Acoustical studies of ternary liquid mixtures of 2-aminothiazole in DMF-Water at different temperatures

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Keywords: Thermo-acoustic, 2-aminothiazole, ultrasonic velocity, density, adiabatic compressibility, ternary mixture, DMF, molecular interaction.

ABSTRACT. The nature and the relative strength of the intermolecular interaction between the components of the liquid mixtures have been successfully investigated by ultrasonic method. In present study, the densities (ρ), ultrasonic velocities (u), viscosity(η) and refractive index(n₀) in a ternary liquid mixture of 2-aminothiazole with N,N-dimethylformamide (DMF) in water have been measured at 303.15, 308.15, 313.15,318.15 and 323.15 K respectively, over the entire composition range by using densitometer, ultrasonic interferometer, viscometer and refractometer respectively. The measured data have been used to compute the various thermo-acoustic parameters using the standard relations namely, adiabatic compressibility (β), intermolecular free length (Lᵢ), specific acoustic Impedance (Z),Wada constant (W), molar sound velocity(R), relative association (Rₖ), apparent molar compressibility(Φₚ),apparent molar volume(Φᵥ) viscosity relaxation time(Γ), absorption coefficient, internal pressure (n),free volume (Vᵢ),Gibb's free energy(∆G) and specific refraction (r), etc. The results have been analyzed on the basis of variation in thermodynamic parameters. These parameters are useful for explaining the molecular association and interaction between the components of ternary liquid mixtures. The variation in densities and ultrasonic velocities with concentrations in the system show similar trends for evaluated parameters of the constituents in ternary mixture at different temperatures. The results have been interpreted in terms of solute-solvent and solvent-solvent interaction.

1. INTRODUCTION

In the historical development of organic chemistry, nitrogen and sulphur heterocyclic compounds have maintained the interest of researchers in the last decades, some most common nitrogen heterocycles are thiazoles and thiadiazoles. The wide spread applications of thiazole in the agrochemical industries and medicinal chemistry proves this moiety is an important bioactive class of heterocycles⁵. Different methods like di-electric, magnetic resonance, infrared and Raman effect are used to study molecular interaction and different solution properties. Likewise, ultrasonic method has been extensively used by many workers to study the molecular interactions and physicochemical behavior in liquid mixtures⁵.⁶.⁷. The study of thermodynamic properties of mixtures provides good measure of solute –solvent interactions. The experimental data of thermo acoustical properties of liquid and liquid mixtures are fascinating and highly fundamental and practically important in chemical industry and engineering design⁸.⁹. In continuation of our research work, present study reported the results of ultrasonic study of the ternary mixture of 2-aminothiazole with DMF-water solvent in entire composition range, at five different temperatures. 2-aminothiazole is used as an intermediate for dyestuff, in photographic chemicals, in medicinal chemistry and its derivatives can find application for treatment gastric ulcer and cancer etc⁵. DMF is an aprotic protophilic and polar solvent used widely in industrial processes, including manufacturing of synthetic fibers, leather, films and in surface coating⁶.⁷. DMF is of particular interest solvents because any significant structural affects are absent due to lack of hydrogen bond. DMF is a stable compound having strong electron-pair donating and accepting capacity, It has large dipole moment and high dielectric constant (μ=3.24 Debye and ε=36.71at 298K)⁹. It is widely used in studies of solvent reactivity relationships¹⁰.¹¹. An exhaustive survey of literature has shown that a few attempts
have been made for ultrasonic velocity data for ternary liquid mixtures in DMF solvent. However, no effort have been made for the ultrasonic and thermodynamic studies for ternary mixture of 2-aminothiazole with DMF in water. The present study was undertaken in order to have deeper understanding of the intermolecular interaction between the components of ternary liquid mixtures. Using the experimental values of ultrasonic velocity (u), density (ρ), viscosity(η) various thermo acoustical parameters such adiabatic compressibility (βs), Intermolecular free length (Lf), Specific acoustic Impedance (Z),Wada constant (W), molar sound velocity(R), relative association (RA), apparent molar compressibility (Φβ), apparent molar volume ( Φυ) viscosity relaxation time(Γ), absorption coefficient, internal pressure (π),free volume (Vf),Gibb’s free energy(ΔG) and specific refraction (r), have been estimated using standard relations. These thermo-acoustic parameters of pure components and mixtures are being used to investigate the molecular packing, molecular motions and various intermolecular interactions and their strength, influenced by the size in pure components and in the mixtures. The results are interpreted in predicting nature and strength of molecular association between the components of the liquid mixtures.

