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Acoustics and thermal studies of conventional heat transfer fluids mixed with ZnO nano flakes at different temperatures

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ABSTRACT

The present investigation is mainly focused upon the effect of adding zinc oxide nano particles in the conventional heat transfer fluid involving the mixture of ethylene glycol & water (EG-water). Efforts have been made to synthesize zinc oxide nano particles by precipitation technique and to blend with EG-water to prepare nanofluids using ultrasonication. From the characterization, the shapes of the particles were found to be agglomerated nano flakes. The ultrasonic velocity and transport properties have been measured to characterize the nanofluids and they also shed light on the molecular environment of the dispersed phase at different temperatures. The dynamic viscosity shows the nanofluids to be non-Newtonian fluids at very less shear rate. In addition, the stability of the nanofluid is being interpreted from UV-visible study.

1. Introduction

Nanotechnology is considered to be the emerging technology that has brought a revolutionary step towards all scientific research for the last two decades. In the area of synthetic research, this technology is extended to prepare various metals and their oxides for the utilization in different fields like sensors [1], optoelectronics [2, 3], catalysis [4], ultraviolet light emitter, piezoelectric devices, solar cells, chemical gas sensors etc,. [5, 6]. Especifically in the area of material sciences, zinc oxide has gained significant importance due to its high thermal and mechanical stability. This oxide is considered to be a semiconductor with a broad energy band (3.37 eV) and this property makes it suitable to use in the area of catalysis, electronics, photochemical and optoelectronics [7, 8]. Besides, ZnO nanoparticles were described as promising, versatile and strategic inorganic material with an extensive range of applications [9]. As nano fertilizers, few colloidal sols of ZnO were reported to be used in the field of agriculture to increase the food crop growth and yield [10, 11, 12]. In addition, due to eco-friendly in nature, these nano structured oxides are also found to be actively utilized in environmental science for water treatment [13]. Based on these desirable properties, ZnO nano materials have gained enormous interest in all innovative field of research [14, 15]. Another class of material in the field of nano science that gained a great importance in research is nanofluid, where these fluids are supposed to have the potential to enhance the thermal conductivity of conventional heat transfer fluids. Choi and his team were the first to introduce this class of fluids [16, 17, 18]. Besides heat transfer, nanofluids have many other applications such as engine cooling and vehicle thermal management [19], nuclear system [20], fuel cells [21, 22], solar collectors [23], space [24] and even in domestic refrigerators [25]. Different types of nanofluids based on various dispersed phases and base fluids have been reported earlier [26, 27, 28, 29, 30]. Few reports have also been reviewed based on zinc oxide nanofluids [31, 32].

Varieties of heat transfer fluids both organic and aqueous origins were being used to meet the operating needs of diverse applications. Apart from water, ethylene glycol has better heat transfer properties including high density and low viscosity in comparison to many other fluids. And, a mixture of 60% EG-water has also been found to use as heat transfer fluid in many arctic and sub-arctic regions [33]. In the present investigation, 60% EG-water has been considered as the base fluid for the preparation of nanofluids.

Ultrasonic velocity measurement in liquid systems is an important technique to understand the intermolecular association between the components present in them [34, 35, 36]. The physico-chemical behaviour and inter-molecular interations involving different binary liquid

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mixtures have been reported earlier involving ultrasonic velocity [37, 38, 39, 40]. The molecular environment of nanofluids can also be studied by considering the acoustic properties. Very few studies related to ultrasonic velocity on nanofluids have been reported so far [30, 41, 42, 43]. Apart from these, another important aspect of nanofluid is its stability. UV-visible spectrophotometry is one of such techniques for the study of stability in nanofluids. Some researchers have studied the stability based on UV-visible spectrophotometry for different nanofluids [44, 45].

In continuation of our earlier work on nanofluids [43, 46], presently we report the synthesis and characterization of ZnO nanofluid in EG-water. ZnO nano particles were synthesized by systematic precipitation technique [47] and subsequently the nanofluid was prepared by dispersing nano particles in base fluid, 60% EG-water mixture. Physico-chemical properties such as viscosity, density and ultrasonic velocity were measured for the nanofluid at three different temperatures.

