivity of base lubricant and nanolubricants at different volume concentrations are measured with KD2 Pro thermal property analyser (Fig. 2). The principle of operation is the transient hot wire. Thermal conductivity with an accuracy of ± 0.001 could be measured by this device. The apparatus meets international standards such as ASTM D5334-14 and IEEE 442 – 1981. A constant temperature circulator with an accuracy of 0.01 °C is used to maintain the temperature of the sample constant. Measurements are conducted repetitively. Based on the deviation between the thermal conductivity of calibration standard fluid and the measured thermal conductivity, the measurement error is estimated and the thermal conductivity data reported in the study represents an average of fifteen measurements with an estimated standard error of ± 1.2%. The thermal conductivity of nanolubricant at different volume fractions is determined (0.07 to 0.6 vol %) in the temperature range of (20 °C to 50 °C).

Fig. 2. KD2 Pro thermal property analyser.

2.3. Rheological characterization

The rheological behaviour of the pure lubricant and nanolubricants are observed with a Brookfield rotational type rheometer (Fig. 3), having a measurement range between 1.0 and 2000 cP. ASTM D2196-10 standard is followed for the investigations. The amount of viscous drag is proportional to the amount of torque required to rotate the spindle. Rheocalc software along with a PC based data acquisition system is employed to record the measurements. The maximum uncertainty in the measurement is found to be 1.6%. The experiments are conducted with different volume fractions (0.07 to 0.6 %), and temperatures (20–90 °C). The shear rate is varied (3.75 s^{-1} to 225 $s^{-1)}$ to explicate the effect of shear rate on the viscosity and flow behaviour of the nanolubricant. Five sets of data corresponding to each test condition are logged and average values are considered for the representation of experimental results.

2.3.1. The power law model

The flow behaviour of the pure lubricant and graphene-PAG nanolubricant are predicted through Ostwald De Waele or power-law model, which is the most generalized model for the flow prediction of non-Newtonian fluids. According to Ostwald–De Waele power-law model (Eqn.1) the shear stress and rate of shear can be correlated as

$$\tau = m\gamma^n \quad (1)$$

The Power Law model is described by two parameters, consistency coefficient or consistency index (m) and flow behaviour index (n). The consistency index pertains to the consistency of the fluid at a particular moment, which portrays how much viscous the fluid is. If the magnitude of n < 1, the fluid is known as shear-thinning or pseudoplastic fluid. This means that the apparent viscosity decreases with an increase in shear rate. When n greater than 1, it is shear-thickening or dilatant fluid, i.e. their apparent viscosity increases as shear rate increases. In order to obtain these indices, rheogram of shear stress and the shear rate has been drawn and the indices are calculated.

2.4. Modelling of thermo physical properties of graphene-PAG nanolubricant

Genetic algorithm-based regression correlations have been generated to predict the thermal conductivity and viscosity of graphene-PAG nanolubricants, using Eureqa Data Robot, the A.I. powered modelling engine. The evolutionary algorithms generate solutions according to the theory, survival of the fittest, to optimize problems and predictive regression correlations using methods motivated by natural evolution, such as inheritance, mutation, selection, and crossover. A population of individuals is maintained within search space and each representing a possible solution to a given problem. These individuals are similar to chromosomes and the variables are analogous to genes. During the successive generation, a proportion of the existing population is selected to breed a new generation. The various steps involved in the genetic algorithm are shown in Fig. 4. The regression curves are fitted with R-squared values greater than 0.998.

3. Results and discussion

3.1. Dispersion stability of nanolubricants

Fig. 5 shows the UV–vis spectra of the nanolubricant after the 1st and 5th days of preparation. The maximum absorbance is obtained within a wavelength range of 200 to 320 nm. It is clear

Fig. 3. Brookfield DV-II + Pro rheometer.