2. MATERIALS AND METHODS

Materials: In present study, used solute 2-aminothiazole (Hi-Media) and solvent DMF( Fisher Scientific) were analytical (AR) and spectroscopic reagent (SR) grade. They were used without further purification.

Method: The mass of sample was measured using digital electronic balance (Model SHIMADZU AUY-220, Japan,) with precision of ±0.1 mg. The required ternary mixtures were prepared over the entire range of compositions in DMF-water solvent and kept in air tight flask. The densities of pure liquids and ternary mixtures were measured with portable digital densitometer (Anton Paar, DMA-35, Austria). The average uncertainty in measurement in the measured density is ±5 x 10^{-3} kg m^{-3}. The ultrasonic velocity of pure and liquid mixtures was measured using multi-frequency ultrasonic interferometer (Model: F-81S, Mittal Enterprises, New Delhi,) operating at 2 MHz’s frequency at five different temperatures and overall accuracy of ±2 m/s. The instruments was calibrated by measuring the velocity of benzene and carbon tetrachloride. In the present work, Ostwald viscometer (10 ml) is used to measure the viscosity of pure liquid and mixtures and efflux time was measured using a RACER digital chronometer within a precision of ±0.01 sec. R.I. was measured with Abbe refractometer (Atago DR-A-1 Japan). The measured values are agreed closely with literature values.

The density (ρ), ultrasonic velocity (u) viscosity (η) and R.I. of pure liquids and ternary mixtures were measured at 303.15, 308.15, 313.15, 318.15 and 323.15K temperatures. The temperature was controlled through the water circulating around the liquid cell using thermostatically controlled High Precision water bath MSW-274 (Macro scientific work pvt. Ltd. Delhi) with an uncertainty of ±0.3°C.

3. THEORY AND CALCULATIONS:

The data of density (ρ), ultrasonic velocity (u) viscosity (η) and R.I. has been used to evaluate many acoustical parameters by using the following standard expressions for understanding solute-solvent, solvent-solvent interaction and structural changes.

(i) Adiabatic compressibility:- \[ \beta_s = \frac{1}{\rho u_s^2} \]  (1)

Where \( \rho_s \) = density of solution, \( u_s \) = sound velocity.

(ii) Intermolecular free length (Lf) :- \[ L_f = K\sqrt{\beta_s} \]  (2)

Where ‘K’ is a temperature dependent constant known as Jacobson constant\(^{(m)}\).

(iii) Specific acoustic impedance(Z):- \[ Z = u_\rho \]  (3)
Wada’s constant \( W = \left( \frac{M}{\rho_r} \right) \beta^\frac{1}{2} \) \hspace{1cm} (4)

Where ‘M’ molar mass of the solution.

Molar sound velocity or Rao’s constant \( R = \left( \frac{M}{\rho_r} \right) u_r \frac{1}{2} \) \hspace{1cm} (5)

Relative association \( R_d = \left( \frac{d_u}{d_o} \right) \frac{u_o}{u_s} \) \hspace{1cm} (6)

Where \( \rho_o = \) density of solvent, \( u_o = \) velocity of solvent

Apparent molar compressibility \( \phi = \frac{1000}{m \rho_o} (\rho_o - \rho) + \left( \frac{\beta M}{\rho_o} \right) \) \hspace{1cm} (7)

Apparent molar volume \( \phi_v = \frac{1000}{m \rho_o} (\rho_o - \rho) + \left( \frac{M}{\rho_o} \right) \) \hspace{1cm} (8)

Relative viscosity \( \eta_r = \left[ \frac{d_o d_u}{d_o d_w} \right] \frac{u_o}{u_w} \) \hspace{1cm} (9)

Viscosity relaxation time \( \Gamma = \frac{4 \eta}{3 \rho u^2} \) \hspace{1cm} (10)

Absorption coefficient \( \frac{a}{f^2} = \frac{8 \pi^2 \eta}{3 \rho \times u^2} \) \hspace{1cm} (11)

Internal pressure \( \pi = bRT \left( \frac{K \eta}{u} \right) \frac{1}{2} \left( \frac{\rho^\frac{2\gamma}{3}}{M^\frac{2\gamma}{3}} \right) \) \hspace{1cm} (12)

Free volume \( V_f = \left( \frac{M_{eff} u}{K \eta} \right)^\frac{1}{2} \) \hspace{1cm} (13)

‘k’ is temperature independent constant equal to 4.28x10^9 for all liquids.

