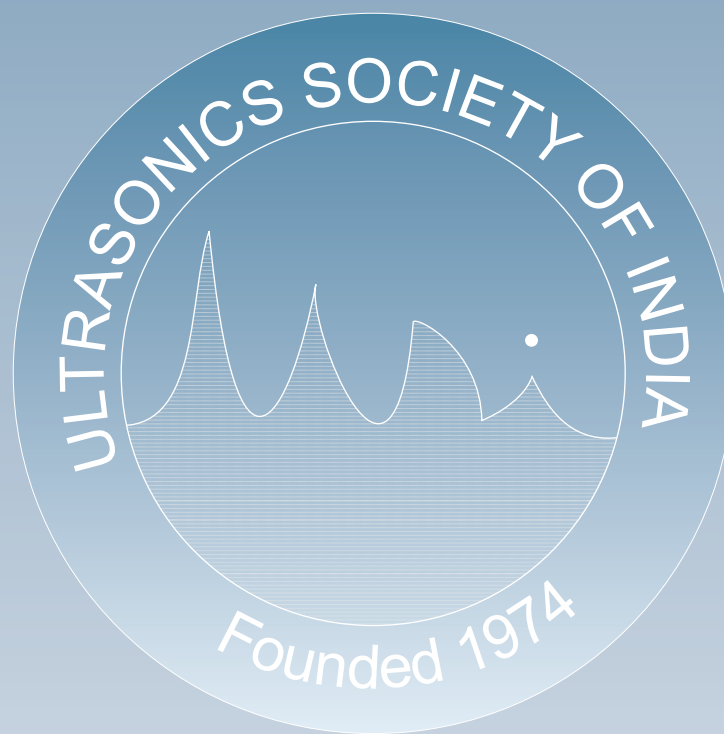
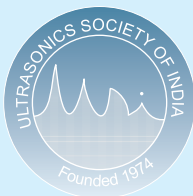


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Ultrasonic speed measurements in binary mixtures containing esters at temperatures from 303.15 to 318.15 K

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Density and ultrasonic speed have been measured in pure and binary mixtures containing o-anisidine and butyl acetate or propyl acetate over the whole composition range at 303.15, 308.15, 313.15 and 318.15 K. From these values, isentropic compressibility and intermolecular free lengths as well as excess quantities for sound speed, isentropic compressibility and intermolecular free length have been calculated. The results have been fitted to Redlich-Kister polynomial equation. The results have been explained on the basis of intermolecular interactions.

Keywords: o-anisidine, butyl acetate, propyl acetate, ultrasonic speed, thermodynamic parameter.

Introduction

This paper is a continuation of our earlier work related to the study of thermodynamic properties of binary and ternary mixtures¹⁻⁵. In recent years, measurements of the thermodynamic and transport properties have been adequately employed in understanding the nature of molecular systems and physic-chemical behavior in binary and ternary mixtures. Esters are one of the best candidates that exist as dipolar associates in their pure liquid state, available with not only aliphatic, aromatic and even acrylic nature but also with a variety of general structures.

o-Anisidine is a clear, yellowish to reddish liquid with an amine odor. It is used in the manufacturing of dyes. Butylacetate is a clear, moderately volatile liquid with a characteristic ester odor. It is miscible with all conventional solvents. It is an important solvent in the paint industry on account of its versatility. Because of its low water absorption, its high resistance to hydrolysis and its high solvency, n-butyl acetate can also be employed as an extractant in the manufacture of pharmaceutical preparations and as an ingredient of cleaners, essences and fragrances. Propylacetate is a colourless, volatile solvent with good solvent power for numerous natural and synthetic resins. It is used as a

solvent in the coatings, printing inks and chemical downstream industries.

Here, we have measured densities, ρ , and ultrasonic speeds, U , for the binary systems formed by {o-anisidine + butylacetate, o-anisidine + propylacetate} at 303.15 - 318.15 K for the liquid region at the whole composition range. Isentropic compressibility and intermolecular free length have been calculated by using the measured data. The excess parameters such as excess sound speed, excess isentropic compressibility and excess intermolecular free length values have been fitted to the Redlich-Kister equation to determine the coefficients.

Experimental

The mass fractions of o-anisidine, butylacetate and propylacetate obtained from SDFCL are 0.98, 0.97 and 0.978 respectively. All the liquids obtained from the suppliers were further purified by standard procedure⁶. A comparison of the experimental values of density and ultrasonic velocity obtained in the present study with the values reported in literature shows good agreement. Job's method of continuous variation was used to prepare the mixtures in the required proportions. The mixtures were preserved in well-stoppered conical flasks. After thorough mixing of the liquids, the flasks were left

undisturbed to allow them to attain thermal equilibrium.

An electronic balance with a precision of ± 0.1 mg have been used for the measurements of mass. The densities of pure liquids and liquid mixtures have been measured using specific gravity bottle.

The ultrasonic speeds have been measured by using a single crystal ultrasonic pulse echo interferometer, at a fixed frequency of 3 MHz. The temperature has been controlled through water circulation around the liquid cell using thermostatically controlled constant temperature water bath.

From the experimental values of density, speed of sound, the parameters like isentropic compressibility, intermolecular free length and their excess properties have been calculated using the standard equations⁷.

Result and Discussion

The experimental values of density, ρ , and speeds of sound, U , for all the mixtures over the entire range of composition and at temperatures 303.15, 308.15, 313.15

and 318.15 K are presented in Table 1. The variations of excess ultrasonic speed with mole fraction of o-anisidine at different temperatures are shown in Figs.1 (a, b). From the figures can be observed that as the mole fraction of o-anisidine increases, U^E values first

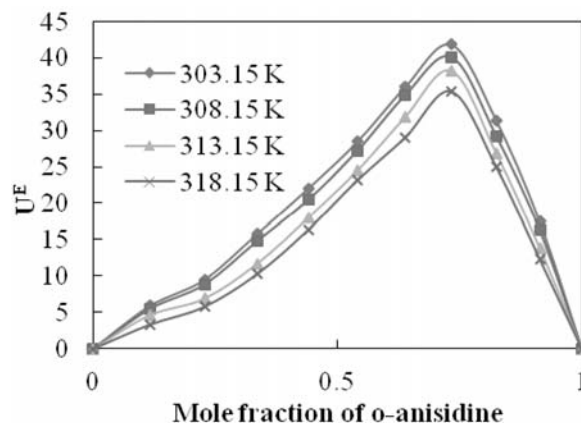


Fig. 1(a) Variation of U^E with mole fraction of o-anisidine for binary mixture, o-anisidine + butylacetate at different temperatures.

Table 1 – Density, ρ , and speed of sound, U , of binary mixtures at different temperatures

