Acoustic and solvation behaviour of potassium thiocyanate in mixed solvents

J Ishwara Bhat & H R Shiva Kumar

Department of Chemistry, Mangalore University, Mangalagangothri 574 199

Department of Chemistry, KVG College of Engineering, Sullia 574 239

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Measurement of ultrasonic velocity and density at different concentrations of potassium thiocyanate in water, dimethylsulphoxide (DMSO), dimethylformamide (DMF) and their mixtures have been investigated at 298K using ultrasonic interferometer at frequency 2MHz. These data are used to evaluate adiabatic compressibility ($\beta_{ad}$), intermolecular free length ($L_o$), specific acoustic impedance ($Z$), relative association ($R_A$), solvation number ($S_e$), apparent molar compressibility ($\phi_a$) and molar volume ($V_o$).

These results are used to calculate limiting molar compressibility ($\phi_{\infty}$) and molar volume ($V_{\infty}$). Gucker and Mason’s relations have been verified. Formation of complex at 60 % DMSO and DMF has been identified in the system. These data are utilised in the qualitative study of ion-ion and ion-solvent interaction.

1 Introduction

Studies on acoustic parameters in mixed solvents have been widely used to investigate the process of ion-ion and ion-solvent interaction. Potassium thiocyanate (KSCN) is water soluble, highly stable and finds a number of industrial, analytical and biological applications. In the present investigation the study of acoustic velocity and density of potassium thiocyanate in water-DMSO and water-DMF systems at 298K has been undertaken.

2 Experimental Details

Potassium thiocyanate (Ranbaxy-AR) was dried at 100-120°C for 3 hrs and stored in vacuum desiccator. Doubly distilled water and purified DMSO and DMF were used in the present work. Solution of potassium thiocyanate (1.0M) was prepared in different compositions of water-DMSO and water-DMF mixtures. The ultrasonic velocity was measured using ultrasonic interferometer [M-81, Mittal Enterprises, New Delhi] with a measuring frequency tolerance ± 0.3% at 25°C. The temperature of the liquid was maintained constant to an accuracy of ±0.01°C by circulating water from a thermostat. The system was calibrated with pure water. The density of the solvent and solutions was measured by using pyknometer to an accuracy of ±0.001 gm.

3 Results and Discussion

Different thermodynamic properties such as adiabatic compressibility, apparent molar compressibility ($\beta_{ad}$), intermolecular free length ($L_o$), specific acoustic impedance ($Z$), relative association ($R_A$) and solvation number ($S_e$) have been calculated for potassium thiocyanate solution in pure and mixed solvents at 298K using ultrasonic velocity and density values. The following equations as given by Nikam et al. were used for the calculations:

$$\beta_{ad} = \frac{1}{U^2 d}$$

$$\phi_a = \frac{1000}{d/d_0} (d_0 \beta_{ad} - d \beta_{ad,d}) + \beta_{ad} \frac{M}{d}$$

$$L_o = K \sqrt{\beta_{ad}}$$

$$Z = U \times d$$

$$R_A = \frac{d}{d_0} \left[ \frac{U_0}{U} \right]^{1/3}$$

$$S_e = \left[ 1 - \frac{\beta_{ad}}{\beta_{ad,d}} \right] \frac{n_1}{n_2}$$

where $d$, $d_0$ and $U$, $U_0$ are the densities and ultrasonic velocities of solution and solvent respectively. $M$ is the molecular weight of the solute, $\beta_{ad}$ and $\beta_{ad,d}$ are the adiabatic compressibilities of solvent and solution, $K$ the Jacobson constant, $n_1$ and $n_2$ are the number of moles of solvent and solute respectively.

Ultrasonic velocities of KSCN in different compositions of DMSO and DMF with water (V/V) have been determined at 298K and from these data adiabatic compressibility was calculated using Eq. (2). These values are presented in Tables 1 and 2. The sound velocity
increase on the addition of co-solvents, reaches a maximum at 60% of co-solvents and then bringing about minima in adiabatic compressibility in the same region.

The decrease in compressibility may be due to the fact that 60% of interstitial spaces of water molecules are filled by the organic co-solvent. It causes increase in density with a later decrease, as there will be lesser association of these solvents. These results are in conformity with the characteristic structural changes associated with gradual addition of DMSO to water and consequent changes in physical properties of DMSO-water mixtures. The addition of DMSO increases the 3D-polymeric structure of water till 60% of DMSO. This indicates the formation of complex at this region and hence there is no change in velocity and compressibility with increase in concentration of potassium thiocyanate at that composition.

The increase in sound velocity and decrease in $\beta_{ad}$ with increase in concentration of KSCN at all compositions of water-DMSO and water-DMF except at 60% is
Table 2 — Experimentally determined ultrasonic velocity ($U$) and adiabatic compressibility ($\beta_{ad}$) at different concentrations ($C$) of KSCN in water-DMF mixtures at 298K.