Gibb’s free energy \( \Delta G \) was calculated from the relation;

\[ \Delta G = KT \log \left( \frac{K.T.\Gamma}{\hbar} \right) \] \hspace{1cm} (14)

‘K’ Boltzmann constant, ‘\( \hbar \)’ is Planck constant and \( \Gamma \) relaxation time.

The specific refractive is defined and calculated by the following equation;

\[ r = \frac{n^2 - 1}{n^2 + \frac{1}{\rho}} \] \hspace{1cm} (15)

5. RESULTS AND DISCUSSION

The experimental values of density, ultrasonic velocity, viscosity and R.I. for 2-amino thiazole with DMF in water are presented in Table 1. The some of the calculated thermo-acoustics parameters are given in Table 2-3. In order to understand reaction kinetics of ternary mixture, tabulated values of thermo-acoustic parameters are graphically represented in Figures 1-17.
Table 1 Density, velocity, viscosity, refractive index of 2-aminothiazole with DMF in water at 303.15, 308.15, 313.15, 318.15 and 323.15 K.

| Conc. | \( \rho \) kg m\(^{-3}\) | \( u \) m s\(^{-1}\) | \( \eta \times 10^{-3} \) Nsm\(^{-2}\) | \( n_D \) |
|-------|----------------|----------------|------------------|-------|
| T=303.15K | | | | |
| 0.000 | 985.47 | 1627.6 | 1.7451 | 1.4073 |
| 0.002 | 985.9 | 1629.6 | 1.8523 | 1.4071 |
| 0.004 | 986.5 | 1631.4 | 1.7891 | 1.4072 |
| 0.006 | 987.4 | 1633.8 | 1.7556 | 1.4072 |
| 0.008 | 987.9 | 1635.3 | 1.7262 | 1.4075 |
| 0.01 | 988.6 | 1636.8 | 1.6806 | 1.4077 |
| T=308.15K | | | | |
| 0.000 | 979.9 | 1605.5 | 1.69 | 1.4054 |
| 0.002 | 981.9 | 1610.3 | 1.73 | 1.4052 |
| 0.004 | 982.92 | 1612.8 | 1.71 | 1.4055 |
| 0.006 | 983.58 | 1614.9 | 1.67 | 1.4056 |
| 0.008 | 984.31 | 1617.2 | 1.61 | 1.4056 |
| 0.01 | 984.96 | 1618.9 | 1.58 | 1.4057 |
| T=313.15K | | | | |
| 0.000 | 974.78 | 1580.6 | 1.4443 | 1.404 |
| 0.002 | 975.97 | 1583.2 | 1.4943 | 1.4035 |
| 0.004 | 976.92 | 1587.2 | 1.4796 | 1.4037 |
| 0.006 | 977.65 | 1591.6 | 1.4478 | 1.4038 |
| 0.008 | 978.32 | 1595.6 | 1.4158 | 1.4039 |
| 0.01 | 978.89 | 1598.8 | 1.3684 | 1.4044 |
| T=318.15K | | | | |
| 0.000 | 968.8 | 1576.5 | 1.2006 | 1.4021 |
| 0.002 | 969.89 | 1579.6 | 1.2621 | 1.4018 |
| 0.004 | 970.93 | 1584.2 | 1.2626 | 1.4019 |
| 0.006 | 971.78 | 1586.3 | 1.2333 | 1.4021 |
| 0.008 | 972.4 | 1587.6 | 1.1888 | 1.4022 |
| 0.01 | 972.97 | 1588.7 | 1.1295 | 1.4023 |
| T=323.15K | | | | |
| 0.000 | 964.27 | 1569.6 | 1.1351 | 1.4008 |
| 0.002 | 964.87 | 1577.2 | 1.1661 | 1.3993 |
| 0.004 | 965.93 | 1580.6 | 1.1512 | 1.3994 |
| 0.006 | 966.89 | 1582 | 1.1221 | 1.3996 |
| 0.008 | 967.98 | 1585.6 | 1.0782 | 1.3997 |
| 0.01 | 968.84 | 1586.6 | 1.049 | 1.4001 |