2. Experimental

All chemicals were procured from Sigma-Aldrich with purity of >99% and were used without any further purification. Deionized double distilled water was used throughout the experiment.

2.1. Preparation of ZnO nanofluids

Zinc oxide nano particles were synthesized by precipitation method as reported earlier by our research team [47] using the aqueous solutions 0.1M of zinc sulphate and 0.2M Sodium hydroxide as precursors with constant stirring to get a white coloured dense precipitation. This was then centrifuged and calcined at 400 °C for 2hours. Fine powders of zinc oxides nano particles were obtained after grinding the samples. The nano particles were characterized for their shape, size and arrangement.

ZnO nanoparticles were dispersed in base fluid (60% EG-water mixture) for 0.02% (w/V) concentration to get nanofluids. This solution was mixed properly for 15–20 min using magnetic stirrer before subjecting to ultra-sonication for 2 h to get a dispersed phase.

2.2. Structural characterization

The structural characterization for the shapes, sizes and arrangement of the synthesized nano particles was carried out from XRD, EDX and SEM analysis. The XRD spectra of ZnO nano particles were taken by using Rigaku smart lab X-ray Diffractometer using CuKα radiation $\lambda = 1.5405$ Å and X-rays generator operating at 40kV. The scanning range was maintained within 20–100deg with the scanning speed of 5^0min^{-1}. The XRD patterns are presented in Fig. 1. The elemental composition of ZnO nano particles has been studied from EDXA using GEMINI ULTRA 55 instruments. The EDX image for the sample has been presented in Fig. 2. The SEM morphologies and the analysis of the compositions were carried out by using GEMINI instrument at different resolutions of 50K and 100K. The images are shown in Fig. 3.

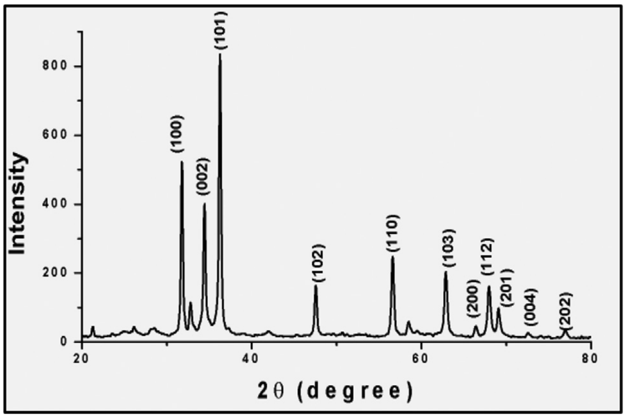


Fig. 1. X-Ray diffractogram of ZnO nano particles.

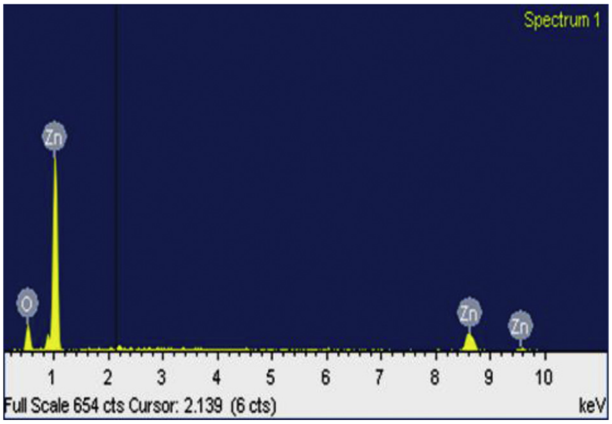


Fig. 2. EDXA image of ZnO Nano particles.

