Frequency Dependence Studies of Acoustical and Thermo-Dynamical Parameters of The Binary Mixtures of Isopropyl Sulphide And Acetic Acid

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Abstract: - Using thermal expansivity and ultrasonic velocity data for the binary mixtures of isopropyl sulphide and acetic acid, at room temperature, a number of acoustical and thermo-dynamical parameters such as Moelwyn-Hughes parameter (C_1), reduced volume ($\tilde{v}$), fractional free volume (f), repulsive exponent (n), Sharma's constants (S_0), isobaric (Γ_b), isochoric (Γ_c), isothermal Gruneisen parameters (Γ_T), Bayer's non-linear parameter (B/A), Lattice Gruneisen parameters (Γ), cohesive energy density (ε_i) and interaction parameter (ζ) has been evaluated. The frequency dependence of these parameters has been investigated at various concentrations of the binary mixture under study, within the frequency range 1-7 MHz. The variations of these parameters with change in the composition of the mixture and variation of frequency of the ultrasonic waves, propagating in the medium, have been used to explain the molecular interactions, anharmonicity and structural information about the systems under study. The obtained results indicate a decrease in intramolecular modes of vibration and anharmonicity in liquid mixture at higher frequency. These also suggest that binary mixture of isopropyl sulphide and acetic acid becomes less associative and hence weak interaction forces dominate at higher frequency.

Keywords: - Thermal expansivity, molecular interactions, anharmonicity, structural information.

1. INTRODUCTION

The thermo acoustical parameters play an important role in understanding the significance of microscopic factors such as molecular order, molecular cohesion and intermolecular interactions in solids, liquids and their binary solutions. These parameters are useful quantities in studying the internal structure and anharmonicity. Several approaches have been proposed to evaluate thermo-acoustical parameters for a wide variety of liquids, polymers and liquid crystals. Some of these parameters are of fundamental significance in the equation of state calculations, which serves as an effective guide in determining the mechanism of ultrasonic absorption in liquids and their mixtures.

The investigation of thermo-dynamical and ultrasonic velocity data for a binary mixture helps us

to understand the microstructure and thermodynamic properties of materials. The change in acoustical properties of a system has been found to be directly related to nature, type and strength of molecular interactions in the system [1].

The variation in Grüneisen parameter provides information about the change in the harmonicity of inter-atomic interactions [2-4]. Sharma's constants, another useful tool for understanding molecular interactions in a mixture, have been found closely related to isothermal Gruneisen parameter, Molecular constant (r), isochoric thermo-acoustical parameter, linear thermal expansion coefficient [5-7], are other structurally important parameters. The Moelwyn–Hughes parameter has a close relationship with the Gruneisen parameter and some of the thermo-acoustical parameters like Bayer's non-linearity parameter, Rao's acoustical parameter, and fractional available volume can be evaluated with help of Moelwyn–Hughes parameter [8-9].

Di-isopropyl sulphide is present in simulated meat flavours derived from autolysed baker's yeast and used as good flavorings agent in food additives [10]. Acetic acid is widely used chemical, having wide spread uses in food and drug industry. Acetic acid form binary mixture with many liquids including isopropyl. An exhaustive review of the literature reveals that only a few reports are available on the frequency response of the thermo-dynamical and acoustical parameters of the binary mixtures of isopropyl sulphide and acetic acid binary mixture [11].The main objective of the present paper is to compute various thermo-acoustical parameters, and to test the interrelationship between Moelwyn–Hughes parameter, Bayer's non-linearity parameter and available volume. In addition, the effect of increase in frequency on the variation of various thermo-dynamical and acoustical parameters viz; Moelwyn-Hughes parameter (C_1), reduced volume ($\widetilde{V}$), fractional free volume (f), repulsive exponent (n), Sharma's constants (S_0), isobaric (Γ_b), isochoric (Γ_c), and isothermal (Γ_T) Gruneisen parameters, Bayer's non-linear parameter (B/A), Lattice Gruneisen parameters (Γ') cohesive energy density (ε_i) and interaction parameter (ζ) at specific concentrations of the binary liquid mixtures of isopropyl sulphide and acetic acid at room temperature has been investigated.