Table 2 Adiabatic compressibility, free length, acoustic impedance, Wada const., molar sound velocity, relative association and app. mol. compressibility of 2-aminothiazole with DMF in water at different temperatures.

| Conc. | \( \beta_s \times 10^{30} \) N\(^{-1}\) m\(^2\) | \( L_x \times 10^{-13} \) m | \( Z \times 10^6 \) kgm\(^{-2}\)s\(^{-2}\) | \( W \times 10^{1} \) mPa\(^{-1}\)s\(^{2}\)mol\(^{-1}\) | \( R \) (m\(^3\))mol\(^{-1}\) | \( R_A \) | \( \Phi \) |
|-------|-------------|----------------|----------------|------------------|--------|--------|------|
| T=303.15K | | | | | | | |
| 0.000 | 3.8306 | 4.0611 | 1.6040 | 9.8988 | 0.5258 | 1.0000 | - |
| 0.002 | 3.8195 | 4.0553 | 1.6066 | 9.8985 | 0.5258 | 1.0000 | -6.3941x10\(^{-1}\) |
| 0.004 | 3.8087 | 4.0496 | 1.6094 | 9.8965 | 0.5257 | 1.0003 | -3.6901x10\(^{-1}\) |
| 0.006 | 3.7941 | 4.0418 | 1.6132 | 9.8929 | 0.5255 | 1.0007 | -3.6837x10\(^{-1}\) |
| 0.008 | 3.7852 | 4.0371 | 1.6155 | 9.8912 | 0.5254 | 1.0009 | -2.2741x10\(^{-1}\) |
| 0.010 | 3.7756 | 4.0319 | 1.6181 | 9.8878 | 0.5252 | 1.0013 | -2.1601x10\(^{6}\) |
| T=308.15K | | | | | | | |

International Letters of Chemistry, Physics and Astronomy Vol. 59 191
| Conc. | $\Phi_V \times 10^{10}$ m$^3$/mol$^{-1}$ | $\text{Abs}_{	ext{r}x\text{p}(\omega)} \times 10^{-10}$ Np.m$^{-1}$ | $\Gamma_{10}$ x 10$^3$ Nm$^{-2}$ | $V_i \times 10^6$ m$^3$/mol$^{-1}$ | X10$^{-10}$ | rx10$^{-4}$ |
|-------|---------------------------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| 0.00  | -                              | 3.5610 x10$^{-11}$ | 4.2916          | 2.8830          | 2.6476          | 2.3438          |
| 0.01  | 1.2857                         | 1.2703           | 1.6506          | 1.5698          | 2.4996          | 2.4974          |
| 0.00  | -1.5946                        | 1.6013           | 1.5691          | 2.4974          | 2.4996          | 2.4974          |
| 0.01  | -1.6506                        | 1.5698           | 2.4996          | 2.4974          | 2.4996          | 2.4974          |
| 0.00  | -1.5988                        | 1.6013           | 1.5691          | 2.4974          | 2.4996          | 2.4974          |
| 0.01  | -1.6506                        | 1.5698           | 2.4996          | 2.4974          | 2.4996          | 2.4974          |
| 0.00  | -1.5946                        | 1.6013           | 1.5691          | 2.4974          | 2.4996          | 2.4974          |
| 0.01  | -1.6506                        | 1.5698           | 2.4996          | 2.4974          | 2.4996          | 2.4974          |
| 0.00  | -1.5988                        | 1.6013           | 1.5691          | 2.4974          | 2.4996          | 2.4974          |
| 0.01  | -1.6506                        | 1.5698           | 2.4996          | 2.4974          | 2.4996          | 2.4974          |
| 0.00  | -1.5946                        | 1.6013           | 1.5691          | 2.4974          | 2.4996          | 2.4974          |
| 0.01  | -1.6506                        | 1.5698           | 2.4996          | 2.4974          | 2.4996          | 2.4974          |

Table 3 app. Molar volume, viscosity relaxation time, Abs. coefficient, internal pressure, free volume, Gibb’s free energy and specific refraction of 2-aminothiophene with DMF in water at different temperatures.
The specific acoustic impedance is the parameter related to the elastic properties of the medium. The specific acoustic impedance is the impedance offered to the sound wave by the components of the mixture. In our present investigation (Fig.6), specific acoustic impedance increase with increase in concentration. This trend further supports the possibility of molecular interaction due to H-bonding between solute-solvents and solvent-solvent molecules which restrict the free flow of sound waves. The specific acoustic impedance is directly proportional to density, ultrasonic velocity and inversely proportional adiabatic compressibility.

