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Research Article

Insights of physico-chemical and thermal conductivity enhancement of polyvinylidene fluoride (PVDF) mixed znonanoparticles for heat transfer applications

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ABSTRACT

The conventional heat transfer liquids such as water, oil, etc., having lesser thermal properties cannot be fulfill the requirements of some of the heat exchangers system. To enhance the thermal properties of the liquid, nanoparticles are dispersed. Nanofluids (NF) have potential application in thermal transfer systems. The motto of the present work is devoted to studying thermo-physical studies on ZnO with different weight percentages of Polyvinylidene Fluoride (PVDF)-added nanocomposite-based nanofluids at various concentrations in water for heat transfer applications. The prepared nanocomposites were dispersed into the distilled water to prepare nanofluids and were characterized by ultrasonic velocity measurements to figure out the molecular interaction and thermal behavior of the nanofluids. Various physicochemical Characteristics, such as adiabatic compressibility, intermolecular free length, free volume, internal pressure, specific acoustic impedance, relaxation time, and surface tension, were determined. The mechanical properties of the PVDF ZnO NFs were analyzed based on the findings of the bulk modulus. Moreover, the thermal conductivity of the PVDF ZnO nanofluids was theoretically analyzed based on density and ultrasonic velocity measurements. The PVDF mixed with ZnO nanoparticle shows better thermal properties compared with conventional liquid say water.

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INTRODUCTION

The improved heat exchanger is important in many areas where a huge amount of heat is generated such as power plants and coolants. An effective heat exchanger with the wanted cooling performance and an effective coolant with pass-on heat are required. Hence, there is a need for better coolants with excellent thermo physical characteristics

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of the coolants such as thermal conductivity and viscosity is essential. However, conventional coolants like water and glycol having poor thermal conductivity hinders their practical applications. The fluids have less viscosity is easy to circulate the system or device without using any additional pumping technology. So, there is a need for alternative better coolants with greater thermal conductivity and lesser viscosity to be the better choice of coolants. Recently, the dispersion of solid materials such as alumina, titanium dioxide and zinc oxide into the conventional liquid water to improves its heat exchange characteristics of flat plate water solar collectors in shell and tube type heat exchangers [1-3]. The several metal oxides like $Al_2O_{3,}$ CuO and TiO_2 are the most commonly reported materials for preparing nanofluids [4, 5]. But, a scan of literature survey showed that there are few reports on ZnO based nanofluids for their use in heat transfer applications. Long term stability of a nanofluid remains a big challenge in colloidal science due to Van der Waals forces existing between the nanoparticles leads to agglomeration of nanoparticles reduce its dispersion stability and hence increase its sedimentation rate. The nanoparticle-liquid interaction plays a vital role in enhancing the thermo-physical properties of the nanofluids. Polyvinylidene fluoride (PVDF) is a well-known semi-conducting polymer with a wide range of applications. It shows several crystalline phases like α, β, γ, δ, and ε. Its properties lead to various scientific and technological applications in flexible optoelectronic devices. The polymer PVDF is a well-known material for the applications of piezoelectric nanogenerator as well as used as a binder for electrodes preparing in battery and supercapacitors applications. So, it is essential to understand the microstructure and molecular interactions between solute-solute and solute-solvent interaction play a vital role in heat transfer as well as dispersion stability. Hence, the present work focuses on studying the heat transfer properties of Zinc Oxide with PVDF nanocomposites-based nanofluids and their stability.

EXPERIMENTAL DETAILS

Preparation of Nanofluids

Initially, we prepared Zinc Oxide nanoparticle by wet chemical method. Adding 10%, 20%, and 30% of PVDF into ZnO materials and grind well for homogeneous ZnO with PVDF nanocomposites (ZnO PVDF1, ZnO PVDF2, and ZnO PVDF3). Then the prepared ZnO sample was dissolved in triple distilled water at different concentrations 0.025M, 0.05M, 0.1M, and 0.2M and sonicated for 15 minutes at room temperature. Similarly, other nanofluids such as ZnO PVDF1 NFs, ZnO PVDF2 NFs, and ZnO PVDF3 NFs were prepared.

Characterization Techniques

An ultrasonic interferometer (F-81 type, MITTAL manufacture, 2 MHz frequency, accuracy: ±0.01 m/s) at 303.15 K was used to detect ultrasonic velocity. The viscosity of the NFs was measured at 303.15 K with a digital viscometer (accuracy: $\pm 0.01 \times 10^{-3}$ Ns/m^2; BROOKFIELD manufacture, USA). The NFs' density was measured using a specific gravity bottle. We used a SHIMADZU electronic balance to determine the liquid's mass, which has a precision of ±0.001 gm. To measure the refractive indices, an Abbe refractometer with an accuracy of ±0.001 was utilized. All the measurements were found at room temperature using an electronically controlled thermostat.