x ₁	o-Anisidine + Butylacetate							
	303.15 K		308.15 K		313.15 K		318.15 K	
	U /m.s ⁻¹	ρ/Kg.m ⁻³	U /m.s ⁻¹	ρ/Kg.m ⁻³	U /m.s ⁻¹	ρ/Kg.m ⁻³	U /m.s ⁻¹	ρ/Kg.m ⁻³
0.0000	1176.6	871.0	1160.9	866.8	1139.4	860.1	1104.2	855.9
0.1157	1222.8	887.5	1205.6	884.1	1185.2	877.9	1151.1	873.7
0.2274	1265.3	908.5	1246.8	904.5	1227.1	898.5	1195.6	894.1
0.3353	1309.3	932.1	1289.4	927.9	1270.3	922.0	1240.8	917.1
0.4397	1351.9	956.0	1330.4	951.2	1313.6	945.1	1286.2	939.6
0.5407	1393.6	979.3	1371.4	973.5	1356.0	967.2	1331.1	961.1
0.6384	1435.2	1002.7	1412.2	996.4	1398.1	989.9	1373.8	983.1
0.7331	1474.1	1025.8	1449.6	1018.9	1438.1	1012.5	1415.8	1005.2
0.8248	1495.6	1048.1	1469.8	1040.9	1459.4	1034.6	1440.0	1027.0
0.9138	1512.6	1068.5	1487.0	1061.0	1478.0	1054.9	1460.9	1046.8
1.0000	1525.3	1087.1	1500.0	1080.0	1494.8	1074.6	1481.1	1066.5
o-Anisidine + Propylacetate								
0.0000	1140.0	856.8	1106.7	845.2	1076.6	835.3	1026.0	823.3
0.1045	1190.1	871.7	1154.9	862.1	1126.4	853.5	1078.0	842.9
0.2079	1236.1	894.3	1200.9	885.2	1175.5	877.0	1131.5	866.5
0.3103	1277.9	919.8	1243.8	910.5	1220.8	902.6	1180.8	892.0
0.4118	1317.0	945.9	1284.0	936.5	1263.8	928.5	1227.7	918.2
0.5122	1356.2	971.8	1325.6	962.1	1307.6	953.6	1275.4	942.9
0.6116	1396.0	998.8	1366.6	988.9	1351.0	980.2	1322.3	969.2
0.7101	1435.8	1024.2	1406.9	1014.5	1393.6	1006.1	1368.3	995.6
0.8077	1468.5	1048.4	1441.2	1038.9	1431.1	1031.5	1409.6	1021.7
0.9043	1497.0	1068.4	1470.7	1059.8	1463.3	1053.1	1446.2	1044.1
1.0000	1525.3	1087.1	1500.0	1080.0	1494.8	1074.6	1481.1	1066.5

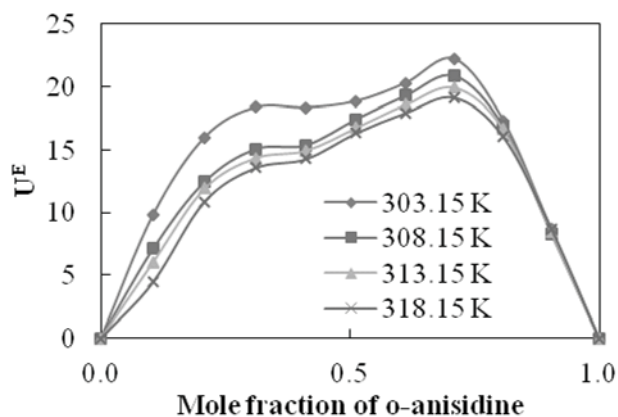


Fig. 1(b) Variation of U^E with mole fraction of o-anisidine for binary mixture, o-anisidine + propylacetate at different temperatures.

increases reach a maximum value (at 0.7 mole fraction) and then decreases. The same trend is observed in both the systems. From the figures it is also observed that as the temperature increases the values of U^E decreases. The speed of sound is related to the isentropic compressibility and the density. A change in the structure resulting from mixing affects the compressibility and/or the density which in turn influences the speed of sound. In this way, the compaction or the expansion effects will have more influence on the values of the speed of sound. Thus, if strong interactions arise among the components of a mixture leading to the formation of molecular aggregates and more compact structures, the sound will travel faster through the mixture by means of longitudinal waves and the speed of sound deviations with respect to linear behavior will be positive.

The variation of excess isentropic compressibility, k_s^E , with mole fraction of o-anisidine at different temperatures for the two systems are shown in Figs.2 (a, b).

For the two systems k_s^E values are observed to be negative. These values are observed to be more negative with increase in temperature. The observed negative values of k_s^E may be due to close packing arrangements⁸. The k_s^E is more negative in case of o-anisidine + propylacetate mixtures when compared to o-anisidine + butylacetate mixture. U is generally higher when the structure has high rigidity.

Figures 3 (a, b) shows that L_f^E are negative for the two systems over the whole composition range. The very large negative values for o-anisidine + propylacetate as compared to o-anisidine + butylacetate has been observed. The intermolecular free length depends on isentropic compressibility and shows similar behavior

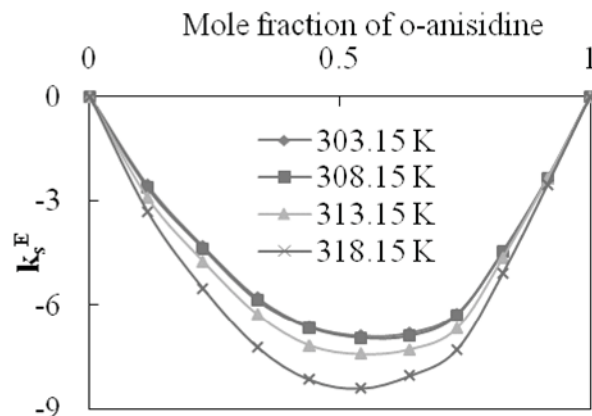


Fig. 2(a) Variation of k_s^E with mole fraction of o-anisidine for binary mixture, o-anisidine + butylacetate at different temperatures.

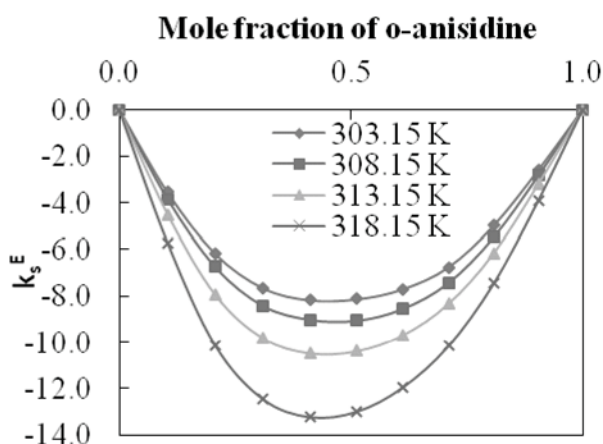


Fig. 2(b) Variation of k_s^E with mole fraction of o-anisidine for binary mixture, o-anisidine + propylacetate at different temperatures.

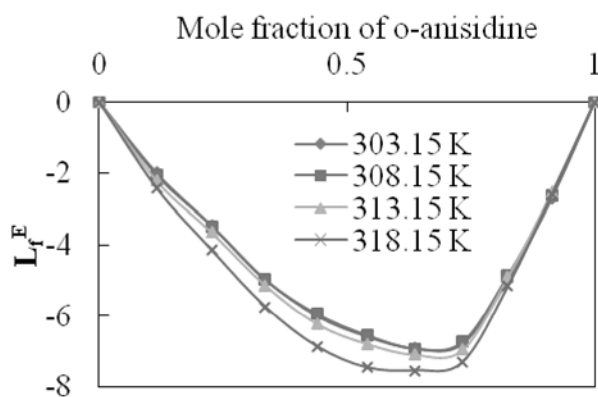


Fig. 3(a) Variation of L_f^E with mole fraction of o-anisidine for binary mixture, o-anisidine + butylacetate at different temperatures.

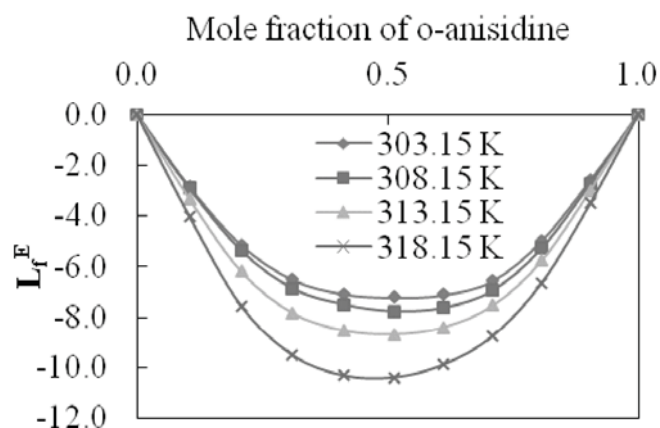


Fig. 3(b) Variation of L_r^E with mole fraction of o-anisidine for binary mixture, o-anisidine + propylacetate at different temperatures.

to that of isentropic compressibility and inverse to that of speed of sound.

