

EXPERIMENTAL STUDIES OF THERMAL CONDUCTIVITY, VISCOSITY AND STABILITY OF ETHYLENE GLYCOL NANOFUIDS

Tony John, T. S. Krishnakumar

T.K.M College of Engineering, Kollam, Kerala-691005, India

T.K.M College of Engineering, Kollam, Kerala-691005, India

ABSTRACT

Nanofluids are colloidal solutions of nanometer sized particles in a base fluid. They exhibit enhanced thermal conductivity and viscosity compared to the base fluid from which they are prepared. The variation of thermal conductivity and viscosity of ethylene glycol based alumina and copper oxide nanofluids with respect to particle volume concentration and temperature were investigated. The increase in thermal conductivity values were slightly greater than the prediction of Hamilton Crosser model and the effective viscosity of nanofluids were much higher than the values predicted by Einstein-Batchelor model. No consistent trend was observed for temperature dependence of relative effective viscosity of nanofluids. The stability of nanofluids increased with increase in viscosity of the base fluid and decreased with increasing particle volume concentration. These results are helpful in extending the use of nanofluids to various fields such as industrial cooling and lubrication.

Keywords—cooling; nanofluids; thermal conductivity; viscosity; stability

NOMENCLATURE

k_{eff} Thermal conductivity of nanofluid
 k_p Thermal conductivity of nanoparticle
 k_f Thermal conductivity of base fluid
 μ_{eff} Viscosity of nanofluid
 μ_f Viscosity of base fluid
 ϕ Volume Fraction

1. INTRODUCTION

Nanofluids are colloidal solutions prepared by suspending nanometer sized particles in a base fluid [1]. The term “nanofluid” was proposed by Choi in 1995 [2]. Lee et al. discovered that nanofluids with unprecedented stability of suspended nanoparticles, despite huge differences in density of nanoparticles and fluid, have thermal conductivity enhancement much better than theoretically predicted values [3]. In spite of having solid particles, these fluids retain their Newtonian characteristics at low volume concentrations and hence can be employed as heat transfer fluids in practical applications as substituents for conventional fluids due to their enhanced thermo-physical properties. These exciting results on nanofluid thermal conductivity make nanofluids promising for applications in thermal management

systems.

Further research on the hydrodynamic properties of nanofluids resulted in concluding that they show abnormal increase in viscosity, which is an important parameter in determining the pumping power and other heat transfer characteristics such as convective heat transfer co-efficient. These revelations makes nanofluids suitable for applications in micro channel flow passages with miniaturized (smaller and lighter) heat exchangers, reduced heat transfer fluid inventory and reduced emissions. Recent experiments have also demonstrated that nanofluids have attractive properties for applications in the area of heat transfer, drag reduction, binding ability for sand consolidation, gel formation, wettability alteration, and corrosive control. The tribological performance of lubricating oils can also be significantly improved by dispersing carbon and metallic-based nanoparticles in these lubricants. They also find application in numerous fields including tribology, chemistry, surfactants and coatings, pharmaceutical and medical applications [4].

