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SYNTHESIS AND THERMO-PHYSICAL CHARACTERIZATION OF GRAPHENE BASED TRANSFORMER OIL

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Abstract

Transformer oils serve as coolants and an insulation medium to avoid overheating. Due to low thermal conductivity of transformer oils which may result in overheating and hence nanoparticles are introduced to overcome this challenge. In this research, graphene nanoparticles (GNPs) were dispersed in transformer oil at a concentration of 0.01-0.1 wt% respectively. The stability of the graphene-transformer oil nanofluids (GTNFs) is studied using UV-Vis spectrophotometer.Optical microscope is used to study the dispersion quality and the uniformity of clusters formed at different weight fractions qualitatively. Thermal conductivity and viscosity of the samples were measured over a range of temperature from 20-100 °C. The thermal conductivity and viscosity of GTNFs were observed to be strongly dependent on the temperature and concentration of GNPs. The enhancement in thermal conductivity obtained was from 2.63-69.31 % as the temperatures varies from 20-100 °C, for 0.01-0.1 wt% of GNPs. It was also further observed that the viscosities of the GTNFs were reduced by 8.33-23.97% as the temperature rises allowing more convectional heat flow. It is thus concluded that the increase in thermal conductivity and decrease in viscosity as temperature rises enhances the overall performance of the transformer oil.

Keywords: Transformer oil, Graphene nanofluids, Thermo-physical properties, Thermal conductivity, Viscosity

1. Introduction

Nanotechnology has been expanding widely over the years and was predicted to have the usage of 1 trillion per year starting from year 2015 [1]. It is a technology

Nomenclatures

g	Gravitational acceleration
r_p	Particle radius
v	Sedimentation rate

Greek Symbols

μ	Fluid viscosity
ρ_f	Density of the fluid
ρ_p	Density of the particles

Abbreviations

Ag	Silver
Ag_2Al	Silver Alumina
Al_2Cu	Aluminum Copper
AlN	Aluminium Nitride
Al_2O_3	Aluminium Oxide
Au	Gold
CNT	Carbon Nanotube
CTAB	Cetyltrimethyl Ammonium Bromide
Cu	Copper
CuO	Copper Oxide
$CuSO_4.5H_2O$	Copper(II) Sulfate Pentahydrate
CVD	Chemical Vapor Deposition
EG	Ethylene Glycol
Fe_3O_4	Iron Oxide
GA	Gum Arabic
GNPs	Graphene Nanoparticles
GNS	Graphene Nanosheet
GO	Graphene Oxide
GTNFs	Graphene- Transformer Oil Nanofluids
HCl	Hydrochloric Acid
H_2O_2	Hydrogen Peroxide
H_3PO_4	Phosphorus Acid
H_2SO_4	Sulphuric Acid
$KMnO_4$	Potassium Permanganate
MWCNTs	Multiwalled Carbon Nanotubes
$NaH_2PO_2.H_2O$	Sodium Hypophosphite Monohydrate
NaOH	Sodium Hydroxide
OA	Oleic Acid
$Sb_2O_5:SnO_2$	Antimony-tin Oxide
TiO_2	Titanium Dioxide
ZnO	Zinc Oxide
NACA	National Advisory Committee for Aeronautics
WHO	World Health Organization

which comprises microscopic particles known as nanoparticles with size ranging from 1-100 nm. The five major types of nanoparticles are Carbon Nanotubes (CNT) and Fullerenes, Semiconductors, Metals, Ceramics, and Polymeric. These nanoparticles are exceptionally prominent for its thermal, mechanical, electrical and optical properties. In terms of heat transfer, the thermal properties of the nanoparticles play an important role in enhancing the performance of heat transfer fluids. Heat transfer fluids such as water, transformer oil and ethylene glycol (EG) which are widely used in heating systems, transportation, electronic cooling, petroleum refineries and many more. However, these conventional fluids have considerably low thermal conductivities which cause them to have limited range of applications. Hence, nanoparticles with higher thermal conductivities are dispersed into the heat transfer fluids producing nanofluids with higher heat transfer capabilities [2]. Nanofluids in general are suspensions of nanoparticles dispersed in a base fluid. The addition of micro-sized particles into base fluids is not efficient due to its heavier weight and the gravitational force which causes the sedimentation of particles leading to a degradation in thermal properties [3]. The sedimentation can be significantly reduced or even prevented with nanoparticles which are lighter and smaller in size and thus enhancing the thermal properties of the fluid [4].

