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Effect of Temperature Change on Ultrasonic Velocity and Some Acoustic Parameters of Ternary Liquid Mixture of Methanol+Ethanol+1-Propanol

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Abstract
The density, viscosity and ultrasonic velocity have been measured for ternary mixture of Methanol+Ethanol+1-Propanol of fixed equal volumes of the components at temperatures of 303 K, 308 K, 313 K, 318 K, 323 K and 328 K. The experimental data have been used to calculate some acoustic and thermodynamic parameters: adiabatic compressibility, free length, free volume, and internal pressure. It was observed that adiabatic compressibility (β), free length (L_f), and free volume (V_f), increased with increase in temperature, whereas internal pressure (π_i) decreased with increase in temperature. Some probable reasons on the increase or decrease of acoustic and thermodynamic parameters with temperature change are presented.

Keywords: Ternary mixtures, Ultrasonic velocity, Acoustic/Thermodynamic Parameters temperature.

1. Introduction

Ultrasonic investigation of liquid mixture containing components is of considerable importance in understanding intermolecular interaction between the component molecules as that finds application in several industrial and technological processes. Ultrasonic velocity and the derived acoustical parameters like adiabatic compressibility, free length, relaxation time, acoustic impedance, etc., with their excess values, provide valuable information about the molecular environments. This has been studied for various binary and ternary mixtures with respect to variation in concentration of the liquids and temperatures [Manoj *et al*, 2013], [5] [Vasantharani *et al*, 2009] [8].

The ultrasonic study of liquids is of immense important in understanding the nature and strength of molecular interactions. The biological activity of drug molecules and the activation energy of the metabolic process basically depend on the type and strength of the intermolecular interactions. Thermodynamic and transport properties of liquid mixtures have been extensively used to study the departure of a real liquid mixture behavior from ideality. From the literature, the nature and degree of molecular interactions in different solutions depend upon the nature of solvent, the structure of solute molecule and extent of solutes taking place in the solution [Bedare *et al*, 2014] [1].

In recent years ultrasonic investigations find large number of applications in characterizing of thermodynamic and physiochemical aspect of ternary liquid mixtures. The acoustical and thermodynamic parameter have been used to study different kinds of associations, molecular motion and various types of interaction and their strengths influenced by the size of pure component and the mixtures [Dange and Chimankar, 2013] [2].

The accurate thermodynamic properties of alcohols, in particularly 1-propanol are of interest for different branches of science and engineering. Propan-1-ol is an important industrial chemical fluid. Propan-1-ol is used as a solvent in the pharmaceutical industry. Hydrogen bonding is one of the most important types of intermolecular interactions play an important role in various physicochemical, biological and industrial processes [Vasantharani *et al*, 2009], [7] [Furniss *et al*, 1989] [4]

In this paper, variation of some parameters of ternary mixture containing methanol, ethanol and 1-propanol with temperature have been studied for a fixed concentration of equal volumes of the individual liquids making up the mixture.

Materials and Methods

A concentration in volume fraction of mixture was prepared by taking liquids of methanol, ethanol (BDH grades, 99.4% v/v) and distilled water. The volume fractions of the component liquids making the mixture were kept constant in the ratio of 1:1:1 throughout the variation of temperature. The density, viscosity, and ultrasonic velocity were measured as a function of temperature of the ternary liquid mixture at 4 MHz and at temperatures of T = 303 K, 308 K, 313 K, 318 K, 323 K and 328 K.

The density of the various systems at different temperatures were measured using relative measurement method and the viscosity of the mixture was measured using an Ostwald`s viscometer. The flow time was determined using a digital stopwatch with an accuracy of ±0.01s. The ultrasonic velocity of the liquid mixture was measured using a single crystal variable path interferometer at 4 MHz. The selected temperature of the liquid mixture was maintained constant by circulating water from a thermostatically controlled water bath with an accuracy of ±0.1 K.