Certain properties of the liquid state can be studied to advantage as a function of the free length between the molecules. These properties include compressibility, surface tension, viscosity and diffusion constants *i.e.*, those that are mainly dependent on the forces between the molecules.

The reason for choosing the intermolecular free length for these studies are as follows. The intermolecular forces, which in one way or the other determine the said properties of liquids, consists of attractive forces and repulsive forces.

In the present investigation, the excess isentropic compressibility and excess intermolecular free length exhibit negative values over the entire range composition in all the two liquid systems clearly indicating the presence of strong interactions between unlike molecules.

Conclusion

From the data's of speed of sound and density related acoustical parameters and some of the excess parameters such as excess speed of sound, excess isentropic compressibility and excess free length for the binary liquid mixtures of butylacetate, propylacetate with o-anisidine at 303.15, 308.15, 313.15 and 318.15 K, it is

very obvious that there exist a strong molecular interaction existing between the unlike molecules.

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Study of physical and acoustical properties of silver nanofluid

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Nanoparticles (1-100 nm) in base fluids have numerous exceptional physiochemical properties absolutely different from those of bulk metal due to their extremely small size and large superficial area to volume. A one-step chemical method has been developed for the preparation of stable, non agglomerated silver nano fluid without using any surfactant. Silver nanofluid is prepared by using silver nitrate as a source for silver nanoparticles, distilled water as a base fluid, and tannic acid (tannin) as reducing agent of Ag^+ ions. The formation of colloidal silver was followed by X-ray diffraction topography, UV-visible absorption spectroscopy monitoring the growth of the nanoparticles. Investigation of sample was carried out to study physical properties like absorbance; electrical conductance. The ultrasonic study has been done in different concentration of silver nano fluid at different temperature. The different acoustical parameters are calculated using the experimental data of ultrasonic velocity, density and viscosity. The characteristic behaviors of the acoustic parameters are observed at the particular temperature/particle concentration. The results are discussed in correlation with the thermophysical properties predicting the enhanced thermal conductivity of the samples. The absorbance values and the electrical conductance of the samples increase with the gradual addition of silver nitrate solution to tannic acid.

Keywords: Nanofluid, colloidal solution, ultrasonic velocity, acoustical parameter, absorbance, electrical conductance.

Introduction

The nanoscale dimension and high surface area to volume ratio of nanoparticles makes their physicochemical and acoustical properties quite different from those of the bulk materials¹⁻². This makes nanomaterials capable of being potentially applied in diverse fields including photonics and electronics, sensing, imaging, information storage, environmental remediation, drug delivery, and biolabelling¹. Again, to take care of growing demand of energy density heat transfer capacity needs to be increase and this can be attained by use of fluid with higher thermo physical properties. Innovative heat transfer fluids-suspended by nanometre sized solid particles are called 'nanofluids'³ including metal oxides, chemically stable metals and several allotropes of carbon with thermal conductivities, typically an order of magnitude higher than those of the base fluids and with sizes significantly smaller than 100 nm. The heat transfer enhancement using nanofluids may be affected by several mechanisms such as Brownian

motion, sedimentation, dispersion of the suspended particles, thermophoresis, diffusiophoresis, layering at the solid/liquid interface, ballistic phonon transport. However, the proper physical mechanism of heat transfer enhancement has not been established till date. Ultrasonication is an accepted technique for study of such Brownian motion and dispersing aggregated nanoparticles for the preparation of aqueous nano suspensions. Most of the studies⁴ have focused on the effect of sonication time on the stability of nanofluids and all of them used continuous ultrasonic waves to sonication. The ultrasonic property of solid-liquid suspensions in micrometer size of particles causes a change in ultrasonic velocity which affects the thermal conductivity of the nanofluids⁵. Therefore, the investigation of the influence of nanoparticles on the properties of a base liquid is necessary in order to be able to better predict the final properties of the complex fluids. In the present work we have made the study of the propagation of ultrasonic wave in a colloidal solution with dispersed Ag-metal nanoparticles. We prepared

stable nanofluids containing silver metal nanoparticles with tannic acid as reducing and stabilizing agent suspended in the base fluid like water. Even though reports are available on the thermal conductivity and viscosity of nanofluid, very little work is reported on the acoustical parameters of nanofluid. Hence, an attempt is made in this work to prepare nanofluid of Ag and study their acoustical parameters with the aid of ultrasonic wave.

Experimental

Materials

Silver nitrate (AgNO_3 , 99.9%), tannic acid ($\text{C}_{76}\text{H}_{52}\text{O}_{46}$, Mw = 1701), K_2CO_3 were all purchased from Sigma Aldrich and used without further treatment. All of the solutions were freshly made for the synthesis of silver nanoparticles, especially the freshly made tannic acid aqueous solution, which was ice bathed before use. All solutions were prepared using distilled water.

Methods

Preparation of Samples

In a typical synthesis, Ag nanoparticles are prepared by wet chemical method⁶. During the preparation of nanofluid, aqueous solutions of tannic acid and K_2CO_3 powder are prepared in distilled water. Then the solution of tannic acid and K_2CO_3 was heated at a temperature 30°C . Silver nitrate solution was added to the above heated solution under constant stirring for different molar concentrations such as 1ml, 2ml, 3ml, 4ml and 5ml. Ultrasonication process is used to suppress the formation of particle clusters and to obtain stable suspensions. The absorption spectrum of synthesized silver nanofluids for different concentration was observed by ultraviolet-visible spectrophotometer. Ultrasonic velocity measurements of the prepared nanofluids are carried out using Nanofluid interferometer at a fixed frequency of 2 MHz. Density of the fluids are measured using specific gravity bottle (10cc). Viscosity of the fluids are measured using Ostwald viscometer. The absorbance of the nanofluid was measured using colorimeter CL157 and electrical conductance with the help of Conductivity Meter.

Results and discussion

The crystalline structure of the particle is characterized by X-ray diffraction (XRD) analysis using Shimadzu XRD-6000 diffractometer. The X-ray diffraction patterns

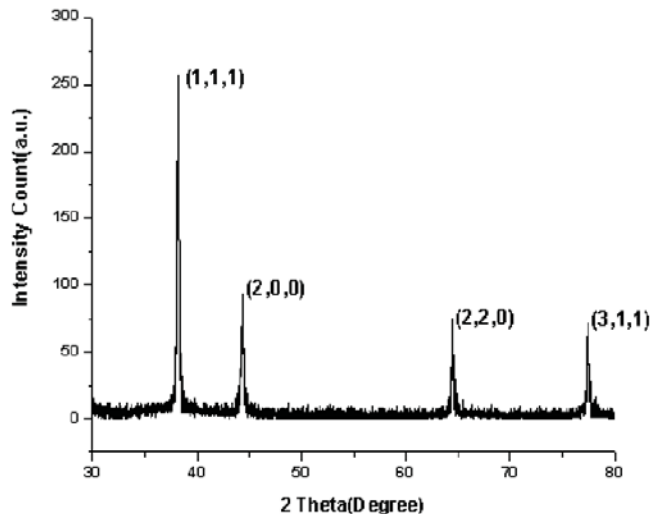


Fig. 1 X-ray diffraction pattern of Ag nanoparticles

are recorded in 2θ range between 30° and 80° with a scanning rate of $0.6^\circ/\text{sec}$.

Fig.1 shows the X-ray diffraction pattern of synthesized silver nanoparticles. The diffraction pattern shows four sharp and well defined diffraction lines at $2\theta = 38.15^\circ$, 44.34° , 64.5° and 77.46° , which can be assigned to the (111), (200), (220) and (311) reflections of the face centered cubic (fcc) structure of metallic silver, respectively. The well defined intense peaks in diffraction pattern confirm excellent crystallinity of silver nanoparticles.