Nanostructured carbon materials such as GNPs have been attracting attention due to its high thermal conductivity of 5000 W/mK which is much higher than that of CNT (3000 W/mK) and diamond (2000 W/mK) [5-6]. Graphene is a monolayer graphite which consist of one-atom-thick planar sheet with bonded carbon atoms densely packed together in a honeycomb crystal lattice [7]. Due to the ability of GNPs to enhance the thermo-physical properties of heat transfer fluids, it can be added to transformer oil to increase the overall performance of the transformer. Transformers are used to regulate the voltage supply to homes, processing plants, factories and all other places which use electricity. While performing its duty, transformers also generate electricity which causes an increase in temperature which leads to overheating and failure to operate. Transformer oil which is stored in the transformer tank is used as a source of cooling and electrical insulation of the electrical appliances inside the tank. Thus, it is highly desirable for the oil to have a higher thermal conductivity to enhance the cooling effect. The addition GNPs into base oil has high potential to increase the thermal conductivity which in turn increases the heat transfer rate from the electrical appliances to the oil. Consequently, overheating which causes transformers failure can be prevented.

As of today, there are generally two methods of producing nanofluids. The method used to prepare nanofluids is vital as the thermo-physical properties of the nanofluids can only be studied if the nanoparticles are well dispersed in the base fluid. The one-step method can be divided into chemical methods and physical methods respectively. In 2001, Eastman et al. [8]developed a one-step physical vapour condensation method to reduce agglomeration by direct condensation of metallic vapor into nanoparticles by allowing contact with a low vapour pressure liquid. However, this method is not appropriate for large scale synthesis of nanofluids due to its high cost. Hence, a one-step chemical method is developed. Zhu et al. [9] prepared copper nanofluids using the one-step chemical method by reducing Copper(II) Sulfate Pentahydrate ($CuSO_4.5H_2O$) using Sodium Hypophosphite Monohydrate ($NaH_2PO_2.H_2O$) in EG under microwave irradiation. The copper nanofluids produced were well dispersed and stable. Despite its stability, it has a major disadvantage in which the residues of reactant left in the nanofluids due to incomplete reaction. Apart from the one-step a method, the two-step method is the more widely used to prepare nanofluids. The nanoparticles are first prepared as dry powders by mechanical or chemical exfoliation before dispersing the nano-sized powders into a base fluid. The second processing step of dispersing the powder into the fluid is aided with agitation, ultrasonic agitation, homogenizing or ball milling. The two-step method is considered as the most economic method in producing large scale nanofluids. Rashmi [10] prepared CNT-water nanofluid using the two step methods where CNT was obtained in powder form before dispersing it into water in the presence of dispersant. The CNT-water nanofluids were homogenized at 28000 rpm and sonicated for four hours at room temperature. Gum Arabic (GA) was used to aid the dispersion of CNT and the nanofluids produced were found to be stable for more than 6 months.

With a stable nanofluid produced, its thermo-physical properties such as the thermal conductivity and viscosity are important properties of nanofluid to be studied. It is generally expected to show high thermal conductivity enhancement and lower viscosities. In 2010, Weiyu et al.[11]developed graphene-EG nanofluid which resulted in a thermal conductivity enhancement of 86% with 5.0 vol% of GNPs proving significant increase in thermal conductivity compared to a conventional EG fluid. Choi et al. [12] work showed 20% enhancement in thermal conductivity at 4 vol% of Al_2O_3 nanoparticles. GNPs which has a thermal conductivity of 5000 W/mK is much higher than that of Al_2O_3 (30 W/mK) has a high potential to further enhance the thermal conductivity. In the work done by Madhusree et al. [13], the viscosity of copper oxide (CuO)-gear oil nanofluid decreases significantly with the increase in temperature. Similar observations were also reported by Praveen et al. [14] on CuO-EG nanofluids. The viscosity of an ideal transformer oil should be kept low so that it has less resistance to the convectional flow of oil allowing higher rate of heat transfer.

This study aims to synthesize and evaluate the stability of GTNFs with concentration of GNP ranging from 0.01 to 0.1 wt%, respectively. The thermo-physical properties (thermal conductivity and viscosity) of the stabilized GTNFs were also investigated as a function of temperature and GNPs concentration. Enhanced thermo-physical properties could considerable extend the lifetime and performance of transformer.