Abnormal thermal conductivity increase relative to the base fluid was reported by Eastman et al. [5]. They obtained a 40% increase in the thermal conductivity of ethylene glycol with 0.3 vol. % copper nanoparticles of 10nm diameter. Das et al. [6] have observed increases of 10-25% in water with 1-4 vol. % alumina nanoparticles. They also proposed that thermal conductivity of nanofluids is a strongly increasing function of temperature, much more so than that of pure liquids. Numerous experimental studies have been carried out to measure the thermal conductivity of nanofluids using different techniques such as transient hot wire, steadystate parallel plates, and temperature oscillation. Despite numerous studies motivated by the significant benefits of utilizing nanofluids in various applications, the pertinent mechanisms of the thermal conductivity enhancement of nanofluids have not been well understood. Moreover, published results demonstrate thermal conductivity enhancement varying from anomalously large to small values. Al₂O₃ and CuO are the most common nanoparticles used in the literature. The result of these studies shows that the effective thermal conductivity increases with an increase in the volume fraction. In addition, the size of the particles is found to have a significant effect on the thermal conductivity enhancement. It should be noted that smaller particles exhibit larger surface area to volume ratio than the larger particles. As such, smaller particle diameters can possibly result in a larger augmentation in the effective thermal conductivity [7]. Compared with the experimental studies on thermal conductivity of nanofluids, there are limited rheological studies reported in the literature. Models of the effective viscosity of nanofluids based on the experimental data are limited to certain nanofluids. Murshed et al. [8] measured relative viscosity data for TiO₂ and Al₂O₃/water-based nanofluids, and reported a maximum enhancement of 80% at 4% and 5%, respectively. Masuda et al. were the first to measure the viscosity of several water-based nanofluids for temperatures ranging from room condition to 67^o C. Wang et al. obtained some data for the dynamic viscosity of Al₂O₃/water and Al₂O₃/ethylene glycol mixtures at various temperatures. As most of the theories developed to predict the value of effective viscosity of the nanofluids underestimated them when compared to the measured data, Maiga et al. [9] performed a least-square curve fitting of some experimental data of Wang et al. [7] including Al₂O₃ in water and Al₂O₃ in ethylene glycol. The concept of nanofluids attracted lot of attention from the scientific world since its introduction and numerous researches are being conducted to shed light on various thermo-physical properties such as thermal conductivity, viscosity, heat transfer coefficient, boiling heat transfer characteristics, etc. But even after a couple of decades of research, scientists have failed to reach a consensus in explaining the reasons that contribute to abnormal enhancement in properties shown by nanofluids.

2. PREPARATION OF NANOFLUIDS

Nanofluids were prepared using ethylene glycol as base fluid and Copper oxide [Cu(II)O] (procured from SIGMA ALDRICH Co., U.S.A) and Titanium dioxide [TiO₂] (purchased from Kerala Metals and Minerals Limited) nanoparticles at different particle concentrations. Measured quantity of nanopowder

was added to measured volume of base fluid. No additives or surfactants were used in the preparation as they may affect the thermo-physical properties of the nanofluids. The nanoparticles were accurately measured using a high precision (up to 0.00001 g) electronic weighing machine. The mixture of nanoparticles and base fluid

were continuously agitated for 6 hours in a high frequency sonicator (POWER SONIC 410) at maximum frequency (40 kHz). The purposes of the agitation are breakdown or de-agglomeration of clustered nanoparticles, to facilitate even particle distribution and to minimize nanoparticle sedimentation. The following nanofluids were synthesized using the above procedure i) Copper oxide/ Ethylene glycol nanofluids of volume fractions 0.1% and 0.5% ii)

Titania/ Ethylene glycol nanofluids of volume fractions 0.1%, 0.5% and 1%.

3. MEASUREMENT OF THERMAL CONDUCTIVITY

The thermal conductivity of the synthesized nanofluids were measured using a KD2 PRO thermal conductivity probe (Decagon Devices Inc., US). This instrument works on the classical transient hot wire method which is the most widely used technique in measurement of thermal conductivity of liquids in general and nanofluids in particular. The instrument was then tested for uncertainty using distilled water, glycerine (provided as test fluid by manufacturer) and ethylene glycol and was found to be less than $\pm 1\%$. Care was taken to ensure vertical positioning of the probe without any shake or vibration, which may cause uncertainty in readings due to convection heat transfer.

4. MEASUREMENT OF VISCOSITY

The viscosity measurement of the nanofluids were carried out using a Brookfield DV II+ PRO Cone/Plate Rheometer (Viscometer). It is a precise torque meter which is driven at discrete rotational speeds. The torque measuring system, which consists of a calibrated beryllium-copper spring connecting the drive mechanism to a rotating cone, senses the resistance to rotation caused by the presence of sample fluid between the cone and a stationary flat plate. The resistance to the rotation of the cone produces a torque that is proportional to the shear stress in the fluid. This reading is easily converted to absolute centipoise units (mPa.s)

from pre-calculated range charts. Alternatively, viscosity can be calculated from the known geometric constants of the cone, the rate of rotation, and the stress related torque. The correct relative position of cone and plate is obtained by following a simple mechanical procedure without the need for external gauges or supplementary instrumentation. The stationary plate forms the bottom of a sample cup which can be removed, filled with .5 ml to 2.0 ml of sample fluid (depending on cone in use), and remounted without disturbing the calibration. The sample cup is jacketed and has tube fittings for connection to a constant temperature circulating bath. The system is accurate to within $\pm 1.0\%$ of full scale range. Reproducibility is to within $\pm 0.2\%$. Working temperature range is from 0°C to 100°C . The equipment was operated in external mode, which uses a computer program (Rheocalc) to display the viscosity values of the fluid being tested. The temperature of the sample was kept constant during the measurement process by circulating mineral oil through the jacketing of the cup of the instrument. A JULABO F 25 constant temperature bath/ circulator was used for circulating fluid. The instrument was tested with pure fluids (Distilled water and ethylene glycol) and the uncertainty limits were found to be inside the manufacturer specified limit.

