Study of molecular interactions of 2-amino-5-nitrothiazole in NNDMF, acetonitrile, and ethanol using acoustical parameters

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Abstract: Density, ultrasonic velocity of binary mixture of 2-amino-5-nitrothiazole + N,N-dimethylformamide (NNDMF), 2-amino-5-nitrothiazole + acetonitrile, and 2-amino-5-nitrothiazole + ethanol were measured at different concentrations of 2-amino-5-nitrothiazole and at different temperatures (303.15, 308.15, 313.15, 318.15 and 323.15) K. Acoustical parameters such as adiabatic compressibility, intermolecular free length, acoustical impedance, sound velocity number, and relative association were determined from experimental data of density and ultrasonic velocity. The effect of temperature and concentration variations in the strength of molecular interaction has been studied. Effective correlation was observed in terms of solute-solvent and solvent-solvent interaction at all temperatures and concentration.

Subjects: Chemistry; Material Science; Materials Science; Physics

Keywords: density; ultrasonic velocity; acoustical parameter; molecular interaction; 2-amino-5-nitrothiazole; NNDMF; acetonitrile and ethanol

1. Introduction
Thiazole and its derivatives are of biological significance (Carbone et al., 2013; Gupta & Kant, 2013; Jalhan, Jindhal, & Gupta, 2012; Saundaneanand, Walmik, Kirankumar, & Annapurna, 2014; Shah, 2014)
and the study of molecular interaction in the solution is useful to understand their biological applications. Recently, Otutu, Osabohien, and Efuruhevwe (2011) synthesized and studied the spectral properties of heteraryl monoazo dye which is a derivative of 2-amino-5-nitrothiazole. Sulfur- and nitrogen-containing heterocyclic compounds represent an important class of drugs in the therapeutic chemistry and also contributed to the society from biological and industrial point which helps to understand life processes (Shah, 2012). The knowledge of densities, ultrasonic velocities, and various acoustical parameters are useful for the studies of physicochemical properties of a system. Study of molecular interaction in liquid provides valuable information regarding internal structure molecular association, complex formation, internal pressure etc. The studies of solution properties of liquid solution of polar and non-polar components have great applications in industrial and technological process (Talegaonkar, Burghate, & Wadal, 2013). The recent publications (Chauhan, Kumar, & Patial, 2013; Pradhan & Roy, 2014; Singh, Shakya, Shakya, & Yadav, 2012; Stepanov & Minchenko, 2014; Xie, Dong, Zhang, Lu, & Ji, 2014) in this area shows that the many researchers give attention toward study of ultrasonic velocity measurement and study of acoustical properties.

Different liquids flow with different rates depending upon the nature of molecules present in the substance and how easily they slide over one another during their flow. Nagargun, Rao, and Rambabu (2013) have studied speeds of sound and density for binary mixtures of ethyl benzoate (EB) with N,N-dimethylformamide (NNDMF), N,N-dimethylacetamide (NNDMAc), and N,N-dimethylaniline (NNDMA) as a function of mole fraction at different temperature and atmospheric pressure. Recently, Naik, Narwade, Bodakhe, and Muley (2014) reported the molecular interactions in substituted pyrimidines-acetonitrile solutions at different temperature. Measurement of ultrasonic study of an organic ligands solutions provide an excellent method of obtaining data on the ion solvent and solvent–solvent interaction and structure breaking and making properties of solutes. The ultrasonic velocities, densities, and relative association of 2-amino-5-nitrothiazole used in this investigation in polar aprotic–polar protic mixed media at different temperature was lacking and therefore in the present work, we investigate sound velocities and densities for (2-amino-5-nitrothiazole + NNDMF), (2-amino-5-nitrothiazole + acetonitrile), and (2-amino-5-nitrothiazole + ethanol) of different concentrations at temperatures 303.15, 308.15, 313.15, 318.15, and 323.15 K in order to know the effect of temperatures on various acoustical properties. The applications of 2-amino-5-nitrothiazole in different fields of science developed our interest in the measurement of their velocities and densities, and computes the acoustical properties to understand their interaction with non aqueous solvents mixture at different temperature.

2. Materials and experimental

The solvents N,N-dimethylformamide (NNDMF) (Fine-lab, minimum wt. 98%), acetonitrile (Sigma-Aldrich minimum wt. 99%), and ethanol (Sigma-Aldrich minimum wt. 99%) of analytical grade used without purification. The ligand 2-amino-5-nitrothiazole (Hi-MEDIA, minimum wt. 97%) used are of synthesis grade. The ligand solutions in different solvents were prepared by dissolving an accurate amount in an organic solvent in standard flask with airtight caps, and the mass measurements were performed using high-precision digital balance (Adair Datta of accuracy ±0.01 mg). The ultrasonic velocities of pure component and their mixtures were measured by ultrasonic interferometer (Mittal enterprises, model F-81s) at 2 MHz having accuracy ±1 m s⁻¹ in velocity. It consists of high-frequency generator and a measuring cell. The densities of NNDMF, acetonitrile, ethanol, and ligand solutions were measured by digital density meter (Anton Paar DMA 35 of accuracy ±0.001). A thermostatically controlled well-stirred water bath whose temperature was controlled to ±0.1 K was used for all the measurements.