2. THEORETICAL FORMULATION

The thermo-dynamical and acoustical parameters *viz* Moelwyn-Hughes parameter (C_1), reduced volume ($\widetilde{V}$), Sharma's constants (S_0, S^*), isobaric (Γ_b), isochoric (Γ_c), isothermal (Γ_T), and lattice (Γ') Gruneisen parameters, fractional free volume (f), repulsive exponent (n), Bayer's non-linear parameter (B/A) and cohesive energy density (ε_i) are calculated using standard relations available in literature [12].

Moelwyn-Hughes defined the C_1 parameter as the pressure coefficient of the isothermal bulk modulus, which can be written as [8];

$$C_1 = \left[\left(\frac{\partial}{\partial P}\right)\left(\frac{1}{\beta}\right)\right] = \left(\frac{1}{\beta}\right)\left(\frac{\partial ln\beta}{\partial P}\right)_T = \left(\frac{\partial ln\beta}{\partial lnV}\right)_T \quad (1)$$

Sharma has obtained an expression for the parameter C_1 in terms of the thermal expansion coefficient (α) as [7]

$$C_1 = \left(\frac{13}{3}\right) + \left(\frac{1}{\alpha T}\right) + \left(\frac{4\alpha T}{3}\right) \quad (2)$$

where,

$$\alpha = \left(K_T \times 0.0191\right)^{1/4}, \quad (3)$$

$$K_T = \frac{1.71 \times 10^{-3}}{T^{4/9} U^2 d^{4/3}} \quad (4)$$

Reduced molar Volume ($\widetilde{V}$) can be expressed in terms of α, in the zero pressure limit as [13, 14]

$$\widetilde{V} = \left[\frac{1+\alpha T}{3(1+\alpha T)}\right]^3 \quad (5)$$

Reddy *et al.* have established the correlation between Moelwyn-Hughes parameter C_1 and other thermo-acoustical parameters [9, 15]. The derived relations are as follows:

$$\frac{B}{A} = C_1 - 1 \quad (6)$$

$$f = \frac{V_a}{V} = (C_1 + 1)/2 \quad (7)$$

$$n = 3C_1 - 12 \quad (8)$$

where (B/A) is Bayer's nonlinearity parameter for the closest packed volume; V_a/V is the fractional (free) available volume (f); and n are the exponents describing the magnitude of attractive and repulsive force, respectively.

Several authors attempted the estimation of n and highlighted its importance. The magnitude of m is recognized as 6 in the literature [2, 13, 16]. Assuming the sound velocity U as a function of both volume V

and temperature T, the isobaric, isothermal and isochoric acoustical parameters are related as [5, 17]

$$K_{ith} = K_{iba+} K_{ich} \quad , \tag{9}$$

in which,

$$K_{ich} = \frac{5}{3} + (2\alpha\alpha)^{-1} + \frac{2\alpha\alpha}{3} \ , \tag{10}$$

$$K_{ib} = -\frac{1}{\tilde{V}^{C_1}}\left[(2\alpha\alpha)^{-1} - \tilde{V}^{C_1} + 1\right] \tag{11}$$

Sharma [18] introduced three dimensional an harmonic parameters for isothermal, isobaric and isochoric Gruneisen parameters for liquid state parameters are identical to the corresponding acoustical parameters, so one can write

$$\Gamma_T = \Gamma_b + \Gamma_c, \tag{12}$$

The lattice Gruneisen parameter (Γ') is evaluated using the value of C_1 is given as

$$\Gamma' = (C_1 - 1)/2 \ , \tag{13}$$

Sharma [18] proposed a theoretical method to establish a relation between the molecular constant r and the isochoric thermo-acoustical parameter Δ as,

$$r = \frac{P_i}{\varepsilon} = (1 - \Delta)^{-1}, \tag{14}$$

where,

$$\Delta = -\frac{\left[2 + (\alpha T)^{-1}\right]\left[(-2\alpha)\left(V^{C_1}\right)^{-1}\right] T}{2} \tag{15}$$

Where P_i and ε are the internal pressure and cohesive energy density respectively.

