J. Indian Chem. Soc.,
Vol. 97, June 2020, pp. 895-902

Acoustical studies of nickel nanoparticles based nanofluids: Concentration and temperature dependent approach

Vijayta Gupta*, Samriti Sharma, Amit Kumar Sharma and Meena Sharma

Physical Chemistry Laboratory, Department of Chemistry, University of Jammu, Jammu-180 006, Jammu & Kashmir, India

E-mail: vijayta1gupta@gmail.com

Manuscript received online 20 December 2019, revised and accepted 12 May 2020

Thermodynamic, acoustical and transport properties of fluids are extremely significant for explaining the character of molecular relations, which affect the physico-chemical behaviour of liquids. The present investigation was aimed to determine the acoustical parameters of nickel (Ni) nanofluids. The Ni nanoparticles were synthesized and characterized by XRD, TEM, SEM and EDX. The various concentrations (0.05%, 0.1%, 0.15%, 0.2%, and 0.25%) of Ni nanoparticles were dispersed in gum arabic-ethylene glycol (EG) mixture followed by ultrasonication to obtain Ni-EG nanofluids. Speed of sound, density, viscosity and different acoustical parameters of nanofluids were measured at different concentrations and temperatures. The Ni-EG nanofluids showed increase in speed of sound and acoustic impedance upto 0.2 wt% and later on get decreased. Viscosity and density of nanofluid increased with concentration and decreased with temperature used in this study. The intermolecular free length, adiabatic compressibility, Gibb's free energy, relaxation time and absorption coefficient first decreased upto 0.2 wt% and then increased. The nanofluids showed decrease in Gibb's free energy, relaxation time, absorption coefficient, acoustic impedance and speed of sound, and increased for adiabatic compressibility as well as intermolecular free length with increase in temperature. In conclusion, Ni-EG nanofluids upto 0.2 wt% concentration have significant nanoparticle-fluid interaction and could be highly suitable for different industrial applications.

Keywords: Ni-EG nanofluids, speed of sound, acoustical studies.

Introduction

In recent years, technology for fluids i.e. nanofluids has been considered as one of the emerging techniques, which cope up the drawbacks to some extent for the conventionally used base fluids and bulk fluids[1]. Nanofluids are a new class of colloidal mixtures consisting of solid nanoparticles with size 1–100 nm and base fluids such as water, glycols, lubricants and light oils[2]. In general, nanoparticles of metals, oxides, carbides, or carbon nanotubes are used to prepare nanofluids, and studies have point out that nanoparticles can favourably modify thermophysical or transport properties of the base fluid[3]. Nanofluids, with improved thermophysical properties, have wide applications in several areas like electronics, industrial cooling, air conditioning, engines, nuclear system cooling, micro manufacturing, antibacterial activity, nano-drug delivery etc.[4,5]. These makes nanofluids as promising material for heat transfer applications in many industrial processes. Ultrasonic study of liquids is a valuable technique for comprehending its physico-chemical properties. Ultrasonic properties of a liquid are connected to the uniting forces among atoms as well as molecules. Derived parameters from ultrasonic properties measurement represent qualitative knowledge concerned to character and potency of connections in liquid[6]. Ultrasonic study of liquids and liquid mixtures has become powerful technique during last two decades for the examination of character of molecular connections and investigation of physico-chemical features.

Understanding of mechanisms of heat transfer enhancement is based on molecular interactions (fluid-particle and particle-particle interactions) within the fluid, and ultrasound wave propagation is considered one of the commonly used methods to study these changes[7]. Studies of the variation in thermophysical properties are also helpful to interpret various important features about the molecular interactions in binary mixtures. Determinations of acoustical parameters have also used to study the physico-chemical behavior and inter-molecular interactions in different binary liquid mixtures in recent years[8]. However, scanty of research has been con-

ducted on acoustical studies of nanofluids. Acoustical properties of nanofluids are important to determine their effects on flow and heat transport features of nanofluids[9]. Thus, there is an utmost requirement for the systematic investigations for concentration and temperature dependent effects on molecular relations in nanofluids.

Recently, Ni nanoparticles have become appealing nanomaterials in diverse research fields because of their multiple appliances in the area of catalysis and magnetism. The modern applications of Ni nanomaterials also include base-metal-electrode multilayer ceramic capacitor and high density magnetic recording media because of its various features like high melting point, excellent electrical conductivity and reasonably priced[10,11]. Different heat exchange fluids are used in heat management systems and ethylene glycol (EG) is one of them, which is frequently used as a coolant due to its lower freezing point. EG is considered better than water as well as other fluids due to its better heat transfer properties. EG has an also vast application like carriers, solvents, binders, lubricants, bases, coupling agents etc. in various industrial areas[12]. Although, a significant literature exists for nanofluids related to their thermal conductivity and viscosity, but, only scanty of information is available for acoustical properties of Ni nanofluids at different concentrations and temperatures. In view of the available literature and importance of the nanofluids[13–16], the present investigation was aimed to study the effect of various concentrations and temperatures on acoustical parameters Ni-EG nanofluids for understanding of particle-fluid, particle-particle interactions.