2.3. Acoustics and optical studies of nanofluids

The Ultrasonic velocity of all the nanofluids was measured at three different temperatures, 303.15, 308.15 and 313.5K by using a Mittal make single-crystal variable-path ultrasonic interferometer operating at 2MHz frequency. The temperature was controlled by circulating water from a thermostatically regulated bath around the sample holder (within ±0.01 K). The velocity measurements were précised to ±0.001 ms^{-1}. A bicapillary pyknometer was used to measure the densities of the nanofluids, which was calibrated with deionized doubled distilled water for 0.9923×10^3 kg m^{-3} as its density at temperature 303.15 K. The precision of density measurement was within ±0.0003 kg m^{-3}. A BROOKFIELD viscometer (DV-III Ultra) was used for the measurement of dynamic viscosity [48, 49] whereas the kinematic viscosities of the solutions were measured by a calibrated Ostwald viscometer. This viscometer was immersed in a constant temperature water bath maintained within ±0.01 K, and the time of flow was determined at three different temperatures. A KD-2 Pro KS-1 sensor instrument was used to determine the thermal conductivity of the samples, where it uses transient hot wire source method for the conductivity measurement. In addition, UV-Visible spectral studies have been taken for the nanofluids by using ELICO made single beam UV-Vis Spectrophotometer [Model: SL159]. Apart from this, ultrasonic velocity and density data were used to calculate isentropic compressibility, β_s, Intermolecular free length, L_f of the fluid samples by following relations [50]:

$$\beta s = 1/\rho U^2 \tag{1}$$

$$L_f = K\,(\beta s)^{1/2} \tag{2}$$

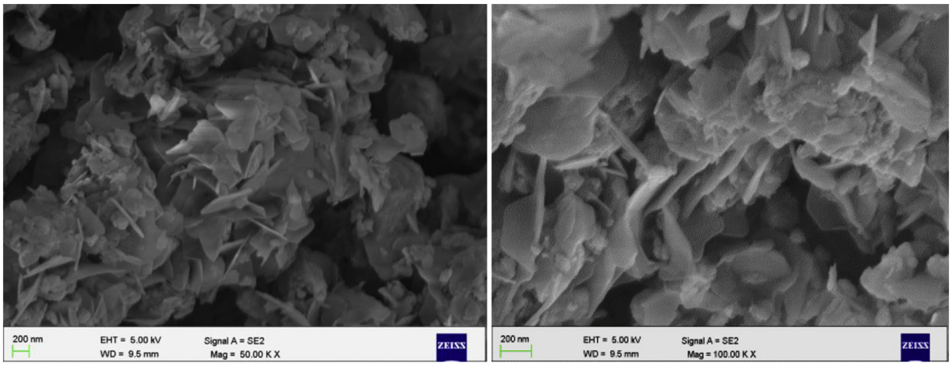
where ρ and U are the density and ultrasonic velocity for the fluid samples, K is the Jacobson's temperature-dependent constant [$=(93.875 + 0.375\,T) \times 10^{-8}$],

In addition, the thermal conductivity, k of pure liquids was calculated by using Bridgman's relation [51] and that for nanofluids, k_{nf} was done by using modified Bridgman's equation [52] as below.

$$k = 3.0\left(\frac{\rho N_A}{M}\right)^{2/3} k_B U \tag{3}$$

$$k_{nf} = 3.0\left(\frac{\rho_{nf} N_A}{M_{nf}}\right)^{2/3} k_B U \tag{4}$$

N_A is the Avogadro's number, M and M_{nf} are the molar mass of the



(a) **(b)**

Fig. 3. SEM morphology of ZnO Nano particles (a) at 50K resolution, (b) at 100K resolution.

liquid and relative molar mass of nano fluids, respectively. ρ_{nf} and k_B are the density of nanofluid and Boltzmann's constant. The measured values of ultrasonic velocity, viscosities of nanofluids and calculated values of isentropic compressibility and thermal conductivity have been presented graphically as shown in Figs. 4, 5, 6, 7, 8 and 9. Besides, the comparative studies of thermal conductivity of pure liquids are presented in Table 1.

