Research article

Investigations on the Thermal and Electrical Conductivity of Polyethylene Glycol-based CuO and ZnO Nanofluids

Swaminathan Ponmani,[a,b] Pawan Gupta,[a] Prashant Jadhawar,[c] R. Nagarajan,[d] Jitendra Sangwai[a,*]

[a]Gas Hydrate and Flow Assurance Laboratory, Department of Ocean Engineering, Indian Institute of Technology Madras, Chennai- 600036, Tamilnadu, India

[b]Department of Petroleum Engineering, AMET University, kanathur, Chennai 603112

[c]School of Engineering, University of Aberdeen, UK

[d]Department of Chemical Engineering, Indian Institute of Technology Madras, Chennai- 600036, Tamilnadu, India

*Corresponding Author:

Jitendra S. Sangwai

Email: jitendrasangwai@iitm.ac.in

Phone: +91-44-2257-4825

Fax: +91-44-2257-4802

Abstract

In this experimental work, three different types of nanofluids were evaluated for their stability using dynamic light scattering (DLS) and particle morphological study using scanning electron microscopy (SEM). The nanofluids used in this study are zinc oxide (ZnO) nanoparticle in water and 5 wt% polyvinylpyrrolidone (PVP) as a dispersant, and ZnO with polyethylene glycol (PEG 600) and CuO with PEG 600 with 5wt% PVP at different concentration of 0.1, 0.3 and 0.5wt %. Thermal and electrical conductivities were determined by KD-2 Pro® and PC 700 Eutech®. The result shows better enhancement in the thermal and electrical conductivity in the ZnO+PVP+Water system, followed by the CuO+PVP+PEG and ZnO+PEG systems. The highest percentage enhancement in thermal conductivity found to be 35.5 % of ZnO+ PVP+water systems. The thermal conductivity results were compared with a theoretical model and show good agreement with results predicted by the model. The proposed model of Nan et al. (1997) is based on a hypothesis regarding the physical mechanism in heat transfer for nanofluids. This study is expected to form the basis for the development of nanofluid-based technologies with PEG as the primary additive in the upstream oil and gas industry especially in gas hydrates and drilling technology.

Keywords: Electrical conductivity; Nanofluids; Stability; Thermal conductivity; Nan's model.

1. Introduction

Nanofluids have shown various interesting properties, and the distinguishing features offer exceptional potential for different industrial applications, such as electronic, transportation, improve recovery in oil and gas well, industrial cooling applications, nuclear systems cooling, etc. Nanofluids are essentially two-phase systems, viz., solid phase in the liquid phase. Several industries are in need of a cooling medium to improve the heat transfer performance, and of new technology to overcome persistent challenges. A solid has a higher thermal conductivity than a liquid, and hence, to increase the thermal conductivity, nanosized particles suspended in a base fluid, known as 'nanofluids', are used. Nanoparticles have good electrical, magnetic and optical properties. Accordingly, the mechanism for thermal conductivity enhancement is believed to be the responsible parameter for enhancing heat transfer in engineering applications. However, the research on the electrical properties of the nanofluid is very rare. The factors responsible for enhancing heat transfer of nanofluids are the types of nanoparticle, particle size, aggregation, Brownian motion of the particles and temperature of the nanofluids. Various mechanisms and models, based on various assumptions, have been recently developed for explaining the unusually high thermal conductivity of nanofluids [1-3]. Two significant requirements for measuring the thermal conductivity are preparation of a homogeneous mixture and long-term stability which can withsatand the initial equilibrium conditions until measurement. The electrical conductivity of a suspension depends on the background electrolyte, particle size, charge, and volume fraction [4-6]. Choi and his group [7] were the first to report that the suspended particles in the base fluid can significantly enhance the heat transfer and give rise to improvement in the heat exchange systems. Yu et al. [8] studied the thermal conductivity of copper oxide (CuO) nanofluids with ethylene glycol and polyvinylpyrrolidone (PVP) as dispersants. The results showed about 46% enhancement in the thermal conductivity in about 0.5 vol% of particle concentration at 50 °C, and demonstrated that the temperature and Brownian motion of nanoparticles play an important role in the thermal conductivity enhancement. Mehrali et al. [9] showed that by using graphene nanoplatelets, a stable nanofluid with distilled water could be prepared without surfactant by ultrasonic probe dispersion technique. The researchers showed about a 28% enhancement in thermal conductivity and demonstrated that the stability of nanofluids found to be enhanced due to ultrasonication. These nanofluids can act as an advanced heat transfer fluid in a medium temperature applications in solar and heat exchangers. Xie et al. [10] conducted studies using different nanoparticles, such as silicon carbide (SiC), zinc oxide (ZnO), carbon nanotubes (CNT) and aluminum oxide (Al_2O_3) with base fluids, such as deionized water, glycerol, ethylene glycol and the mixture of water and ethylene glycol. The results showed that the thermal conductivity enhancement has been influenced by the volume fraction of the particle and due to temperature. Fedele et al. [11] studied CuO, titanium oxide (TiO_2) and single-walled carbon nanohorns (SWCNHs) with water as a base fluid. They used polyethylene glycol (PEG) and sodium dodecyl sulfate (SDS) as dispersing agents. They investigated three dispersion techniques, such as sonification, high-pressure homogenization and ball milling for the formation of nanofluids. The high-pressure homogenization method was found to yield better stability of the nanofluids.