Experimental

Synthesis and characterization of Ni nanoparticles:

Nickel chloride hexahydrate ($NiCl_2.6H_2O$) was used for synthesis of Ni nanoparticles. Hydrazine hydrate was used in this process for reduction. The solution of 2 g of nickel chloride and ethylene glycol was heated to 60°C followed by the addition 20 ml hydrazine to form Ni-hydrazine complex. To the above reaction mixture, 6 ml of 1 *M* NaOH solution dissolved in ethylene glycol was added under stirring and this solution was magnetically stirred for 1 h at 60°C to get black colored nickel nanoparticles. These nanoparticles were collected, washed several times by triply distilled water and ethanol and dried in an oven at 60°C.

The X-ray diffraction (XRD), transmission electron microscopy (TEM), scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDX) was done for the characterization of synthesized Ni nanoparticles. The Cu Kα radiation (λ = 1.541 Å) for 2θ value (10°–100°) in X-ray diffractometer (Model: Bruker, AXS D8 Advance) was used to identify the crystallite size and structure of nanoparticles. The ultraviolet-visible-NIR spectrophotometer (Lambda 750 Perkin Elmer) was used in UV-Vis spectroscopic study. The particle size and morphology of Ni nanoparticles was determined using TEM [Model Hitachi (H-7500) microscope] operating at 80 kV. The SEM (Make: JEOL; Model: JSM-6390LV) and EDS (Make: JEOL; Model: JED-2300) was used for surface morphology and elemental analysis.

Synthesis of Ni nanoparticles based nanofluids:

The synthesized nanoparticles of Ni at various concentrations (0.05%, 0.1%, 0.15%, 0.2%, and 0.25% w/w) were dispersed in 1 wt% gum arabic-EG solution by employing ultrasonication for 4 h to obtain stable homogenous Ni-EG nanofluids. Gum arabic was added to stabilize the Ni-EG nanosuspension.

Acoustical studies of Ni-EG nanofluids:

A multi frequency ultrasonic interferometer (±0.05% accuracy and 6 MHz frequency) was employed for measuring speed of sound through Ni-EG nanofluids. Specific gravity bottle (5 cc; with ±2.0 parts in 10^4 accuracy) was used for determination of density of Ni-EG nanofluids. Ostwald's viscometer (±0.001 Nsm-2 accuracy) was employed to measure viscosity of Ni-EG nanofluids. These all parameter were achieved for various concentrations at three different temperatures of 298.15 K, 303.15 K and 308.15 K. These experimentally measured values of density (ρ), speed of sound (U) and viscosity (η) were used in evaluation of different acoustical parameters such as adiabatic compressibility (β_{ad}), intermolecular free length (L_f), relaxation time (τ), acoustic impedance (Z), absorption coefficient (α/f^2) and Gibb's free energy (ΔG) were measured by using the following standard relations:

Adiabatic compressibility (N^{-1} m^2) was determined from the following Newton-Laplace's equation[17,18]:

$$\beta_{ad} = 1/U^2\rho$$

U and ρ represents velocity and density of nanofluid.

Following formula was used for intermolecular free length (*m*) calculation[19]:

$$L_f = K_T \beta_{ad}^{1/2}$$

K_T represents Jacobson's constant. It is temperature reliant and its value is 2.0568×10^{-6}, 2.0756×10^{-6} and 2.0943×10^{-6} at 25, 30 and 35ºC, respectively.

Following relation was used for the determination of relaxation time (s)[20]:

$$\tau = \left(4/3\right)\beta\eta$$

Absorption coefficient ($s^2\ m^{-1}$) was calculated by using the following relation:

$$\left(\alpha/f^2\right) = 4\pi^2\tau/2U$$

Acoustic impedance (Nsm^{-3}) was determined from the following equation[21]:

$$Z = U \times \rho$$

Gibbs free energy ($J\ mol^{-1}$) was determined from the following given relation:

$$\Delta G = kT \ln\left(kT\tau/h\right)$$

Results and discussion

Structural and morphological assessment of Ni nanoparticles:

The patterns recorded in the XRD for Ni nanoparticles indicated five characteristic peaks at 2θ = 44.680, 52.008, 76.548, 93.056 and 98.625° (Fig. 1). These sharp and strong peaks indicated the crystalline nature of the sample and are indexed to face centered cubic (fcc) structure of nickel (PDF