The strong absorption peaks of UV-visible spectrum also revealed the presence of silver nano particles as shown in Fig. 2. The UV-visible spectrum shows strong absorption peaks at 410 nm and 416 nm due to the

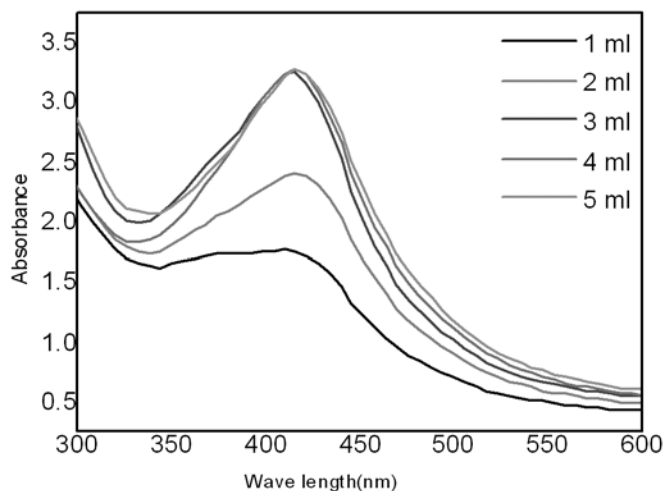


Fig. 2 Absorption spectra of silver nanofluids

plasmon oscillation modes of conduction electrons in the colloidal nanoparticles liquid suspensions. This indicates that size of Ag nanoparticles increases with increase of concentration. The peak shifting towards the shorter wavelength region was observed indicating the decrease in the size of nanoparticles.

The absorbance of visible light increases with increase of concentration of nanofluid as per Beer and Lambert law and principle of colorimetry as shown in Fig. 3. This indicates that with increase of concentration more and more agglomeration of silver nano particles takes place with the base liquid which increases the density of the medium and hence increases in absorption. Again, the conductance of the silver nano fluid increase with concentration up to 4ml and then decreases for higher concentration (Fig. 4). The electrical conductance of the

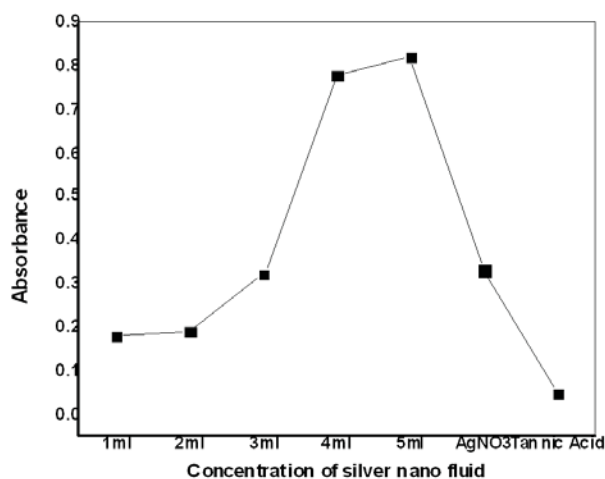


Fig. 3 Variation of absorbance with concentration of silver nano fluid

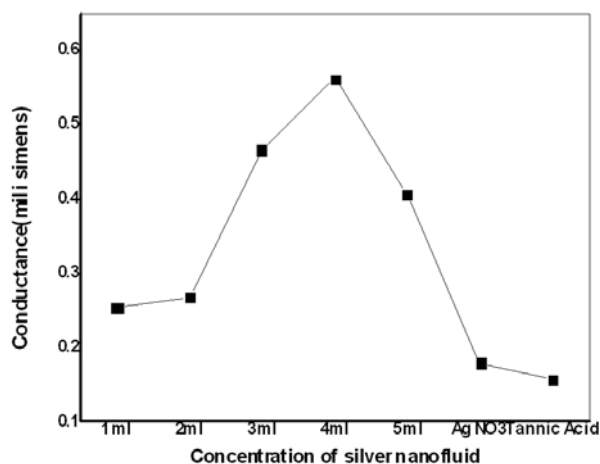


Fig. 4 Variation of conductance with concentration of silver nano fluid

solutions is a summation of contribution from all the ions present in unit volume of the solution and the velocities with which the ions move under the influence of the EMF. Thus, for an applied EMF, maintained at constant but exceeds decomposition voltage of the electrolyte, the current flowing between the electrodes immersed in the electrolyte will vary inversely with the resistance or directly with the conductance of the electrolyte solution.

The measured ultrasonic velocity data and density of nano fluid are used to compute the acoustical parameter such as acoustic impedance (Z) adiabatic compressibility (β) and intermolecular free length (L_f) and using relations⁷. Fig.5 shows that the ultrasonic velocity in silver nano fluid increases with temperature and for higher temperatures it is almost constant.

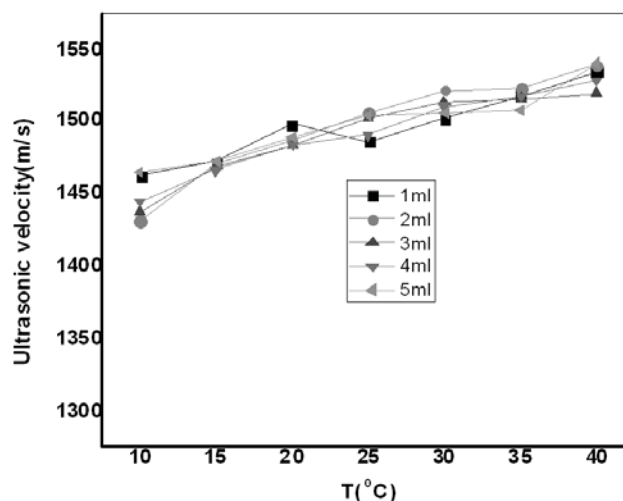


Fig. 5 Variation of ultrasonic velocity with temperature

The ultrasonic velocity in liquids generally decreases with temperature due to having negative temperature gradient of velocity. The anomalous behavior of velocity can be interpreted as the nanosized Ag particles have more surfaces to volume ratio and formation of hydrogen bonds with H_2O molecules can absorb more H_2O molecules on its surface, hence making the transport easy from one point to another point, which enhances the velocity. It is observed that acoustic impedance values increases with increase in concentration of particles and then it decreases with further increase in concentration. From the profile it is observed that acoustic impedance (Fig. 6) increases with increase in Ag nano particles in water. But there is a decrease of Z -value at 2 ml concentration which shows weak interactions. The higher

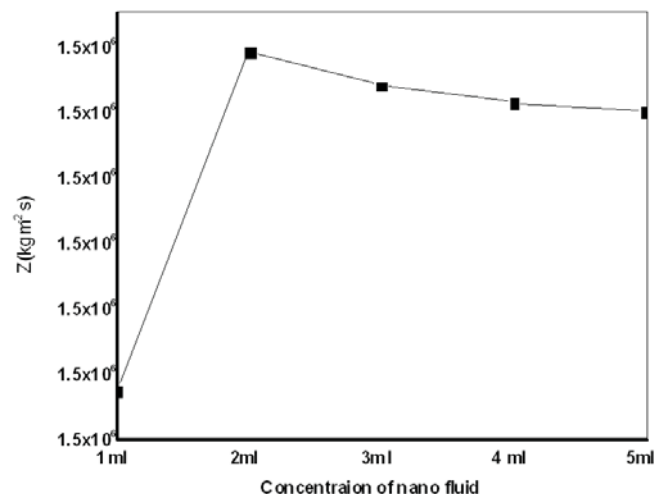


Fig. 6 Variation of acoustic impedance with concentration of silver nano fluid

values of acoustic impedance indicate that there is a significant interaction between the particle and base fluid molecules which may affect the structural arrangement. The interaction between particles and base fluid molecules increases the intermolecular distance between the molecules which in turn causes impedance in the propagation of ultrasonic waves. It is also found that the adiabatic compressibility and free length decreases with increasing concentration of particles. The decrease in adiabatic compressibility and free length shows (Fig. 7 and Fig. 8) the weaker force of interaction between particles and base fluid molecules. Compressibility decreases due to the fact that Ag^+ ions form a core compact structure with the solvent molecules through hydrogen bonding.