RESULTS AND DISCUSSION

Viscosity Measurements

Viscosity is a key characteristic of NFs that affects their overall performance. It plays a crucial role in determining pumping power, pressure drop during laminar flow, heat transfer by convection, and volume fraction. Various factors influence the viscosity of NFs, such as the shape and size of particles, the concentration of nanomaterials in the fluid, the type of surfactants, shear rate, particle clumping, and the fluid's pH level.

The shape of the dispersed nanomaterials significantly affects the viscosity of NFs. Timofeeva et al. [4], have studied the viscosity analysis of different morphology of alumina based nanofluids. It was noticed that elongated particles exhibit higher viscosity compared to those with spherical nanoparticles. The viscosity of nanofluids greatly impacts their flowability of nanofluids [5]. Additionally, the fluid's capacity to adapt to various flow regimes and buoyancy forces is determined by its viscosity and density.

In this study, viscosity measurements were performed for pure metal oxide ZnO NFs, along with ZnO PVDF1 NFs, ZnO PVDF2 NFs, and ZnO PVDF3 NFs, as shown in Figure 1. Based on Figure 1, it can be detected that the viscosity enhances almost linearly as the concentration rises for all ZnO PVDF samples (ranging from 0.025% to 0.2% mass concentration). At the higher concentration (0.2%), a 1.05% increase was observed when comparing ZnO NPs with ZnO PVDF NPs. The rise in viscosity is associated with a higher concentration of nanoparticles in the base fluid, leading to increased internal viscous shear stresses. [6-7]. The significant variations suggest that interactions between the constituents and the base fluid are attributed to the direction of the higher surface area of the NPs [8].

Density Measurements

The NFs density is defined as the mass per unit volume of a fluid containing suspended nanoparticles. It is a critical property that significantly influences their behaviour and performance in various applications. Density is a key factor in thermal conductivity and heat transfer coefficients [9], which are critical for applications such as cooling systems and heat exchangers. It also affects viscosity [10], Reynolds

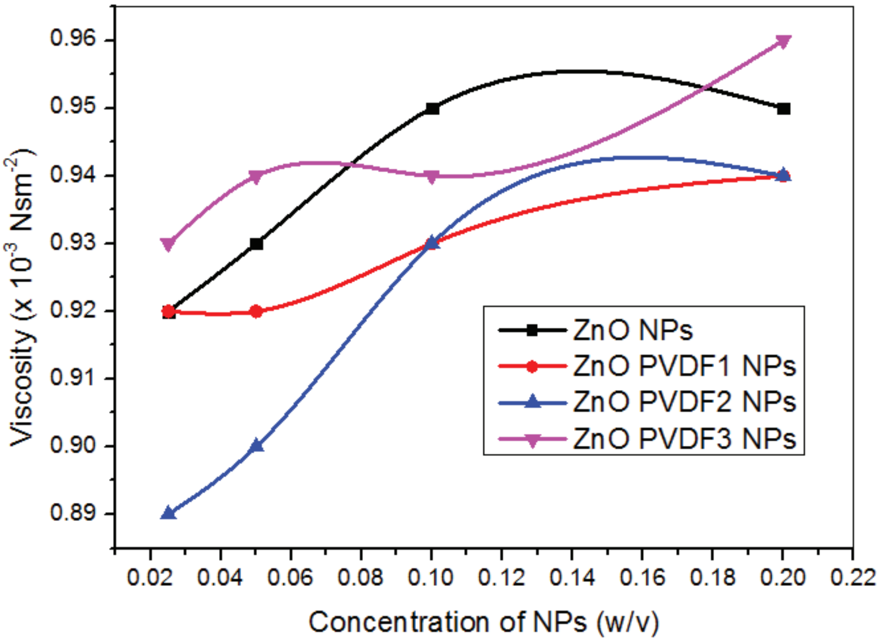

Figure 1. Viscosity studies of ZnO and ZnO PVDF NPs.

number [11] and pressure drop in fluid flow [12], which influences the design and efficiency of pumping systems. Understanding density is essential for sustaining the stability of nanoparticle suspensions and blocking sedimentation. Changes in density can result in buoyancy-driven flows, impacting heat transfer patterns in systems that use nanofluids. Reliable density data are essential for creating models that predict the performance of nanofluids in various applications.

In this study, we measured the density of pure metal oxide ZnO NFs, as well as ZnO PVDF1 NFs, ZnO PVDF2 NFs, and ZnO PVDF3 NFs, as presented in Figure 2. The results indicate that the density increases with concentration for all ZnO PVDF samples linearly due to more number of nanoparticles [13]. At a lower concentration (0.025%), the density values follow this order: ZnO NFs < ZnO PVDF2 NFs < ZnO PVDF3 NFs < ZnO PVDF1. Conversely, at a higher concentration (0.2%), the order is: ZnO PVDF1 < ZnO PVDF2 NFs < ZnO NFs < ZnO PVDF3.