3. Results and discussion

3.1. Structural analysis

The X-ray Diffraction patterns confirms the synthesis of high purity hexagonal wurtzite structure for ZnO nanoparticles [Ref:JCPDS file no. 36–1451] and the well defined peaks located at Bragg angles (2θ) = 31.75°, 34.34° and 36.23° are of higher intensity and corresponding to planes having Miller indices of (100), (002) and (101) respectively. The average crystallite sizes (τ) were calculated by using Debye-Scherrer equation [35, 53]

$$\tau = \frac{K\ \lambda}{\beta\ \cos\theta} \tag{5}$$

where K is Scherrer constant and know as the crystallite shape factor. λ represents the wavelength of X-ray (1.5405 Å), β is full width at half maximum of diffraction peak, and θ is the Bragg angle of intense peak. The average crystallite size of the nano particles was found to be 37nm. The EDX spectrum is a good supplement to the results of XRD analysis, where the weight % composition of zinc was found to be 69.28% and that for oxygen was 11.87%. The SEM images for ZnO nanoparticles have been taken at different resolutions, where the particles of ZnO were found to be well-defined flakes shape arranged in agglomerated form in the range of 49–179nm.

3.2. Acoustics and optical analysis

Viscosity and ultrasonic velocity are two important parameters to provide information about the molecular environment in a fluid system. And in case of nanofluids, they are dependent on the density, temperature, size and dispersion of nano particles in it [54]. In the present study, dynamic viscosity is found to be reduced initially with increased shear rate but there is not any remarkable changes with the further increase in shear rate as shown in Fig. 4. This trend in viscosity is supposed to be shear-thinning type at lower shear rate [55, 56] indicating non-Newtonian flow and the fluids show Newtonian motion at high shear

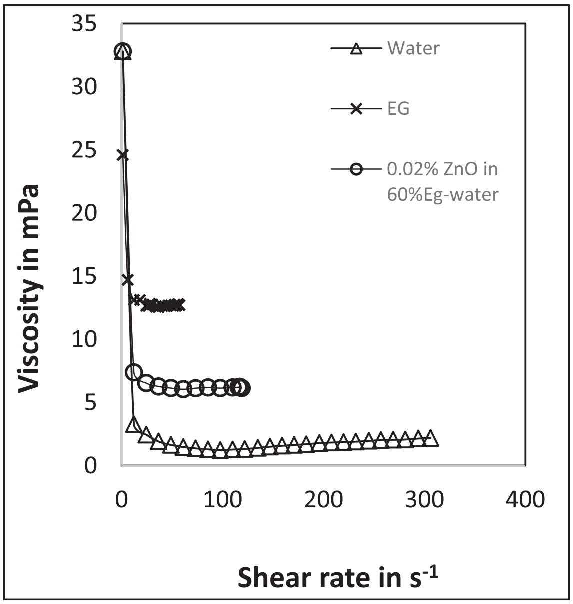


Fig. 4. Dynamic viscosity vs shear rate of only water, only EG and 0.02% ZnO nanofluids in 60% EG-water systems.

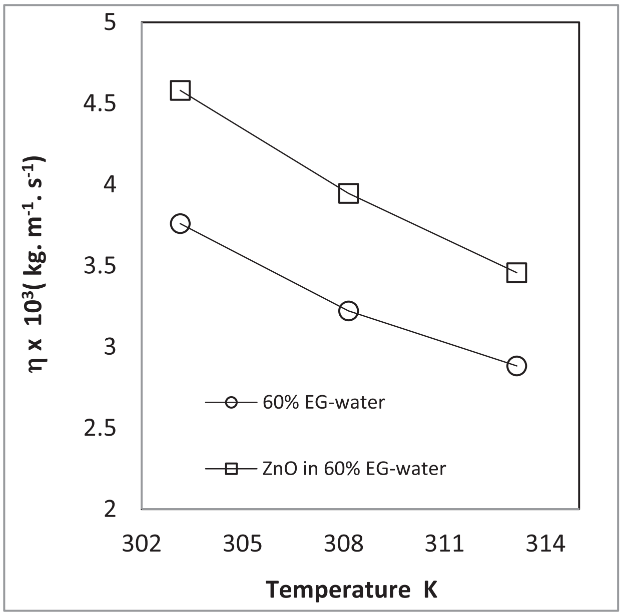


Fig. 5. Variation of viscosity, η vs Temperature for 60% EG-water and ZnO nano particles in EG-water.