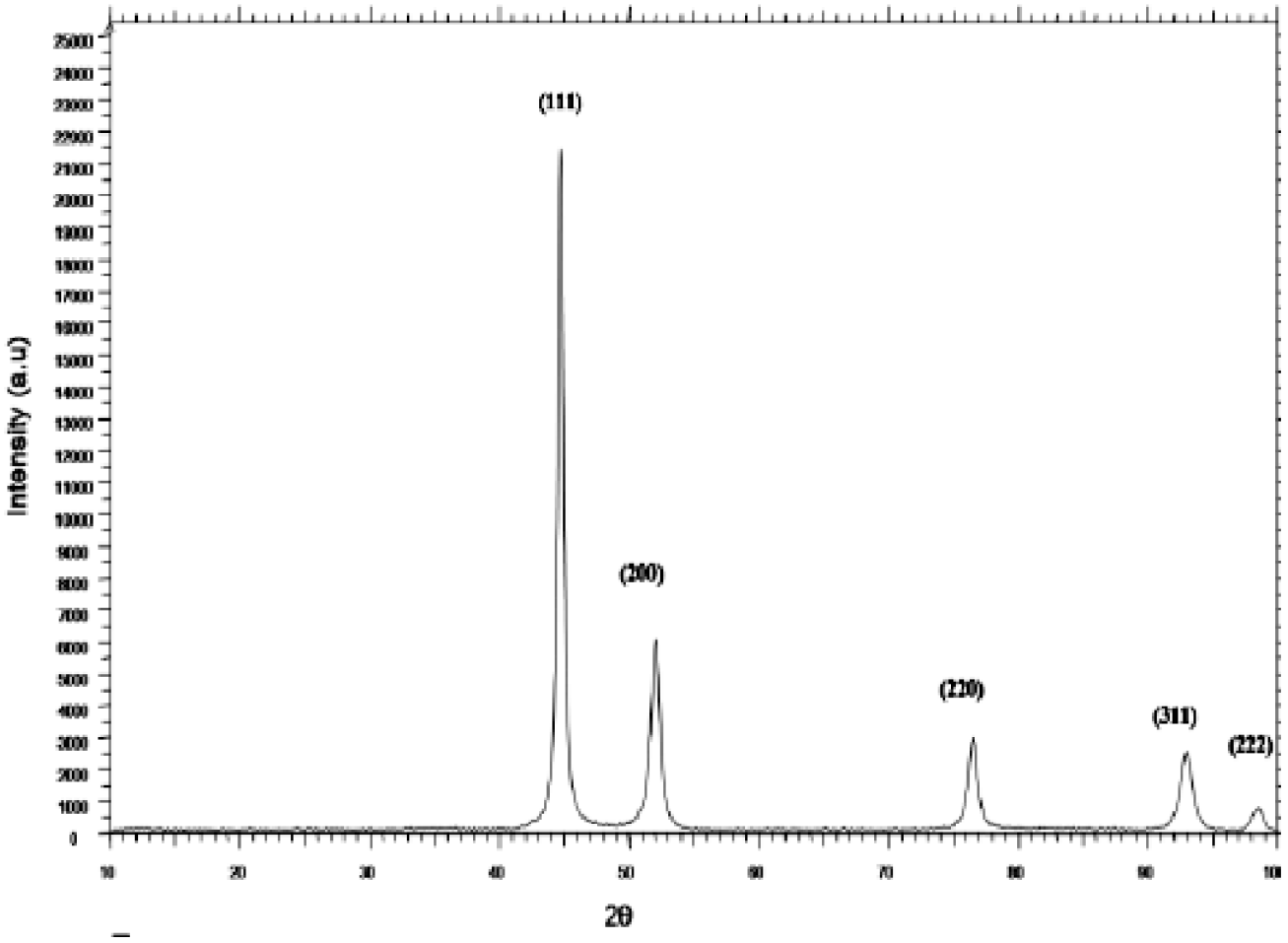

Fig. 1. The representative image of XRD for Ni nanoparticles.

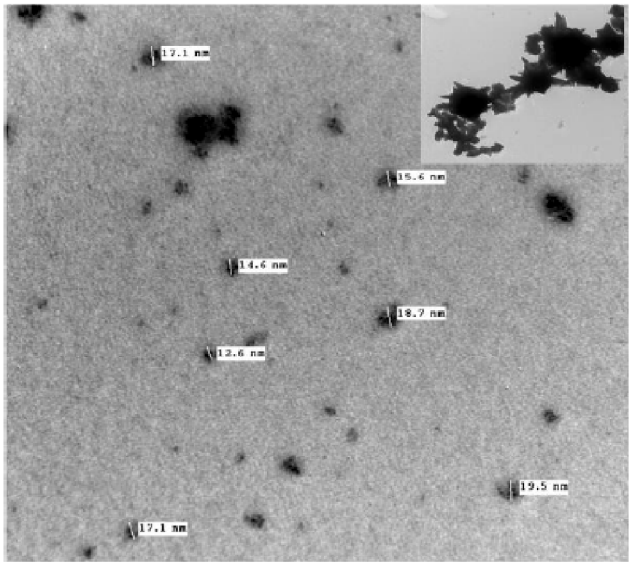

Fig. 2. The representative TEM image of of Ni nanoparticles. Inset shows aggregated nanoparticles.