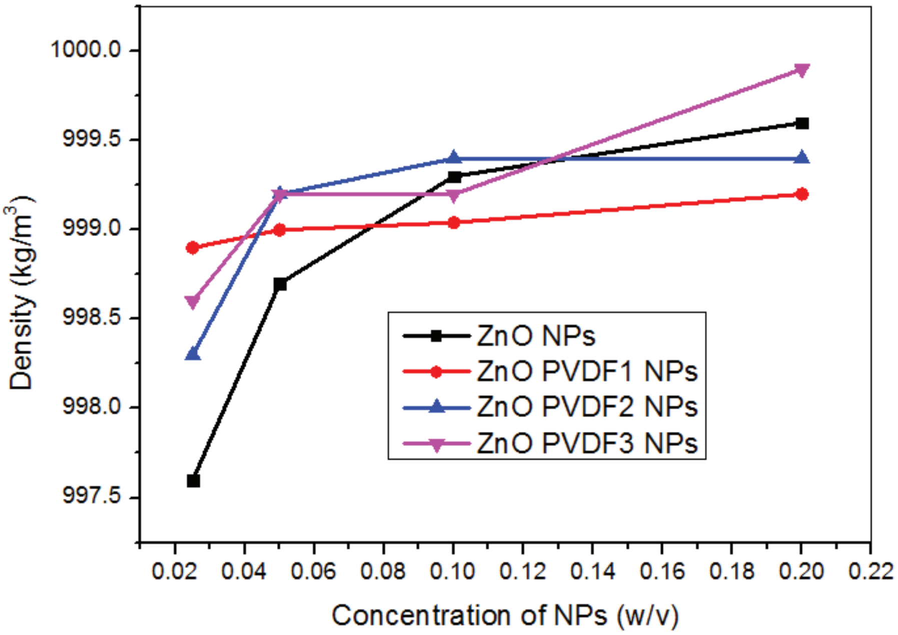

Figure 2. Density studies of ZnO and ZnO PVDF NPs.

Refractive Index (RI) Analysis

The refractive index offers valuable insights into the optical properties of nanofluids, which are crucial for applications related to light propagation, including optical devices, sensors, and photonic technologies [14]. By understanding the RI, one can characterize nanofluids and assess their composition, revealing the impact of nanoparticle addition on the optical behavior of the base fluid. It can also serve as a quality control measure during the synthesis and formulation of nanofluids; consistent values indicate the uniformity and stability of the nanoparticle suspension. Additionally, it can correlate with other characteristics, like thermal conductivity and density, providing a comprehensive understanding of the fluid's behavior and performance [15]. It can help estimate the concentration of NPs in a nanofluid, which is particularly useful for monitoring and controlling concentrations in various applications. Changes in the RI may indicate the stability of NFs, with significant variations over time suggesting issues like particle aggregation or sedimentation that could affect fluid performance. Moreover, the refractive index affects how NFs interact with light, including their absorption and scattering properties, which is particularly important for applications such as solar energy harvesting, where optical characteristics directly influence efficiency [16]. If the refractive index of the base liquid is uncertain, it is difficult to calculate the absorption coefficient from the transmission spectrum [17].

Figure 3 illustrates the relationship between refractive index and concentration for ZnO NPs, ZnO PVDF1 NPs, ZnO PVDF2 NPs, and ZnO PVDF3 NPs at 303 K. At a lesser concentration of 0.025%, the refractive index is at its lowest, with values of 1.334, 1.333, 1.333, and 1.334 for ZnO NPs, ZnO PVDF1 NPs, ZnO PVDF2 NPs, and ZnO PVDF3 NPs, respectively. Conversely, at the higher concentration of 0.2%, the refractive index reaches its peak, with values of 1.335, 1.335, 1.3355, and 1.336 for ZnO NPs, ZnO PVDF1 NPs, ZnO PVDF2 NPs, and ZnO PVDF3 NPs, respectively.

Ultrasonic Velocity (U) Measurements

The ultrasonic technique has become a crucial tool for investigating the physicochemical characteristics of materials and has widespread applications in fundamental science, industry, and biochemical technology [18-20]. Measuring ultrasonic velocity and associated parameters in solvents is key to understanding the physicochemical behavior of solutions. This allows for the calculation of important thermodynamic properties, shedding light on molecular interactions [21]. Additionally, ultrasonic velocity helps assess properties like density, compressibility, and viscosity, which are critical for analyzing the thermal and mechanical performance of NFs in applications such as heat transfer, lubrication, and drug delivery. Variations in ultrasonic velocity can also reflect changes in nanoparticle concentration, size, and distribution within the fluid, making it an essential tool for characterizing NFs.