Kole and Dey [12] prepared stable ZnO-ethylene glycol nanofluids by prolonged sonification of >62 hours and showed that extended time for sonification gives better fragmentation and dispersion of the particles. In their studies, they considered both temperature and nanoparticle concentration for thermal conductivity enhancement. The results showed that approximately 40% of thermal conductivity enhancement is achieved at 30 ^{o}C and with 3.75vol% of ZnO. Suganthi et al. [13] investigated a colloidal dispersion of ZnO-propylene glycol. Thermal conductivity was measured in the temperature range of 10-60 ^{0}C and for various aggregate sizes.

The researchers observed that the thermal conductivity enhancement depends on temperature and that higher enhancement is possible at a lower temperature. The result shows that the temperature and aggregation of particles are major factors in the formation of a solvation layer on the ZnO nanoparticle surfaces. Jeong et al. [14] showed that for ZnO nanoparticles, the thermal conductivity and viscosity enhancement depend on the particle shape. The researchers used spherical and rectangular particles with concentration range of 0.05 to 5 vol%. The results show about 12% and 18% enhancement in thermal conductivity, respectively, for spherical and rectangular particles at 5vol%. Moattar and Cegincara [15] used PEG to prepare stable nanofluids of ZnO which was later characterized using dynamic light scattering. They observed the effect of ZnO nanoparticles concentration and temperature on the volumetric and transport properties of the aqueous solution of PEG but did not report information on enhancement in the thermal and electrical property. Ponmani et al. [5] studied experimentally the thermal and electrical conductivity of ZnO and CuO nanofluids in xanthan gum. They observed an approximately 25 and 50 % enhancement in thermal and electrical conductivity, respectively. White et al. [16] investigated the electrical conductivity of propylene glycol-based ZnO nanofluids. The result showed that a higher volume fraction of the particle gives better enhancement in electrical conductivity. They observed that for about 7 % volume fraction of nanoparticles, electrical conductivity showed up to 100-fold increase over the base fluid. Kim et al. [17] prepared stable nanofluids using aluminum oxide, zinc oxide and titanium oxide nanoparticles with water and ethylene glycol as a base. They showed that the enhancement in the thermal conductivity increases linearly in lesser size of the particles when suspended in the base fluids. Khedkar et al. [18] investigated the thermal conductivity of CuO nanofluids in monoethylene glycol and water. The results show that the enhancement in thermal conductivity is due to the concentration of nanoparticles in the base fluids and also the interaction between the particles. The experimental measured values of thermal conductivity were compared with different existing models which were accurately fitted with models. Sahooli et al investigated the nanofluids that were prepared using CuO nanoparticles with PVP. The results shows that PVP act as surfactant and resulted in good stability for a week with optimum pH and enhancement of thermal conductivity from 17% to 31% at the temperature of 25 ^{0}C and 50 ^{0}C [19]. Manasrah et al. have observed the enhancement of thermal and physical properties of PEG with carbon nanotubes. The results show that the viscosity of the nanofluid is one of the most dependent parameter for the concentration of the nanoparticles suspended in the solution [20].

Nanoparticles and their nanofluids are of interest to the upstream industry include CuO and ZnO because of their good thermal and electrical properties. However, information regarding the formation and characterization of thermal and electrical properties of nanofluids in a base fluid PEG is scarily available in open literature. PEG has low toxicity and is used in several industrial applications such as in pharmaceutical, cosmetic, lubricants, binders, bases and coupling agents. It is especially useful in the separation and purification of biological materials. One of the commonly used additive PVP, which is a water-soluble polymer, is made from the monomer N-vinylpyrrolidone. This is used as an emulsifier, stabilizer, and medicine and food-additive and also used in the oil and gas industry as inhibitors in gas hydrates. Using the polymer as base fluid for nanoparticles will help ensure better distribution throughout. Formation of CuO and ZnO nanofluids in above mentioned stabilizers and the information on their thermal and electrical properties will add values to some of the application of these nanofluids for various industrial application including upstream oil and gas industry.

In the present paper, the formulation of ZnO and CuO nanofluids using deionized water, and PEG with and without dispersant PVP is being presented. Stable nanofluids containing CuO and ZnO nanoparticles were prepared through a two-step method. The thermo-physical properties, such as thermal conductivity and electrical conductivity, were measured. The effects of particle concentration, dispersant, and stability over time (1, 3 and 7days) in static condition were observed, and the stability was measured by a DLS instrument. The thermal conductivity was compared between theoretical model predictions and experimental data, and electrical conductivity was investigated in detail and reported. In addition, CuO has high thermal conductivity as compared to ZnO, so therefore, one of the objective is also to observe the enhancement in the thermal and electrical properties in the presence of ZnO nanoparticles.

2. Materials and Methods

2.1. Materials

CuO and ZnO nanoparticles were used in this study. The CuO nanoparticles were supplied by Sigma-Aldrich with a spherical shape, diameter of <50nm and with purity of >97%. The ZnO nanoparticles were purchased from Sigma-Aldrich chemicals with a spherical shape, diameter of <50nm and with purity of >97%. Deionized water was used as a base fluid along with PEG and PVP. PVP was used as a dispersant to improve the stability. The properties of nanofluids used in this work are summarized in Table 1, which shows the dispersant, nanoparticles used and the concentration of particles.