From Fig. 7-8, the molar compressibility and Molar sound velocity nonlinearly decreases with increase in concentration which indicates that the magnitude of molecular interaction is enhanced in the system, which indicate interaction between solute-solvent molecule increases. This leads to tight packing of the medium by increases the molecular interactions.
Relative association is the measure of extent of association of components in the medium. The relative association is depends on either breaking up of the solvent molecules on addition of solute to it or the salvation of present ions. From Fig. 9 the relative association increases with increase in concentration. The increasing trend indicates there is a salvation of present solute ions.

From Fig.10-11, the apparent molar compressibility and apparent molar volume nonlinearly decreases and increases with increase in concentration which indicate interaction between solute-solvent molecules enhanced. Some values are positive due to the compressibility of solvent due to the weak electrostatic force in the vicinity of ions. This trends supports that the availability of more number of components in a given regions of space. This leads to tight packing of the medium and there by increases the interactions.

The viscosity relaxation time is the time required for the excitation energy to appear as translational energy. In present work (fig.12) viscosity relaxation time non-linearly varies with increase in molar concentration and decreases with increases in temperature. Where, with increase in temperature, it shows the instantaneous conversion of excitation energy to translational energy. This indicates strong molecular interaction between the solute and solvent molecules, where it show the instantaneous conversion of excitation energy to translational energy.

From table 3 and fig.13 absorption coefficient decreases with increase in concentration and this trend suggest that the extent of complexity decreases with increase in concentration.

The internal pressure is a measure of cohesive forces between the constituent’s molecules in liquids. The internal pressure is an inverse function of free volume. The internal pressure for a given system (fig.14) decreases with increase in concentration of solute, which indicate decrease in London force (cohesive forces) which leading to breaking the structure of solute. This suggests there is a weak interaction between the solute and solvent molecules or there is a decrease in the extent of complexation with increase in concentration.

The free volume increases (fig.15) with increase in concentration of solute and temperature. This increasing trend is due to stronger intramolecular interaction than intermolecular interaction which attribute to lose packing of molecules inside the shield, this suggest weak molecular interaction in components of mixtures.

The Gibbs free energy (fig.16) decrease with increase in molar concentrations of 2-aminothiazole and increases with increase in temperatures. The increase in Gibbs free energy ($\Delta G$) with temperature suggests longer time for rearrangement of molecules in the mixture. The decreasing positive values of Gibbs free energy ($\Delta G$) suggest the molecular dissociations.

Specific refraction depends on molecular weight and nature of liquids. The specific refraction fig.17 decreases with increase in molar concentration and with increase in temperatures. That means inter molecular interactions among the components are very weak.
Figure 3. Viscosity Vs Concentration

Figure 4. Adiab. Comp. Vs Concentration

Figure 5. Free length Vs Concentration

Figure 6. Acoustic impedence Vs Concentration

Figure 7. Molar compr. Vs Concentration

Figure 8. Mol. S. velocity Vs Concentration
Figure 9. Rel. asso. Vs Concentration

Figure 10. App.mol.compr. Vs Concentration

Figure 11. App.mol.vol Vs Concentration

Figure 12. Visco.relx.time Vs Concentration

Figure 13. Abs. coeff. Vs Concentration

Figure 14. Int. pressure Vs Concentration
6. CONCLUSIONS

In the present investigation experimental values of density, ultrasonic velocity, viscosity and R.I. and related acoustic parameter values indicate that thermodynamic parameters are sensitive to molecular interactions for ternary liquid mixtures at different concentrations and at varying temperatures. Thus it is conclude that in mixture of studied compound, both solute-solute and solute-solvent interaction is existing. Some parameters specially, free length and adiabatic compressibility indicate strong interaction between solute-solvent molecules in the studied system.

Acknowledgments:
The author thanks Head, Department of Chemical Technology, SGBA University, Amravati, India, for providing necessary facilities and also thanks Principal Dr. G. P. Patil, Vidya Bharti College, Karanja (Lad) for their constant support.

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