2. Methods

2.1. Chemical

GNPs is obtained from Graphene supermarket, USA with purity of 99.2% with average flake thickness of 12 nm and average particle (lateral) size of 4500 nm, respectively. Complimentary naphtha based transformer oil was obtained with the collaboration of Lube World Holdings Sdn. Bhd. Naphtha based oil was also chosen as the oil allows natural convection circulation as sludge present (if any) will not precipitate at the bottom and obstruct the transformer cooling system [15].

2.2. Synthesis of nanofluids

GTNFs were prepared using the two-step method in which a measured amount of dry GNPs were dispersed into measured amount of transformer oil. Different amount of GNPs ranging from 0.01 to 0.1 wt% were selected based on the similarity of weight percentage used by other researchers on other GNPs or other carbon-based nanofluids [10, 16, 17]. The GNPs of different weight percentage were dispersed into transformer oil to produce nanofluids with the total weight of 40 grams. Table 1 shows the amount (in wt. %) of GNPs in each sample.

Table 1. Amount of GNP and transformer oil used in the present study.

Samples	GNPs (wt%)	Transformer Oil (g)
G1	0.01	39.996
G2	0.02	39.992
G3	0.04	39.984
G4	0.08	39.968
G5	0.10	39.960

After the GNPs were dispersed in the transformer oil, the samples were sonicated for 4 hours using a water bath sonicator (Elma Transsonic TI-H-15) at room temperature and 35 kHz. The sonication time was set as a fixed variable based on the stability studies done by Rashmi et al. [18] in which 4 hours of sonication time can maintain the stability of nanofluids for over six months signifying its high stability. Sonication helps to entangle the nanoparticles which are in clusters to prevent agglomeration by means of physical vibrations. Despite that, prolonged sonication may cause deformation of the natural GNPs structure which leads to the inability to fully utilize the properties of the structure [18].

2.3. Stability studies on nanofluid

In this research, the stability of nanofluids prepared was studied using a UV-Vis spectrophotometer (Shimadzu UV-1800, Japan) in which the concentration measurement is based on Beer-Lambert’s law that states that the absorbance is directly proportional to the concentration of the sample.Stability studies were performed for a period of 3 weeks. This method used evaluates the stability of nanofluids prepared quantitatively by measuring the concentration of nanoparticles in the suspension versus sedimentation time. Before the start of the stability tests, the UV-Vis absorbance curve and a linear calibration curve were obtained as shown in Figs. 1 and 2 respectively by exposing light energy with wavelength between 190 nm to 900 nm to the samples. The UV-Spectra obtained for this research was at 570 nm (highest peak). With the wavelength of 570nm for the GTNF, a linear calibration curve was obtained. For the next 3 weeks, the concentrations of samples were measured and the consistency of the concentration readings indicates how well the GNPs dispersed in transformer oil.

2.4. Characterization of nanofluids

The dispersion of the GTNFs were further studied using an optical microscope (Swift M10D Series Digital Microscope) using 40X magnification lens. The usage of the optical microscope is limited as it is an ordinary light microscope and thus it does not give accurate image of the clusters formed. However, it allows qualitative assessment to be done in which the uniformity of clusters formed can be observed.

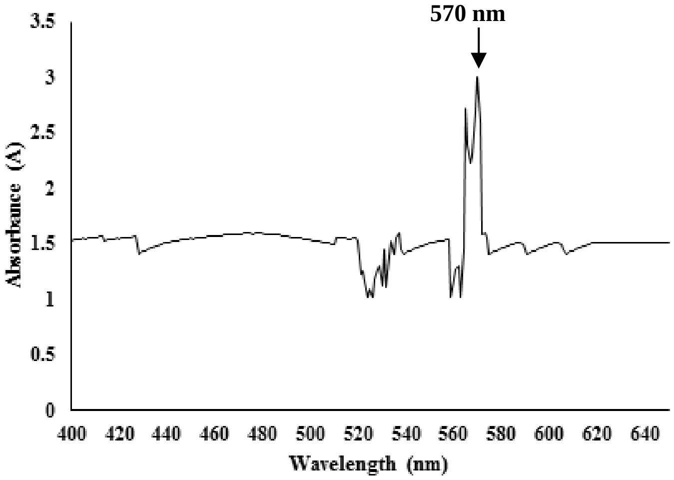


Fig. 1. UV-Vis absorbance curve.