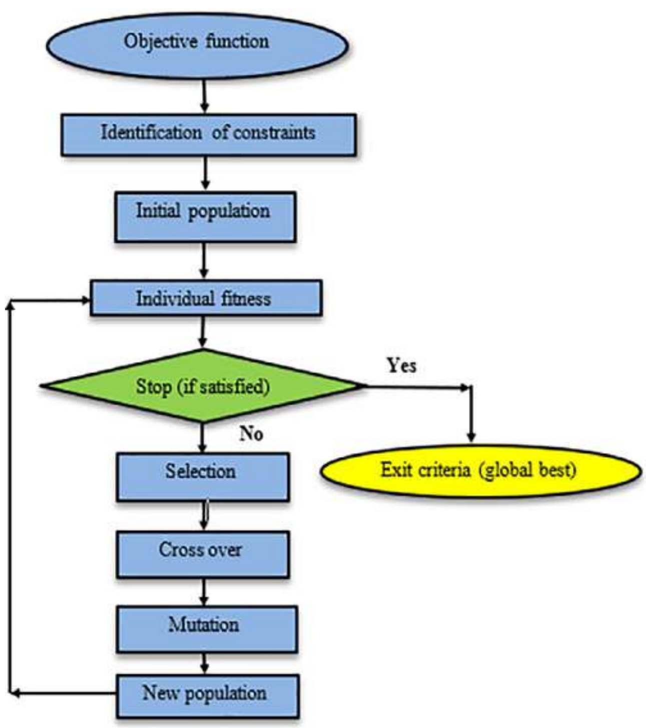

Fig. 4. Steps involved in a genetic algorithm.

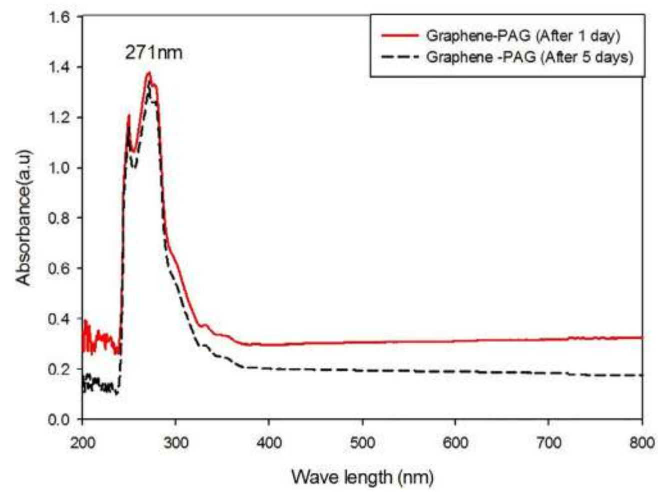

Fig. 5. Uv–*vis* spectrum of graphene nanolubricant (0.6 vol%).

from the figures that the absorbance value is increased first to a peak at a particular wavelength and thereafter gradually decreases from Uv to the near-infrared region. Here the peak absorbance obtained for graphene nanolubricant is almost the same (1.39) and which is an indication of stability. This is the sign of the presence of a higher population of nanoparticles in the base lubricant to interact with the light.

3.2. Thermal conductivity

Fig. 6 depicts the thermal conductivity of nanolubricant as a function of nanoparticle volume fraction at various temperatures. The nanolubricants exhibit higher thermal conductivity than that of pure lubricant at all particle volume fractions. The thermal conductivity of the colloid increases with increase in graphene particle concentration. The maximum enhancement in the thermal conductivity ratio obtained was 1.48 at a volume fraction of 0.6% and temperature 20 °C. There major reasons for this enhancement are :(i) The Brownian motion effect. The heat is carried by phonons by propagating lattice vibrations in the crystalline solids suspended in fluids. Such phonons are propagating in a random direc-

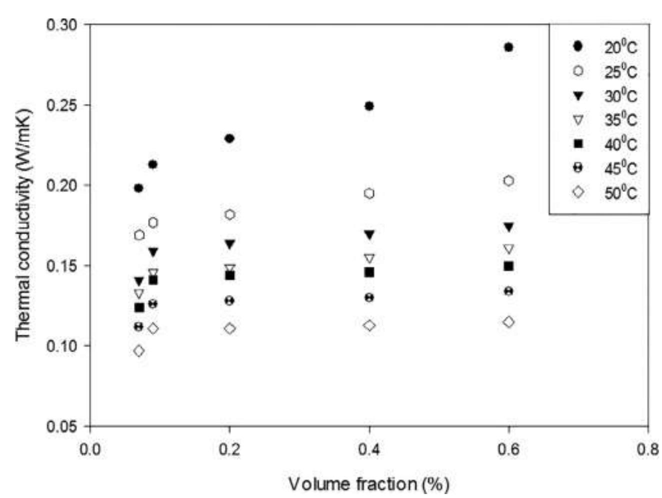

Fig. 6. Effect of volume fraction on thermal conductivity of graphene-PAG nanolubricant.