#04-0850), which is in agreement with previous report[22]. The average crystallite size was 16.65 nm by the use of Debye-Scherrer's equation. TEM image indicated that the prepared Ni nanoparticles have spherical morphology (Fig. 2). The inset indicated the self-assembled flower like structures might be due to magneto dipole attractions. This result is in agreement with previous results[23]. The particle size measured from the TEM images is in close agreement to the calculated result from XRD pattern. SEM micrograph in Fig. 3a revealed the formation of well defined spherical particles. EDS analysis indicated that the only element present in these nanoparticles was nickel (Fig. 3b). Zero valence state of nickel was proved due to the absence of oxygen.

Acoustical studies:

Studies of thermodynamic, acoustical and transport properties of fluids are highly significant to explain the nature of their molecular connections, which further affect the physico-chemical characteristics of liquids. Speed of sound is considered as one of the valuable property in understanding the physico-chemical characteristics of liquids. In present investigation, it was observed that speed of sound in Ni-EG nanofluids increased upto 0.2 wt% and afterwards decreased (Fig. 4a, Table 1). Further with increase in temperature, speed of sound decreased. The increase in speed of sound at lower concentrations of Ni nanoparticles might be due to intermolecular interactions between Ni nanoparticles and ethylene glycol molecules (particle-fluid) as a result overall compressibility of nanofluid decreases. Therefore, it results in increase

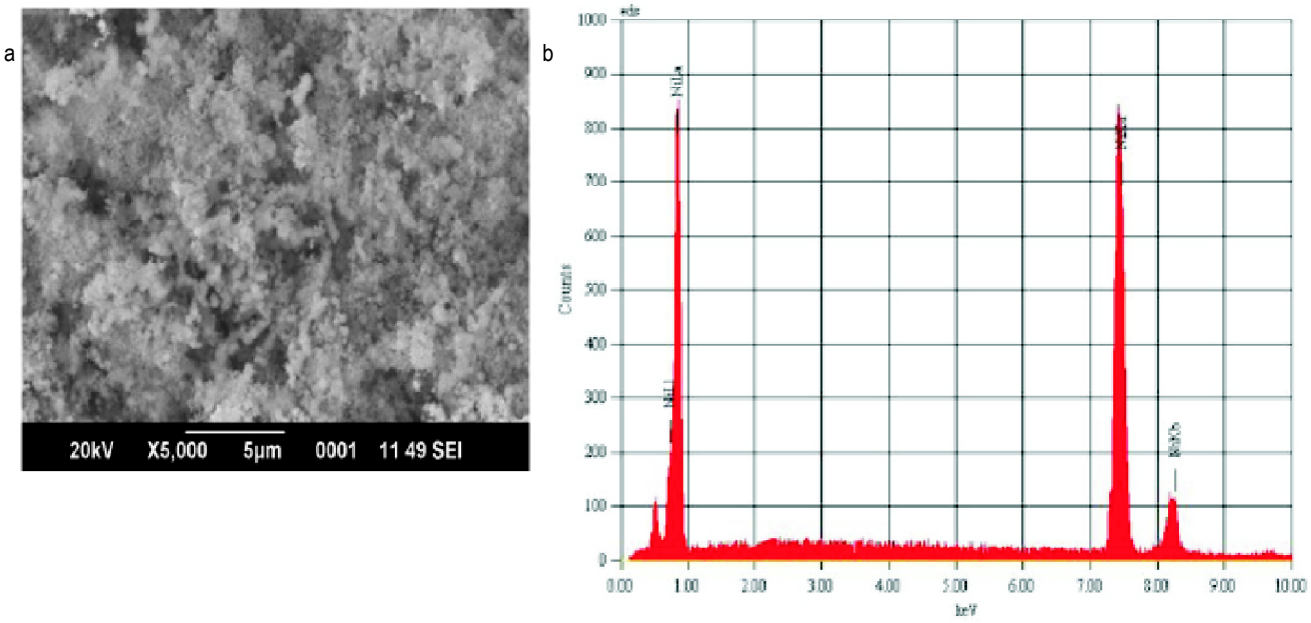

Fig. 3. The representative images of (a) SEM and (b) EDS of Ni nanoparticles.

of speed of sound. However, the interparticle interactions (Ni-Ni) predominate the particle-fluid interations at >0.2 wt%. This resulted in increase in overall compressibility of nanofluid, which lead to decrease in speed of sound[24]. Fast movement of suspended molecules in the fluid take place with rise in temperature, which results in decrease of speed of sound. This confirms that detioration of the particle-fluid interaction establish at elevated temperatures.