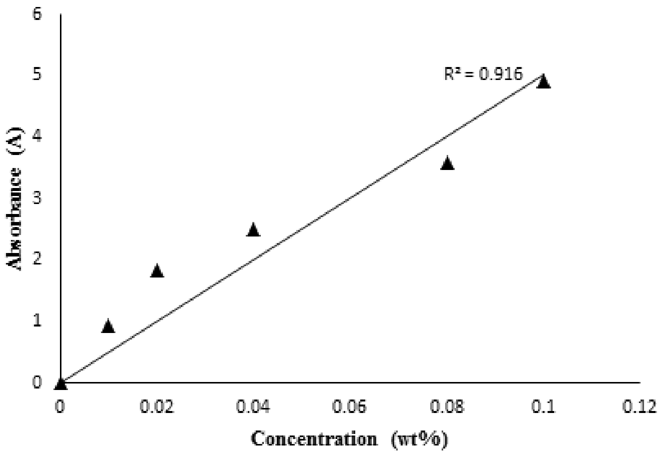


Fig. 2. Calibration curve for different GNPs concentration at 570 nm.

2.5. Thermo-physical properties

2.5.1. Thermal conductivity measurements

A thermal conductivity analyser (KD2 Pro Decagon Device, USA) was used to measure the thermal conductivity of the stable nanofluids of different GNPs concentration (0.01-0.1 wt%) against different temperature. The variation in temperature (20-100 °C) of the nanofluids is done by immersing the nanofluids into an oil bath at 20 °C intervals. This range of temperature was chosen based on the operating temperature of transformer oil at 65 °C which may go up to 95 °C. The thermal conductivity meter was first calibrated using the standard glycerine at room temperature. The probe (60 mm long single needle) from the meter was inserted into the nanofluid samples prepared. The readings are automatically taken at a 15 minutes interval. Only readings with the error with value less than 0.01 were recorded. Approximately 10 readings results were recorded per sample and the average readings were then obtained The percentage enhancement in thermal conductivity of each sample compared to pure transformer oil was calculated using Eq. (1) [18]:

$$Percentage\ Enhancement\ (\%) = \frac{k_{nanofluid} - k_{base\ fluid}}{k_{base\ fluid}} \times 100\% \tag{1}$$

2.5.2. Viscosity measurements

The viscosities of the nanofluids were measured using the viscometer (Brookfield DV-II+ Pro EXTRA) for G1, G3 and G5 against different temperature of GTNFs. Spindle model LV-1 (61) was used based on its wide range of viscosity. Similarly to the measurements taken for the thermal conductivity, the temperatures were varied between 20-100°C with 20°C interval. All results obtained from the experiments were analysed using graphs plotted of different concentration of GNPs against temperature of the nanofluids.

3. Results and Discussion

3.1. Stability of nanofluid

The stability of the GTNFs is measured based on the GNPs concentrations which corresponds to the absorbance at 570 nm as shown in Fig. 1. The stability of each sample (G1-G5) is shown in Fig. 3. From Fig. 3, it is clear that all the samples prepared were stable in the three weeks of UV-Vis measurement. It is also observed that the stability of G5 was slightly decreased with increasing sedimentation time. This is due to the higher amount of GNPs added to the oil forming a denser solution. It is a distinguished fact that denser particles tend to sediment quicker than less dense particles. However, even the largest change in concentrations of G5 is very small of about 5.1% which occurred at 170 hour before the concentrations of G5 stabilizes again.