2.2. Nanofluids Formulation

The nanofluids were prepared by dispersing the nanoparticles in a base fluid by a two-step method. The base fluid was mixed with a measured quantity of nanoparticles in a beaker covered with aluminum foil to ensure that there is no evaporation during sonification. The polymer was thoroughly dispersed using a magnetic stirrer to avoid lump formation and to get a stable nanofluid system with a polymeric base. The beaker containing the sample is submerged in the water. For this study, ultrasonication (Crest Ultrasonic, 25 KHz, and 450 W) was performed at high frequency of 25 KHz and 450 W for one hour. After 30 minutes, the water was changed and sonification then continued for the next 30 minutes, to ensure that the water did not overheat.[5] Sonification time of one hour is employed for all suspensions prepared homogeneously as seen by visual observation. Also, it is expected that longer sonification time will not substantially change suspended particle morphology.

2.3. Scanning Electron Microscopy

Topography, morphology, and arrangement of agglomerated particles were observed in the dispersed state using SEM. Hitachi S-4800^{TM} Scanning Electron Microscope (SEM) was used for morphological characterization of nanoparticles, and micrographs of ZnO and CuO nanoparticles were obtained.

2.4. Dynamic Light Scattering

DLS measures particle size distribution in a range of nanofluids. The Brownian diffusivity of particles is measured using DLS and is related to their size. The particle size is measured by illuminating the particles with a laser and analyzing the intensity fluctuations in the scattered light. The DLS measurements were carried out using 90 Plus^{TM} Nanoparticle Size Analyzer by Brookhaven Instruments. It is capable of measuring particle size distributions in the range of 1nm to 6µm with a precision of ±1%. Measurements were made at 90° scattering angle and at 25° C.

DLS works on the principle that when the sample is illuminated by a laser beam, the fluctuations of the scattered light are detected at a known scattering angle by a detector.

2.5. Thermal Conductivity and Electrical Conductivity Meter

The thermal conductivities of the nanofluids prepared in this study have been measured using the transition hot wire method with KD2 pro®. A temperature-controlled bath was used to maintain the temperature of the nanofluids at 25° C. The unit was calibrated using standard samples. The KS-1 sensor applies a very small amount of heat to the needle, which helps to prevent free convection in liquid samples. Thirty seconds are allowed for temperature equilibration before heating starts, after which heat is applied for thirty seconds, and measurements are taken over the full time. Electrical conductivity was measured using the PC 700 Eutech ® Instrument. The PC 700 includes an electrode with a nominal cell constant of k = 1.0, a built-in temperature sensor, and 1 meter cable. The instrument was auto-calibrated with 0.01 N KCl [5, 19].

3. Results and Discussion

Table 1 shows the combinations of CuO and ZnO nanofluids prepared in this work. PVP was used as a dispersant at a concentration of 5 wt% in the base fluid to prepare some of these nanofluids. The nanoparticle concentration in the base fluid (water and PVP) was varied as 0.1, 0.3, and 0.5 wt% on water and polyethylene glycol basis. Nanofluids made up of CuO and ZnO nanoparticles were prepared separately using the ultrasonication technique in deionized water with PVP and PEG, with PVP as a dispersant. SEM images of dried samples were obtained after preparation of nanofluids. Figure 1(a) shows ZnO nanofluid preparation with 5 wt% of PVP in water. The SEM results show that the ZnO nanoparticles interact with water-soluble polymers, resulting in loosely-packed and random distribution of spherical-shaped particles in the polymer solution. Figure 1(b) shows ZnO+PEG nanofluid preparation with polyethylene glycol. This nanofluid exhibits reduced agglomeration due to the interaction of polymer chains surrounding the ZnO particles. Figure 1(c) shows CuO+PVP+PEG nanofluid preparation, which also exhibits reduced agglomeration compared to ZnO+PEG. The size of the ZnO nanoparticles increases with concentration. ZnO also tends to reduce the size by interacting with PEG concentration to form polymers (packing effect) [21,22].

DLS measurements were carried out to characterize the dispersion of nanoparticles in the nanofluid. For 0.5wt % ZnO in 5wt% PVP, it is observed from Figure 2 that the particles range in size from 70 nm to 350 nm on the first day. Comparisons of the systems of PEG versus PVP show that the PVP system offers better dispersion and less agglomeration with time. This is based on the overall size of the particles and their agglomerating tendency. For 0.5 wt% ZnO in PEG (Figure 3), it is observed that the particles range in size from 100 nm to 500 nm on the first day. The agglomeration did not happened on second day but up to seventh day the DLS graph shift shows the particle size agglomeration. This experiment was continued upto 23 days and it has been observed that the particle agglomeration during seventh day is almost similar at the end of 23rd day. The DLS graph for nanofluid system of 0.5 wt% CuO with base fluid as 5wt% PVP in PEG is shown in Figure 4. The dispersion of the nanoparticles is more or less even up to the seventh day. This system stability is good, and the DLS graph indicates that the stability can even be maintained over a longer period of time. 0.5 wt% ZnO with base fluid containing 5 wt% PVP offers better stability among all the systems studied. The Brownian motion of nanoparticles was indicated by several authors as a significant factor for the observed enhancement.