tion. Some ballistic phonon effects could lead to an increase in thermal conductivity. If the ballistic phonons initiated in one particle can reach a nearby particle. (ii) Interfacial molecular layering of the liquid: Existence of a 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 fluid and solid particles. (iii) The organized chain like clustering of particles with in the fluid and the increased thermal conductivity of suspended solid particles. The thermal conductivity of pure lubricant and nanolubricants at all particle concentrations decreases with an increase in temperature. This is due to the following phenomena:(i) at lower concentrations and at elevated temperatures, the mean path between the nanoparticle increases and thus reduces the probability of collision between particles and may also leads to decline the near-field radiation effect .

3.3. Modelling of thermal conductivity of graphene nanolubricant

The correlations proposed to forecast the thermal conductivity of graphene-PAG nanolubricants by considering various parameters such as temperature, particle concentration and thermal conductivity of base fluid and particle is shown in Eqn.2. A comparison of experimental thermal conductivity and that predicted from the available models in literature and the proposed model is presented in the subsequent sections. The proposed can predict the thermal conductivity of graphene-PAG nanolubricant for various temperature and volume fractions. The model is valid for $0.07 \leq \phi \leq 0.6\%$ and $20 \leq T \leq 50$ °C.

$$k_{NL} = 150 + 1.06e4\Phi + \frac{-0.00164}{\Phi} + 0.00468T^2 + 0.654\Phi T^2 + 1.12e - 6\Phi T^4 - 1.45T - 136\Phi T - 5.03e - 6T^3 - 0.0014\Phi T^3 \quad (2)$$

Where Φ the particle concentration in %, T is is the temperature in K, kp, the particle thermal conductivity in W/mK, and kNL is the thermal conductivity of the nanolubricant. The comparison of experimental thermal conductivity and that predicted from the present correlation is portrayed in Fig. 7. The proposed model for graphene-PAG nanolubricant well predicts the experimental data within a residual standard error range of ± 3.6%. (a) shows the comparison of thermal conductivity obtained from experiments and that predicted form present correlation. Fig. 7(b) portrays a comparison between proposed correlation and few models available in the literature (Maxwell [20], Hamillton and Crosser [21], Yu and Choi [22], Timofeeva [23]). From figure it is obvious that the classical models are not good enough to predict the thermal conductivity of graphene-PAG nanolubricants. The classical models considered only the particle concentration and the shape effects as the influencing parameters on the effective thermal conductivity of nanofluids, from this study, it is evident that temperature plays significant role in the thermal conductivity of colloidal suspensions. Moreover, the classical models are formulated to predict the thermal conductivity of low viscous base fluids suspended with metals and oxide which are in spherical shapes. Models to predict thermal conductivity of nanolubricant, in particular with glycol based lubricants suspended with platelet shaped graphene are scarce.

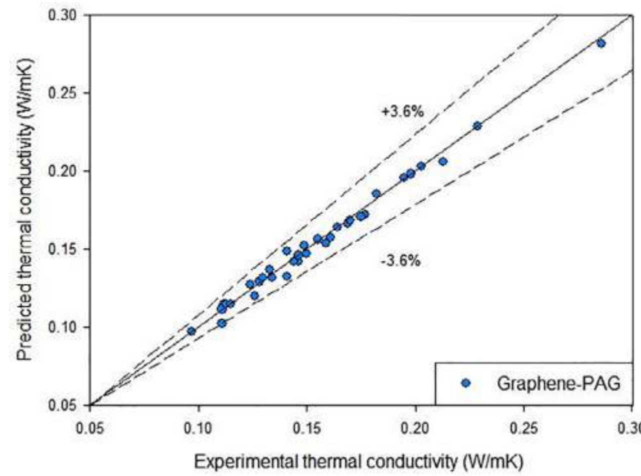

Fig. 7. Comparison of thermal conductivity (a) experimental vs correlation results (b) experimental vs model predictions.