Some acoustic and thermodynamic parameters were calculated [Bedare *et al,* 2014], [1] [Manoj *et al*, 2013] [5]

(i) Adiabatic compressibility (β)

$$\beta = 1/U^2\rho \quad (1)$$

(ii) Intermolecular free length (L_f)

$$L_f = K_T\beta^{1/2}, \quad (2)$$

(iii) Free volume (V_f)

$$V_f = (M_{eff}U/k\eta)^{3/2}, \quad (3)$$

(iv) Internal pressure (πi)

$$\pi_i = bRT(k\eta/U)^{1/2}\left(\rho^{2/3}/M_{eff}^{7/6}\right), \quad (4)$$

where *U* is ultrasonic velocity, *ρ* is density of the mixture, K_T is the temperature-dependent constant known as Jacobson's constant (K_T = 2.131 × 10−6 at 318K), *Meff* is the effective molecular weight of the mixture (*Meff* = *ΣmiXi)*, where *mi* and *Xi* are the molecular weight and mole fraction of individual constituents, respectively), *k* is a temperature-independent constant which is equal to 4.281 × 10^9 for all liquids, *η* is the viscosity of the mixture, *b* stands for cubic packing, which is assumed to be 2 for all liquids, *T* is the absolute temperature in Kelvin, *,* *R* is the universal gas constant, k_B is Boltzmann's constant, and *h* is Planck's constant, *f* is the frequency of ultrasonic wave.

Results and Discussion

The data obtained from experiment relating to density, viscosity, and ultrasonic velocity at indicated temperatures for frequency 4MHz, for the given mixture, and calculated values of adiabatic compressibility, free length, free volume, internal pressure have been presented in Table 1.

Table 1: Density (ρ), viscosity (μ) velocity (U), adiabatic compressibility (β), free length (L_f), free volume (V_f) and internal pressure (π_i) of methanol + ethanol+1-propanol mixture

T(K)	ρ(kg/m³)	μ(×10³ Nsm⁻²)	U(msec⁻¹)	β (×10¹⁰Pa⁻¹)	L_f (×10¹¹m)	V_f ×10⁷m³mol⁻¹	π_i ×10⁻⁸Pa
303	782.3	1.4931	1301.8	7.4138	5.4102	0.7321	5.2713
308	779.6	1.2425	1282.4	7.7311	5.6101	0.8434	5.1516
313	777.2	1.1536	1261.1	7.9426	5.7621	0.9364	5.0487
318	775.5	1.1011	1237.3	8.1121	6.0951	1.0023	4.9013
323	772.4	1.0523	1216.4	8.2814	6.4352	1.0453	4.7002
328	770.1	1.0034	1199.2	8.4652	6.8345	1.1054	4.4142

Looking at Table 1and Figure 1 & 2, it can be seen that the density, viscosity and ultrasonic velocity decrease with increase in temperature. The decrease of values with temperature shows a decrease in intermolecular forces due to the increase in the thermal energy of the system. The velocity decreases with the increase in temperature because the fact that free length increases with the increase of temperature. Since the association of the interacting molecules varies with the temperature of the ultrasonic wave, cohesive force as well as internal pressure increases with the increase of temperature [Manoj *et al*, 2013], [5] [Furniss *et al*, 1989] [4].

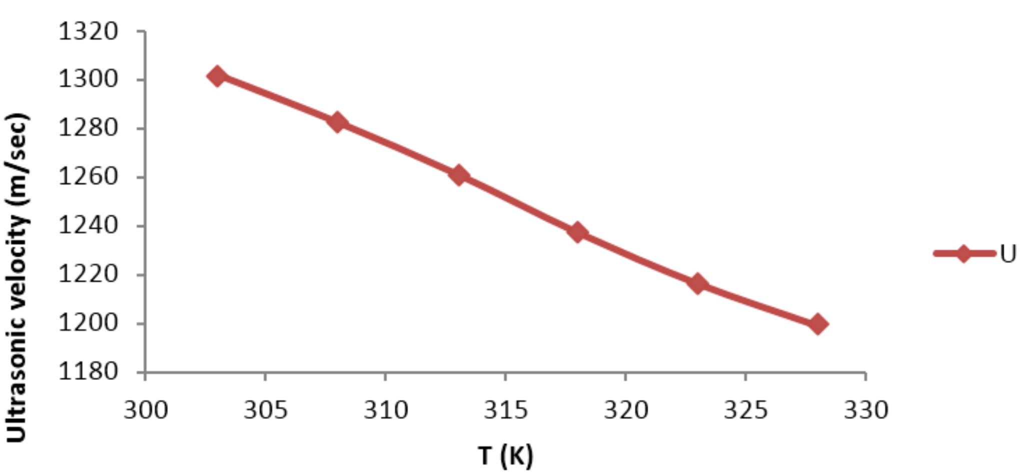

Fig 1: Variation of ultrasonic velocity U ($msec^{-1}$) with temperature T(K)