Figure 5 shows the variation of thermal conductivity ratio (k_{nf}/k_{bf}) with nanoparticle concentration. From the results, it can be seen that the CuO and ZnO nanofluids with higher particle concentrations exhibit higher thermal conductivities. The extent of increase in thermal conductivity depends upon the nature of the base fluid. All the experiments for thermal and electrical conductivity were carried out in replicates of three each.. The error in the experimental data can be given as the total of the instrumental and the measurement error. The measurement error is attributed to the deviation from the actual results (from repeatability) due to experimental conditions. It is found to be ±1%. The instrumental uncertainty for electrical conductivity and temperature measurement is ±1% (full scale + 1 digit) and 0.1 K in the conductivity range is 0–1999 µS/cm. Thus, the total experimental error is limited to 2%. The percentage increase in the thermal conductivity over the base fluid is shown in Table 2. It can be seen that the system that exhibits the highest thermal conductivity is ZnO+PVP. When using PEG as the base fluid, the increase in thermal conductivity is not significant. This is confirmed in the presence of CuO, which has a high thermal conductivity but is unable to cause a large change. The reason of lesser conductivity in case of ZnO+ PEG 600 and CuO + PEG 600 + 5 wt% PVP is due to the presence of polymers (PEG and PVP) which may have reduced the Brownian motion of the dispersed nanoparticles due to increased viscosity of the solution, and creates much more heat and mass transfer limitations.

Theoretical models have been developed for thermal conductivity of nanoparticle-suspended fluids considering only thermal conductivities of the base fluid and particles, and the volume fraction of particles. Particle size, shape, and the distribution and motion of dispersed particles have also had a significant impact on the thermal conductivity enhancement, and this has not been fully appreciated. Nan et al. [23] developed a model for thermal conductivity of nanofluids which is based on the mechanisms of the formation of a nanolayer at the solid–liquid interface, and nanoparticle aggregation According to Nan et al.[23], the thermal conductivity of the nanofluid can be calculated as follows:

$$k_{nf} = k_{bf}\frac{3+\phi(2\beta_{11}(1-L_{11})+\beta_{33}(1-L_{33}))}{3-\phi(2\beta_{11}L_{11}+\beta_{33}L_{33})} \tag{1}$$

where, k_{nf} and k_{bf} are the thermal conductivity of nanofluids and that of base fluids, L_{ii} and ϕ are the geometrical factor and the volume fraction of particles, respectively. β_{ii} is defined as,

$$\beta_{ii} = \frac{k_p - k_{bf}}{k_{bf} + L_{11}(k_p - k_{bf})} \tag{2}$$

K_P is the thermal conductivity of the particle, which is 27 W/mK for CuO and 10 W/mK for ZnO. For spherical particles, the shape factor and aspect ratio may be taken as 1. The model parameters are obtained as: $L_{11}= 0$ and $L_{33} = 1$ for ZnO+PVP, $L_{11}= 0$ and $L_{33} = 1$ for ZnO+PEG, and $L_{11}= 0$ and $L_{33} = 1$ for CuO+PEG+PVP, respectively. Figures 5 (a), (b) and (c) shows the validation of the Nan's model with the experimental data for ZnO+PEG, ZnO+PVP+water, and CuO+PVP+PEG nanofluid systems. It should be mentioned that the thermal conductivity determined here by Nan's model incorporates the effects of particle geometry and finite interfacial resistance. The experimental results agree with the correlated values of theoretical models.

Figure 6 and Table 3 shows the information on the electrical conductivities of prepared nanofluids and percentage enhancement of the electrical conductivities of these nanofluids over the base fluid, respectively. It has been observed that the increase in the nanoparticle concentration enhances the electrical conductivity of the base fluid. The reason for the enhancement in the electrical conductivity is primarily due to the effective dispersion of the nanoparticles in the various base fluids, which, thus help to form the electrical double layer (EDL) on the surface of the nanoparticle. The formation of the EDL is because of the ionic concentration of the base fluid surface charge size, and the size of the nanoparticles. These nanoparticles get migrated towards the opposite charge in the influence of electrical field along with EDL. The extent of increase in electrical conductivity depends upon the nature of the base fluid. ZnO+PVP+water exhibits the highest electrical conductivity, followed by CuO+PEG nanofluid systems and then ZnO+PEG 600 nanofluid system. From Table 3, it has been observed that an increase in the nanoparticle concentration gives rise to an increase in electrical conductivity. The highest percentage increase in electrical conductivity has been shown in ZnO+PVP+water systems.

4. Conclusion

Water-based nanofluids have been formulated, characterized and investigated for thermal and electrical conductivities by dispersing CuO and ZnO nanoparticles, and prepared by a two-step method using PVP as a dispersant. The morphology characterization of nanoparticle was performed by using scanning electron microscopy. The stability of prepared nanofluids was investigated by allowing the system to settle in a static condition for 1, 3 and 7 days, followed by measurement of particle size distribution by DLS apparatus. The enhancements in thermal conductivity and electrical conductivity were measured at different concentrations. ZnO+PVP+water system shows greater enhancement compared to the other two systems. This study has demonstrated the feasibility of formulating stable nanofluids of CuO and ZnO in PEG and provided with the information on thermal and electrical properties of these nanofluid systems, which is not available in the literature.