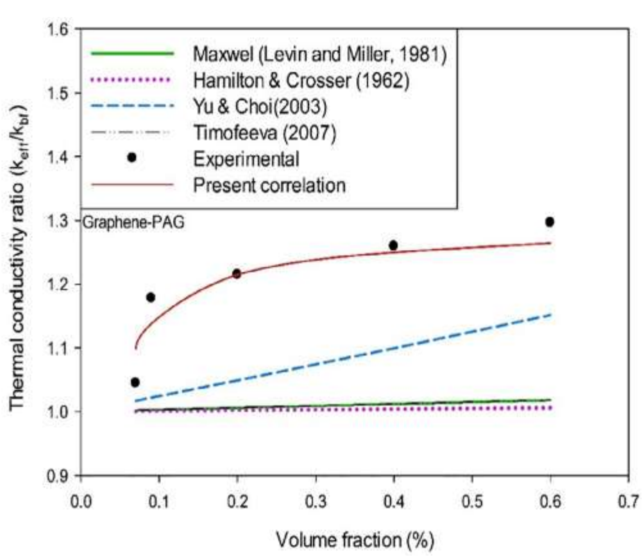

Fig. 7 (*continued*)

3.4. Effect of particle concentration on viscosity of nanolubricants

Fig. 8(a) shows the variation of viscosity with volume fraction at different shear rates. From figure, it is inferred that volume fraction and shear rate have crucial impact on the viscosity of the nanolubricant. As the shear rate increases, the relative viscosity is found to be decreasing. Besides, the nanolubricants sustain adequate relative viscosity at elevated temperatures and moderate shear rates and proves that they are more appropriate to use at elevated temperature and shear rates. This is in line with the operating conditions in refrigerant compressors. Fig. 8(b) provides a better understanding of viscosity variation with temperature. The results reveal that irrespective of the shear rate and volume fraction, the viscosities nanolubricants diminish with an increase in temperature. A similar trend was displayed by pure lubricant as well. This possible reason is, 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 a decrease in viscosity.

3.5. Rheological characterization

The rheological studies were performed over the range of shear rate from 3.75 to 225 s^{-1} covering a wide range of temperatures. Fig. 8 Fig. 9 (a) shows the variation of viscosity with the shear rate for pure lubricant. As expected, pure lubricant exhibits Newtonian behaviour within the shear rate range considered.

The same measurement ranges of shear rates are considered to measure the viscosity of the nanolubricant as well and is represented in Fig. 9(b). The figure portrays the shear and temperature dependent viscosity variation of graphene based nanolubricants at different particle concentrations. Irrespective of the temperature and concentrations, the apparent viscosity diminished with increase in shear rate. The alteration in viscosity of nanolubricant is found to be trivial at lower particle concentrations. Though, significant decrease in viscosity is manifested at higher volume fractions. That is the nanolubricant exhibits the severe non-Newtonian behaviour. The organisation and structural alterations of the intermingling particles can be related to the shear thinning characteristics of the evenly dispersed colloids. The application of shear will make the nano-platelets to orientate in the flow direction. This probably segregates the flocculation of particles in the colloidal suspension and eventually decreases the amount of fluid which immobilized by the nano-racemes. The force of interaction then be decreases and which in turn reduces the resistance against the flow and hence the apparent viscosity. The shear dependent viscosity of the nanolubricant largely decreases for a shear rate lower than 50 s^{-1}. Over this shear rate value, the apparent shear viscosity of the nanolubricants tends to a Newtonian plateau. Fig. 10(a) and 10 (b) show rheogram of pure lubricant and nanolubricant respectively. The curves are fitted with an R-squared value higher than 0.99. Here, n and m represent the power law and consistency indices respectively. From the figures, it is clear that regardless of temperature nanolubricant behave as non-Newtonian fluid with a power-law index less than unity. Meanwhile, the pure PAG oil behaves like a Newtonian fluid with power-law index approximately equal to one.