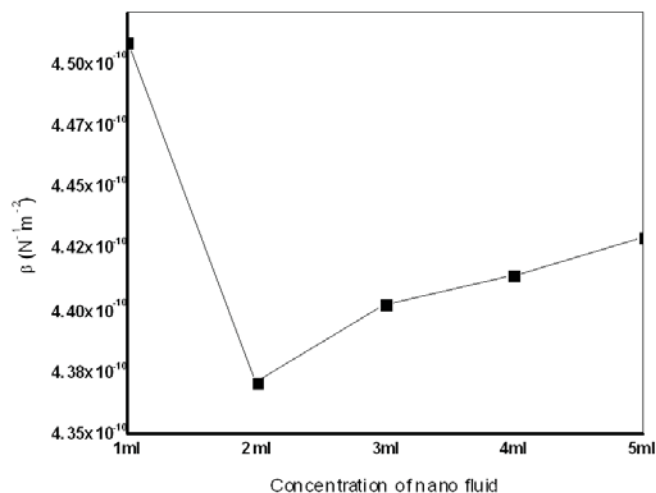


Fig. 7 Variation of adiabatic compressibility with concentration of silver nano fluid

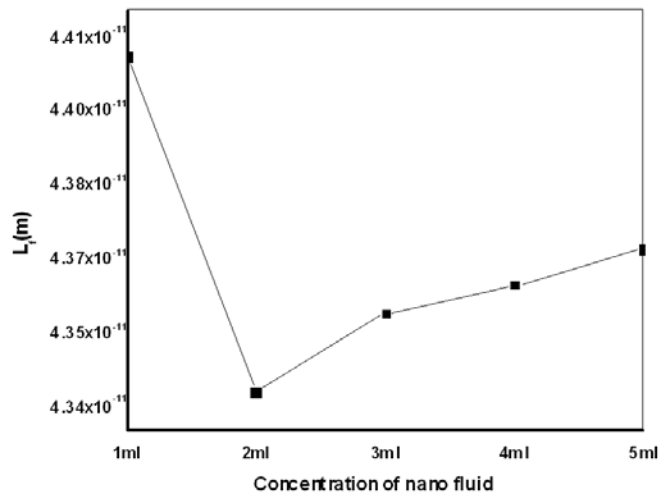


Fig. 8 Variation of intermolecular free length with concentration of silver nano fluid

Deviations in the values of adiabatic compressibility and intermolecular free length exhibit in the system may be due to weak forces operating with some specific interaction between molecules.

Conclusion

The ultrasonic velocity in Ag nanofluid has been investigated for different temperatures vary from below room temperature to above it. Various acoustical parameters were evaluated using the experimental data. Interaction between particles and water molecules was analysed using acoustical parameters. From the analysis of all acoustical parameters, it is evident that particle - particle interaction becomes predominant after 2ml due to agglomeration. It is observed that there is particle-fluid interaction which favors increase in velocity. Such particle-fluid interaction studies are helpful to understand the reasons behind unusual enhancements in physical properties of nanofluids and to comprehend the mechanism of fluid flow in nanoscale. It may be concluded that ultrasonic velocity is higher for nanofluids compared with base liquid for better enhancement of nanosuspension that could be used for industrial applications.

Acknowledgements

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Ultrasonic characterization of thymine in aqueous solution by non-destructive technique

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Ultrasonic characterization of thymine in aqueous solution by non-destructive technique is essential for utilizing them in biomedical technology. In biological sciences, nitrogenous bases are increasingly termed nucleobases because of their role in nucleic acids, their flat shape is particularly important when considering their roles as the building blocks of DNA and RNA. The present paper reports the thermo-acoustic analysis of thymine in aqueous solution by ultrasonic non-destructive technique at different molar concentrations and temperatures. The non-linear and complex behaviour of thymine in aqueous medium helps to detect phase separation and strength of intermolecular interactions between the constituents in the aqueous solution of thymine.

Keywords: Non-destructive technique; thymine; thermo-acoustic parameters; material characterization.

Introduction

Ultrasonic wave velocity in a medium provides valuable information about the physical properties of the medium¹⁻³. It also provides important information about various inter and intra-molecular processes such as relaxation of the medium or the existence of isomeric states or the exchange of energy between various molecular degrees of freedom⁴⁻⁶. Ultrasonic parameters are extensively being used to study molecular interactions in pure liquids binary liquid mixtures and ionic interactions in single and mixed salt solutions of bio-liquids⁷⁻⁸. The experimental investigations have shown that derived parameters provide a better insight into molecular processes.

Experimental

Materials and Methods

The liquids used were of BDH analar grade and were redistilled in the laboratory. In this study the measurements have been made in the temperature range 293 K-308 K. The temperature of the liquid mixture was kept constant by the use of thermostat U-10 with ± 0.01 K accuracy. Density measurement was carried out by

using hydrostatic sinker method with an accuracy $\pm 0.01\%$. A monopan electrical balance of least count as 0.0001 gm was used to record change in plunger weight dipped in the solutions correct to fourth place of decimal. Ultrasonic velocity measurements were made with an ultrasonic multi-frequency interferometer at frequency 3 MHz with an accuracy of $\pm 0.1\%$.

Result and Discussion

Figure 1 contains the plot of ultrasonic velocity versus

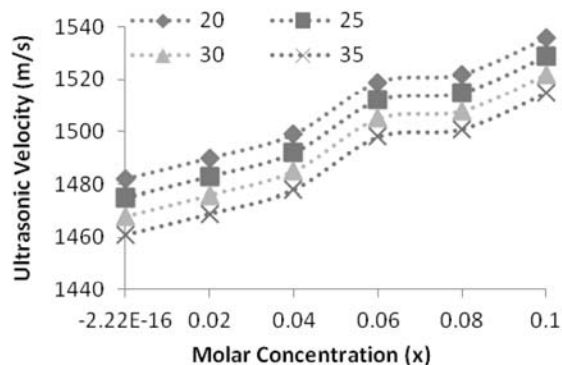


Fig. 1 Ultrasonic velocity vs conc. of thymine in aqueous solution.

molar concentration of thymine in aqueous solution at different temperatures. It is observed that ultrasonic velocity increases with increase in molar concentration of thymine and there is complex formation at molar concentration 0.06 due to molecular aggregation.

The association in the constituent molecules may be due to hydrogen bonding or due to dipole-induced dipole interaction between the constituent molecules. Amino group in thymine act as hydrogen bond acceptor or donor, hence association may be possible through hydrogen bonding.

Figure 2 shows the variation of density with molar concentration of thymine in aqueous solution. It is observed that density increases with increase in concentration of thymine in aqueous solution. Increase in density decreases the volume indicating association in component molecules due to structural reorganization.

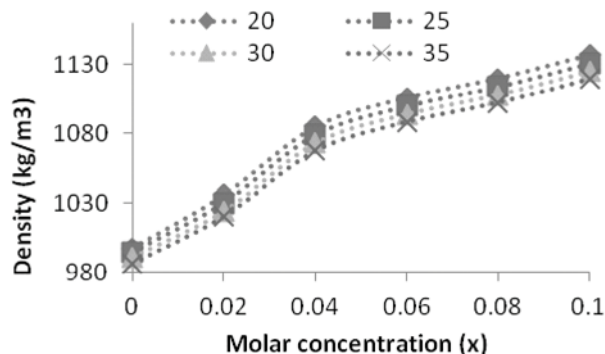


Fig. 2 Density vs conc. of thymine in aqueous solution.

Figure 3 contains the plots of adiabatic compressibility versus molar concentration. It is observed that adiabatic compressibility decreases with increase in molar concentration indicating strong molecular interaction in the component molecules of thymine in aqueous solution, showing association tendency of constituents.