Sharma [19] and Reddy *et al*. [20] have introduced dimensionless parameters S_0

$$S_o = \Delta(3 + 4\alpha T) = 3\Delta S^* , \tag{16}$$

$$S^* = \left[1 + (4/3)\alpha T\right] \ , \tag{17}$$

where Δ is given by equation (15)

The interaction parameter (ζ) for the system is determined using the relation:

$$\varsigma = \frac{U^2}{U^{im}} - 1 \ , \tag{18}$$

where U is the experimental value and $U^{im} = X_1U_1 + X_2U_2$ is ideal mixing value of the ultrasonic velocity. X_1, X_2, U_1 and U_2 are the mole fractions and ultrasonic velocities of isopropyl sulphide and acetic acid, respectively.

3. RESULTS AND DISCUSSION

The necessary experimental data X_1, T, U, ν and α for the system under consideration have been taken from the literature are reported in table 1[21].

Table 1: Experimental values of density (d), ultrasonic velocity (U) and theoretically calculated value of coefficient of thermal expansion (α) for isopropyl sulphide and acetic acid binary solution as a function of mole fraction of isopropyl sulphide at different frequencies range 1MHz -7MHz.

X_1	d	U			
		1 MHz	3 MHz	5 MHz	7 MHz
0	997	1064	1090	1110	1123
0.0419	979	1276	1289	1298	1305
0.0897	999	1297	1307	1315	1321
0.1445	985	1306	1316	1324	1331
0.2081	935	1320	1331	1338	1346
0.2827	989	1332	1341	1348	1354
0.3715	1005	1342	1351	1357	1363
0.4791	1150	1353	1361	1368	1374
0.6119	1152	1363	1370	1377	1381
0.7801	999	1369	1377	1384	1389
1	1102	1372	1381	1386	1391
X_1	d	$\alpha \times 10^{-3}$			
		1 MHz	3 MHz	5 MHz	7 MHz
0	997	1.231	1.216	1.205	1.198
0.0419	979	1.131	1.125	1.121	1.118
0.0897	999	1.114	1.110	1.107	1.104
0.1445	985	1.116	1.111	1.108	1.105
0.2081	935	1.129	1.124	1.121	1.118
0.2827	989	1.103	1.099	1.097	1.094
0.3715	1005	1.093	1.089	1.087	1.085
0.4791	1150	1.041	1.038	1.035	1.033
0.6119	1152	1.036	1.034	1.031	1.030
0.7801	999	1.084	1.081	1.079	1.077
1	1102	1.048	1.045	1.043	1.041

Using standard thermo-dynamical and acoustical relations available in literature, various parameters evaluated for the binary mixture of isopropyl sulphide and acetic acid at room temperature are reported hereafter. The frequency dependence of these parameters, at various compositions of the binary mixture is reported in the table 2 and table 3. The Moelwyn-Hughes parameter is of great importance due to its close relationship with Gruneisen parameter and several other thermo-acoustic parameters. A perusal of table 2 reveals that C_1 values increase by < 0.4 % with increase in frequency. The slight increase in C_1 values with rise in frequency signifies readjustment of constituents of the binary system, leading to a closer packing. This fact is complemented by slight decrease in $\tilde{V}$ values and the

constant behavior of the fractional free volume (f) for the system under investigation for a similar variation. Sharma's constant (S_0) register a constancy in their values with rise in frequency as well as rise in mole fraction of isopropyl sulphide in the binary solution.