In this study, we measured the ultrasonic velocity of pure ZnO nanofluids (NFs), as well as ZnO PVDF1, ZnO PVDF2, and ZnO PVDF3 NFs, as illustrated in Figure 4. It is evident from Figure 4 that the ultrasonic velocity enhances as the concentration of NPs rises. This indicates that structural changes are taking place in the liquid system, leading

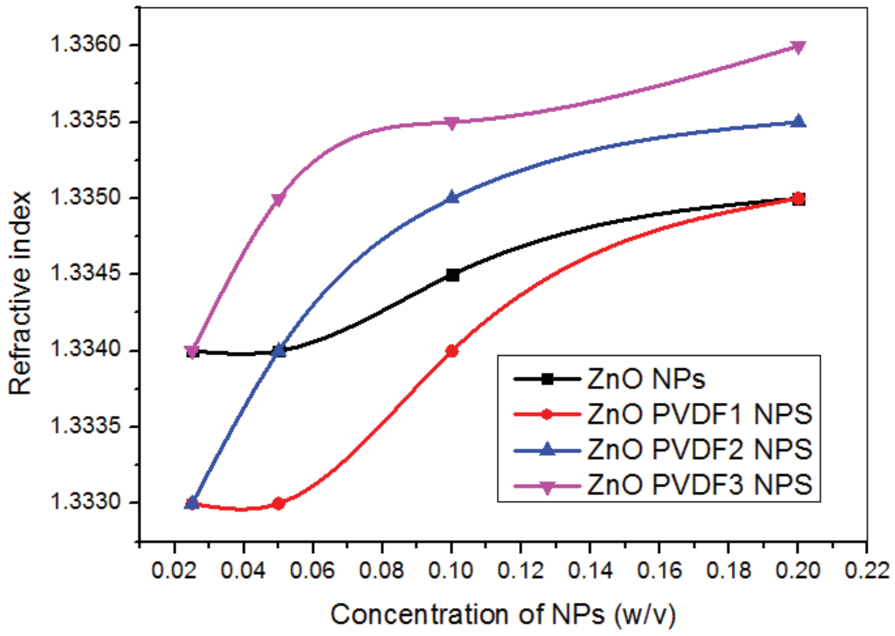

Figure 3. Refractive index studies of ZnO and ZnO PVDF NPs.

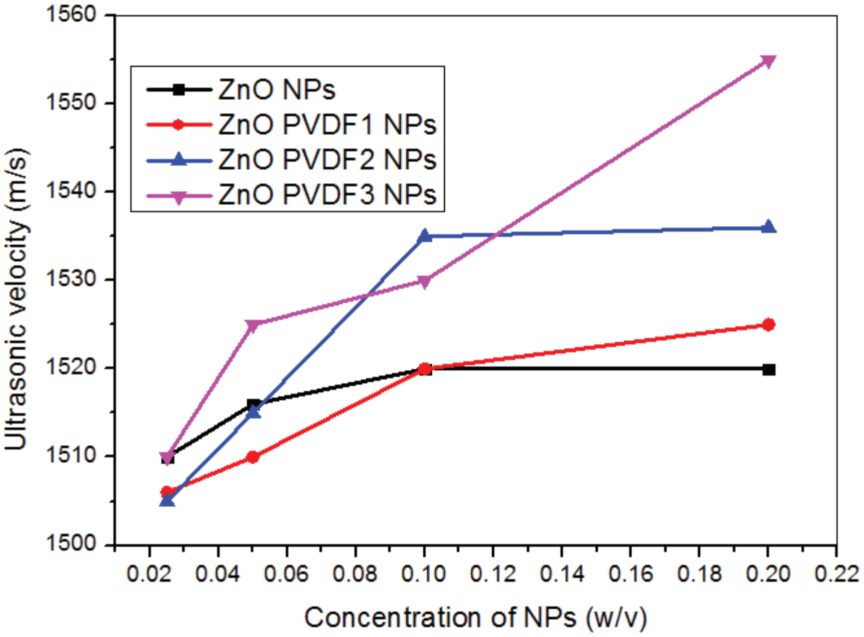

Figure 4. Ultrasonic velocity studies of ZnO and ZnO PVDF NPs.

to a reduction of intermolecular forces. The increase in velocity suggests that the interaction may be caused by surface effects. Consequently, particle-fluid interactions likely contribute to the rise in velocity values [22].

Determination of Adiabatic Compressibility (β_{ad}) and Bulk Modulus (K)

Adiabatic compressibility describes how much the volume of a substance decreases with each unit rise in sound pressure, without any heat or mass being transferred to the environment. The adiabatic compressibility (β_{ad}) and bulk modulus (K) of ZnO NFs and ZnO PVDF NFs can be derived from the following formulae 1 and 2 [20]:

$$\beta_{ad} = 1/U^2\rho \quad m^2N^{-1} \tag{1}$$

$$K = 1/\beta_{ad} \quad Nm^{-2} \tag{2}$$

Where ρ represents the density and U denotes the velocity of the NFs.