3. Theory

Numerous methods are available in the literature for measuring ultrasonic velocity in solid and liquids. The ultrasonic interferometer is considered as more reliable and precise instrument. The expression used to determine ultrasonic velocity using ultrasonic interferometer is:

\[ u = \frac{v}{\lambda} \]

where \( u \) is ultrasonic velocity and \( \lambda \) is wavelength.
The isentropic compressibility $\beta_s$ was calculated from following equation:

$$\beta_s = 1/\rho u^2$$

where $\rho$ is density of solution and $u$ is speed of ultrasonic velocity.

The intermolecular free length $L_f$ is calculated using the standard expression:

$$L_f = K\rho_s^{1/2}$$

where $K$ is temperature-dependent constant known as a Jacobson constant (Syal, Patial, & Chouhan, 1999).

The acoustic impedance $Z$ is obtained by equation

$$Z = u\rho$$

The relative association $R_A$ was calculated by the following equation:

$$R_A = (\rho/\rho_o)(u_o/u)^{1/3}$$

where $\rho_o$ is density of solvent and $u_o$ is velocity of solvent. Also, the sound velocity number is calculated from following equation:

$$[U] = u - u_o/u_o c$$

$[U]$ is sound velocity number; $c$ is concentration of the solute.

4. Results and discussion

The calibration of the ultrasonic interferometer was done by measuring the ultrasonic velocities and densities of the pure NNDMF, acetonitrile, and ethanol, respectively. The measured value is found to be in good concordance with literature values which are shown in Table 1. Small difference may occur due to difference in purity of chemicals, measurements, techniques, and calibrations.

The values of densities and ultrasonic velocities for systems 2-amino-5-nitrothiazole + NNDMF, 2-amino-5-nitrothiazole + acetonitrile, and 2-amino-5-nitrothiazole + ethanol respectively are listed in Table 2 and the values of acoustical parameters such as adiabatic compressibility, relative association, acoustical impedance, linear free length, and sound velocity number are listed in Tables 3–5, respectively. In this investigation, the value of density and ultrasonic velocity increases with increase in the concentration of 2-amino-5-nitrothiazole and decreases with increase in the temperature at any particular concentration for all the three systems (Figures 1–5). The values from Table 2 show that ultrasonic velocity values decrease with increase in temperature due to the breakage of heteromolecular clusters at high temperatures (Nagargun et al., 2013). The decrease in ultrasonic velocity and density with increase in temperature indicates that cohesive forces decreased (Godhani, Dobariya, Sanghani, & Mehta, 2012). The increasing temperature has two opposite effects namely increase in molecular interaction (structure formation) and destruction of structure formed previously. When the thermal energy is greater than the interaction energy, it causes the destruction of previously formed structure. Thus, the increase in temperature favors the increase in kinetic energy and volume expansion and hence, results in the decrease in $\rho$ and $u$. Also, these results indicate that there is a significant interaction between the solute and solvent molecules because the presence of amine group of 2-amino-5-nitrothiazole and alcoholic group of ethanol creates the possibility of hydrogen bonding between molecules.
When there is an ultrasonic wave incident on the solution, the molecules get perturbed. The reason is medium has some elasticity and due to this, perturbed molecules regain their equilibrium positions (Godhani et al., 2012). When a solute is added to a solvent, its molecules attract certain solvent molecules toward them; this phenomenon is known as compression. Every solvent has a limit for compression and is known as limiting compressibility. The increase in isentropic compressibility ($\beta$) with increase in temperature might be due to molecular interaction in solution which supports solvent-solute interactions. Figures 6–8 it clearly indicate that the isentropic compressibility increases with increase in temperature and decreases with increase in concentration of solute. The decrease in isentropic compressibility with increasing concentration of 2-amino-5-nitrothiazole clearly indicates the presence of solute–solvent interactions due to aggregation of solvent molecules around solute molecules (Thirumaran & Rajeswari, 2011). The variation in ultrasonic velocity in a solution depends on the intermolecular free length on mixing. On the basis of a model for sound propagation given by Kincaid and Eyring (1938), ultrasonic velocity increases with decrease in free length ($L_f$) and vice versa. Intermolecular free length is a predominant factor for determining the variation in ultrasonic velocity, in liquids and their solutions (Landge, Badade, & Kendre, 2013). Intermolecular free length ($L_f$) is the distance between the surfaces of the neighboring molecules and indicates a significant interaction between solute–solvent and solvent–solvent molecules (Yadav, Kumar, & Yadav, 2014). From graph, we say that increase in $U$ and decrease in $L_f$ indicate close association between solute and solvent molecules, whereas reverse of these suggest solute–solute interactions. Hence, it is also a good tool to investigate the molecular interactions in the binary solvent mixture. When the ultrasonic wave travels through a solution, some part of it travels through the medium and remaining part of ultrasonic wave gets reflected by the ion (Kharkale, Bhaskar, Agarwal, & Paliwal, 2013); it means ions restricts free flow of sound wave. The character that decreases this restriction or backward movement of sound waves is known as acoustic impedance ($Z$). From Figures 9–11 it is clearly observed that the value of acoustic impedance decreases as temperature increases. If the temperature increases, ultrasonic velocity decreases and value of