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Tables

Table 1: CuO and ZnO nanofluids studied in the present work[+]

Nanoparticle (<50 nm)	Concentration of nanoparticles, wt %	Dispersant: Polyvinylpyrrolidone, wt %	Base fluid
ZnO	0.1	-	PEG 600
	0.3		
	0.5		
ZnO	0.1	5	Deionized Water
	0.3		
	0.5		
CuO	0.1	5	PEG 600
	0.3		
	0.5		

[+] The percentage is on the basis of base fluid, PEG 600 and water.

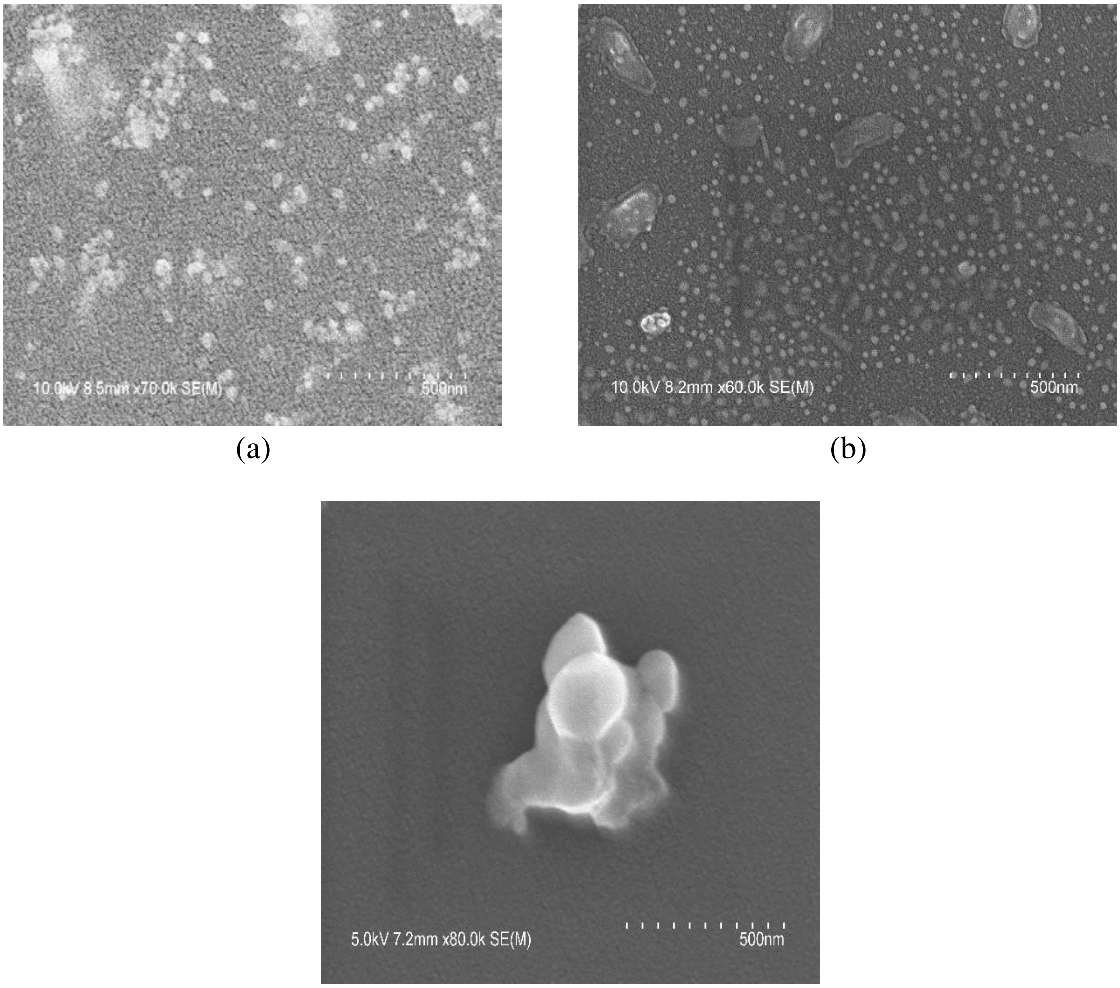
Table 2. Percentage increases in thermal conductivity of the prepared nanofluids

Concentration of nanoparticles (wt %)	Thermal conductivity (W/mK)			Percentage increase in thermal conductivity over the base fluid		
	ZnO + water+ 5wt% PVP	ZnO+ PEG 600	CuO + PEG 600 + 5 wt% PVP	ZnO + Water + 5 wt % PVP	ZnO+PEG 600	CuO + 5wt % PVP +PEG 600
0	0.512	0.201	0.201	---	---	---
0.1	0.612	0.221	0.241	19.53	9.95	14.76
0.3	0.649	0.228	0.253	26.76	13.43	20.48
0.5	0.693	0.239	0.275	35.35	18.91	30.95

Table 3. Percentage increases in electrical conductivity of the prepared nanofluids

Concentration of nanoparticles (wt %)	Electrical conductivity (µS/cm)			Percentage increase electrical conductivity in over the base fluid		
	ZnO + water+ 5wt% PVP	ZnO+PEG 600	CuO + PEG 600 + 5 wt% PVP	ZnO + Water + 5 wt % PVP	ZnO+PEG 600	CuO + 5wt % PVP +PEG 600
0.0	170	3.48	3.56	---	---	---
0.1	185	5.33	5.68	26.99	53.16	53.51
0.3	215	5.36	5.80	40.16	54.02	64.86
0.5	261	5.50	5.90	70.14	65.70	75.68

Figures



(a) (b)

(c)

Figure 1. SEM image for: (a) 0.5 wt % ZnO+5 wt% PVP nanofluid; (b) 0.5 wt% ZnO+PEG 600 nanofluid; (c) 0.5 wt % CuO+5 wt % PVP+ PEG 600 nanofluid.