Viscosity describes internal resistance to the flow of fluid. The results showed that viscosity and density of nanofluid increased as the concentration of nanoparticle was increased (Table 2). The increased viscosity was might be because of greater viscous dissipation with the increase in nanoparticles concentration. Moreover, the viscosity as well as density of nanofluids decreased with rise in temperature. This revealed deterioration of intermolecular forces because of increased thermal energy of system at higher temperatures[25].

Adiabatic compressibility (Fig. 4b, Table 1) and intermolecular free length (Fig. 4c, Table 1) initially decreased upto 0.2 wt% concentration of Ni nanoparticles and afterwards it started increasing with increase in concentration. Due to significant particle-fluid interactions at lower concentrations of Ni nanoparticles, adiabatic compressibility and intermolecular free length decreased but above 0.2 wt% concentration

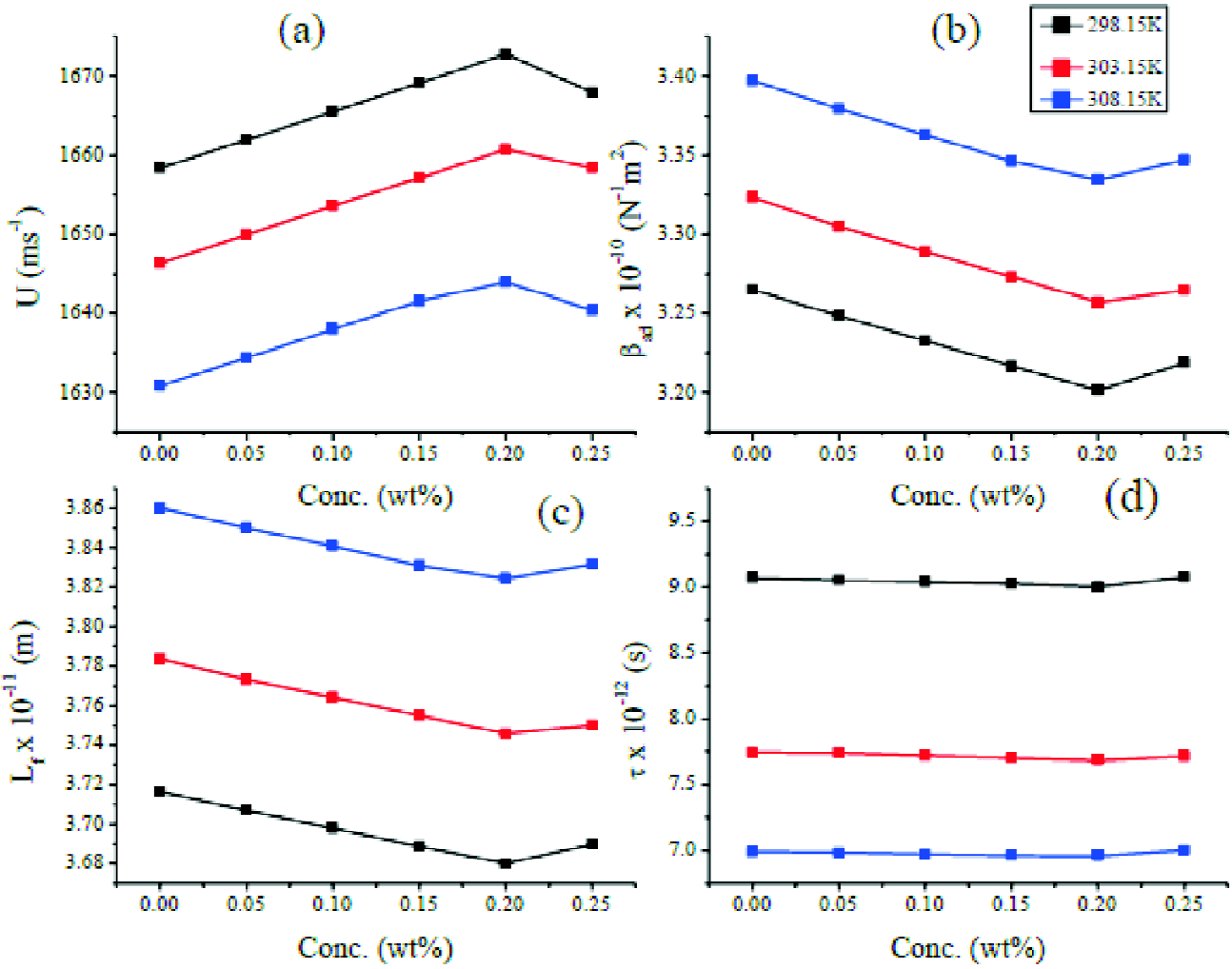

Fig. 4. Graphs of (a) speed of sound, (b) adiabatic compressibility, (c) intermolecular free length and (d) relaxation time for Ni nanofluids at different concentrations and temperatures.