The slight increase in the repulsive exponent (n) values with rise in frequency is being compensated by the increasing associative tendency of isopropyl sulphide and the acetic acid molecules (Dipole moment =1.74D), leading to slight decrease in $\tilde{V}$ values and the constant behavior of the fractional free volume (f) for the system. The average value of f (~0.20) for all the samples under study are consistent with the reported f values for saturated hydrocarbons [22], quasi spherical molecular liquids [3], polymers [23], and are greater than those reported for fluorocarbons (f = 0.15) [24] and ternary mixtures (f = 0.17) [25]. The average value of repulsive component (n) lies in the range 11.413 to 11.490, as the frequency for the system is changed from 1 MHz to 7 MHz. These values are well within the range ~ 4.6-19, as reported for the liquids [23] but are less than that reported for polymers and fluorocarbons. The fluctuating behavior of n with increase in concentration (table 2) shows the competition between associative nature of the polar molecules of acetic acids, (due to increase in its mole fraction), and the steric effects of isopropyl molecules (impacted by the rise in frequency) in the system. The value of the S_0 as reported in table 2, remains fairly constant within the acceptable range of experimental error. The estimated value of the S_0 in general lies in the range 1.11 ± 0.01. These values are in good agreement with the results reported by Sharma [18, 19]. It has also been established that the value of S_0 is independent of increase in frequency (1 MHz to 7 MHz) for a given system. This confirms the earlier conclusion that the constant S_0 which turns out to be remarkably constant for any system existing either in a liquid or in solid state. This suggests that S_0 may be regarded as a molecular constant for the liquid phase.

The Grüneisen parameter describes the effect of changing the volume of a crystal lattice on its vibrational properties. It also reports the effect of the change of temperature, on the size or dynamics of the lattice. For a model, in which compaction is assumed to take place, by an increase of long-range positional order and growth of clusters, the pseudo-Grüneisen parameter is predicted to be temperature independent[26]. Isobaric Grüneisen parameter (Γ_b) and Isochoric parameter (Γ_c), as calculated by using lattice-dynamical method, include the effect of dispersion and crystal anharmonicity.

Table 2: Frequency dependence of various thermo-dynamical parameters of the binary mixture of isopropyl sulfide and acetic acid at room temperature

ν	C_1	n	S_0	$\tilde{V}$	f
X_1 = 0.0420					
1	7.733	11.24	1.115	1.275	0.200
3	7.746	11.23	1.115	1.274	0.200
5	7.755	11.26	1.115	1.273	0.200
7	7.761	11.28	1.116	1.273	0.200
X_2=0.0897					
1	7.771	11.31	1.116	1.272	0.200
3	7.781	11.34	1.116	1.271	0.200
5	7.788	11.36	1.116	1.27	0.200
7	7.794	11.38	1.116	1.27	0.200
X_3=0.1445					
1	7.768	11.3	1.116	1.272	0.200
3	7.777	11.33	1.116	1.271	0.200
5	7.785	11.35	1.116	1.271	0.200
7	7.792	11.37	1.116	1.27	0.200
X_4= 0.2081					
1	7.737	11.21	1.115	1.275	0.200
3	7.748	11.24	1.115	1.274	0.200
5	7.754	11.26	1.115	1.273	0.200
7	7.762	11.28	1.116	1.273	0.200
X_5=0.2827					
1	7.796	11.39	1.116	1.27	0.200
3	7.805	11.41	1.116	1.269	0.200
5	7.812	11.43	1.116	1.269	0.199
7	7.818	11.45	1.116	1.268	0.199
X_6 = 0.3715					
1	7.82	11.46	1.116	1.268	0.199
3	7.829	11.49	1.116	1.267	0.199
5	7.834	11.5	1.116	1.267	0.199
7	7.84	11.52	1.116	1.266	0.199
X_7 = 0.4791					
1	7.952	11.85	1.117	1.257	0.198
3	7.96	11.88	1.117	1.257	0.198
5	7.968	11.9	1.117	1.256	0.198
7	7.974	11.92	1.118	1.256	0.198
X_8 = 0.6119					
1	7.964	11.89	1.117	1.256	0.198
3	7.971	11.91	1.117	1.256	0.198
5	7.978	11.93	1.118	1.255	0.198
7	7.983	11.95	1.118	1.255	0.197
X_9 = 0.7801					
1	7.841	11.52	1.116	1.266	0.199
3	7.849	11.55	1.116	1.265	0.199
5	7.855	11.57	1.117	1.265	0.199
7	7.86	11.58	1.117	1.265	0.199