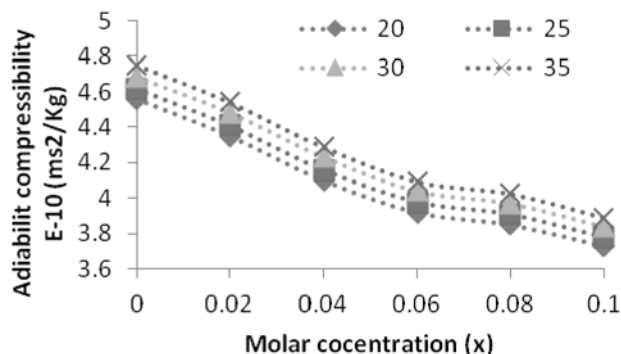


Fig. 3 Adiabatic compressibility vs conc. of thymine in aqueous solution

The observed decrease of adiabatic compressibility with molar concentration indicates the enhancement of degree of association in the constituents. Hence the intermolecular distance decreases with increase in molar concentration. It is primarily the compressibility that changes with structure which leads to change in ultrasonic velocity.

Figure 4 contains the plot of acoustic impedance (Z) versus molar concentration. It is observed that, the values of acoustic impedance increases with increase in the molar concentration of component thymine molecules in aqueous solution. It is in good agreement with the theoretical requirements because ultrasonic velocity increases with increase in molar concentration.

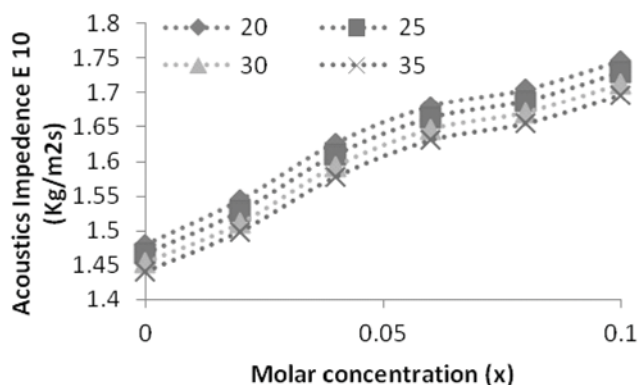


Fig. 4 Acoustic impedance vs. conc. of thymine in aqueous solution.

Conclusion

1. The observed molecular association may be due to the formation hydrogen bond or due to interstitial accommodation or due to induction or due to London dispersion forces in the constituent molecules.
2. Decrease in adiabatic compressibility with increase in molar concentration is due to molecular aggregation.
3. Thermo-acoustic parameters indicate the strength of molecular interactions.

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A piezo-resonator method for shear attenuation measurement of rubber

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Determination of shear wave attenuation coefficient in rubbers has significance in many applications, for example in non-destructive evaluation of bonding of rubber linings, acoustic absorbing/transparent coatings in air and underwater acoustic devices, shear wave attenuation tissue characterization, *etc.* Generally an elaborate and expensive acoustic test setup is used for measurement of acoustic properties of rubber and other acoustic materials. In the present work, a piezoelectric resonator method based on bandwidth of a radial mode piezoelectric disc is proposed. Piezoelectric resonators like quartz controlled microbalance (QCM) are well-known for their use in thin film thickness monitor in vacuum coating units based on resonance of quartz element. Likewise, quartz oscillator has also been deployed for viscosity determination. The present work describes a radial mode piezoelectric resonator coupled with rubber pads, wherein the radial oscillations excite shear waves into the rubber. The bandwidth of the resonator is related to the penetration depth (attenuation coefficient) of the shear waves. Bandwidth based on data from literature on decay time constant of a damped piezoelectric resonator has been used to compute the shear wave attenuation coefficient in rubbers. A good degree of agreement is observed between the calculated attenuation coefficient and the experimental value from the literature. The proposed method would provide a simple solution to measure shear wave attenuation coefficient in rubbers or other visco-elastic media at discrete frequencies.

Keywords: Piezoelectric resonator, radial mode, shear wave attenuation.

Introduction

The present work reports on a method based on a piezoelectric resonator for determination of shear wave attenuation coefficient in rubbers. Piezo-electric resonators are piezoelectric solid structures, with or without sandwiched metallic layer, having a definite geometry and size, which can vibrate in a particular mode at a particular frequency. Generally piezoelectric resonators vibrate with high Q, *i.e.* with a narrow resonant frequency and thus can be used to sense physical effect which changes its frequency. A number of methods based on piezoelectric resonators are known for various applications of sensing in research and industry. Among these mention can be made of Quartz Crystal Microbalances (QCM) which are used for monitoring thickness of thin films in vacuum coating units and their viscoelastic properties¹. QCMs are based on change of resonant frequency and Q of a quartz crystal vibrating in thickness-shear mode, due to the deposited mass of the

coated material. Now-a-days QCMs are used for sensing mass in the realm of pico- to femto-gram. Micro-machined cantilevers, capacitive resonant sensors, *etc.* have also been successfully used for mass detection in this range. Resonant piezoelectric sensors such as those based on tuning forks have been used for measurement of force².

Recently, a low force sensor based on the damped vibration characteristic of a radial mode piezoelectric resonator disc interfaced with a rubber medium has been reported for application in load measurement^{3,4}. In the present paper an attempt has been made to extend the physical principle and reported measurements of this work to deduce shear wave attenuation in rubbers. Shear wave attenuation measurement in rubbers is significant for evaluation of bonding and acoustic damping characteristics of rubber linings, acoustic absorbing/transparent coatings in air and underwater acoustic devices, tissue characterization, *etc.*

Piezoelectric Resonator Model

The resonator is based on PZT (lead zirconate titanate ceramic) material. Thin circular disc of diameter 30 mm and thickness 2 mm, poled along thickness with electrodes on both the sides, has been considered. Under a.c. excitation the disc gets longitudinally deformed along thickness due to the piezoelectric coupling constant d_{33} . Simultaneously, the disc gets radially deformed due to Poisson's ratio. The lowest resonance occurs at the fundamental radial mode having frequency constant $N_r = 2100$ Hz-m. This corresponds to 70 kHz for the diameter of the disc. The next higher mode is the longitudinal thickness resonance having the frequency constant $N_t = 2020$ Hz-m, which corresponds to 1 MHz for the thickness of the disc. At the radial mode resonance, the disc primarily vibrates in transverse radial direction with practically insignificant vibration in thickness direction. These vibrations excite shear waves in to the interfaced rubber disc.

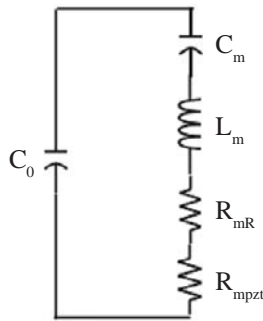


Fig. 1 A series LCR branch in parallel with the blocked capacitance of the PZT. C_m , L_m , for motional capacitance and inductance, R_{mR} for motional resistance for radiative load and R_{mpzt} for motional loss in the PZT.

The authors in reference³ have considered the vibration of the PZT disc at resonance, the equivalent circuit of which is shown in Fig. 1, comprising of a series LCR branch in parallel with the blocked capacitance of the PZT disc. C_m , L_m represents motional capacitance and inductance. R_{mR} for motional resistance for radiative load and R_{mpzt} for motional loss in the PZT.

Resonator Decay Time Constant

When excited by a pulse, the vibrations of the disc decay after the cut-off excitation voltage with a time constant τ governed by the LCR circuit and can be given by³

$$\tau = 2.L_m / (R_{mR} + R_{mpzt}) \quad (1)$$

The loss resistance is predominantly due to radiative losses R_{mR} in the medium, the losses within the PZT being considerably smaller. This is also clear from the difference in the values of mechanical Q of the loaded and unloaded PZT disc.

For static force measurement, the earlier work³ has considered that the decay time constant τ decreases almost linearly with applied stress and can be treated for force measurement over a small range. No explanation has been given for this response of the decay time constant with applied stress. An explanation of this response of the resonator is given as follows. The rubber-piezoelectric ceramic combination makes a rough interface where rubber makes contact with the ceramic initially at few points. The acoustic coupling (or loading of the piezo-ceramic) is weak at low stress and the pulse decay time constant is large. As the applied stress increases the point of contact becomes wider, the acoustic coupling into rubber increases and the decay time constant decreases. Clearly, this process shall stabilize when the rough interface becomes smooth interface at large stress. The decay time constant vs applied stress graph should actually saturate at high stress level.