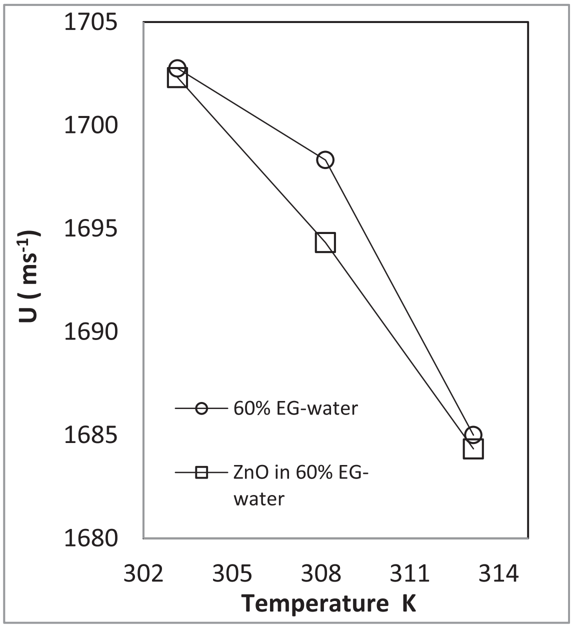


Fig. 6. Variation of ultrasonic velocity, U vs Temperature for 60% EG-water and ZnO nano particles in 60% EG-water.

rate. Again, the effective dynamic viscosity has a higher value for only EG than the nanofluid with ZnO. This is indicative of the presence of dispersed phase that facilitate the flow by reducing the viscosity. Besides, the kinematic viscosities decrease with temperature as shown in Fig. 5. This, however, indicates the increase in disturbance in particulates with temperature and makes the fluids to move fast. The ultrasonic velocity decreases slightly for nanofluid than that of base fluid. However, it decreases sharply with the increase in temperature as indicated in Fig. 6. It may be due to the increased randomness of dispersed particles in the fluid, which restrict the ultrasonic propagation in the nanofluid resulting in the reduction of the sound velocity. Isentropic compressibility, βs, that arises due to the compactness of particles [57] in fluids shows reduction with temperature in the present investigation. This decrease in compressibility of nanofluids from its corresponding base fluid as shown in Fig. 7 is due to the increased dispersion in the medium and possible association among the particulates. This makes the nanofluid to be less compressed. However, βs increases with temperature due to increased particle-mobility. Intermolecular free length, L_f, is the distance between the surfaces of the neighbouring molecules and is dependent on compressibility. This was found to be reduced in nanofluids than that of base fluids and shows similar trends with temperature as that of βs, which has been presented in Fig. 8. This is due to the increase in inter-particulate distance with temperature. In addition to this

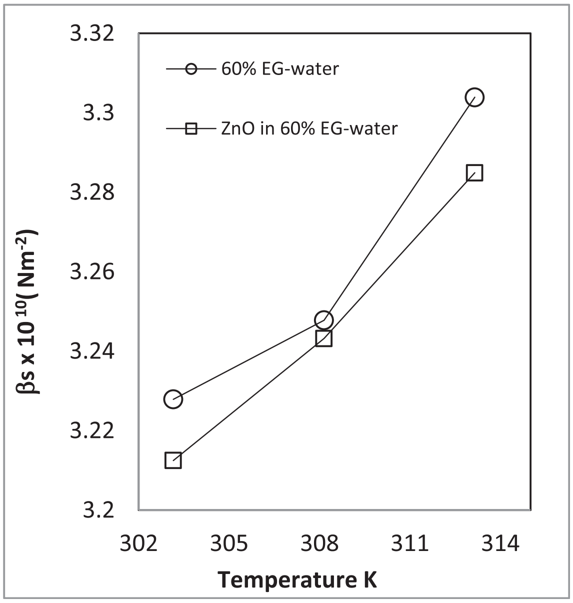


Fig. 7. Variation of isentropic compreesibility, βs vs Temperature for 60% EG-water and ZnO nano particles in 60% EG-water.