It is observed that isothermal Grüneisen parameter (Γ_T) is the sum of isobaric and isochoric parameters [27]. The composition and frequency dependence values of Γ_b, Γ_c and Γ_T, for the binary solution under study, are reported in Table 3. Isochoric Gruneisen parameter Γ_c is a negative quantity for polymers and a positive quantity for liquid crystals[27]. This conclusion of Tolpadi [27] is authenticated for the

present binary liquid system as the relation Γ_c = -(Γ_b-Γ_T) [28], applies fairly well for the present investigation (table 3).The perusal of table 2 indicates that Gruneisen parameters Γ_b and Γ_T show a slight linear increase with the rise in frequency from 1 MHz -7 MHz. Whereas Γ_c shows an opposite trend as expected. The change in composition of the binary mixture, also reports a trend of variation in Γ_b, Γ_T, and Γ_c values, similar to the trends of variation in the values for these parameters, with frequency change. For values of Γ', a trend of variation similar to the variation of Γ_b and Γ_T is observed.

As is well established that changes in Γ'values are governed by the relative changes in molecular vibrations frequency, molecular order, and structure, these trends of variations in the values of Γ' complement the conclusions arrived at earlier. Bayer's nonlinearity parameter (B/A) is strongly sensitive [29] to frequency and composition changes for a system. The increasing value of B/A and Γ' are attributed to increase in ultrasonic velocity with concentration of isopropyl sulfide. In present binary solution, the increase in B/A and Γ' with concentration shows decrease in intermolecular modes of vibration and harmonicity in liquid mixtures. It also indicates the associating nature of the binary solution. The increase in B/A and Γ' with increase in frequency (1 MHz to 7 MHz) is prevalence of associating nature of the intermolecular forces in the given binary solution. A gradual increase in the magnitude of cohesive energy density (ε_i) with increase in frequency as well as increase in mole fraction of isopropyl sulphide is observed for the binary system under study (table 3). The slight increase in ε_i values, with increase in frequency, indicates greater associative interactions among the molecules at higher frequency.

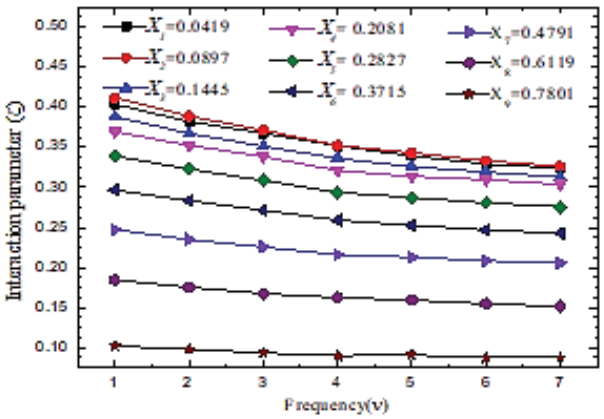

Figure 1: Variation of interaction parameter with frequency at various mole fraction (X) of isopropyl sulphide

Table 3: Frequency dependence of some thermo-dynamical parameters of the binary mixture of isopropyl sulfide and acetic acid at room temperature