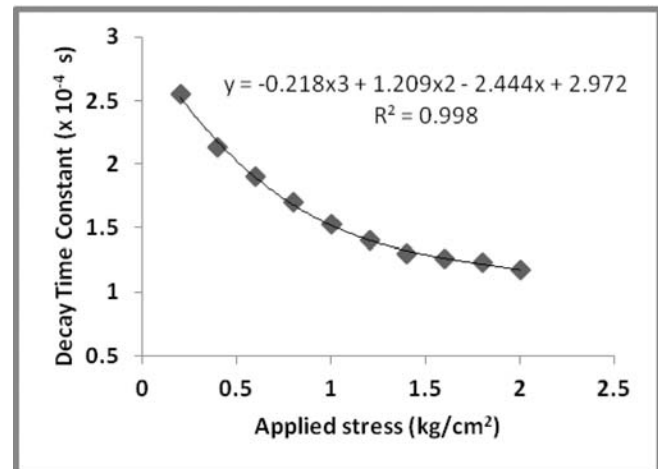


Fig. 2 Variation of decay time constant taken from [3] with applied stress between the piezoelectric resonator and the rubber interface.

A least square polynomial fit of the experimental data of reference [3] with correlation coefficient 0.9976 (Fig.2) shows that the decay time constant saturates at $\sim 1.15 \times 10^{-4}$ s. Under this condition, the decay time constant would be due to the radiated shear wave in the rubber with smooth interface. From this value of τ , the bandwidth of the resonator can be determined as :

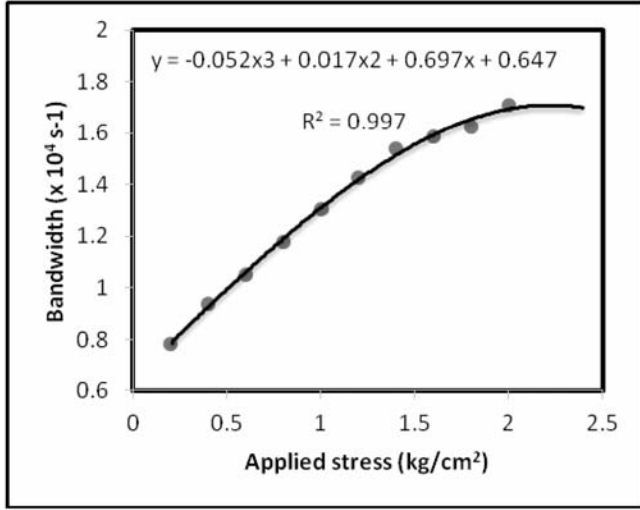


Fig. 3 Variation of band width of piezoelectric resonator interfaced with rubber with applied interfacial stress.

$$\Delta f \cdot \tau/2 = 1 \quad (2)$$

The variation of the bandwidth with applied stress also shows a polynomial fit with a high degree of correlation coefficient equal to 0.997. It is to be noted that the decay time constant above is for the case where piezo-resonator is sandwiched between two rubber layers. For one rubber layer on one side this decay time constant would be double, *i.e.* 2.3×10^{-4} s and the corresponding limiting value of bandwidth would be half *i.e.* 8.5×10^3 s⁻¹.

Evaluation of shear wave attenuation coefficient

From the theory of QCM radiating into a viscous medium the half-power bandwidth has been shown⁵ to be related to the shear wave penetration depth (attenuation coefficient). The quartz crystal half-power half bandwidth increases to $\Delta\Gamma$ as follows.

$$\Delta\Gamma/f = 1/(\pi \cdot Z_q) \cdot [\rho_{liq} \cdot \omega \cdot \delta/2] \cdot [1 + 2 \cdot h_r^2/\delta^2] \quad (3)$$

Where Z_q represents the shear acoustic impedance of quartz, ρ_{liq} is the density of the viscous liquid having viscosity η . δ is the shear wave penetration depth given by $(2\eta/\rho_{liq} \cdot \omega)^{1/2}$, which is related to the attenuation coefficient (α) as $\delta = 1/\alpha$. h_r is the surface roughness parameter.

This theory can be applied to the present case where, instead of quartz crystal and viscous medium, a PZT disc and the rubber medium is involved Z_q shall be radial mode acoustic impedance of piezo-ceramic Z_c equal to the product of density ($=7.6$ g/cm³) and radial mode

Table 1 – Material parameters of rubber and PZT disc.

Sl.	Parameter	Value
1	Density of rubber	1.7 g cm ⁻³
2	Density of piezoelectric element (PZT)	7.6 g cm ⁻³
3	Radial velocity in PZT	2100 m/s
4	Longitudinal velocity in PZT	4040 m/s
5	Dielectric loss in PZT (tan)	0.004
6	Mechanical loss (tan)	0.002
7	Mechanical Q of a unloaded piezo-resonator	500
8	Mechanical Q of a loaded piezo-resonator	16.1

velocity (2.1×10^5 cm/s) of piezo-ceramic⁶. ρ_{liq} shall be the density of rubber $\rho_r (=1.7$ g/cm³). Under the condition that the piezo-ceramic-rubber sandwich is under large applied stress, the roughness term h_r at the interface can be neglected. The bandwidth Δf of the piezo-ceramic resonator can thus be written as double of $\Delta\Gamma$ as :

$$\Delta f/f = 2 \cdot [\rho_r \cdot \omega]/(2\pi \cdot Z_c \cdot \alpha)$$

$$\text{Or,} \quad \alpha = 2 \cdot \rho_r \cdot f^2/(Z_c \cdot \Delta f) \quad (4)$$

Substituting the values of various quantities in Eq. (4), the shear wave attenuation coefficient α at the frequency 70 kHz is computed as ~ 18 dB per cm. From the literature⁷ the value α is seen quite close to this value.

Conclusion

A method using a piezoelectric resonator is proposed to determine shear attenuation coefficient at specific frequencies in rubbers or similar materials. The model used is based on expression of bandwidth of the radial mode PZT disc radiating shear waves in rubber medium. A close agreement is observed between the computed value and the literature value of shear wave attenuation coefficient at 70 kHz in natural rubber.

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PhD Thesis Summary

Ultrasonic Study of Smart Materials for Engineering Applications

(Awarded 2016 by NIMS University, Jaipur to Rajkumar, GITM, Gurgaon)

This Ph.D. thesis focused on thermo-acoustic properties of smart materials. The thesis presented theoretical studies of higher order elastic constants, mechanical parameters like Young modulus, bulk modulus, shear modulus, tetragonal modulus, Cauchy's relations, Zener anisotropy, ultrasonic velocity, ultrasonic Grüneisen parameters, non-linearity parameters and ultrasonic attenuation coefficients. The Coulomb and Born-Mayer potential approach was applied for the computation of second and third order elastic constants. The lattice parameter and hardness parameter was used to find out the second and third order elastic constants.

The thesis was divided into six chapters. Chapter 1 described in detail the introduction. Chapter 2 represented the review and literature. Chapter 3 elaborated complete theory for different materials of higher order elastic constants, sound velocity, ultrasonic Grüneisen parameters and ultrasonic attenuation due to different phenomenon. Chapter 4 provided the results of elastic and ultrasonic investigations of different NaCl-type materials like americium monpnictides AmX (X: N, P, As, Sb and Bi), californium monpnictides CfX (X: N, As and Sb), strontium monochalcogenides SrX (X: S, Se and Te), praseodymium monpnictides, cerium arsenide (CeAs) and Samarium monosulfide over a large temperature range 10-500K along $\langle 100 \rangle$, $\langle 110 \rangle$ and $\langle 111 \rangle$ crystallographic directions. Chapter 5 presented the discussion of the results obtained from the of present work. These results are discussed with previous findings on the similar types of materials. In discussion of the results, future performance of these materials has been discussed. Chapter 6 presented a complete summary of the work and major conclusions.