3.6. Modelling of viscosity of graphene nanolubricants

Most of the viscosity models available in the literature are based on spherical nanoparticles. Suitable models to predict the viscosity of carbon-based nanolubricants are scarce in the literature. Hence new correlations are proposed to predict the viscosity

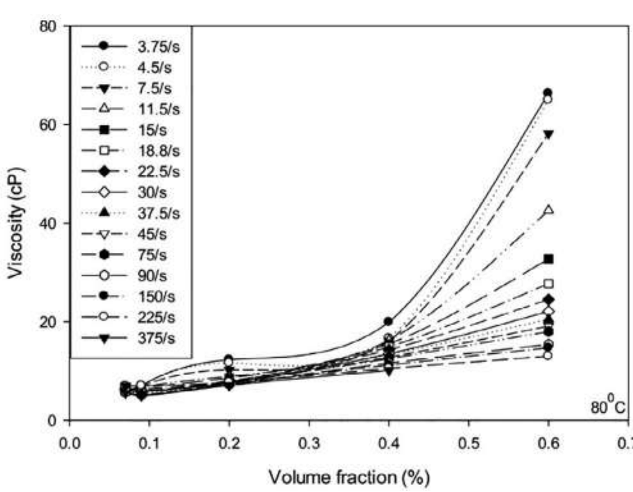

Fig. 8. (a) Variation of viscosity with volume fractions at different shear rates (b) Effect of temperature on viscosity, of nanolubricant.

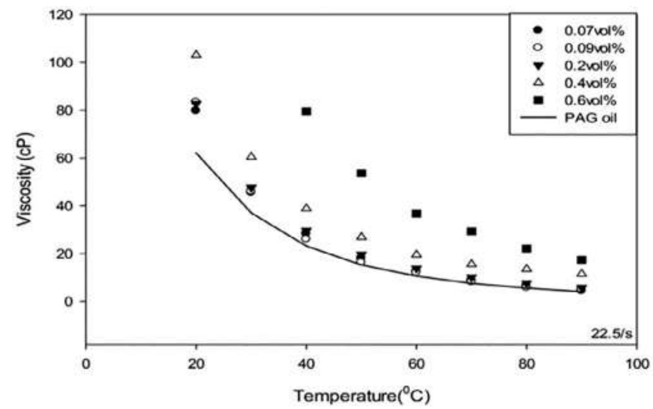

Fig. 8 (*continued*)

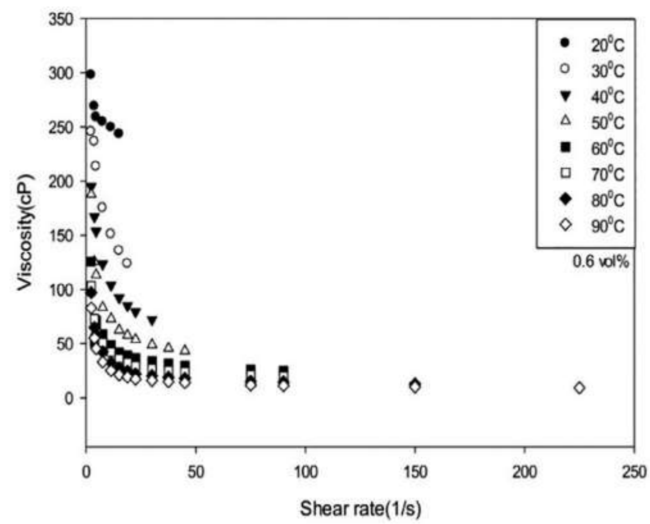

Fig. 9 (*continued*)