5. Comparison of Stability of Nanofluids

Stability of nanofluids were studied using visual comparison at regular intervals. The time taken for visible settling was noted as a measure of stability. The condition of ethylene glycol based nanofluids were monitored every 8 hours for a period of 5 days.

6. RESULTS AND DISCUSSIONS

6.1 Thermal Conductivity of Nanofluids

The effective thermal conductivities of ethylene glycol increased due to addition of nanoparticles. The increase in thermal conductivity was proportional to increase in particle volume concentration. Fig. 1 represents the increase in thermal conductivity of ethylene glycol due to addition of nanoparticles. The increase in thermal conductivity is attributed to the fact that the thermal conductivities of metal oxides are several order of magnitude higher than that of liquids used as base fluids.

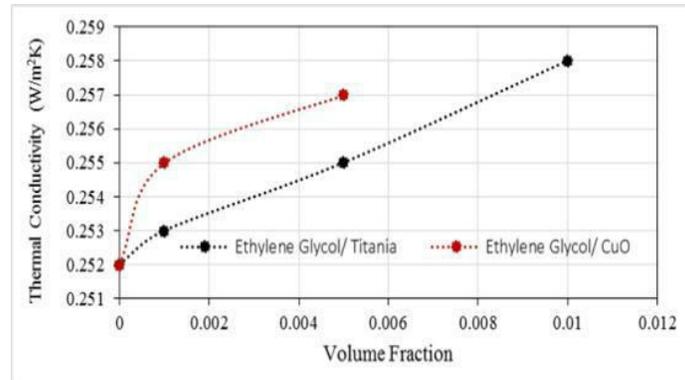


FIGURE 1. THERMAL CONDUCTIVITY INCREASE IN ETHYLENE GLYCOL BASED NANOFLUIDS

The effective thermal conductivities of these nanofluids relative to their base fluids as a function of their volume fraction are shown in Fig. 2. The data points corresponds to the average of 5 readings. These results were compared with values predicted by the Maxwell model which can be represented by Eqn. 1a:

$$k_{eff} = \frac{k_p + 2k_f + 2\phi_p(k_p - k_f)}{k_p + 2k_f + \phi_p(k_p - k_f)} k_f \quad (1)$$

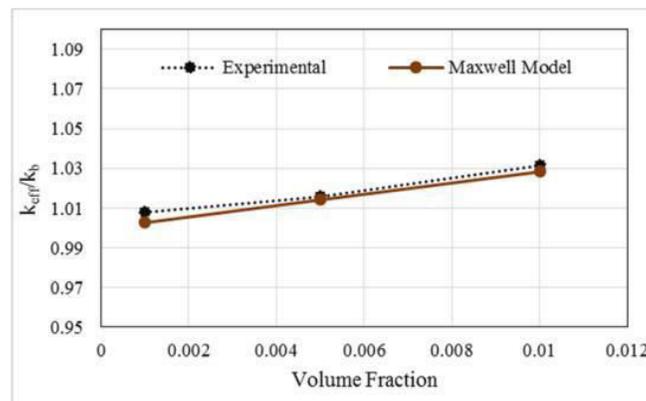


FIGURE 2. RELATIVE THERMAL CONDUCTIVITY OF TITANIA/ ETHYLENE GLYCOL NANOFLUID AT 35°C

The experimental values measured were generally higher than those predicted by Maxwell. However, the difference between the predicted and measured values were less than 2.5%. This contradicts many of the literature data which reports that the relative thermal conductivity of nanofluids was much higher than those predicted by Maxwell model. This may be due to the

fact that all the nanofluids prepared in our work were of low volume fraction ($\leq 1\%$). The Maxwell equation was originally derived for low volume fraction solid suspensions. Another factor may be the absence of surfactants or additives which may affect the effective thermal conductivity of nanofluids. It may be noted that recent studies by Utomo et al. also reported that the variation of their measured thermal conductivities from Maxwell equation were within $\pm 5\%$ [2].