On the basis of the obtained results the thesis conduct that the high order elastic constants provide a connection between the mechanical and dynamical performance of the materials and supply useful knowledge regarding the nature of forces operating in them. Evaluated values of SOECs and TOECs of the investigated materials also provide mechanical properties like Young modulus, shear modulus, bulk modulus, Zener anisotropic factor and tetragonal moduli, which in

turn provide future performance of particular materials and nature of the same. The values of higher order elastic constants provided information on the mechanical stability and stiffness of the materials. The ultrasonic velocities of the investigated materials provided information about crystallographic texture and are key parameters to characterize the materials. The Grüneisen parameter is another one important parameter, which was used to characterize investigated materials. The Grüneisen parameters solely depend on values SOECs and TOECs, which provide thermoelastic behaviour of the material. The Grüneisen parameter was used to explain the thermal properties of the materials like thermal expansion, thermal conductivity, specific heat, energy density and many more parameters. The acoustic coupling constants of samarium monosulfide was used to measure acoustic energy and thermal energy conversion through phonon-viscosity mechanism. Another important investigated parameter was ultrasonic attenuation, which was obtained by Mason's approach by means of phonon-phonon interaction, electron-phonon interaction and thermo elastic mechanisms. The electron-phonon mechanism was seen to play important role in low temperature region, where mean free path of electron is equal to mean free path of phonon. At room temperature (300K) and above, the phonon-phonon interaction and thermo elastic mechanisms plays important role. The ultrasonic attenuation due to electron-phonon interaction was seen to have similar behaviour as of reciprocal of resistivity at different lower temperatures and decrease with temperature. The ultrasonic attenuation due to phonon-phonon interaction increase with temperature.

The outcome of the research, which is expected from this thesis work, may be beneficial to various scientists and engineers, who are engaged in the field of material science and engineering. The obtained results may be applied for society in various forms.

It was concluded that the obtained results will be useful for finding various thermo physical properties of the materials at different higher temperatures which have being or various application of these materials.

Dr. KUPPILI TRINATH : A PROFILE



(1954-2016)

Dr. Kuppili Trinath obtained Ph.D (Physics) degree in the field of Acoustic Sensors for Underwater Applications from Andhra University, Visakhapatnam. He joined DRDO at Naval Physical and Oceanographic Laboratory (NPOL), Kochi and worked in Naval Science and Technological Laboratory (NSTL), Visakhapatnam since 1981. He was Associate Director (Scientist G) and Head of the Sensors and Acoustic Division. He was responsible for the Design and Development of Acoustic Sensors for Underwater vehicles and was in-charge of the Underwater Acoustic Transducer Testing Facility as well.

He was Life Fellow of the USI and ASI and Life Member of many professional bodies. He was editor of "Cutting Edge" magazine of NSTL and published more than 100 papers in National and International Journals.

He was honoured by DRDO AGNI award for the development of Acoustic Sensors, Sir C V Raman Award of ASI, Viveknanda's Excellence Award, Laboratory Scientist Award etc. He was co-chair for Transducers, Electro-Acoustics and MEMS Session of Indo-French Acoustic Conference 2013. He visited USA twice as a member of DRDO Delegation for mutual Co-operation with USA and presentation of papers.

The community of the Ultrasonics Society of India paid rich tribute to Dr. Trinath on his passing away as a distinguished researcher who has promoted and honoured our discipline in DRDO and as well as in India.

Dr. TUSHYA KUMAR SAXENA : A PROFILE



(1955-2014)

Dr. T.K. Saxena, obtained degrees in B.Sc. (Hons.) and M.Sc. both in Physics from Delhi University in 1974 and 1976 respectively. He obtained his Ph.D. Degree also from Delhi University in 1981 for his work in the area of Semiconductor Physics, done under the guidance of Prof. P. C. Mathur. After working as Research Associate at Delhi University during 1981-82, he joined NPL as Scientist 'B' 1982. He was Head, Acoustics, Ultrasonics and Vibrations (AUV) standards and Electronic and Instrumentation Cell at CSIR-National Physical Laboratory, New Delhi.

He contributed extensively during his life in the fields of Electronic Circuitry, Computer Programming, Automation & Software Development, the Computer Aided Measurements & Control. He Published a good number of papers in Indian Journal of Engineering and Materials Science, Journal of Instruments Society of India, Journal of Physics D: Applied Physics, Mapan-JMSI and NPL Technical Bulletin.

He was a life member of Ultrasonics Society of India, Metrology Society of India, Computer Society of India and Instrument Society of India. He was a recipient of NPL Technology Day Award (2006), CSIR Technology Innovation Award (2012) and Merck Millipore India Innovation Award (2012).

He expired in 2014 after completion of 32 years of service in CSIR-National Physical Laboratory, New Delhi.

The community of the Ultrasonics Society of India paid tribute to Dr. Saxena on his passing away as a distinguished Scientist and administrator, who has promoted and honoured our discipline in India and as well as in abroad.

Journal of Pure and Applied Ultrasonics

INFORMATION FOR AUTHORS

1. Type of Contribution

JOURNAL OF PURE AND APPLIED ULTRASONICS welcomes contributions on all aspects of ultrasonics including ultrasonic studies in medical ultrasonics, NDT, underwater, transducers, materials & devices and any other related topic. Contributions should fall into one of the following classes.

Paper - These should be on original research work contributing to scientific developments. They should be written with a wide readership in mind and should emphasize the significance of the work.

Reviews and Articles - Includes critical reviews and survey articles.

Research and Technical notes - These should be short descriptions of new techniques, applications, instruments and components.

Letters to the editor - Letters will be published on points arising out of published articles and papers and on questions of opinion.

Miscellaneous - Miscellaneous contributions such as studies, interpretive and tutorial articles, conference reports and news items are also accepted. Recommended contribution lengths are: Papers 2000-4000 words. Reviews and Surveys 2000-5000 words; Conference Reports 500-1500 words; News Items, Research and Technical Notes up to 1000 words.

2. Manuscripts

Manuscripts should be typed on one side of the paper

in double spacing with wide margins on A4 size paper. The originals of text and illustrations plus two copies should be provided to facilitate the process of reviewing. Complete manuscript in MS WORD or Page Maker would ensure no mistake. Floppy containing article should be sent along with the manuscript.

Title - Titles should be short and indicate the nature of the contribution.

Abstract - An abstract of 100-200 words should be provided on the title page of paper and review article. This should indicate the full scope of the contribution and include the principal conclusions.

Mathematics - Mathematical expressions should be arranged to occupy the minimum number of lines consistent with clarity e.g., $(x^2+y^2)/(x-y)^{1/2}$.

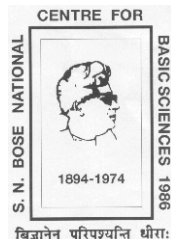
Illustration - The line illustrations along with captions should be clearly drawn with black Indian ink. Figures in Excel are preferred.

References - References should be referred to in the text by number only. The reference number should be given as superscript. The corresponding reference shall contain the following information in order; names and initials of author (s)(bold), title of the work, journal or book title (italic), volume number (bold), year of publication in brackets, page number, e.g., **Giri R. and Nath G.**, Ultrasonic study of solvent extractant in nuclear technology, *J. Pure Appl. Ultrason.*, **37** (2015) 44-47.

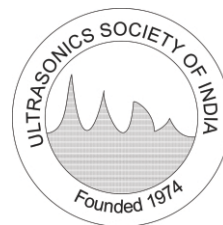
Units and Abbreviations - Authors should use SI units wherever possible.

XXIst National Symposium on Ultrasonics (NSU-2016)

08 – 10 November, 2016



Organised by
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Important dates :

- Abstract submission- from 1st July, 2016 to 31st August, 2016
- Confirmation of abstract acceptance - 15th September, 2016
- Registration ends- 30th September, 2016