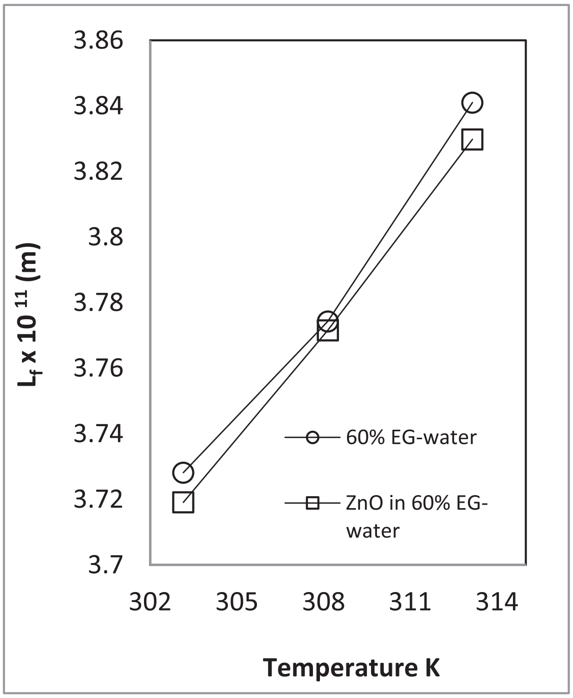


Fig. 8. Variation of Intermolecular free length, L_f vs Temperature for 60% EG-water and ZnO nano particles in 60% EG-water.

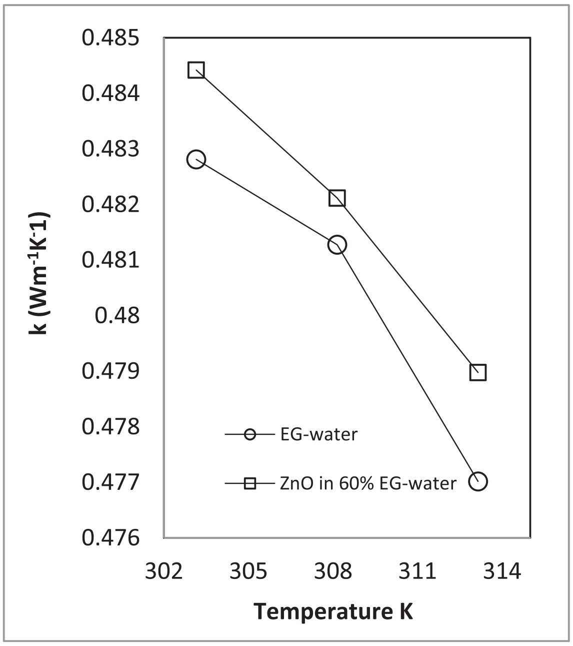


Fig. 9. Variation of thermal conductivity, k vs Temperature for 60% EG-water and ZnO nano particles in 60% EG-water.

Table 1

Comparative studies of literature, experimental and theoretical values of Thermal Conductivity of liquids.

Sl No.	Liquid samples	Thermal conductivity W.m^{-1}.K^{-1}		
		Literature	Experimental	Theoretical
1	Ethylene Glycol	0.254	0.247	0.334
2	Water	0.606	0.664	0.638
3	60% EG + Water	—	0.349	0.482

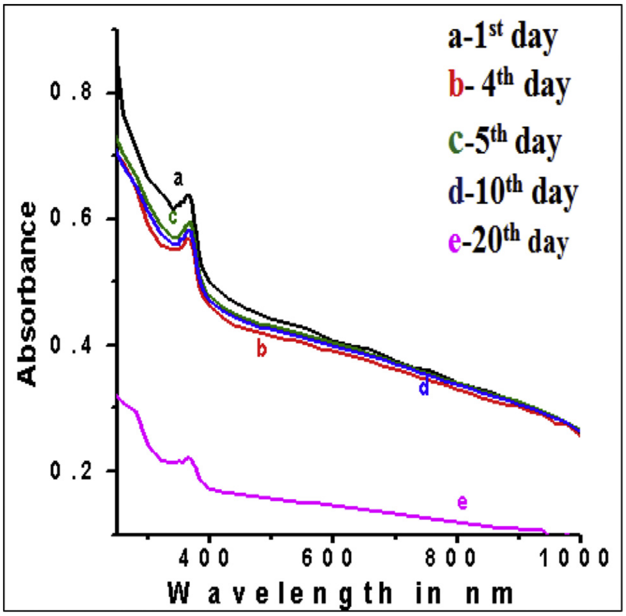


Fig. 10. Variation of UV-visible absorption spectrum of ZnO in 60% EG-water.