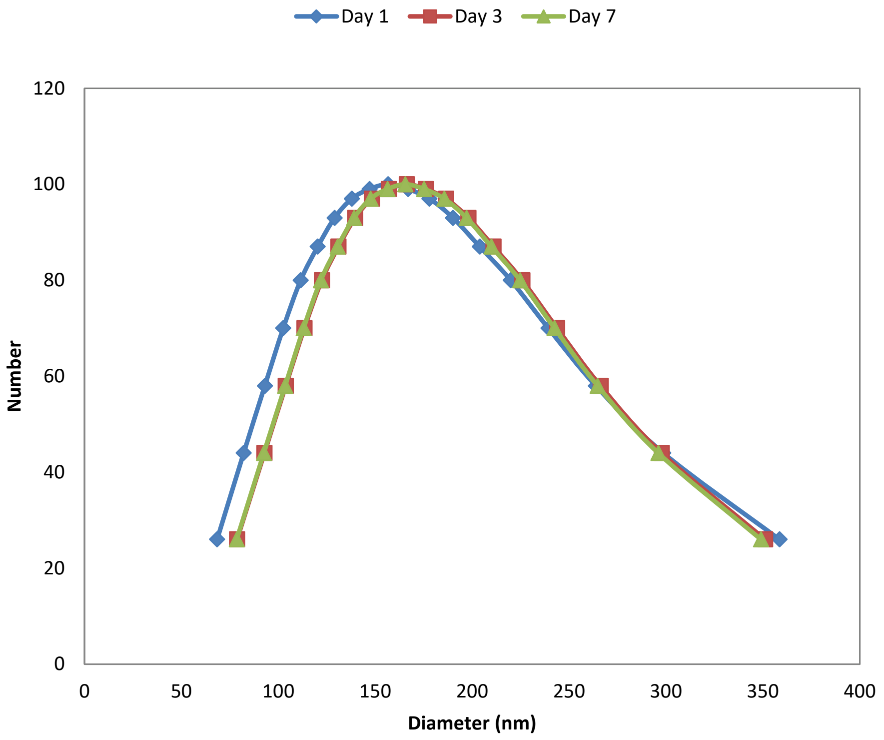


Figure 2. DLS curve for 0.5 wt% ZnO+5 wt% PVP

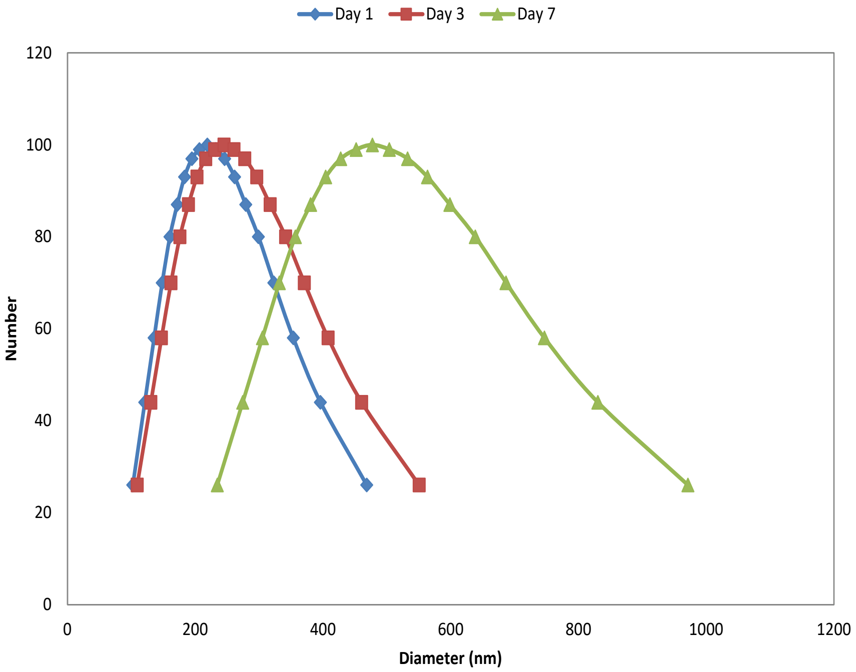