Adiabatic compressibility is directly influenced by density and ultrasonic velocity, showing an inverse relationship with velocity [23]. In this study, the adiabatic compressibility decreases as the concentration of nanoparticles (NPs) increases in all cases [Fig. 5]. The primary reason for this reduction in adiabatic compressibility with rising NP concentration is the corresponding increase in nanofluid (NF) density. As more NPs are dissolved in the base fluid, the NF density rises, leading to a decrease in the value of adiabatic compressibility, since it is inversely proportional to density [24]. Figure 6 demonstrates a linear increase in bulk modulus across all cases. At a 0.2% concentration of ZnO in PVDF3 NFs, the bulk modulus reaches 2.42 GPa, indicating a significant increase because of dipole-induced dipole and dipole-dipole interactions, which results in a positive shift in velocity and a negative shift in compressibility [25–26]. The adiabatic compressibility is an important parameter that changes with structure. The decrease in adiabatic compressibility with increase in concentration of the solute indicates that a close packing of solute molecules resulting structure making effects of PVDF mixed with ZnO by the water solvent. Hence, there is an increase in the bulk modulus values due to less change of volume.

Thermal Conductivity Analysis

Thermal conductivity serves as a measure of a material's capacity for heat transfer. Researchers aiming to find the thermal conductivity of nanofluids (NFs) through experimental methods have developed and documented various techniques in the literature [27]. This property in NFs is influenced by several factors, including NP characteristics (such as shape, concentration, and size) [28, 29]; temperature [28]; stability, preparation, and sonication conditions [30, 31]; the presence and quantity of surfactants [28, 29]; base fluid properties [32]; methods for measuring thermal conductivity [33]; and NP alignment [32, 34]. These factors often result in inconsistencies in reported data.

For ZnO-based NFs, including ZnO NFs, ZnO PVDF1 NFs, ZnO PVDF2 NFs, and ZnO PVDF3 NFs, their thermal conductivity is derived from ultrasonic velocity measurements. The calculated values are obtained using a modified Bridgman equation [35].

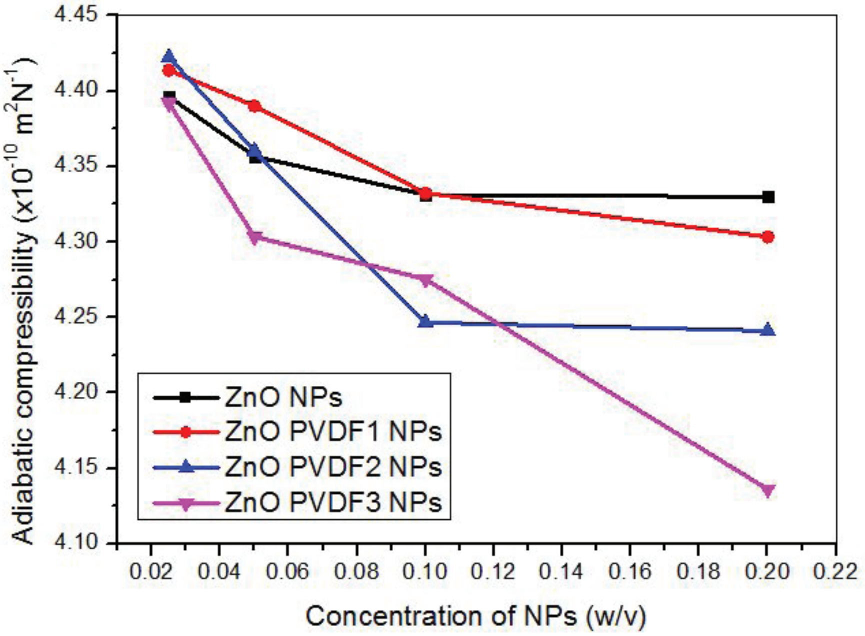

Figure 5. Adiabatic compressibility studies of ZnO and ZnO PVDF NPs.