| Table 1. Experimental and literature values of density and ultrasonic velocity, respectively |
|---|---|---|---|
| $T$ (K) | Expt $\gamma_0$ (ms$^{-1}$) | $\rho_0$ (kg m$^{-3}$) | Lit $\gamma_0$ (ms$^{-1}$) | $\rho_0$ (kg m$^{-3}$) |
| NNDMF | 303.15 | 1458 | 0.9531 | 1456 (Syal, Patial, & Chouhan, 1999) |
| | 308.15 | 1432 | 0.9386 | 1424 (Attri, Reddy, & Venkatesu, 2010) |
| | 318.15 | 1389 | 0.9030 | 1386 (Attri et al., 2010) |
| Acetonitrile | 303.15 | 1276 | 0.7724 | 1278 (Grande, Juliá, Barrero, & Marschoff, 2013) |
| | 308.15 | 1240 | 0.7669 | 1239 (Grande et al., 2013) |
| | 313.15 | 1216 | 0.7591 | 1218 (Grande et al., 2013) |
| | 318.15 | 1198 | 0.7522 | 1198 (Grande et al., 2013) |
| | 323.15 | 1174 | 0.7491 | 1177 (Grande et al., 2013) |
| | 303.15 | 1134 | 0.7786 | 1131 (Naidu & Prasad, 2004) |
| | 308.15 | 1116 | 0.7758 | 1115 (Naidu & Prasad, 2004) |
| | 313.15 | 1096 | 0.7722 | 1099 (Naidu & Prasad, 2004) |
| | 318.15 | 1074 | 0.7687 | 1075 (Gong, Shen, Lu, Meng, & Li, 2012) |
| | 323.15 | 1062 | 0.7643 | 1066 (Naidu & Prasad, 2004) |

Note: Uncertainties in temperature, density and velocity are 0.1 K, 1 m s$^{-1}$ and 0.0005 kg m$^{-3}$. When there is an ultrasonic wave incident on the solution, the molecules get perturbed. The reason is medium has some elasticity and due to this, perturbed molecules regain their equilibrium positions (Godhani et al., 2012). When a solute is added to a solvent, its molecules attract certain solvent molecules toward them; this phenomenon is known as compression. Every solvent has a limit for compression and is known as limiting compressibility. The increase in isentropic compressibility ($\beta$) with increase in temperature might be due to molecular interaction in solution which supports solvent-solute interactions. Figures 6–8 it clearly indicate that the isentropic compressibility increases with increase in temperature and decreases with increase in concentration of solute. The decrease in isentropic compressibility with increasing concentration of 2-amino-5-nitrothiazole clearly indicates the presence of solute–solvent interactions due to aggregation of solvent molecules around solute molecules (Thirumaran & Rajeswari, 2011). The variation in ultrasonic velocity in a solution depends on the intermolecular free length on mixing. On the basis of a model for sound propagation given by Kincaid and Eyring (1938), ultrasonic velocity increases with decrease in free length ($L_f$) and vice versa. Intermolecular free length is a predominant factor for determining the variation in ultrasonic velocity, in liquids and their solutions (Landge, Badade, & Kendre, 2013). Intermolecular free length ($L_f$) is the distance between the surfaces of the neighboring molecules and indicates a significant interaction between solute–solvent and solvent–solvent molecules (Yadav, Kumar, & Yadav, 2014). From graph, we say that increase in $U$ and decrease in $L_f$ indicate close association between solute and solvent molecules, whereas reverse of these suggest solute–solute interactions. Hence, it is also a good tool to investigate the molecular interactions in the binary solvent mixture. When the ultrasonic wave travels through a solution, some part of it travels through the medium and remaining part of ultrasonic wave gets reflected by the ion (Kharkale, Bhaskar, Agarwal, & Paliwal, 2013); it means ions restricts free flow of sound wave. The character that decreases this restriction or backward movement of sound waves is known as acoustic impedance ($Z$). From Figures 9–11 it is clearly observed that the value of acoustic impedance decreases as temperature increases. If the temperature increases, ultrasonic velocity decreases and value of
Table 2. Experimental values of density and ultrasonic velocity of 2-amino-5-nitrothiazole + NNDMF, 2-amino-5-nitrothiazole + acetonitrile and 2-amino-5-nitrothiazole + ethanol at different concentrations and at different 303.15, 308.15, 313.15, 318.15, and 323.15 K respectively

| C (mol/l) | NNDMF + 2-amino-5-nitrothiazole | Acetonitrile + 2-amino-5-nitrothiazole | Ethanol + 2-amino-5-nitrothiazole |
|----------|----------------------------------|----------------------------------------|----------------------------------|
|          | \(\rho\) (g cm\(^{-3}\)) | \(u\) (m s\(^{-1}\)) | \(\rho\) (g cm\(^{-3}\)) | \(u\) (m s\(^{-1}\)) | \(\rho\) (g cm\(^{-3}\)) | \(u\) (m s\(^{-1}\)) |
| 0.00     | 0.9313 | 0.9681 | 0.7781 | 0.7723 | 0.7757 | 0.7757 |
| 0.01     | 0.9348 | 0.9681 | 0.7781 | 0.7723 | 0.7757 | 0.7757 |
| 0.02     | 0.9383 | 0.9681 | 0.7781 | 0.7723 | 0.7757 | 0.7757 |
| 0.03     | 0.9418 | 0.9681 | 0.7781 | 0.7723 | 0.7757 | 0.7757 |
| 0.04     | 0.9453 | 0.9681 | 0.7781 | 0.7723 | 0.7757 | 0.7757 |
| 0.05     | 0.9488 | 0.9681 | 0.7781 | 0.7723 | 0.7757 | 0.7757 |
| 0.06     | 0.9523 | 0.9681 | 0.7781 | 0.7723 | 0.7757 | 0.7757 |
| 0.07     | 0.9558 | 0.9681 | 0.7781 | 0.7723 | 0.7757 | 0.7757 |
| 0.08     | 0.9593 | 0.9681 | 0.7781 | 0.7723 | 0.7757 | 0.7757 |
| 0.09     | 0.9628 | 0.9681 | 0.7781 | 0.7723 | 0.7757 | 0.7757 |
| 0.10     | 0.9663 | 0.9681 | 0.7781 | 0.7723 | 0.7757 | 0.7757 |