Table 1. Speed of sound, adiabatic compressibility, intermolecular free length and relaxation time of Ni nanofluids at different temperatures

Temp. (K)	Concentration of nanoparticles (wt%)	U (ms^{-1})	$\beta_{ad}\times10^{-10}$ ($N^{-1}\ m^2$)	$L_f\times10^{-11}$ (m)	$t\times10^{-12}$ (s)
298.15	0	1658.4	3.2648	3.7164	9.0721
	0.05	1662	3.2483	3.7070	9.0554
	0.1	1665.6	3.2326	3.6980	9.0411
	0.15	1669.2	3.2166	3.6889	9.0285
	0.2	1672.8	3.2013	3.6801	9.0043
	0.25	1668	3.2186	3.6900	9.0781
303.15	0	1646.4	3.3233	3.7838	7.7459
	0.05	1650	3.3049	3.7733	7.7401
	0.1	1653.6	3.2891	3.7643	7.7202
	0.15	1657.2	3.2733	3.7552	7.7038
	0.2	1660.8	3.2571	3.7459	7.6856
	0.25	1658.4	3.2645	3.7502	7.7188
308.15	0	1630.8	3.3973	3.8601	6.9918
	0.05	1634.4	3.3796	3.8501	6.9842
	0.1	1638	3.3632	3.8408	6.9731
	0.15	1641.6	3.3464	3.8311	6.9611
	0.2	1644	3.3348	3.8245	6.9574
	0.25	1640.4	3.3473	3.8317	7.0031

Table 2. Density and viscosity of Ni nanofluids at different temperatures

Temp. (K)	Concentration of nanoparticles (wt%)	$\rho\times10^3$ ($kg\ m^{-3}$)	$\eta\times10^{-3}$ (Nsm^{-2})
298.15	0	1.1137	20.8408
	0.05	1.1145	20.908
	0.1	1.1151	20.9768
	0.15	1.1158	21.0513
	0.2	1.1163	21.0951
	0.25	1.1167	21.1535
303.15	0	1.1101	17.4809
	0.05	1.1114	17.5649
	0.1	1.1119	17.6042
	0.15	1.1124	17.6514
	0.2	1.1131	17.6975
	0.25	1.1138	17.7337
308.15	0	1.1068	15.4355
	0.05	1.1077	15.4994
	0.1	1.1082	15.5501
	0.15	1.1089	15.6015
	0.2	1.1095	15.6473
	0.25	1.1102	15.6911

Table 3. Absorption coefficient, acoustic impedance and Gibb's free energy of Ni nanofluids at different temperatures

Temp. (K)	Concentration of nanoparticles (wt%)	$\alpha/f^2\times10^{-14}$ ($s^2\ m^{-1}$)	$Z\times10^6$ (Nsm^{-3})	$\Delta G\times10^{-21}$ ($J\ mol^{-1}$)
298.15	0	10.7871	1.8470	16.5896
	0.05	10.7440	1.8523	16.5820
	0.1	10.7039	1.8573	16.5755
	0.15	10.6656	1.8625	16.5698
	0.2	10.6144	1.8674	16.5587
	0.25	10.7321	1.8627	16.5923
303.15	0	9.27734	1.8277	16.2761
	0.05	9.2502	1.8338	16.2730
	0.1	9.2063	1.8386	16.2622
	0.15	9.1669	1.8435	16.2533
	0.2	9.1254	1.8486	16.2434
	0.25	9.1781	1.8471	16.2614
308.15	0	8.4543	1.8050	16.1785
	0.05	8.4265	1.8104	16.1739
	0.1	8.3946	1.8152	16.1671
	0.15	8.3618	1.8204	16.1598
	0.2	8.3452	1.8240	16.1575
	0.25	8.4184	1.8212	16.1854