The effect of temperature on thermal conductivity was also studied for a temperature range of 30°C to 60°C for ethylene glycol/ Titania nanofluids. A strong temperature dependence was observed at all particle concentrations and the enhancement in thermal conductivity increased from 1.19% at 30°C to 3.08% at 60°C for 0.5% volume fraction. The results are similar to the temperature dependence studies by Li and Peterson [2]. They obtained a linear relation for temperature dependence with an r squared value of 0.9078. However, the relation observed in present study was of a second degree polynomial nature with an r squared value of 0.946. Fig. 3 shows the variation of effective relative thermal conductivity of ethylene glycol nanofluids with temperature. The strong temperature dependence of nanofluids are due to onset of Brownian motion at higher temperatures. It is to be noted that Maxwell model is insensitive to temperature.

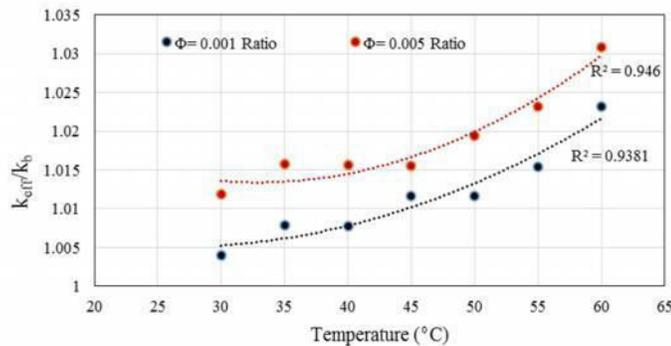


FIGURE 3. TEMPERATURE DEPENDENCE OF RELATIVE THERMAL CONDUCTIVITY FOR ETHYLENE GLYCOL-TITANIA NANOFLUIDS

For ethylene glycol base fluid, addition of CuO nanoparticles gave a higher relative thermal conductivity when compared to Titania nanoparticles. Therefore, from a cooling point of view, CuO nanofluids are superior to Titania nanofluids. This is due to the fact that the thermal conductivity of CuO is greater than that of TiO_2 .

TABLE 1. RELATIVE THERMAL CONDUCTIVITIES OF ETHYLENE GLYCOL NANOFLUIDS

Nanoparticle	Relative Thermal Conductivity (k_{eff}/k_b)		
	$\phi=0.001$	$\phi=0.005$	$\phi=0.01$
TiO_2 (<40nm)	1.0039	1.0119	1.0238
CuO (<50nm)	1.0119	1.01984	-

6.2 Viscosity of Nanofluids

The effective viscosity of base fluids increased due to addition of nanoparticles. The increase was proportional to the concentration of particles. The temperature dependence of base fluids were maintained even after addition of nanoparticles. Fig. 4 shows the increase in viscosity of base fluids due to addition of

nanoparticles at various concentrations.

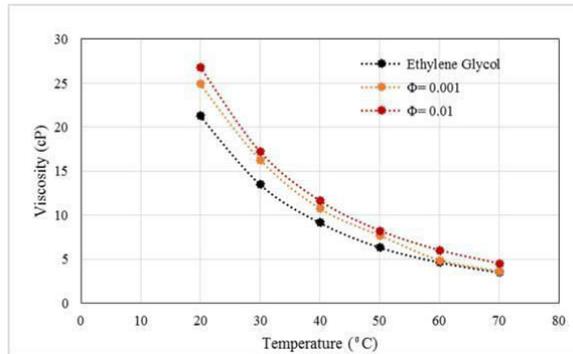


FIGURE 4. INCREASE IN EFFECTIVE VISCOSITY OF TITANIA/ ETHYLENE GLYCOL NANOFLUID AT DIFFERENT PARTICLE CONCENTRATIONS

The effective relative viscosity of nanofluids (ratio of effective viscosity of nanofluid to that of base fluid) are plotted as a function of their volume fraction in fig. 5. The results are compared with the values predicted by Batchelor’s equation for effective viscosity of nanofluids, which is given by Eqn. 2.