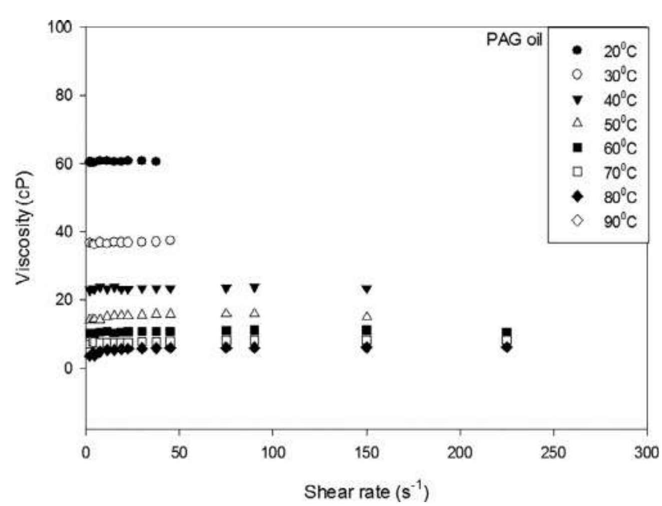

Fig. 9. Viscosity of (a) pure lubricant (b) nanolubricant, with shear rates at different temperatures.

of graphene based nanolubricant.The proposed model for graphene-PAG nanolubricant is shown in Eqn.3

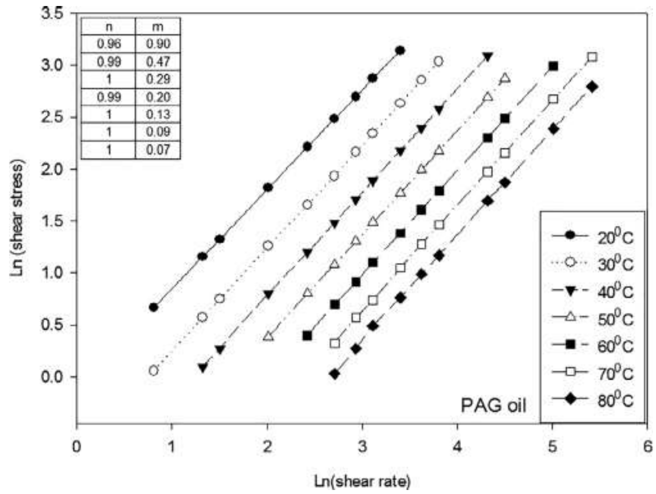

Fig. 10. Rheogram (a) pure lubricant (b) nanolubricant, at different temperatures.

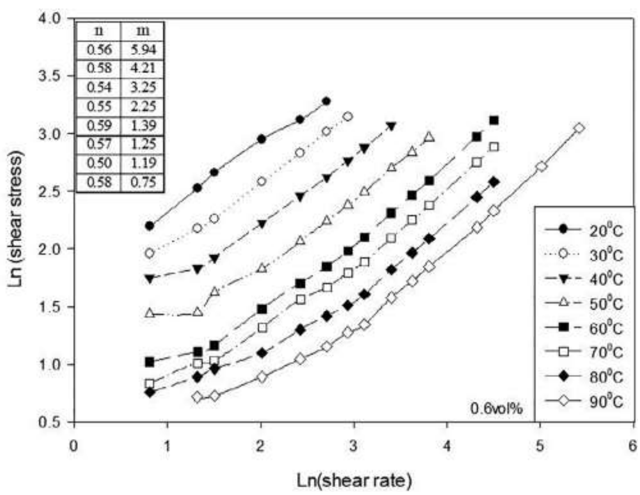

Fig. 10 (*continued*)

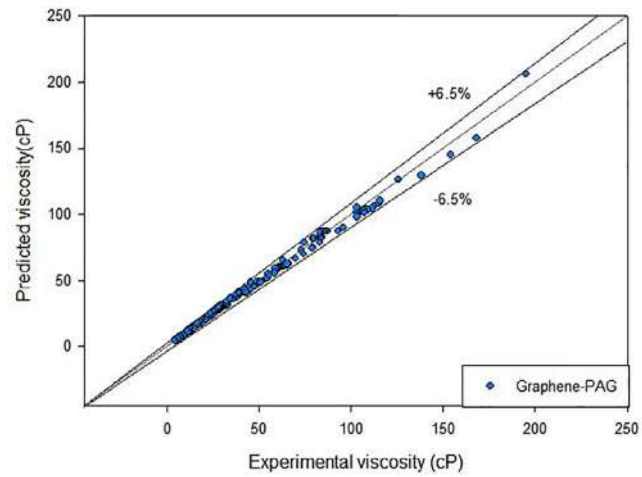

Fig. 11. Comparison between (a) experimental viscosity and predicted viscosity (b) experimental viscosity and model predictions.