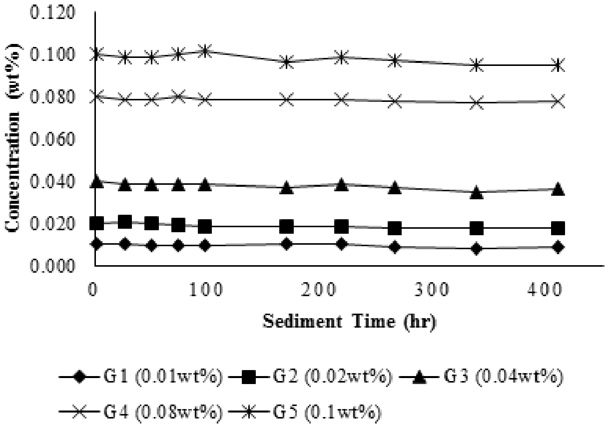
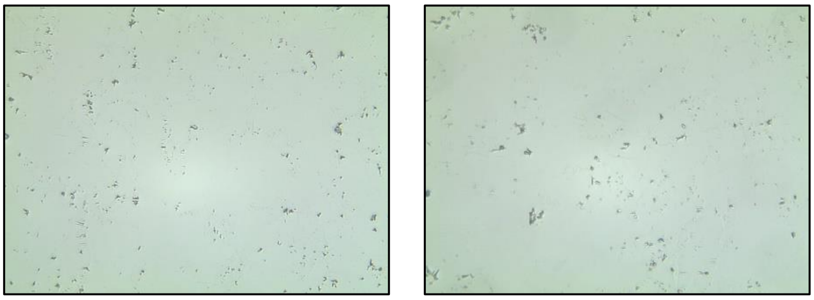


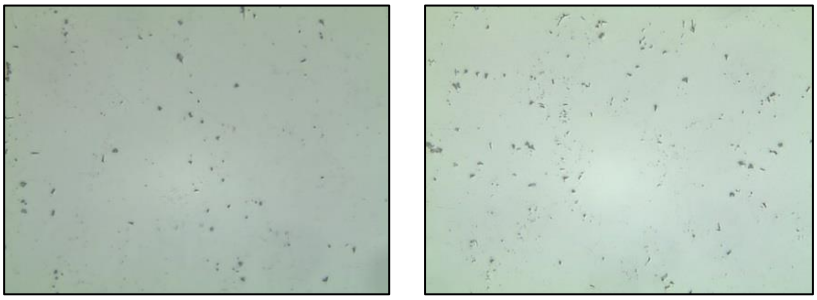
Fig. 3. Effect of concentration and sediment time on the stability of GTNFs.

3.2. Optical microscope

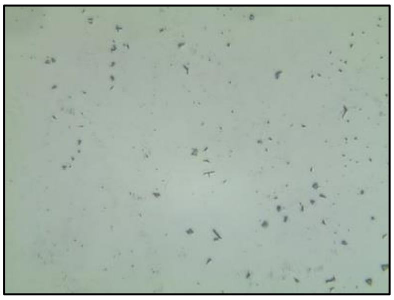
Figure 4 shows images of the dispersion of GNPs in transformer oil as captured using 40X magnification lens of the optical microscope. It is observed that there are good dispersions as the small clusters formed are uniform. GTNFs with higher concentrations such as G5 (0.1 wt%) show more distinct image of the clusters formed as the weight fraction of GNPs is higher causing the nanofluid to have more clusters. All images were taken after two months from the preparation of nanofluid in which agglomerates tend to form.



(a) 0.01 wt % **(b) 0.02 wt %**



(c) 0.03 wt % **(d) 0.04 wt %**



(e) 0.05 wt %

Fig. 4. Optical Microscope Images of (a) G1, (b) G2, (c) G3, (d) G4 and (e) G5.

3.3. Thermo-physical properties

3.3.1. Thermal conductivity

Effects of different GNPs concentrations and temperature are studied on the effective thermal conductivity of GTNFs as shown in Fig. 5. It is depicted that the thermal conductivity increases notably with the increase of GNPs concentrations and temperature. The enhancement in thermal conductivity with respect to concentration obtained at 60 °C for 0.01-0.1 wt % of GNPs as shown in Fig. 6 was from 5.66-29.25 %. The enhancement obtained with respect to temperature for G5 (0.1 wt%) at 20-100 °C was from 13.86-69.31 %.

However, there is a drop in thermal conductivity at 40 °C because of the nature of naphthenic based transformer oil in which its thermal conductivity decreases with increasing temperature [19, 20] which is also observed from Fig. 5. After 40 °C, the effect of Brownian motion of GNPs which is a strong function of temperature overtook the effect of the decreasing thermal conductivity nature of transformer oil. The collision of nanoparticles due to Brownian motion is a slow process which occurs after the thermal diffusion of the nanoparticles suspended in the base fluids [21]. Thus, the effect of Brownian motion is only significant after 40 °C.