Note: Uncertainties in temperature, density and velocity are 0.1 K, 1 m s\(^{-1}\) and 0.001 kg m\(^{-3}\).
Table 3. Isentropic compressibility, relative association, acoustic impedance, linear length and sound velocity number for 2-amino-5-nitrothiazole + NNDMF at different temperature and different concentration

| C (mol/l) | $\beta_s \times 10^{-10}$ (m² N⁻¹) | $R_A$ | $Z \times 10^{-5}$ (kg m² s⁻¹) | $L_f$ (Å) | $[U]$ (kg mol⁻¹) |
|-----------|----------------------------------|-------|--------------------------------|-----------|----------------|
| 303.15 K  |                                  |       |                                |           |                |
| 0.00      | 4.93                             | -     | 13.894                          | 0.4608    | -              |
| 0.01      | 4.89                             | 1.0241| 14.210                          | 0.4542    | 0.6858         |
| 0.02      | 4.73                             | 1.0539| 14.352                          | 0.4514    | 0.4801         |
| 0.03      | 4.74                             | 1.0742| 14.391                          | 0.4518    | 0.4371         |
| 0.04      | 4.62                             | 1.0904| 14.617                          | 0.4461    | 0.3429         |
| 0.05      | 4.54                             | 1.1241| 14.815                          | 0.4422    | 0.3840         |
| 0.06      | 4.46                             | 1.1501| 14.999                          | 0.4383    | 0.4115         |
| 0.07      | 4.42                             | 1.1530| 15.183                          | 0.4363    | 0.3135         |
| 0.08      | 4.31                             | 1.2073| 15.425                          | 0.4308    | 0.3772         |
| 0.09      | 4.26                             | 1.2363| 15.591                          | 0.4283    | 0.3581         |
| 0.10      | 4.20                             | 1.2639| 15.753                          | 0.4253    | 0.3497         |
| 308.15 K  |                                  |       |                                |           |                |
| 0.00      | 5.19                             | -     | 13.432                          | 0.4771    | -              |
| 0.01      | 5.12                             | 1.0266| 13.560                          | 0.4738    | 0.4189         |
| 0.02      | 5.07                             | 1.0780| 13.655                          | 0.4715    | 0.4391         |
| 0.03      | 5.01                             | 1.0966| 13.760                          | 0.4687    | 0.4189         |
| 0.04      | 4.94                             | 1.1287| 13.875                          | 0.4654    | 0.4416         |
| 0.05      | 4.94                             | 1.1540| 13.958                          | 0.4654    | 0.2230         |
| 0.06      | 4.88                             | 1.1936| 14.098                          | 0.4626    | 0.2327         |
| 0.07      | 4.74                             | 1.2239| 14.405                          | 0.4559    | 0.3192         |
| 0.08      | 4.64                             | 1.2531| 14.689                          | 0.4511    | 0.2967         |
| 0.09      | 4.60                             | 1.2783| 14.868                          | 0.4491    | 0.2327         |
| 0.10      | 4.27                             | 1.3111| 15.149                          | 0.4327    | 0.3212         |
| 313.15 K  |                                  |       |                                |           |                |
| 0.00      | 5.47                             | -     | 13.038                          | 0.4942    | -              |
| 0.01      | 5.40                             | 1.0195| 13.160                          | 0.4910    | 0.2853         |
| 0.02      | 5.30                             | 1.0400| 13.324                          | 0.4864    | 0.4992         |
| 0.03      | 5.33                             | 1.0810| 13.296                          | 0.4878    | 0.1902         |
| 0.04      | 5.24                             | 1.1159| 13.456                          | 0.4837    | 0.2853         |
| 0.05      | 5.05                             | 1.1550| 13.747                          | 0.4748    | 0.5135         |
| 0.06      | 4.97                             | 1.1860| 13.920                          | 0.4710    | 0.4992         |
| 0.07      | 4.82                             | 1.2132| 14.213                          | 0.4633    | 0.5706         |
| 0.08      | 4.83                             | 1.2540| 14.253                          | 0.4643    | 0.4279         |
| 0.09      | 4.73                             | 1.3310| 14.510                          | 0.4641    | 0.4121         |
| 0.10      | 4.62                             | 1.3401| 14.795                          | 0.4541    | 0.4279         |