$$\mu_{\text{eff}}/\mu_f = (1 + 2.5\phi + 6.2\phi^2) \quad (2)$$

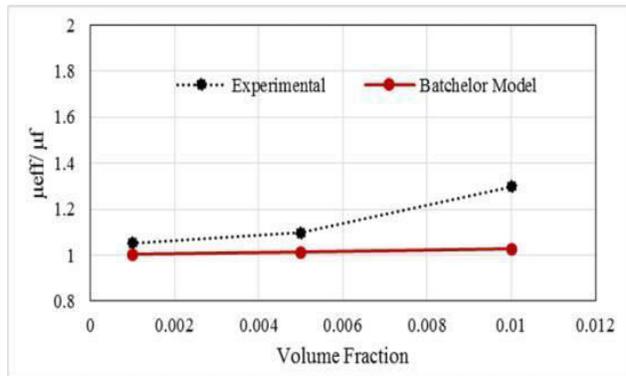


FIGURE 5. RELATIVE EFFECTIVE VISCOSITY OF TITANIA/ ETHYLENE GLYCOL ANOFLUID AT 60° C

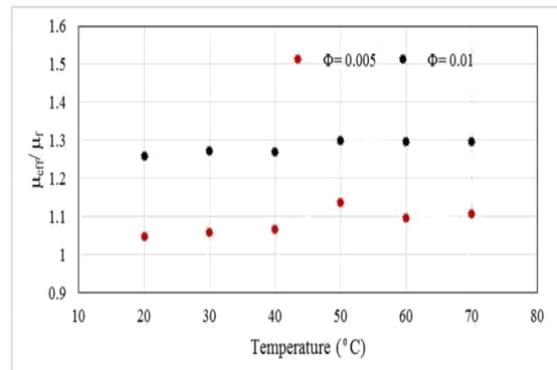


FIG. 5.13 TEMPERATURE DEPENDENCE OF RELATIVE EFFECTIVE VISCOSITY OF TITANIA/ ETHYLENE GLYCOL NANOFLUIDS

It can be seen that the model clearly under predicts the value of effective viscosity for all the nanofluids used for measurement, which is in agreement with results found in the literature. The deviation from the model is more prominent as the volume fraction increases. The temperature dependence of effective relative viscosity of nanofluids were also studied and plotted in fig. 6. A slight increase in effective relative viscosity with increase in temperature was observed for ethylene glycol based nanofluids. The percentage increase in relative viscosity of Titania nanofluids were more when compared to CuO nanofluids.

6.3 Stability of Nanofluids

Ethylene glycol nanofluids settled after two days of preparation when kept stationary. Even though nanofluids give higher relative increase in viscosity and thermal conductivity at higher volume fractions, their practical application may be limited due to stability issues. Addition of surfactants can improve the stability of nanofluids. It is often reported in the literature that the pH can also play an important role in

stability, as agglomeration is maximum at the isoelectric point of a solution. Hence stability can also be increased by adjusting the pH of the nanofluid away from its isoelectric point.

7. CONCLUSIONS

The effective thermal conductivity and viscosity of base fluids increased considerably due to addition of nanoparticles. The relative effective thermal conductivity of nanofluids increased with increasing particle volume concentration and were generally higher than that predicted by the Maxwell model. A strong temperature dependence was established for effective relative thermal conductivity. The nanoparticle thermal conductivity directly influenced the relative effective thermal conductivity of nanofluids. The effective relative viscosity of nanofluids increased with an increase in particle volume fraction. The measured values were much greater when compared to Einstein-Batchelor model prediction. It was also observed that the influence of temperature on the relative effective viscosity was very weak. The stability of nanofluids were found to be low without surfactants and additives. Nanofluids of low particle volume concentrations had better stability.

The parameters considered in this work was limited to particle volume concentration, temperature, nanoparticle and base fluid. Recent studies have identified other parameters such as nanoparticle size, pH value of solution, etc. may affect the effective thermal conductivity and viscosity of nanofluids. The effect of such parameters can be determined experimentally. Experimental studies can be extended to numerous base fluids which are used in the industry as coolants and lubricants. The effect of surfactants in effective thermal conductivity and viscosity are yet to be studied. Stability of nanofluids are a big challenge to be properly addressed even now.

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