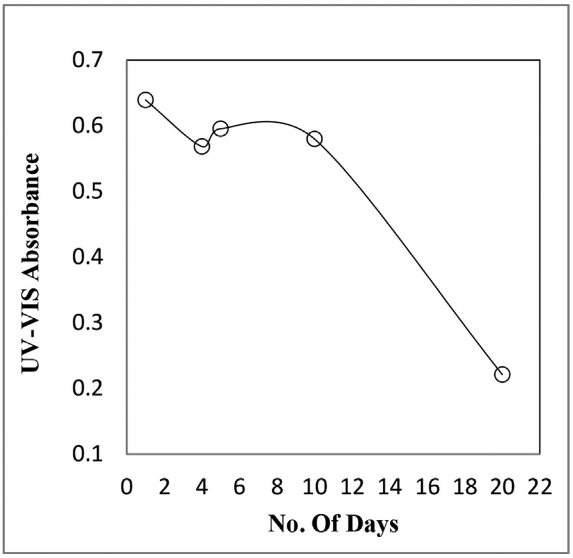


Fig. 11. Variation UV-visible absorption for different days at 365 nm.

particulate-association study, ultrasonic velocity measurement can also be utilized to evaluate thermal conductivity theoretically. For pure liquids like water and ethylene glycol, the theoretical values of thermal conductivity is found to be agreeing with experimental as well as from literature data. But for mixture and nanofluid, the errors are comparatively more. Again, the evaluated value of thermal conductivity for nanofluid is more than that of base fluid. But, however, with temperature, it decreases gradually as indicated in Fig. 9. In the present investigation, both the ultrasonic velocity and density values decrease with temperature and as per Bridgman's relation, thermal conductivity depends on these two factors directly.

The size of the nanoparticles plays an important role in changing the entire properties of materials and UV-visible absorption spectroscopy is being used widely to examine the optical properties of nano-sized particles. The stability of prepared ZnO nanofluid was carried out by studying the optical properties as these particles were found to be revealed by UV–visible spectrum at room temperature. It can also be observed in Fig. 10 that there was intensive absorption in the ultraviolet band of about 200–400 nm and the peak of absorbance was obtained at 365nm for the nanofluid. As per the literature such type of absorption can be referred to excitonic character [36]. The absorbance had also been measured to study the stability responses for a period of 20 days after preparation and presented here in Fig. 11. It has also been observed that the stability of dispersed phase continues to be stable almost for 10 days without much settlement. But after 15 days, the particles start settling down as per the visual observation. The intensity of peak for 20th day clearly indicates the reduced stability of nanofluid [58].

4. Conclusion

Zinc oxide nano particles have been synthesized successfully and the particles were found to be agglomerated flakes type. These nano particles were blended with the heat transfer fluids for the preparation of nanofluids. The acoustic studies reveal the presence of randomness among the particles from compressibility and free length studies. This is indicative of better dispersion and inter-particulate association of suspended particles in fluid. The stability of nanofluids is shown by UV-visible absorption study and as per this analysis, the dispersion phase is stable for at least ten days without much settlement. The rheological studies reveal the nanofluids to be behaved like non-Newtonian fluids at lower shear rate and the behaviour changes as the shear rate increases. Besides, the Bridgman relations for evaluation of thermal conductivity from ultrasonic data are found to be quite impressive for the pure liquids. However, further investigations are required to increase the stability of such nanofluids before it can be utilized for commercialization.

Declarations

Author contribution statement

Venu Gopal V. R.: Performed the experiments.

Susmita Kamila: Conceived and designed the experiments; Analyzed and interpreted the data; Wrote the paper.

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Competing interest statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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