(Continued)
Table 3. Continued

| C (mol/l) | $\beta_s \times 10^{10}$ (m² N⁻¹) | $R_A$ | $Z \times 10^5$ (kg m² s⁻¹) | $L_f$ (Å) | $[U]$ (kg mol⁻¹) |
|-----------|---------------------------------|-------|-----------------|-----------|-----------------|
| 318.15 K  |                                 |       |                 |           |                 |
| 0.00      | 5.60                            | -     | 12.834          | 0.5044    | -               |
| 0.01      | 5.49                            | 1.0249| 13.022          | 0.4994    | 0.5722          |
| 0.02      | 5.38                            | 1.0394| 13.192          | 0.4944    | 0.6839          |
| 0.03      | 5.30                            | 1.0650| 13.324          | 0.4907    | 0.6479          |
| 0.04      | 5.24                            | 1.1061| 13.433          | 0.4884    | 0.5579          |
| 0.05      | 5.12                            | 1.1554| 13.637          | 0.4823    | 0.6047          |
| 0.06      | 5.03                            | 1.2213| 13.819          | 0.4781    | 0.5879          |
| 0.07      | 4.93                            | 1.2515| 14.011          | 0.4733    | 0.5862          |
| 0.08      | 4.84                            | 1.3007| 14.200          | 0.4689    | 0.5669          |
| 0.09      | 4.79                            | 1.3123| 14.312          | 0.4665    | 0.5359          |
| 0.10      | 4.71                            | 1.3694| 14.541          | 0.4626    | 0.5111          |
| 323.15 K  |                                 |       |                 |           |                 |
| 0.00      | 5.84                            | -     | 12.535          | 0.5197    | -               |
| 0.01      | 5.75                            | 1.0155| 12.663          | 0.5156    | 0.5865          |
| 0.02      | 5.56                            | 1.0448| 12.917          | 0.5070    | 0.5026          |
| 0.03      | 5.46                            | 1.0563| 13.085          | 0.5025    | 0.8308          |
| 0.04      | 5.34                            | 1.0896| 13.286          | 0.4969    | 0.8047          |
| 0.05      | 5.26                            | 1.1272| 13.423          | 0.4932    | 0.7624          |
| 0.06      | 5.12                            | 1.1609| 13.651          | 0.4866    | 0.7820          |
| 0.07      | 5.03                            | 1.1964| 13.828          | 0.4823    | 0.7540          |
| 0.08      | 4.93                            | 1.2473| 14.035          | 0.4775    | 0.7331          |
| 0.09      | 4.85                            | 1.2745| 14.210          | 0.4736    | 0.7005          |
| 0.10      | 4.75                            | 1.3304| 14.428          | 0.4687    | 0.6744          |

Table 4. Isentropic compressibility, relative association, acoustic impedance, linear length and sound velocity number for 2-amino-5-nitrothiazole + acetonitrile at different temperature and different concentration

| C (mol/lit) | $\beta_s \times 10^{10}$ (m² N⁻¹) | $R_A$ | $Z \times 10^5$ (kg m² s⁻¹) | $L_f$ (Å) | $[U]$ (kg mol⁻¹) |
|------------|---------------------------------|-------|-----------------|-----------|-----------------|
| 303.15 K   |                                 |       |                 |           |                 |
| 0.00       | 7.95                            | -     | 9.850           | 0.5852    | -               |
| 0.01       | 7.90                            | 1.0212| 9.904           | 0.5833    | 0.5167          |
| 0.02       | 7.75                            | 1.0451| 10.03           | 0.5778    | 0.3918          |
| 0.03       | 7.65                            | 1.0461| 10.13           | 0.5740    | 0.3657          |
| 0.04       | 7.55                            | 1.0764| 10.21           | 0.5703    | 0.3918          |
| 0.05       | 7.38                            | 1.1107| 10.37           | 0.5638    | 0.4388          |
| 0.06       | 7.33                            | 1.1203| 10.43           | 0.5619    | 0.3918          |
| 0.07       | 7.32                            | 1.1550| 10.48           | 0.5615    | 0.2910          |
| 0.08       | 7.18                            | 1.2072| 10.62           | 0.5561    | 0.3330          |
| 0.09       | 6.99                            | 1.2593| 10.80           | 0.5487    | 0.4179          |
| 0.10       | 6.82                            | 1.3253| 10.93           | 0.5420    | 0.4702          |
| 308.15 K   |                                 |       |                 |           |                 |
| 0.00       | 8.49                            | -     | 9.498           | 0.6102    | -               |
| 0.01       | 8.32                            | 1.0264| 9.581           | 0.6066    | 0.4838          |