ν	Γ_b	Γ_c	Γ_T	Γ'	B/A	$\varepsilon_i \times 10^8$
X_1 = 0.0420						
1	3.697	0.623	4.319	7.233	6.733	2.95
3	3.703	0.62	4.323	7.246	6.746	2.99
5	3.707	0.618	4.326	7.255	6.755	3.02
7	3.711	0.617	4.328	7.261	6.761	3.04
X_2 = 0.0897						
1	3.715	0.615	4.331	7.271	6.771	3.08
3	3.72	0.613	4.334	7.281	6.781	3.11
5	3.724	0.612	4.336	7.288	6.788	3.14
7	3.727	0.61	4.338	7.294	6.794	3.16
X_3 = 0.1445						
1	3.714	0.616	4.33	7.268	6.768	3.07
3	3.719	0.614	4.333	7.277	6.777	3.10
5	3.723	0.612	4.335	7.285	6.785	3.13
7	3.726	0.611	4.337	7.292	6.792	3.15
X_4 = 0.2081						
1	3.699	0.622	4.321	7.237	6.737	2.96
3	3.704	0.62	4.324	7.248	6.748	3
5	3.707	0.619	4.326	7.254	6.754	3.02
7	3.711	0.617	4.328	7.262	6.762	3.05
X_5 = 0.2827						
1	3.728	0.61	4.338	7.296	6.796	3.17
3	3.733	0.608	4.341	7.305	6.805	3.20
5	3.736	0.607	4.343	7.312	6.812	3.22
7	3.739	0.606	4.345	7.318	6.818	3.24
X_6 = 0.3715						
1	3.74	0.605	4.345	7.32	6.82	3.25
3	3.744	0.604	4.348	7.329	6.829	3.28
5	3.747	0.602	4.35	7.334	6.834	3.3
7	3.75	0.601	4.351	7.34	6.84	3.32
X_7 = 0.4791						
1	3.806	0.579	4.385	7.452	6.952	3.75
3	3.81	0.577	4.387	7.46	6.96	3.78
5	3.814	0.576	4.39	7.468	6.968	3.81
7	3.819	0.574	4.393	7.478	6.978	3.85
X_8 = 0.6119						
1	3.812	0.577	4.389	7.464	6.964	3.79
3	3.816	0.575	4.391	7.471	6.971	3.82
5	3.819	0.574	4.393	7.478	6.978	3.85
7	3.821	0.573	4.394	7.483	6.983	3.86
X_9 = 0.7801						
1	3.75	0.601	4.352	7.341	6.841	3.33
3	3.754	0.6	4.354	7.349	6.849	3.35
5	3.758	0.598	4.356	7.355	6.855	3.38
7	3.76	0.597	4.357	7.36	6.86	3.40

The variation of the interaction parameter with change in frequency from 1 MHZ to 7 MHz at various mole fraction (X) of the isopropyl sulphide in the binary solution is shown in Figure 1.

It is clear from the graph that the interaction parameter (ζ) is throughout positive and gradually decreases with increase in frequency up to X_I = 0.3715, thereafter it remain almost constant. The positive value of ζ indicates existence of associative interactions between the constituent molecules of the binary solution and supplements the increase in B/A, Γ' and ε_i with increase in mole fraction (X) of isopropyl sulphide in the given frequency range.

4. CONCLUSION

The present determination of several acoustical and thermodynamical parameters of the binary mixtures of isopropyl sulphide and acetic acid, at different compositions of the mixture provides a qualitative as well as quantitative measurement of associative interactions prevalent in the binary system. The effect of the variation in frequency of the ultrasonic wave propagating in the medium, on these parameters have also been investigated. The obtained results indicate a decrease in intramolecular modes of vibration and anharmonicity in liquid mixture at higher frequencies of the ultrasonic wave propagation.

The present analysis also allows us to understand the significance of fractional free volume, the repulsive exponent of the intermolecular potential, and the S_o parameter in describing the anharmonic and non-linear properties, molecular structure order, and interactions in binary liquid mixtures. The universal constancy and frequency invariance of S_o parameter has also been verified for the binary mixture under study.

A significant advantage of the current method of evaluation of various acoustical and thermo-dynamical parameters is its simplicity, as the method depends only on the volume expansivity data, density and ultrasonic velocity.

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