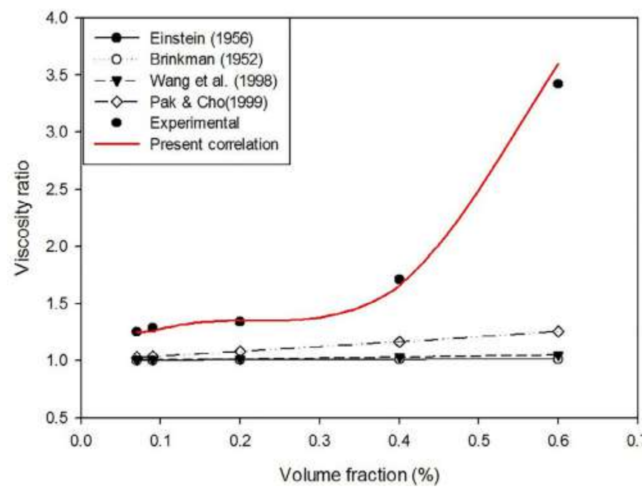

Fig. 11 (*continued*)

$$\mu_{NL} = 9.3\Phi + 0.176T + \frac{2.28e3 + 2.28e3\Phi^2}{T} + \frac{0.000727\left(\frac{3.1e6}{T}\right)^{1.93\Phi}}{Y} - 36 - 0.176\Phi Y \quad (3)$$

The correlation is valid for $0.07 \le \phi \le 0.6\%$, $20 \le T \le 90$ °C and $5 \le \gamma \le 200$ 1/s. where Φ is the particle concentration in %, T is the temperature in K, γ, the shear rate in 1/s and μ_{NL} is the viscosity of the nanolubricant. Fig. 11(a) shows the comparison between the experimental data of viscosity and that predicted from the proposed correlation in the case of graphene-PAG nanolubricant. From the figure, it is obvious that the model is successful in predicting the viscosity of graphene-PAG nanolubricant.

Fig. 11(b) shows the comparison of experimental viscosity with that obtained from a few models available in the literature (Einstein [24], Brinkman [25], Wang et al., [26], Pak and Cho [27]) along with the present correlation. The viscosity at room temperature is used for the comparison. It can be seen from the figure that the classical models fail to predict the viscosity of the graphene nanolubricant, while the present model is in line with the experimental results.

4. Conclusions

Comprehensive experimental investigations on thermal and rheological characteristics of graphene-PAG nanolubricants are carried out. Regression correlations are proposed to forecast the thermal conductivity and viscosity of nanolubricants. The following key conclusions have been drawn from the experimental investigation.

(i) In the range of volume fraction considered, the maximum thermal conductivity ratio obtained for graphene-PAG nanolubricant is 1.48.
(ii) The thermal conductivity of the graphene-PAG nanolubricant increases with an increase in volume concentration and decreases with the intensification of temperature.
(iii) The maximum increase in viscosity is found to be 300 cP at lower shear rate which is about five times higher than the viscosity of pure PAG lubricant.
(iv) Unlike pure lubricant, shear rate plays a vital role in the behavior of the graphene- PAG nanolubricants.

(v) Non-Newtonian shear thinning of nanolubricants was evidenced at lower shear rates. However, as the shear rate exceeds 50/s, shear-thinning is found to be insignificant and CNT nanolubricant behaves almost like a Newtonian fluid.
(vi) Classical models fail to predict the thermal conductivity and viscosity of the graphene-PAG nanolubricants.
(vii) The genetic algorithm based regression models well predict the thermal conductivity and viscosity of nanolubricant.
(viii) Graphene-PAG nanolubricants sustain adequate viscosity than pure lubricant at elevated temperature which is a desirable operating feature for refrigerant compressors.
(ix) Thermal and rheological studies revealed that the potential of Graphene-PAG nanolubricant as an alternative lubricant for refrigerant compressors is excellent.

CRediT authorship contribution statement

S.S. Sanukrishna: Conceptualization, Methodology, Investigation, Software, Writing – original draft. **M. Jose Prakash:** Supervision, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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