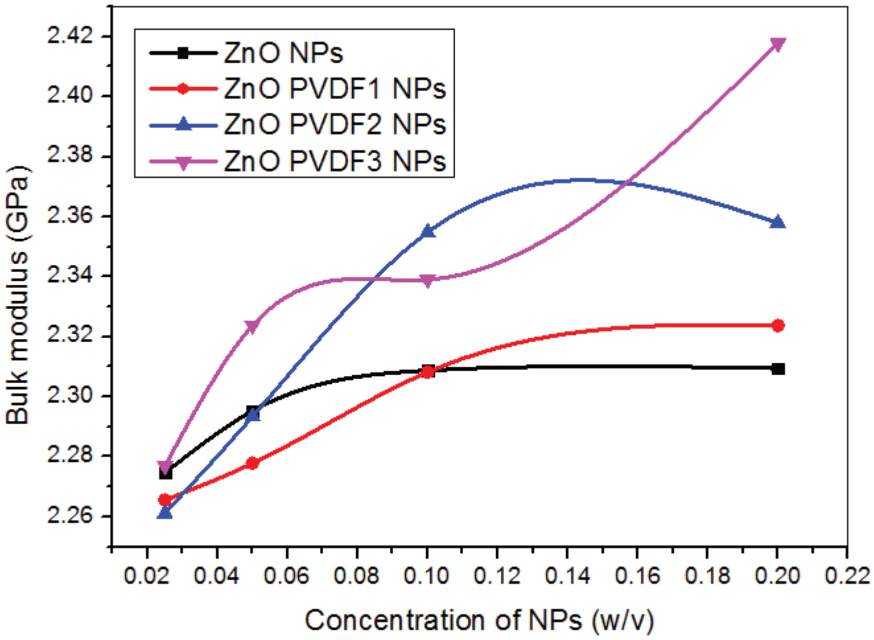

Figure 6. Bulk modulus studies of ZnO and ZnO PVDF NPs.

Figure 7 shows the thermal conductivity of ZnO NPs, ZnO PVDF1 NPs, ZnO PVDF2 NPs, and ZnO PVDF3 NFs. At a higher concentration, when comparing the thermal conductivities of ZnO NPs is less whereas ZnO PVDF3 NPs exhibit better thermal conductivity. Nanofluids demonstrate high thermal conductivities even at very low nanoparticle concentrations, though the precise mechanism behind this enhancement remains unclear. Brownian motion of suspended NPs is considered one of the primary factors contributing to the significant increase in thermal conductivity [36].

Physicochemical Properties

Table 1 represents the various physicochemical properties (intermolecular free length (L_f), free volume (V_f), internal pressure (π), specific acoustic impedance (Z), relaxation time (τ) and Surface tension were evaluated for

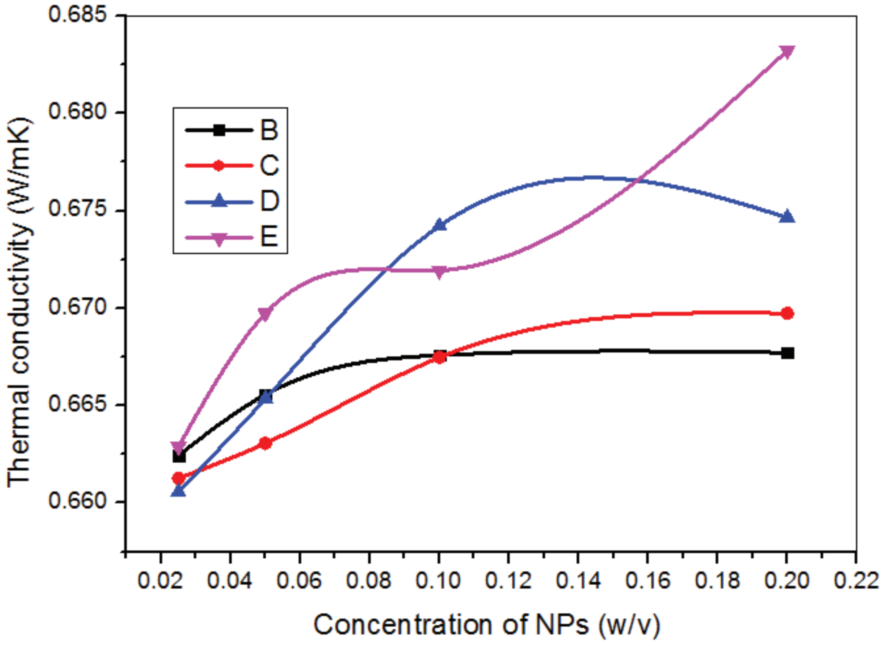

Figure 7. Thermal conductivity studies of ZnO and ZnO PVDF NPs.

Table 1. Various physicochemical properties of NPs at 303.15 K

Name of the sample	Concentration (%)	Intermolecular free length (L_f) (x 10^{-11} m)	Free volume (V_f) (x 10^{-8} m^3 mol^{-1})	Internal pressure (π) (GPa)	Specific acoustical Impedance (Z) (x 10^6 $kgm^{-2}s^{-1}$)	Relaxation time (τ) (x 10^{-13} s)	Surface tension (N/m)
ZnO NFs	0.025	4.3507	1.8135	2.7201	1.5063	5.3928	38.6670
	0.05	4.3311	1.7950	2.7315	1.5140	5.4024	38.8977
	0.1	4.3184	1.7455	2.7581	1.5189	5.4863	39.0518
	0.2	4.3177	1.7455	2.7587	1.5193	5.4846	39.0518
ZnO PVDF1 NFs	0.025	4.3594	1.8063	2.7261	1.5043	5.4144	38.5135
	0.05	4.3476	1.8135	2.7227	1.5084	5.3852	38.6670
	0.1	4.3190	1.8021	2.7285	1.5185	5.3721	39.0518
	0.2	4.3044	1.7822	2.7389	1.5237	5.3935	39.2446
ZnO PVDF2 NFs	0.025	4.3636	1.8965	2.6811	1.5024	5.2480	38.4751
	0.05	4.3329	1.8836	2.6888	1.5137	5.2324	38.8593
	0.1	4.2760	1.8288	2.7158	1.5340	5.2658	39.6313
	0.2	4.2732	1.8015	2.7294	1.5350	5.3155	39.6700
ZnO PVDF3 NFs	0.025	4.3485	1.7843	2.7367	1.5078	5.4459	38.6670
	0.05	4.3044	1.7822	2.7389	1.5237	5.3935	39.2446
	0.1	4.2904	1.7909	2.7344	1.5287	5.3583	39.4378
	0.2	4.2199	1.7780	2.7423	1.5548	5.2941	40.4084