The perfect structure and properties of GNPs being two-dimensional and has low thermal interface resistance attributed the excellent thermal conductivity enhancement at such low concentrations. Also, the thermal conductivity is enhanced due to the large surface area available on the GNPs for heat transfer which is the major contribution of the increase in effective thermal conductivity of the nanofluids prepared.

3.3.2. Viscosity

Viscosities of the GTNFs were measured as function of GNP concentration and temperature as shown in Fig. 7. It can be seen from Fig. 7 that over the measured temperature range, the viscosity of pure oil and GTNFs decreases with increasing temperature. The viscosity of the GTNFs was reduced by 8.33-23.97% as the temperature rises and thus allowing more convectional heat flow. The increasing temperature reduces the intermolecular forces of the particles and fluid and thus decreasing the viscosity [10]. It is also observed that the viscosity of the GTNFs decreases with the increase in GNPs concentration. This is due to the self-lubricating effect of GNPs which increases with increasing concentration. GNPs which is well known as a self-lubricating composite has low shear strength which lowers the viscosity of the GTNFs [22, 23].

The decrease in viscosity as an effect of increasing temperature and concentration of GNPs increases the average velocity of Brownian motion of the nanoparticles [24]. The increase in Brownian motion of the GNPs increases the kinetic energy of the particles and thus increases their collision for heat transfer.

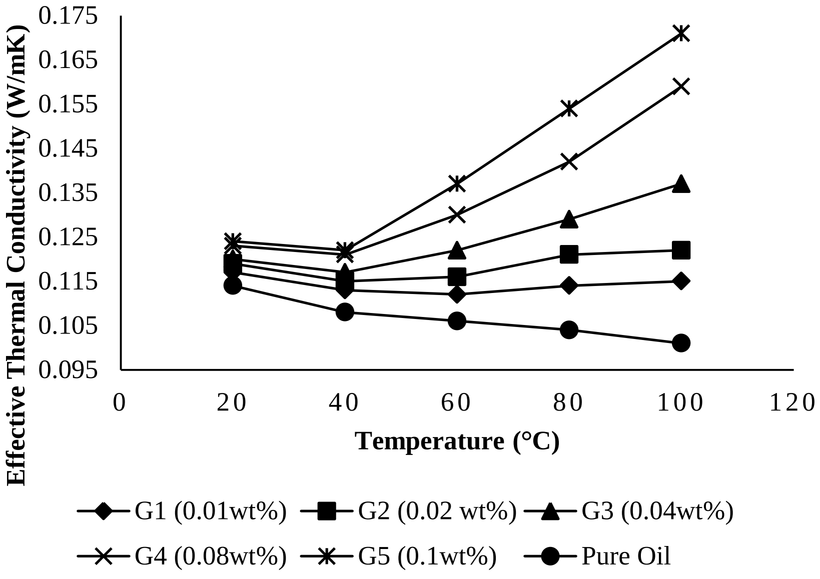


Fig. 5. Effective thermal conductivity of GTNFs.

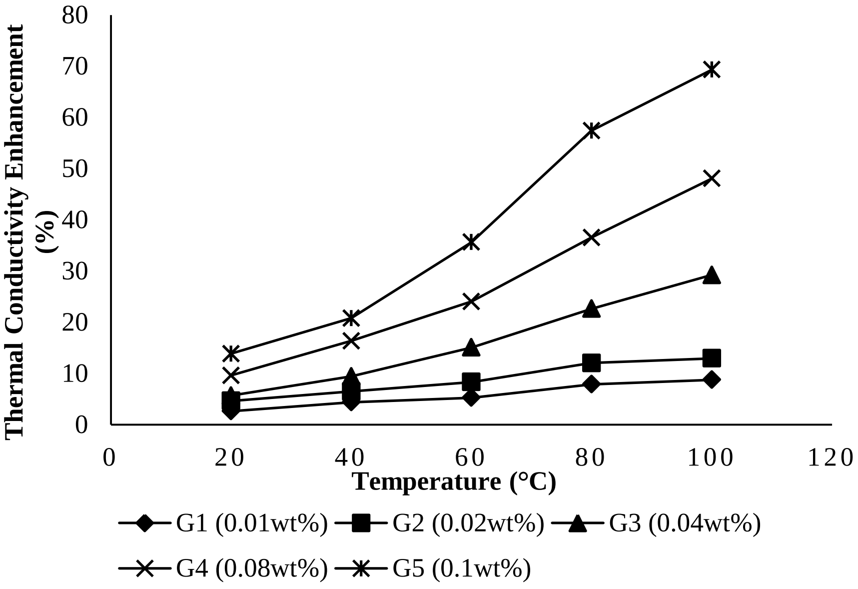


Fig. 6. Percentage thermal conductivity enhancement of GTNFs.