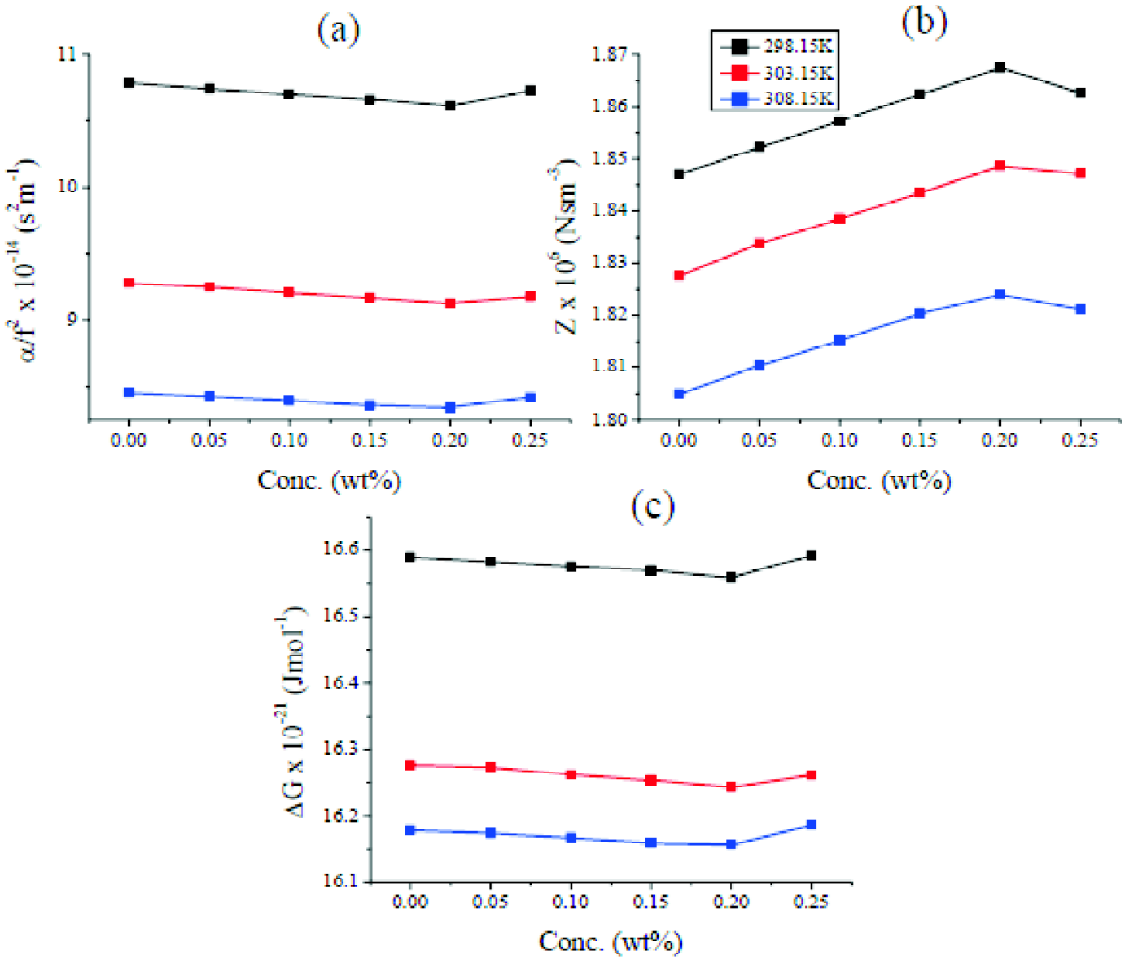

Fig. 5. Graphs of (a) absorption coefficient, (b) acoustic impedance, (c) Gibb's free energy for Ni nanofluids at various concentrations and temperatures.

of Ni nanoparticles, adiabatic compressibility and intermolecular free length increased due to predominance of particle-particle interactions over particle-fluid interactions. The fall in the value of intermolecular free length reveals significant interaction between particles and base fluid molecules[26]. Further, adiabatic compressibility and intermolecular free length increased along with rise in temperature. It might be because of less ordered structure formation with increase in temperature, which resulted to increased spacing among the molecules as a result of increased thermal energy of the system[27].

The initial rise in acoustic impedance was observed as the concentration of nanoparticles increased upto 0.2 wt% and then it decreased with further increase in concentration (Fig. 5b, Table 3). Acoustic impedance of the nanofluids has been decreased with increasing temperature.

The weakening of interactions at higher temperature is responsible for the decrease of acoustic impedance. The results showed that absorption coefficient (Fig. 5a, Table 3), relaxation time (Fig. 4d, Table 1) and Gibb's free energy (Fig. 5c, Table 3) decreased very slightly upto 0.2 wt% concentration of nanoparticles afterwards these parameters increased faintly with further increase in nanoparticles concentration. Further, with increase in temperature, absorption coefficient, relaxation time and Gibb's free energy decreased. The trends of acoustical parameters support the findings for speed of sound, density and viscosity. As, the nanofluids exhibit superior heat transfer properties than conventional heat ex-

change fluids[1]. This is might be due to the enhanced thermal conductivity because of suspended nanoparticles. Moreover, the thermal conductivity of nanofluids is strongly dependent on concentration of nanoparticles. In our present study, the thermal conductivity of Ni nanofluid was higher at 0.2 wt% in comparison to other concentrations as well as base fluid (ethylene glycol). Thus, Ni nanofluid at 0.2 wt% might be applied in different industries as heat exchange fluids after conducting some more investigations.