However, adiabatic compressibility (β), free length (L_f), free volume (V_f) and except internal pressure (π_i) increased with increase in temperature. The free length dependence on the adiabatic compressibility and show a similar behavior to that of the compressibility and inverse to that of velocity. It increased with increase in temperature of mixture, indicating that there is a less interaction between solute molecules. Free volume of the mixture increased as the internal pressure decreased with increase in temperature of mixture. This is most likely because of the loose packing of the molecules inside the shield, which may be brought about by the decreasing magnitude of interactions [Mecke, 1950] [6].

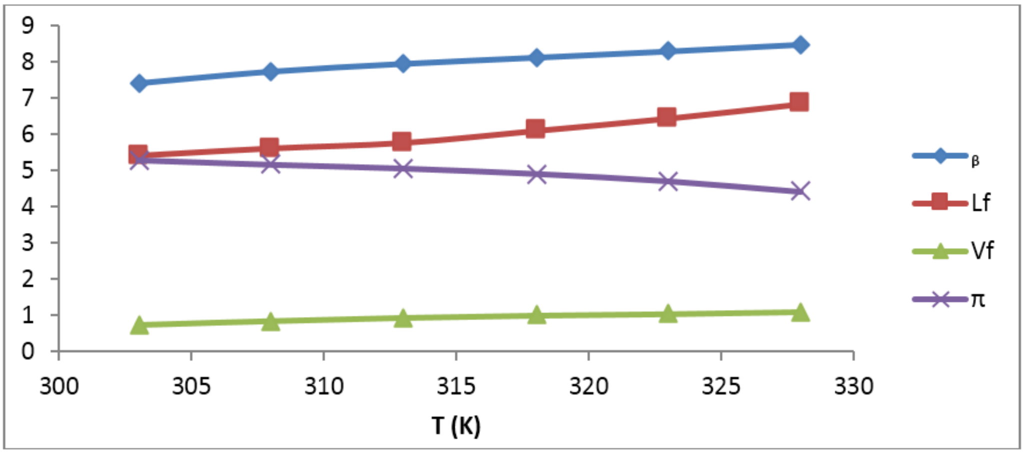

Fig 2: Variation of adiabatic compressibility β ($\times10^{10}Pa^{-1}$), free length L_f($\times10^{11}$m), free volume V_f($\times10^{7}m^3mol^{-1}$) and internal pressure π_i($\times10^{-8}$Pa) of mixture with temperature

From Figure 1 & 2, it can be seen that ultrasonic velocity (U), adiabatic compressibility (β), free length (L_f), and free volume (V_f), increased almost linearly with increase in temperature. While, internal pressure (π_i) decreased almost linearly with increase in temperature. As may be expected, the density and viscosity of the system decreased with increase in temperature. The molecules in a liquid are held together much more strongly than in a gas. A force is needed to overcome the mutual attraction of the molecules so that they can be displaced relative to each other. The more strongly the molecules are held together, the smaller the flow for a given shearing stress. With increasing temperature, the random kinetic energy of the molecules helps to overcome the molecular forces and reduces the viscosity [Dikko, 2014] [3] [Vasantharani *et al*, 2009] [7].

Conclusion
The ultrasonic velocity and the thermodynamic parameters: adiabatic compressibility, free length, free volume, and internal pressure of methanol+ethanol+1-propanol mixture increase as the temperature rises. However, the internal pressure decreases with temperature increase. As usual, the density and viscosity of the ternary mixture decrease with increase in temperature. This could be due to the energy obtained to overcome the resistance to flow. The almost linear variation of acoustical parameters with temperature shows that there exist less intermolecular forces between the components of the ternary liquid mixture.

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