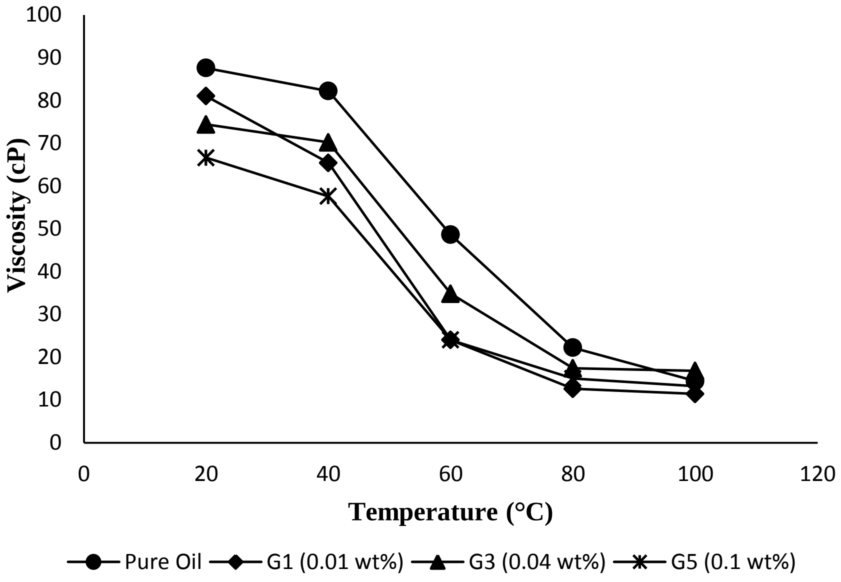


Fig. 7.Viscosity of GTNFs.

4. Conclusion

Transformer oil is used as a source of cooling to prevent overheating in transformers. However, transformer oil being a conventional heat transfer fluid has a low thermal conductivity which leads to low heat transfer rate. GNPs which are widely known for its thermal properties have provided an alternative solution to enhance the thermo-physical characteristics of the fluids. In this work, the GTNFs with GNPs concentration of 0.01-0.1 wt % were stable for more than 3 months after a 4-hour sonication. The thermo-physical properties such as thermal conductivity and viscosity of the nanofluids were proven to increase the heat transfer capability of the oil. From the results obtained, temperature and concentration of GNPs play significant roles in the enhancement of thermo-physical properties. The enhancement in thermal conductivity obtained was from 2.63-13.86 % and 8.77- 69.31 % as the temperatures varies from 20-100 °C, for 0.01 wt% and 0.1 wt% of GNPs. The viscosity of GTNFs were reduced by 8.33-23.97% as the temperature rises allowing more convectional heat flow. The increase in thermal conductivity and decrease in viscosity as temperature rises enhances the overall performance for heat transfer of the transformer oil. Thus, it can be concluded that GNPs can successfully enhance thermo-physical properties of a transformer oil which can be further applied in transformer cooling technologies saving cost and energy.

References

1. Bhandari, A.; Surampalli, R.Y.; Adams, C.D.; Champagne, P.; Ong, S.K.; Tyagi, R.D.; and Zhang, T.C. (2009). Contaminants of Emerging Environmental Concern. *American Society of Civil Engineers (ASCE).*