Figure 3. DLS curve for 0.5 wt% ZnO+PEG

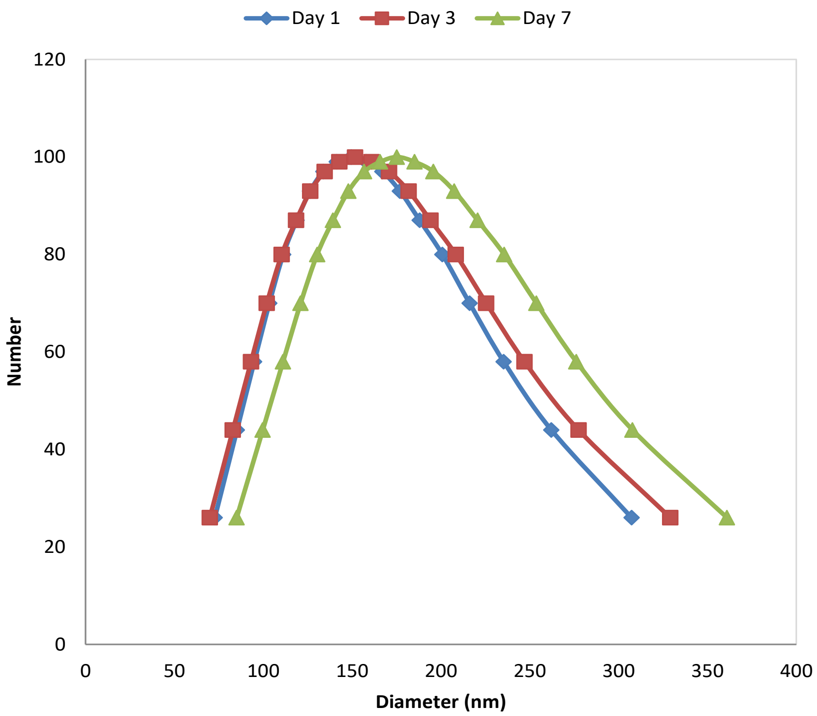
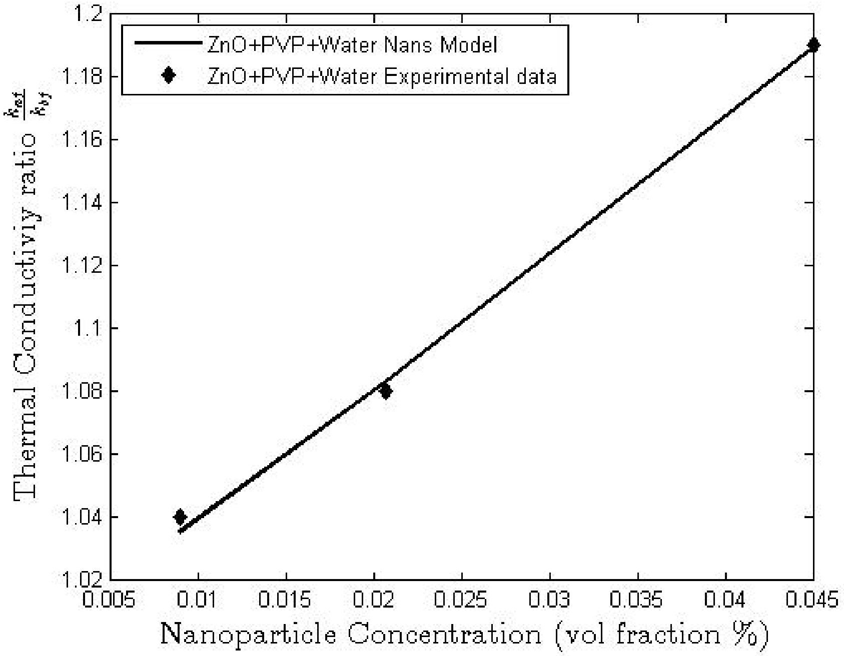
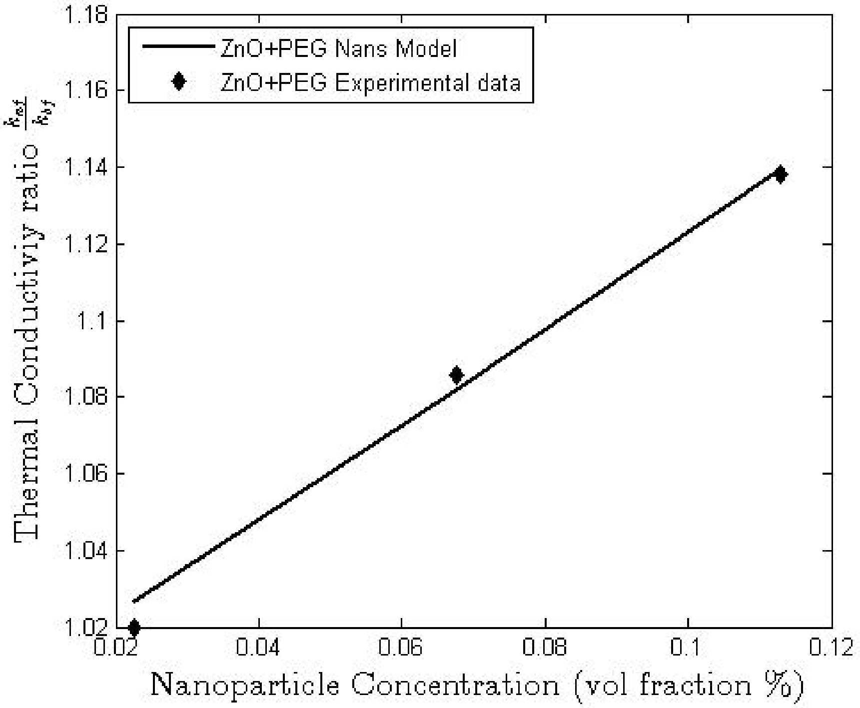