(Continued)
| C (mol/lit) | \(\beta_s \times 10^{-10} \text{ (m}^2 \text{ N}^{-1})\) | \(R_{A} \times 10^{-5} \text{ (kg m}^2 \text{ s}^{-1})\) | \(Z \times 10^{-3} \text{ (kg m}^{-3} \text{ s}^{-1})\) | \(L_{A} (\text{A}^2)\) | \(|U| \text{ (kg mol}^{-1})\) |
|------------|------------------|------------------|------------------|------------------|------------------|
| 0.02       | 8.22             | 1.0440           | 9.690            | 0.6004           | 0.4383           |
| 0.03       | 8.11             | 1.0344           | 9.777            | 0.5964           | 0.5376           |
| 0.04       | 8.03             | 1.0682           | 9.856            | 0.5934           | 0.4435           |
| 0.05       | 7.99             | 1.0910           | 9.979            | 0.5919           | 0.4516           |
| 0.06       | 7.89             | 1.1188           | 10.02            | 0.5882           | 0.3225           |
| 0.07       | 7.74             | 1.1064           | 10.15            | 0.5826           | 0.3686           |
| 0.08       | 7.56             | 1.1427           | 10.29            | 0.5758           | 0.4435           |
| 0.09       | 7.35             | 1.1609           | 10.50            | 0.5677           | 0.4838           |
| 0.10       | 7.22             | 1.1814           | 10.63            | 0.5627           | 0.5001           |
| 313.15 K   |                   |                  |                  |                  |                  |
| 0.00       | 8.91             | –                | 9.229            | 0.6307           | –                |
| 0.01       | 8.85             | 1.0303           | 9.308            | 0.6286           | 0.3289           |
| 0.02       | 8.86             | 1.0212           | 9.415            | 0.6218           | 0.4111           |
| 0.03       | 8.47             | 1.0785           | 9.541            | 0.6149           | 0.5482           |
| 0.04       | 8.32             | 1.0917           | 9.653            | 0.6095           | 0.5756           |
| 0.05       | 8.16             | 1.0845           | 9.778            | 0.6036           | 0.5921           |
| 0.06       | 8.01             | 1.0984           | 9.913            | 0.598            | 0.5756           |
| 0.07       | 8.01             | 0.1427           | 9.944            | 0.598            | 0.4464           |
| 0.08       | 7.83             | 1.1585           | 10.077           | 0.5912           | 0.5139           |
| 0.09       | 7.63             | 1.1584           | 10.227           | 0.5836           | 0.5847           |
| 0.10       | 7.36             | 1.1024           | 10.471           | 0.5732           | 0.6578           |
| 318.15 K   |                   |                  |                  |                  |                  |
| 0.00       | 9.26             | –                | 9.008            | 0.6487           | –                |
| 0.01       | 9.11             | 1.0140           | 9.114            | 0.6434           | 0.5008           |
| 0.02       | 8.96             | 1.0398           | 9.220            | 0.6381           | 0.5069           |
| 0.03       | 8.75             | 1.0294           | 9.348            | 0.6305           | 0.6677           |
| 0.04       | 8.54             | 1.0524           | 9.498            | 0.6229           | 0.7095           |
| 0.05       | 8.30             | 1.0822           | 9.678            | 0.6141           | 0.7679           |
| 0.06       | 8.13             | 1.1340           | 9.837            | 0.6441           | 0.7234           |
| 0.07       | 7.98             | 1.1819           | 9.950            | 0.6022           | 0.7154           |
| 0.08       | 7.82             | 1.1847           | 10.08            | 0.5961           | 0.7303           |
| 0.09       | 7.72             | 1.2100           | 10.16            | 0.5923           | 0.7048           |
| 0.1        | 7.51             | 1.2420           | 10.32            | 0.5842           | 0.7525           |
| 323.15 K   |                   |                  |                  |                  |                  |
| 0.00       | 9.68             | –                | 8.793            | 0.6690           | –                |
| 0.01       | 9.53             | 1.0090           | 8.885            | 0.6638           | 0.5110           |
| 0.02       | 9.41             | 1.0340           | 8.974            | 0.6597           | 0.4258           |
| 0.03       | 9.24             | 1.0523           | 9.071            | 0.6537           | 0.5110           |
| 0.04       | 8.98             | 1.0705           | 9.246            | 0.6444           | 0.6388           |
| 0.05       | 8.72             | 1.1021           | 9.415            | 0.6350           | 0.7495           |
| 0.06       | 8.40             | 1.1087           | 9.628            | 0.6232           | 0.8801           |
| 0.07       | 8.22             | 1.1071           | 9.777            | 0.6165           | 0.8517           |
| 0.08       | 7.99             | 1.1363           | 9.938            | 0.6078           | 0.8943           |
| 0.09       | 7.90             | 1.1824           | 10.02            | 0.6044           | 0.8328           |
| 0.10       | 7.68             | 1.2204           | 10.19            | 0.5959           | 0.8688           |

Table 4. (Continued)
Table 5. Isentropic compressibility, relative association, acoustic impedance, linear length and sound velocity number for 2-amino-5-nitrothiazole + acetonitrile at different temperature and different concentrations