2. Sadeghinezhad, E.; Togun, H.; Mehrali, M.; Sadeghi, N.P.; Tahan, L.S.; Abdulrazzaq, T.; Kazi, S.N.; and Metselaar, H.S.C. (2015). An experimental and numerical investigation of heat transfer enhancement for graphene nanoplatelets nanofluids in turbulent flow conditions. *International Journal of Heat and Mass Transfer*, 81, 41-51.
3. Tessy, T.B.; and Ramaprabhu, S. (2011). Enhanced convective heat transfer using graphene dispersed nanofluids. *Nanoscale Research Letter*, 6, 1-9.
4. Canter, D.N. (2009). Heat transfer fluids. *Tiribology and Lubricant Technology*, 28-35.
5. Balandin, A.; Ghosh, S.; Bao, W.; Calizo, I.; Teweldebrhan, D.; Miao, F.; and Lau, C.N. (2008). Superior thermal conductivity of single-layer graphene. *Nano Letter*, 8(3), 902-907.
6. Ghosh, S.; Calizo, I.; Teweldebrhan, D.; Pokatilov, E.P.; Nika, D.L.; Balandin, A.A.; Bao, W.; Miao, F.; and Lau, C.N. (2008). Extremely high thermal conductivity of graphene: prospects for thermal management applications in nanoelectronic circuits. *Applied Physics Lett*er, 92(15), 151911.
7. Banerjee, S.; Tyagi, A.K.; and Kain, V. (2012). *Functional Materials*. Elsevier.
8. Eastman, J.A.; Choi, S.U.; Li, S.; Yu, W.; and Thompson, L.J. (2001) Anomalously increased effective thermal conductivities of ethylene glycol-based nanofluids containing copper nanoparticles. *Applied Physics Letter,* 78(6), 718-720.
9. Zhu, H.; Lin, Y.; and Yin, Y. (2004). A novel one-step chemical method for preparation of copper nanofluids. *Journal of Colloid and Interface Science*, 227(1), 100-103.
10. Rashmi, W. (2012). Experimental and theoretical studies on convective heat transfer of CNT nanofluids stabilized by gum arabic.
11. Yu, W.; Xie, H.; and Bao, D. (2010). Enhanced thermal conductivities of nanofluids containing graphene oxide nanosheets. *Nanotechnology*, 21(5), 55
12. Choi, C.; Yoo, H.S.; and Oh, J.M. (2008). Preparation and heat transfer properties of nanoparticle-in-transformer oil dispersions as advanced energy-efficient coolants. *Current Applied Physics*, 8(6), 710-712.
13. Kole, M.; and Dey, T.K. (2011). Effect of aggregation on the viscosity of copper oxide-gear oil nanofluids. *International. Journal of Thermal Science*, 50(9), 1741-1747.
14. Namburu, P.K.; Kulkarni, D.P.; Misra, D. and Das, D.K. (2007). Viscosity of copper oxide nanoparticles dispersed in ethylene glycol and water mixture. *Experimental Thermal and Fluid Science*, 32(2), 397-402.
15. Yousef, S.; Liddawi, A.L.; and Ssekasiko, D. (2014). *Properties of transformer oil that affect efficiency*. BEng. Thesis. School of Engineering, Blekinge Institute of Technology, Sweden.
16. Hajjar, Z.; Rashidi, A.M.; and Ghozatloo, A. (2014). Enhanced thermal conductivities of graphene oxide nanofluids. *International Communication in Heat and Mass Transfer*, 57(2) 128-131.
17. Wang, B.; Wang, X.; Lou, W.; and Hao, J. (2012). Thermal conductivity and rheological properties of graphite/oil nanofluids. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 414, 125-131.

18. Rashmi, W.; Ismail, A.F.; Sopyan, I.; Jameel, A.T.; Khalid, M.; Mubarak, N.M.; and Islamic, I. (2011). Stability and thermal conductivity enhancement of carbon nanotube nanofluid using gum arabic. *Journal of Experimental Nanoscience*, 6(6), 37-41.
19. Krawiec, S. Improved heat transfer capability using ISO-Paraffins versus Naphthenic in transformer, *Petrocanada*, 1-10.
20. Wang, H.J; Ma, S.J.; Yu, H.M.; Zhang, Q.; Guo, C.M.; and Wang, P. (2014). Thermal conductivity of transformer oil from 253 K to 363 K. *Petroleum Science and Technology*, 32(17), 177-179.
21. Jang, S.P.; and Choi, S.U. (2004). Role of Brownian motion in the enhanced thermal conductivity of nanofluids. *Applied Physics Letter*, 84(21), 4316.
22. Berman, D.; Erdemir, A.; and Sumant, A.V. (2014). Graphene: a new emerging lubricant. *Material Today*, 17(1) 31-42.
23. Nptel. (2015). Tribology: lubricants & lubrication. *Lubricants*.
24. Khedkar, R.S.; Kiran, S.; Sonawane, S.S.; Wasewar, K.; and Umre, S.S. (2013). Thermo-physical characterization of paraffin based Fe_3O_4 nanofluids. *Procedia Engineering*, 512(2), 342-346.