PVDF-mixed ZnO nanofluids (PVDF ZnO NFs) to explore the effects of component composition and PVDF ZnO concentration on these characteristics.

The free length represents the path length a sound wave covers as it moves between the surfaces of adjacent molecules [37]. The intermolecular free length can be determined using the equation proposed by Jacobson [38]. Intermolecular free length is given by

$$L_f = \frac{K'}{U\sqrt{\rho}}\ (\mathrm{m}) \tag{3}$$

Where K' =2.131 * 10^{-6} is a constant which is temperature dependent. The intermolecular free length measured for pure ZnO NFs and ZnO-PVDF mixtures at room temperature decline with increasing concentrations, is presented in Table 1 signifies a substantial interaction among the particles and the base fluid molecules, [39] as noted by Gupta et al. [40].

Free volume refers to the unoccupied space resulting from the irregular arrangement of molecules. The free volume values for ZnO and ZnO PVDF NFs follow a trend

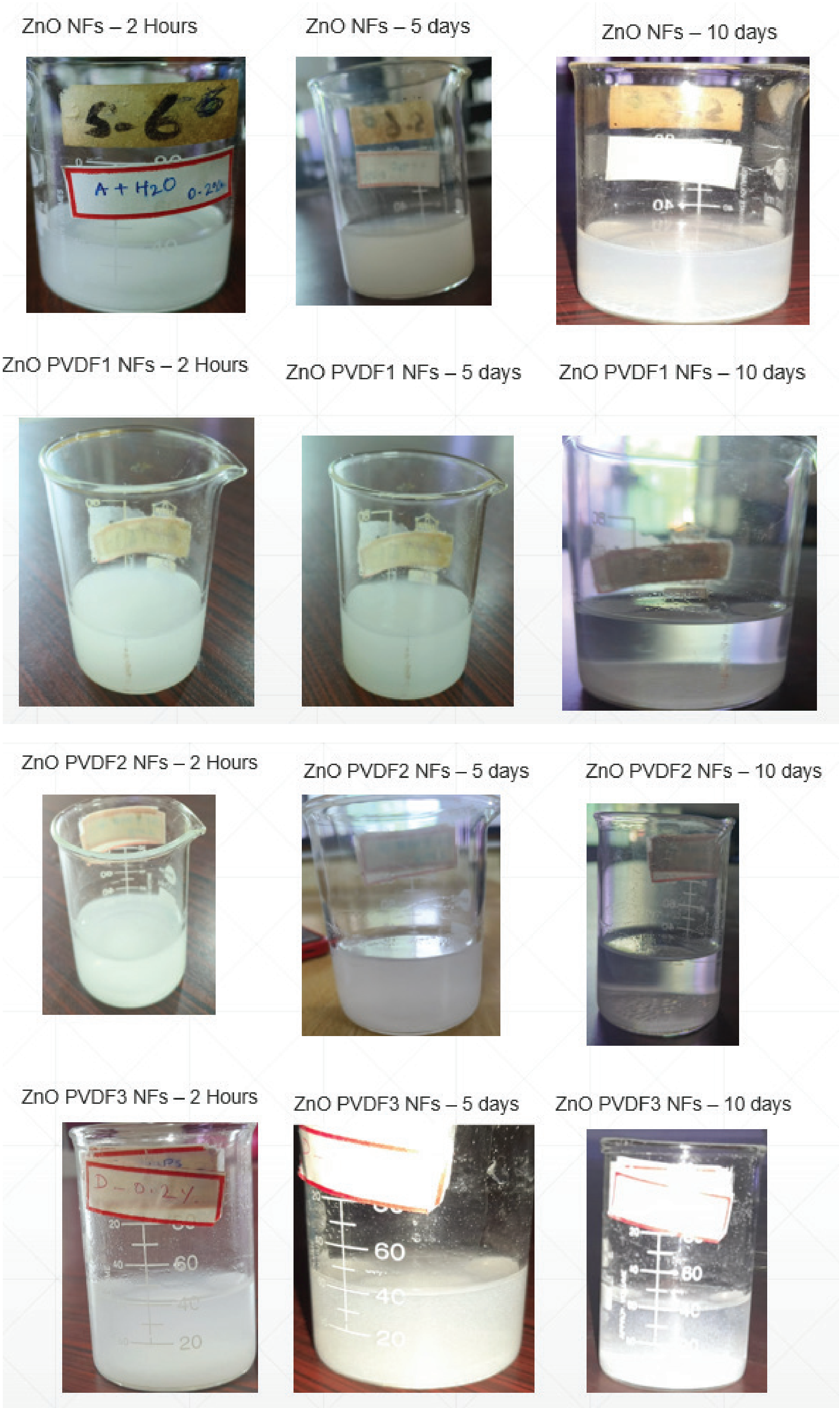
ZnO NFs – 2 Hours
ZnO NFs – 5 days
ZnO NFs – 10 days
ZnO PVDF1 NFs – 2 Hours
ZnO PVDF1 NFs – 5 days
ZnO PVDF1 NFs – 10 days
ZnO PVDF2 NFs – 2 Hours
ZnO PVDF2 NFs – 5 days
ZnO PVDF2 NFs – 10 days
ZnO PVDF3 NFs – 2 Hours
ZnO PVDF3 NFs – 5 days
ZnO PVDF3 NFs – 10 days