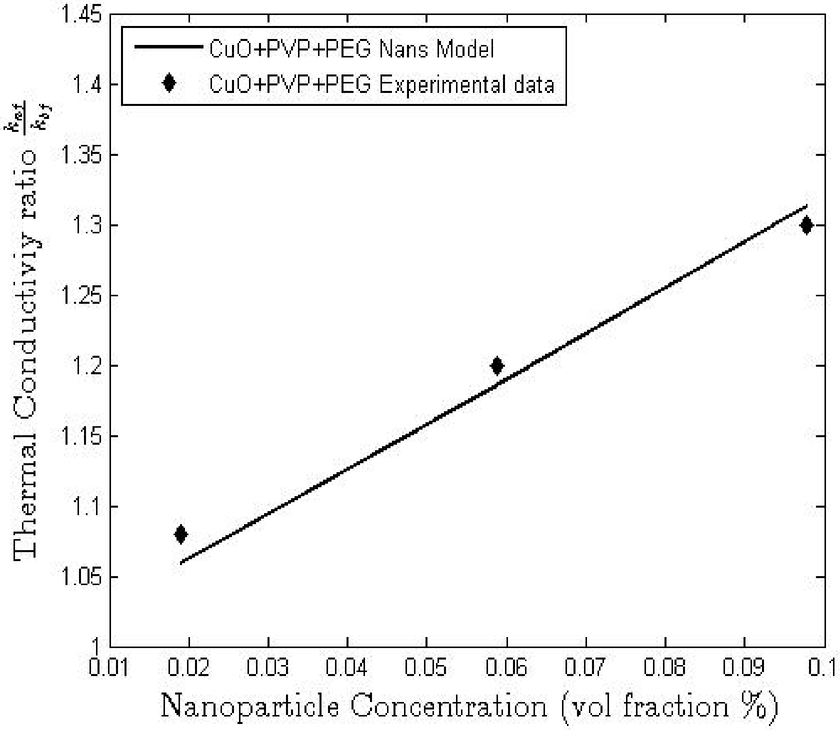
Figure 4. DLS curve for 0.5 wt% CuO+5 wt% PVP in PEG


ZnO+PVP+Water Nans Model
ZnO+PVP+Water Experimental data
Thermal Conductiviy ratio $\frac{k_{nf}}{k_{bf}}$
1.2
1.18
1.16
1.14
1.12
1.1
1.08
1.06
1.04
1.02
0.005
0.01
0.015
0.02
0.025
0.03
0.035
0.04
0.045
Nanoparticle Concentration (vol fraction %)

(a)


ZnO+PEG Nans Model
ZnO+PEG Experimental data
Thermal Conductiviy ratio $\frac{k_{nf}}{k_{bf}}$
1.18
1.16
1.14
1.12
1.1
1.08
1.06
1.04
1.02
0.02
0.04
0.06
0.08
0.1
0.12
Nanoparticle Concentration (vol fraction %)

(b)



(c)

Figure 5. Variation of thermal conductivity with nanoparticle concentration. a) ZnO+PVP+water, b) ZnO+PEG, c) CuO+PVP+PEG.

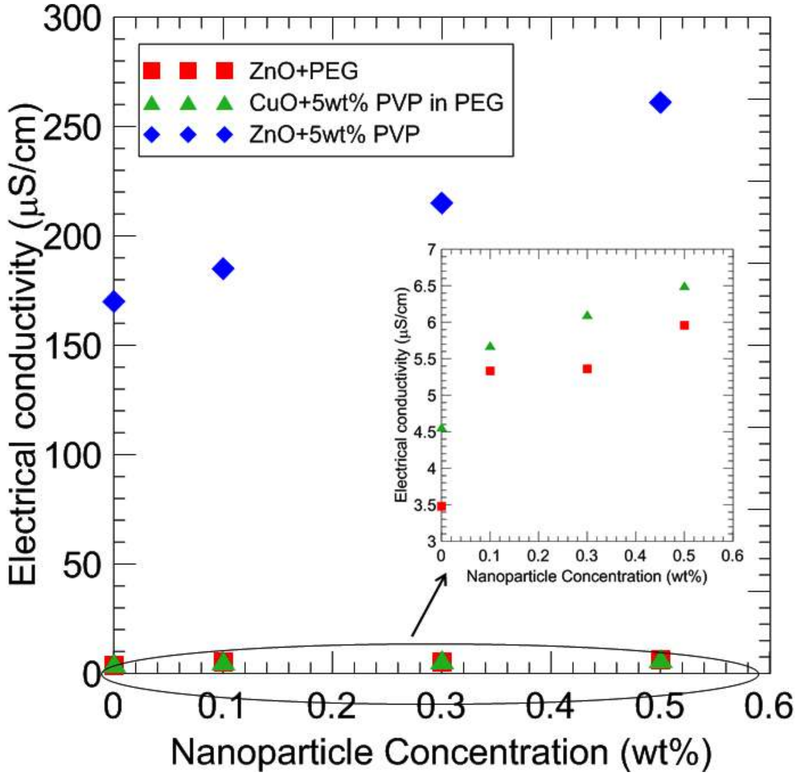


Figure 6. Variation of electrical conductivity with nanoparticle concentration