| C (mol/l) | $\beta_i \times 10^{-10}$ (m$^2$ N$^{-1}$) | $R_A$ | $Z \times 10^{-3}$ (kg m$^2$ s$^{-1}$) | $L_f$ (Å$^2$) | $[U]$ (kg mol$^{-1}$) |
|-----------|-----------------------------------------|-------|---------------------------------|----------------|-----------------|
| 303.15 K  |                                         |       |                                 |                |                 |
| 0.00      | 9.99                                    | -     | 8.8225                          | 0.5602         | -               |
| 0.01      | 9.94                                    | 1.0181| 8.8494                          | 0.6543         | 0.1763          |
| 0.02      | 9.84                                    | 1.0409| 8.9219                          | 0.6510         | 0.1763          |
| 0.03      | 9.62                                    | 1.0632| 9.0648                          | 0.6437         | 0.3527          |
| 0.04      | 9.49                                    | 1.0760| 9.1468                          | 0.6393         | 0.3968          |
| 0.05      | 9.23                                    | 1.1012| 9.3060                          | 0.6305         | 0.5291          |
| 0.06      | 8.92                                    | 1.0963| 9.7420                          | 0.6198         | 0.6466          |
| 0.07      | 8.62                                    | 1.1266| 9.7267                          | 0.6093         | 0.7306          |
| 0.08      | 8.48                                    | 1.1639| 9.8355                          | 0.6044         | 0.7054          |
| 0.09      | 8.26                                    | 1.2056| 10.033                          | 0.5965         | 0.7045          |
| 0.10      | 8.05                                    | 1.2355| 10.255                          | 0.5888         | 0.7407          |
| 308.15 K  |                                         |       |                                 |                |                 |
| 0.00      | 10.3                                    | -     | 8.649                           | 0.6721         | -               |
| 0.01      | 10.2                                    | 1.0152| 8.729                           | 0.6688         | 0.5376          |
| 0.02      | 10.0                                    | 1.0371| 8.805                           | 0.6649         | 0.4480          |
| 0.03      | 9.85                                    | 1.0681| 8.947                           | 0.6572         | 0.5376          |
| 0.04      | 9.87                                    | 1.0948| 8.96                            | 0.6579         | 0.3136          |
| 0.05      | 9.56                                    | 1.1231| 9.122                           | 0.6475         | 0.5376          |
| 0.06      | 9.33                                    | 1.1495| 9.252                           | 0.6397         | 0.6272          |
| 0.07      | 9.10                                    | 1.1460| 9.421                           | 0.6317         | 0.6400          |
| 0.08      | 8.82                                    | 1.1834| 9.586                           | 0.6219         | 0.6272          |
| 0.09      | 8.58                                    | 1.2278| 9.801                           | 0.6134         | 0.7168          |
| 0.10      | 8.35                                    | 1.2890| 10.01                           | 0.6051         | 0.7168          |
| 313.15 K  |                                         |       |                                 |                |                 |
| 0.00      | 10.70                                   | -     | 8.461                           | 0.9912         | -               |
| 0.01      | 10.60                                   | 1.0231| 8.520                           | 0.6879         | 0.1824          |
| 0.02      | 10.40                                   | 1.0186| 8.615                           | 0.6814         | 0.4562          |
| 0.03      | 10.20                                   | 1.0494| 8.744                           | 0.6748         | 0.5474          |
| 0.04      | 10.03                                   | 1.0533| 8.868                           | 0.6692         | 0.6386          |
| 0.05      | 9.82                                    | 1.0110| 8.988                           | 0.6621         | 0.6569          |
| 0.06      | 9.57                                    | 1.1627| 9.083                           | 0.6536         | 0.7290          |
| 0.07      | 9.28                                    | 1.2182| 9.298                           | 0.6437         | 0.8081          |
| 0.08      | 9.12                                    | 1.2448| 9.416                           | 0.6381         | 0.7755          |
| 0.09      | 8.86                                    | 1.2867| 9.596                           | 0.6289         | 0.8110          |
| 0.10      | 8.55                                    | 1.3410| 9.836                           | 0.6178         | 0.8394          |
| 318.15 K  |                                         |       |                                 |                |                 |
| 0.00      | 11.2                                    | -     | 8.248                           | 0.7134         | -               |
| 0.01      | 11.1                                    | 1.0202| 8.311                           | 0.7121         | 0.3724          |
| 0.02      | 10.9                                    | 1.0364| 8.411                           | 0.7038         | 0.4655          |
| 0.03      | 10.8                                    | 1.0681| 8.497                           | 0.7005         | 0.4345          |
| 0.04      | 10.5                                    | 1.0622| 8.614                           | 0.6907         | 0.5121          |

(Continued)
Table 5. (Continued)

| C (mol/l) | $\beta_x \times 10^{-10}$ (m² N⁻¹) | $R_A$ | $Z \times 10^{-3}$ (kg m² s⁻¹) | $L_f$ (Å) | $[U]$ (kg mol⁻¹) |
|----------|----------------------------------|------|--------------------------------|---------|-----------------|
| 0.05     | 10.4                             | 1.1121 | 8.726                          | 0.6874  | 0.5959          |
| 0.06     | 10.2                             | 1.1518 | 8.767                          | 0.6808  | 0.6207          |
| 0.07     | 10.0                             | 1.1647 | 8.908                          | 0.9741  | 0.6384          |
| 0.08     | 9.69                             | 1.2224 | 9.076                          | 0.6636  | 0.7216          |
| 0.09     | 9.50                             | 1.2844 | 9.215                          | 0.6570  | 0.7034          |
| 0.10     | 9.14                             | 1.3293 | 9.456                          | 0.6444  | 0.7635          |
| 323.15 K |                                  |       |                                |         |                 |
| 0.00     | 11.6                             | -     | 8.113                          | 0.7324  | -               |
| 0.01     | 11.4                             | 1.0108 | 8.202                          | 0.7261  | 0.5649          |
| 0.02     | 11.2                             | 1.0370 | 8.312                          | 0.7197  | 0.5649          |
| 0.03     | 11.1                             | 1.0678 | 8.350                          | 0.7164  | 0.3138          |
| 0.04     | 10.9                             | 1.1026 | 8.456                          | 0.7100  | 0.4237          |
| 0.05     | 10.6                             | 1.1334 | 8.611                          | 0.7001  | 0.5273          |
| 0.06     | 10.5                             | 1.1755 | 8.675                          | 0.6968  | 0.5021          |
| 0.07     | 10.4                             | 1.2270 | 8.786                          | 0.6935  | 0.5380          |
| 0.08     | 10.0                             | 1.2556 | 8.894                          | 0.6824  | 0.6355          |
| 0.09     | 9.76                             | 1.3059 | 9.080                          | 0.6718  | 0.6905          |
| 0.10     | 9.41                             | 1.3661 | 9.295                          | 0.6597  | 0.7312          |

Figure 1. Density ($\rho$) plotted against concentration of 2-amino-5-nitrothiazole in binary mixture of 2-amino-5-nitrothiazole + NNDMF (■), 2-amino-5-nitrothiazole + acetonitrile (▲), and 2-amino-5-nitrothiazole + ethanol (×) at 303.15 K.

Figure 2. Ultrasonic velocity ($u$) plotted against concentration of 2-amino-5-nitrothiazole in binary mixture of 2-amino-5-nitrothiazole + NNDMF (■), 2-amino-5-nitrothiazole + acetonitrile (▲), and 2-amino-5-nitrothiazole + ethanol (×) at 303.15 K.
Figure 3. Ultrasonic velocity ($u$) plotted against concentration of 2-amino-5-nitrothiazole in binary mixture of 2-amino-5-nitrothiazole + NNDMF at temperatures 303.15 K (■), 308.15 K (▲), 313.15 K (+), 318.15 K (+), and 323.15 K (●).