Figure 8. Sedimentation analysis of the nanofluids.

opposite to that observed for viscosity (Table 1. A decrease in free volume is observed as internal pressure increases [41], but it is reversed in free volume and internal pressure clearly shown in Table 1.

Specific acoustic impedance (*Z*) is the product of the material's density and the speed of sound in that material. It is related to density and ultrasonic velocity through the following relationship [42].

Specific acoustical impedance

$$Z = U\rho \text{ (kg m}^{-2}\text{ s}^{-1}) \quad (4)$$

The interaction between the NPs and base fluid molecules expands the intermolecular distance, leading to a rise in specific acoustic impedance [43].

Relaxation time (τ) results from the time lag between the passage of the ultrasonic wave and the return of molecules to their equilibrium position. It is calculated using the following relation.

$$\text{Relaxation time } \tau = \frac{4\eta}{3\beta_{ad}} \text{ (sec)} \quad (5)$$

In the current investigation, the relaxation time rises with the concentration of NPs in all cases. This indicates that at higher concentrations the molecular interaction between the base liquid and the NPs is stronger. Typically, an increase in NP concentration can modify the surface tension. The interaction between the NPs and the base fluid can either raise or lower the surface tension, depending on the properties of the nanoparticles and how they interact with the fluid. The chemical composition and surface characteristics of the NPs are key factors in determining the surface tension of the nanofluid. From Table 1, an increasing trend of surface was observed for NFs. The increasing surface tension as the concentration rises; accumulates more particles in the solution. A strong cohesive force develops between the NPs and the liquid molecules, leading to an increase in the surface tension of the nanofluids with higher concentrations [44].

Nanofluids Stability

In this present study, we adopted the sedimentation method to study the stability of the prepared nanofluids. The optical images of the dispersion of ZnO and ZnO PVDF nanoparticles (NPs) in the base fluid at a higher concentration (0.2%) are shown in Figure 8. Based on the observation from Figure 8, it can be seen that the nanofluids (NFs) exhibit moderate stability. The NFs remained stable for 5 days, after which the NPs began to settle at the bottom of the beaker. To obtain a long-term stability of the nanofluids is a big task. It can be achieved by low van der Waals force between the nanoparticle in the fluid medium. This force is responsible for aggregates the nanoparticle and separated out from the liquid and finally settled down due to gravity. However, the addition of dispersant and sonication will improve the stability of the nanofluids. There are several methods are used to analyze the stability of the nanofluids such as sedimentation method, centrifugation method, zeta potential method, etc. Among them, the sedimentation method is a simple and economic method to analyze the stability of the nanofluid.

CONCLUSION

In the present work, we have prepared ZnO and ZnO PVDF nanofluids are synthesized via in two stages with the assistance of a sonicator. The structural and morphological analysis of prepared zinc oxide nanoparticle was analyzed using XRD and SEM, techniques. Then the prepared ZnO and PVDF mixed ZnO nanofluids were prepared by using water as a solvent. Ultrasonic velocity measurements were done and other acoustic parameters were derived. The solute-particle interaction and thermal conductivity enhancement of the nanofluids are investigated based on the obtained acoustic characteristics. The ZnO nanofluids showed better stability than PVDF mixed ZnO PVDF nanofluids. However, there was an enhancement in thermal conductivity was observed in PVDF mixed ZnO nanofluids. So, still there is a challenge to obtain better stability of PVDF mixed ZnO fluids.

AUTHORSHIP CONTRIBUTIONS

Authors equally contributed to this work.

DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

CONFLICT OF INTEREST

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

ETHICS

There are no ethical issues with the publication of this manuscript.

STATEMENT ON THE USE OF ARTIFICIAL INTELLIGENCE

Artificial intelligence was not used in the preparation of the article.

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