Conclusion

The maximum value of speed of sound for Ni-EG nanofluid was observed at 0.2 wt% due to particle-fluid interaction and particle-particle interaction increased after 0.2 wt% due to agglomerations. Further, with increase in temperature particle fluid interaction decreased. In conclusion, Ni-EG nanofluid is highly suitable for nanofluid applications upto 0.2 wt%.

Acknowledgements

We acknowledge the support of Department of Chemistry, University of Jammu, J&K, India for carrying out the present study. We are also thankful to SAIF Chandigarh, CIL Chandigarh, SAIF STIC (Kochi) and GJU S&T (Hisar) for allowing us to utilize the facilities for characterization studies. The first author is highly thankful to CSIR-UGC, New Delhi, India for granting Junior Research Fellowship (JRF) during this study.

References

1. Y. Xuan and Q. Li, *Int. J. Heat Fluid Transfer,* 2000, **21**, 58.
2. R. Wan Meher, R. Yadav, K. L. Yadav and S. B. Yadaw, *Exp. Therm. Fluid Sci.,* 2012, **41**, 158.
3. M. Chopkar, S. Kumar, D. R. Bhandari, P. K. Das and I. Manna, *Mater. Sci. Eng. B,* 2007, **139**, 141.
4. W. Yu and H. Xie, *J. Nanomater.,* 2012, 1. DOI:10.1155/2012/435873
5. J. Hemalatha, T. Prabhakaran and R. P. Nalini, *Microfluid Nanofluid,* 2011, **10**, 263.
6. K. Tamura, T. Sonoda and S. Murakami, *J. Sol. Chem.,* 1999, **28**, 777.
7. R. P. Singh, V. K. Shukla, R. S. Yadav, P. K. Sharma, P. K. Singh and A. C. Pandey, *Adv. Mater. Lett.*, 2011, **2**, 313.
8. S. Kamila and J. K. Dash, *J. Mol. Liq.,* 2012, **172**, 71.
9. H. Chang and H. T. Su, *Rev. Adv. Mater. Sci.,* 2008, **18**, 667.
10. D. P. Wang, D. B. Sun, H. Y. Yu and H. M. Meng, *J. Cryst. Growth*, 2008, **310**, 1195.
11. H. Hu and K. Sugawara, *Mater. Lett.,* 2009, **63**, 940.
12. M. Taghi, Z. Moattar and R. Majdan-Cegincara, *J. Chem. Thermodyn.,* 2012, **54**, 55.
13. I. M. Mahbubul, "Preparation, Characterization, Properties and Application of Nanofluid", ed. W. Andrew, Elsevier Inc., 2019, 374.
14. K. Rahbar, A. Riasi, H. K. B. Sangjoeei and N. Razmjoo, *Energy Convers. Manag.*, 2019, **179**, 373.
15. E. Bellos and C. Tzivanidis, *Energy Convers. Manag.*, 2019, **180**, 171.
16. A. Kaggwa and J. K. Carson, *Int. Nano Lett.,* 2019, **9**, 277.
17. J. S. Rowlinson and F. L. Swinton, "Liquid and liquid mixtures", Butterworths, London, 1982, 16.
18. M. J. W. Povey, "Ultrasonic techniques for fluids characterization", Academic Press, USA, 1997, 25.
19. B. Jacobson, *Acta Chemica. Scand.,* 1951, **5**, 1214.
20. J. H. Hildebrand, *J. Chem. Phys.,* 1959, **31C**, 1423.
21. L. Prigogine and V. Malhot, "The molecular theory of the solution", North Holl Pub, Amster Dam, 1957.
22. J. Tientong, S. Garcia, C. R. Thurber and T. D. Golden, *J. Nanotechnol.,* 2014, 2014, 1. Article ID 193162, http://dx.doi.org/10.1155/2014/193162.
23. K. Singh, K. H. Kate, V. V. Satayanarayana Chilukuri and P. K. Khanna, *J. Nanosci. Nanotechnol.*, 2011, **11**, 5131.
24. R. Kiruba, M. Gopalakrishnan, T. Mahalingam and A. K. S. Jeevaraj, *J. Nanofluids,* 2012, **1**, 97.
25. M. N. Rashin and J. Hemalatha, "Advanced Nanomater. Nanotechno.", New York, Berlin Heidelberg, 2013.
26. V. Gupta, A. K. Sharma and M. Sharma, *Chem. Sci. Trans.*, 2014, **3**, 736.
27. S. Kamila and V. R. V. Gopal, *Heliyon*, 2019, **5**, e02445.