Figure 4. Ultrasonic velocity ($u$) plotted against concentration of 2-amino-5-nitrothiazole in binary mixture of 2-amino-5-nitrothiazole + Acetonitrile at temperatures 303.15 K (■), 308.15 K (▲), 313.15 K (+), 318.15 K (+), and 323.15 K (●).

Figure 5. Ultrasonic velocity ($u$) plotted against concentration of 2-amino-5-nitrothiazole in binary mixture of 2-amino-5-nitrothiazole + Ethanol at temperatures 303.15 K (■), 308.15 K (▲), 313.15 K (+), 318.15 K (+), and 323.15 K (●).
Figure 6. Isentropic compressibility plotted against concentration of 2-amino-5-nitrothiazole in binary mixture of 2-amino-5-nitrothiazole + NNDMF at temperatures 303.15 K (■), 308.15 K (▲), 313.15 K (+), 318.15 K (+), and 323.15 K (●).

Figure 7. Isentropic compressibility plotted against concentration of 2-amino-5-nitrothiazole in binary mixture of 2-amino-5-nitrothiazole + Acetonitrile at temperatures 303.15 K (♦), 308.15 K (■), 313.15 K (▲), 318.15 K (+), and 323.15 K (●).

Figure 8. Isentropic compressibility plotted against concentration of 2-amino-5-nitrothiazole in binary mixture of 2-amino-5-nitrothiazole + Ethanol at temperatures 303.15 K (■), 308.15 K (▲), 313.15 K (+), 318.15 K (+), and 323.15 K (●).
Figure 9. Specific acoustic impedance plotted against concentration of 2-amino-5-nitrothiazole in binary mixture of 2-amino-5-nitrothiazole + NNDMF at temperatures 303.15 K ( ), 308.15 K ( ▲ ), 313.15 K (+), 318.15 K (+), and 323.15 K ( ● ).

Figure 10. Specific acoustic impedance plotted against concentration of 2-amino-5-nitrothiazole in binary mixture of 2-amino-5-nitrothiazole + acetonitrile at temperatures 303.15 K ( ), 308.15 K ( ▲ ), 313.15 K (+), 318.15 K (+), and 323.15 K ( ● ).

Figure 11. Specific acoustic impedance plotted against concentration of 2-amino-5-nitrothiazole in binary mixture of 2-amino-5-nitrothiazole + ethanol at temperatures 303.15 K ( ), 308.15 K ( ▲ ), 313.15 K (+), 318.15 K (+), and 323.15 K ( ● ).
Figure 12. Relative association ($R_A$) plotted against concentration of 2-amino-5-nitrothiazole in binary mixture of 2-amino-5-nitrothiazole + NNDMF at temperatures 303.15 K (■), 308.15 K (▲), 313.15 K (+), 318.15 K (+), and 323.15 K (●).

Figure 13. Relative association ($R_A$) plotted against concentration of 2-amino-5-nitrothiazole in binary mixture of 2-amino-5-nitrothiazole + acetonitrile at temperatures 303.15 K (■), 308.15 K (▲), 313.15 K (+), 318.15 K (+), and 323.15 K (●).

Figure 14. Relative association ($R_A$) plotted against concentration of 2-amino-5-nitrothiazole in binary mixture of 2-amino-5-nitrothiazole + ethanol at temperatures 303.15 K (■), 308.15 K (▲), 313.15 K (+), 318.15 K (+), and 323.15 K.
acoustic impedance ($Z$) decreases (Landge et al., 2013) as acoustic impedance ($Z$) is the product of ultrasonic velocity ($u$) and density ($\rho$). Also, in this present investigation, it is observed that these acoustic impedance ($Z$) value increases with increase in concentration of 2-amino-5-nitrothiazole in acetonitrile, ethanol, NNDMF solutions, respectively. The linear variation in acoustic impedance with concentration confirms the presence of molecular association between the solute–solvent molecules. Such an increasing trends of impedance further support the possibility of molecular interaction between the solute–solvent. Figures 12–14 shows that the relative association ($R_u$) increases with increase in temperature and the increase in concentration of solution. This is due to the solute–solvent interaction that dominates over solvent–solvent interactions. It depends on either the breaking up of the solvent molecules on addition of solute molecules in solvent at certain temperature or the solvation of ions that are present (Ambomase, Tripathy, Tripathy, & Dash, 2011; Meshram, Agrawal, Chandak, & Chapke, 2013). In general, sound velocity number increases with increase in concentration of solute (Chauhan et al., 2013) and increase in temperature, however in present investigation, there is no regular variation observed in sound velocity number.

5. Conclusions

The ultrasonic method is a powerful tool for characterizing physicochemical properties and existence of molecular interaction in the mixture. The result reveals that the density and ultrasonic velocity of 2-amino-5-nitrothiazole + NNDMF, 2-amino-5-nitrothiazole + Acetonitrile, and 2-amino-5-nitrothiazole + ethanol solutions decrease with increase in temperature. It is also seen that the formation of molecular interaction in the mixture. The result reveals that the density and ultrasonic velocity of 2-amino-5-nitrothiazole + NNDMF, 2-amino-5-nitrothiazole + Acetonitrile, and 2-amino-5-nitrothiazole + ethanol solutions decrease with increase in temperature. It is also seen that the formation of molecular interaction in the mixture.
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