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due to the solvation of available ions, indicating the presence of strong ion-solvent interaction.

The density increases while $\beta_{ad}$ decreases with increase in concentration of KSCN solution in all the solvent mixtures (Table 3), suggesting a moderate strong electrolytic nature of potassium thiocyanate. These variations represent the measure of ions capacity to attract solvent molecules.

Intermolecular free length ($L_a$) is a predominant factor in determining the variation of ultrasonic velocity in fluids and their solutions. The linear variation in $L_a$ with concentration of KSCN at different compositions of co-solvents is shown in Fig. 1 (A and B). In the present investigation the intermolecular free length is found to decrease with increase in concentration of KSCN at all compositions indicating a significant interaction between ion and solvent molecule, hence the electrolyte may be considered as a structure promoter in the system. The observed increase in the relative association ($K_A$)
with concentration also supports the above observed fact.

The acoustic impedance \( Z \) is calculated for KSCN solutions in different compositions of water-DMSO and water-DMF system. The variation of \( Z \) with concentration of KSCN at different compositions of co-solvent are shown in Fig. 1 (C and D). The increase in \( Z \) with concentration can be explained on the basis of lyophobic interaction between solute and solvent molecules, which increases the intermolecular distance, making relatively wider gap between the molecules, and becoming responsible for the propagation of ultrasonic waves.

The solvation number, \( S_\text{n} \), is calculated by adiabatic compressibility method using Eq. (6) for \( K^+ \) ion in different compositions of solvent mixtures. These are +ve values (Table 4) suggesting an appreciable solvation of ions. This is an added support for the structure promoting nature of the electrolyte.

The apparent molar compressibility \( \phi_k \) was calculated from ultrasonic velocity and density data using Eq. (2) and the values of, \( \phi_k \) have been plotted against \( \sqrt{C} \) in Tables 5 and 6. These values of, \( \phi_k \) decrease linearly with concentration and are best fitted to the equation suggested by Gucker:

\[
\phi_k = \phi_k^0 + S_k \sqrt{C} 
\]

where, \( \phi_k^0 \) is the limiting apparent molar compressibility and \( S_k \) is the experimental slope. The values of \( \phi_k^0 \) and \( S_k \) are presented in Table 7. In the present study, \( \phi_k^0 \) is found to be +ve and \( S_k \) is -ve for all the studied compositions, which indicates the increase in ion-ion interaction with increase in concentration and suggest structure promoting effect of the electrolyte. The, \( \phi_k^0 \) values are found to be maximum at 60% DMSO and DMF indicating the formation of complex at this range.

The apparent molar volume, \( \phi_v \), was calculated in all the cases at 298K using the following equation:

\[
\phi_v = \frac{M}{d} - \frac{1000(D - D_0)}{d_0 d} 
\]

The obtained, \( \phi_v \) values are shown in Tables 5 and 6. The, \( \phi_v \) data were fitted by a method of least squares as suggested by Mason's following equation:

\[
\phi_v = \phi_v^0 + S_v \sqrt{C} 
\]
Table 3 — Experimentally determined values of density ($d$) at different concentrations ($C$) of KSCN in water–DMSO and water–DMF mixtures at 298K

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Water–DMF System

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\[
\phi_v = \phi_v^0 + S_v \sqrt{C}
\]

...(9)

where, $\phi_v^0$ is the limiting apparent molar volume and $S_v$ is the experimental slope.

The values, $\phi_v^0$, and $S_v$ are presented in Table 7. The +ve values of $\phi_v^0$ suggest the presence of strong ion-solvent interaction. The change of partial molar volume with change in compositions of solvent mixtures has been attributed to variable and preferential solvation of the ions by different species present in the solvent system and the type of interaction between solvent molecules. The -ve values of $S_v$ indicates the presence of weak ion-ion interaction in the system. The sharp decrease in $\phi_v^0$ for the initial addition of DMSO may be due to the solvent-solvent interaction which in turn prevent the preferential solvation of cation by DMSO.\(^{11}\)
Table 4 — Experimentally determined solvation number of K⁺ ion by ultrasonic interferometer at different composition of water–DMSO and water–DMF system at 298K

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Table 5 — Calculated values of apparent molar compressibility ($\phi_a$, m³/mol) and apparent molar volume ($\phi_v$, m³/mol) at different concentrations of KSCN in water–DMSO system at 298K

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Table 6 — Calculated values of apparent molar compressibility ($\phi_k \, m^3 \cdot mol^{-1}$) and apparent molar volume ($\phi_v \, m^3 \cdot mol^{-1}$) at different concentrations of KSCN in water-DMF system at 298K.

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Table 7 — Computed values of limiting apparent molar compressibility($\phi_k^o \, m^3 \cdot mol^{-1}$) and limiting apparent molar volume($\phi_v^o \, m^3 \cdot mol^{-1}$) and their slope $S_k$ and $S_v$ for KSCN in water-DMSO and water-DMF system at 298K.

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References: