International Journal of
Scientific Research in Science and Technology (IJSRST)
Print ISSN : 2395-6011, Online ISSN : 2395-602X
International Conference on Advanced Materials
Held on 14, 15 December 2017, Organized by Department of Physics,
St. Joseph's College, Trichy, Tamilnadu, India

Investigation of Molecular Interactions in Binary Mixtures of Anisole with Methyl Isobutyl Ketone at Different Temperatures

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Abstract

The ultrasonic velocity(U), density (ρ) and viscosity (η) of pure solvents and binary mixtures of Anisole and Methyl Isobutyl Ketone have been investigated at three different temperatures 303.15K, 308.15K and 313.15K and correlated with concentration. The observed data have been used to evaluate some of the thermo acoustical parameters such as adiabatic compressibility (β_a), intermolecular free length (L_f), internal pressure (π_i), acoustic impedance (Z), available volume (V_a), relaxation time (τ), molar volume (V_m) and Gibb's free energy (ΔG), Lennard Jones potential repulsive term exponent(n) to elucidate the molecular association in the mixture. The variation of these parameters with concentration of solute indicates the nature of interaction present in the binary mixture.

Keywords: Anisole, Ultrasonic velocity, adiabatic compressibility, acoustic impedance, molecular association.

Introduction

The study of molecular interactions in liquid mixture provides valuable information regarding internal structure, molecular association, Internal pressure and complex formation. The propagation of ultrasonic waves in liquid is a tool by researcher to probe into the properties of liquids [1-3]. The measurement of ultrasonic velocity in liquid mixtures have greater importance in predicting the physico-chemical properties of liquid mixtures.

In the present study, the density, ultrasonic velocity and viscosity were measured for the binary system of Anisole and Methyl isobutyl Ketone at various temperatures. Anisole is an organic compound with the formula C_7H_8O. It is a colourless liquid and its derivatives are found in natural and artificial fragrances insoluble in water. It is used as a precursor to perfumes and pharmaceuticals. The second component is Methyl isobutyl Ketone, it is a colourless liquid with the formula $C_6H_{12}O$. It is a ketone, used as a solvent for gums, varnishes and nitrocellulose. These liquids have been chosen on the basis of their industrial applications. These applications stimulated the need for extensive information on the thermodynamic and acoustic properties of these mixtures.

From the experimental data, an attempt has been made to study some thermo acoustical properties such as adiabatic compressibility (β_a), intermolecular free length (L_f), internal pressure (π_i), acoustic impedance (Z), relaxation time (τ), molar volume (V_m), available volume (V_a) and Gibb's free energy (ΔG), Lennard Jones potential repulsive term exponent (n). These parameters have been used to understand different kinds of association in pure components, as well as, in the binary mixture.

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Results are used to explain the nature of molecular interactions between mixing Compounds.

2. Experimental Section

The mixtures of various concentrations in mole fraction were prepared by taking analytical reagent grade chemicals with minimum assay of 99.9% and obtained from E. Merck Ltd (India). All the component liquids were purified by the standard methods. The density, viscosity, and ultrasonic velocity were measured as a function of concentration of the binary liquid mixture at 303.15K, 308.15K & 313.15K. Ultrasonic velocity measurements were made using an ultrasonic interferometer with the accuracy of ±0.1m·s–1. An electronically operated digital constant temperature bath operating with an accuracy of ±0.1°C has been used to circulate water through the outer jacket of the double-walled measuring cell containing the experimental liquid. The densities of the mixture were measured using a 10-ml specific gravity bottle by relative measurement method with an accuracy of ±0.01 kg·m–3. An Oswald viscometer (10 ml) with an accuracy of ±0.001 Ns·m–2 was used for the viscosity measurement. The flow time was determined using a digital racer stopwatch with an accuracy of ±0.1s.

Theory

The various thermo acoustical parameters were determined using the following equations [5-11]

Adiabatic Compressibility (β_a):

$$\beta_a = 1/ (U^{2*}\rho) \quad (1)$$

Intermolecular Free Length (L_f):

$$L_f = K_J (\beta_a)1/2 \quad (2)$$

where K_J is the temperature dependent Jacobson's constant but independent of the nature of liquid.

Free Volume (V_f)

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

where, M_{eff} (Effective mass) = $\Sigma\, m_i x_i$, in which mi and xi are the molecular weight and the mole fraction of the individual constituents respectively.

K is the temperature independent constant, which is equal to 4.28 *10^9 for all liquids and η be the viscosity.

Internal Pressure (π_i)

$$\pi_i=(bRT)[(K^*\eta)/(U)]^{½*}[(\rho^{2/3})/(M_{eff}^{7/6})] \quad (4)$$

where, b is the cubic packing which is assumed to be 2 for all liquids and solutions, K is the temperature independent constant ,T is the absolute temperature, R is universal gas constant.

Relaxation Time (τ)

Relaxation time (τ) is the time taken for the excitation energy to appear as translational energy and it depends on temperature and on impurities. The dispersion of ultrasonic velocity in binary mixture reveals information about the characteristic time of the relaxation process that causes dispersion.

$$\tau = (4/3)\, \beta_a{}^*\, \eta \quad (5)$$

Acoustic Impedance (Z)

$$Z = U^*\rho \quad (6)$$

Gibb's Free Energy (ΔG)

The relaxation time for a given transition is related to the activation energy. The variation of relaxation time (τ) with temperature (T) can be expressed in the form of Eyring self-process theory.

$$\Delta G = (K_B T)\log [(K_B T\tau)/h] \quad (8)$$

where K_B is the Boltzmann's constant (1.3806 × 10^{23} Jk^{-1}), h is the plank's constant 6.63*10^{-34} JS), T be the absolute temperature and τ be the relaxation time.

Available volume (V_a)

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$V_a = (M_{eff}/\rho)(1-(U/U\infty))$ (10)

Molar Volume (V_m)

Lennard Jones Potential Repulsive Term Exponent (n)

$V_m = M_{eff}/\rho$ (12)

$n = (6V_m/V_A)-13$ (11)

Results and Discussion

Table 1. Measured Values of Density (ρ), Viscosity (η) and velocity (U) of binary mixture of Anisole with Methyl Isobutyl Ketone at 303.15K, 308.15K & 313.15K

S. No.	X(i) ANI	Y(i) MIK	ρ x 10^{-3} kgm^{-3}			η x 10^{-3} Nsm^{-2}			U ms^{-1}		
			303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K
1	0	1	0.8405	0.7864	0.7850	0.6103	0.5853	0.5695	1177.5	1162.0	1149.3
2	0.1	0.9	0.8689	0.8171	0.8071	0.6459	0.6231	0.6093	1195.8	1180.8	1162.5
3	0.2	0.8	0.8861	0.8353	0.8321	0.6645	0.6523	0.6417	1215.8	1199.8	1189.5
4	0.3	0.7	0.9023	0.8554	0.8523	0.6830	0.6764	0.6634	1233.1	1218.2	1207.1
5	0.4	0.6	0.9306	0.8746	0.8744	0.7175	0.6919	0.6852	1251.3	1236.1	1224.3
6	0.5	0.5	0.9528	0.8967	0.8917	0.7380	0.7106	0.7012	1267.5	1252.5	1237.8
7	0.6	0.4	0.9690	0.9159	0.9099	0.7680	0.7554	0.7252	1288.5	1271.0	1257.8
8	0.7	0.3	0.9913	0.9350	0.9282	0.7904	0.7784	0.7459	1304.3	1287.3	1274.0
9	0.8	0.2	1.0105	0.9523	0.9484	0.8370	0.8168	0.7671	1323.9	1306.9	1291.6
10	0.9	0.1	1.0267	0.9696	0.9618	0.8685	0.8401	0.7924	1339.6	1327.6	1313.5
11	1	0	1.0439	0.9868	0.9791	0.9065	0.8776	0.8599	1357.5	1345.5	1331.1

Table 2. Values of Adiabatic compressibility (β_a), Intermolecular free length (L_f), Acoustic impedance (Z) at 303.15K, 308.15K & 313.15K

S. No.	X(i) ANI	Y(i) MIK	β_a x 10^{-10} m^2n^{-1}			L_F x 10^{-10} m			Z Kgm^{-2} s^{-1}		
			303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K
1	0	1	8.5810	9.4168	9.6434	0.8495	0.9322	0.9547	989.68	913.8	902.2
2	0.1	0.9	8.0473	8.7763	9.1753	0.7966	0.8688	0.9083	1039.1	964.8	937.6
3	0.2	0.8	7.6340	8.3157	8.5001	0.7557	0.8232	0.8415	1077.3	1002.2	989.4
4	0.3	0.7	7.2887	7.8800	8.0535	0.7215	0.7801	0.7972	1112.6	1041.8	1028.2
5	0.4	0.6	6.863	7.4839	7.6331	0.6794	0.7409	0.7556	1164.4	1081.0	1070.1
6	0.5	0.5	6.5328	7.1139	7.3188	0.6467	0.7042	0.7245	1207.6	1122.7	1103.8
7	0.6	0.4	6.2159	6.7581	6.9458	0.6153	0.6690	0.6876	1248.5	1164.1	1144.7
8	0.7	0.3	5.9294	6.4536	6.6372	0.5870	0.6389	0.6570	1292.9	1203.6	1182.3
9	0.8	0.2	5.6458	6.1477	6.3200	0.5589	0.6086	0.6256	1337.8	1244.5	1225.0
10	0.9	0.1	5.4271	5.8511	6.0259	0.5372	0.5792	0.5965	1375.4	1287.2	1263.1
11	1	0	5.1979	5.5972	5.7641	0.5146	0.5541	0.5706	1417.1	1327.7	1303.6

Table 3. Values of Relaxation time(τ),Internal pressure (□i), Gibbs Free energy(ΔG) at 303.15K,308.15K&313.15K

S. No.	X(i) ANI	Y(i) MIK	τ× 10^{-10} (s)			π_i x10^{6} Pa			(-ΔG)x10^{-21} KJ/mole		
			303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K
1	0	1	6.9826	7.3489	7.3226	0.7812	0.6755	0.6729	3.8575	3.9168	3.9795
2	0.1	0.9	6.9304	7.2913	7.4540	0.8229	0.7227	0.7120	3.8580	3.9173	3.9784
3	0.2	0.8	6.7638	7.2325	7.2726	0.8196	0.7364	0.7372	3.8595	3.9178	3.9800
4	0.3	0.7	6.6376	7.1067	7.1236	0.8154	0.7469	0.7458	3.8607	3.9189	3.9813

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5	0.4	0.6	6.5656	6.9042	6.9736	0.8505	0.7455	0.7573	3.8614	3.9208	3.9827
6	0.5	0.5	6.4283	6.7402	6.8426	0.8579	0.7529	0.7551	3.8627	3.9223	3.984
7	0.6	0.4	6.3651	6.8067	6.7162	0.8611	0.7797	0.7586	3.8633	3.9217	3.9852
8	0.7	0.3	6.2488	6.6979	6.6009	0.8689	0.7840	0.7603	3.8645	3.9227	3.9863
9	0.8	0.2	6.3007	6.6952	6.4641	0.8937	0.7975	0.7639	3.8640	3.9228	3.9877
10	0.9	0.1	6.2846	6.5540	6.3666	0.8979	0.7945	0.7574	3.8641	3.9241	3.9887
11	1	0	6.2826	6.5495	6.6088	0.9078	0.8054	0.7980	3.8642	3.9242	3.9862

Table 4. Values of Available volume (V_a), Molar volume(V_m), Lennard Jone Potential repulsive exponent (n) at 303.15K, 308.15K & 313.15K

S. No.	X(i) ANI	Y(i) MIK	V_a m^3/mol			V_m x 10^{-5} m^3/mol			N		
			303.15K	308.15K	313.15K	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K
1	0	1	3.1467	3.4862	3.5937	1.1916	1.2736	1.2759	9.721	8.920	8.302
2	0.1	0.9	2.9346	3.2365	3.4238	1.1619	1.2355	1.2508	10.755	9.905	8.920
3	0.2	0.8	2.7571	3.0466	3.1408	1.1483	1.2181	1.2228	11.990	10.99	10.360
4	0.3	0.7	2.6063	2.8623	2.9554	1.1365	1.1989	1.2032	13.165	12.131	11.428
5	0.4	0.6	2.4204	2.6881	2.7774	1.1105	1.1817	1.1819	14.530	13.375	12.533
6	0.5	0.5	2.2715	2.5258	2.6436	1.0930	1.1614	1.1679	15.872	14.590	13.508
7	0.6	0.4	2.1085	2.3557	2.4662	1.0830	1.1458	1.1534	17.818	16.183	15.060
8	0.7	0.3	1.9712	2.2100	2.3208	1.0667	1.1309	1.1392	19.469	17.704	16.452
9	0.8	0.2	1.8191	2.0492	2.1650	1.0543	1.1188	1.1234	21.775	19.75	18.133
10	0.9	0.1	1.7012	1.8844	1.9981	1.0455	1.1070	1.1160	23.873	22.248	20.512
11	1	0	1.5698	1.7428	1.8560	1.0359	1.0958	1.1044	26.594	24.726	22.703

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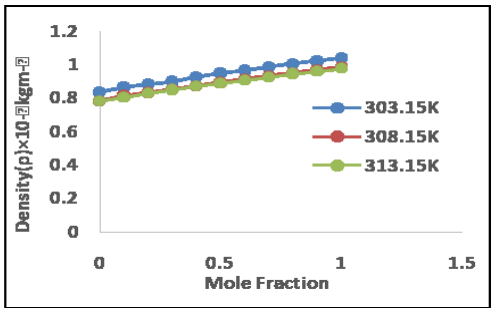

Figure 1. Mole fraction Vs Density

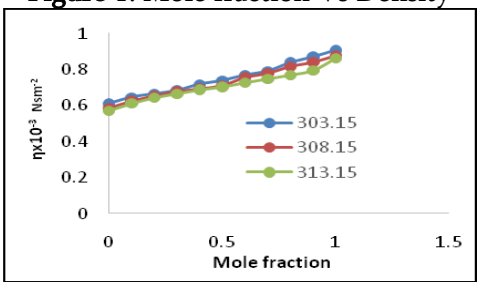

Figure 2. Mole fraction Vs Viscosity

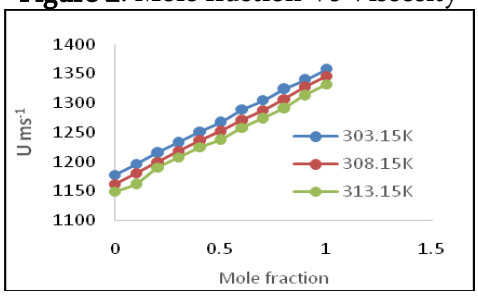

Figure 3. Mole fraction Vs Velocity

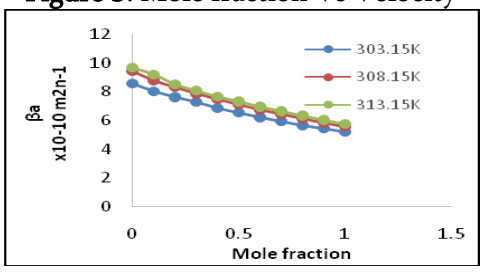

Figure 4. Molefraction Vs Adiabatic compressibility

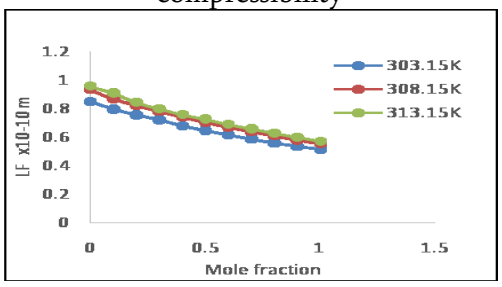

Figure 5. Mole fraction Vs Intermolecular free length

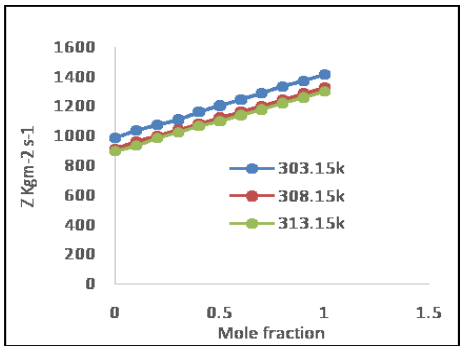

Figure 6. Molefraction Vs Acoustic impedance

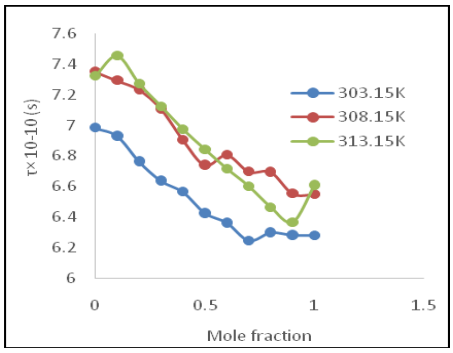

Figure 7. Mole fraction Vs Relaxation time

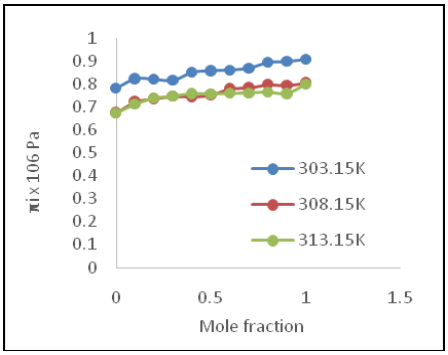

Figure 8. Molefraction Vs Internal Pressure

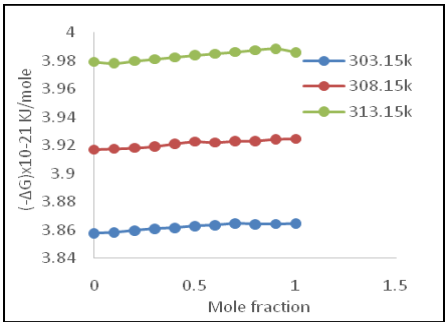

Figure 9. Molefraction Vs Gibb's Free energy

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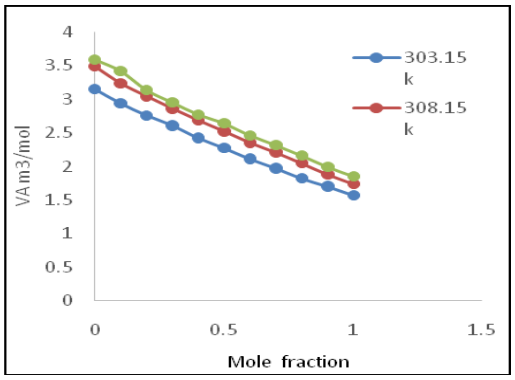

Figure 10. Mole fraction Vs Available volume

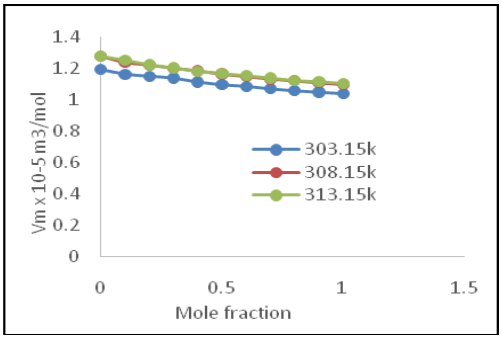

Figure 11. Molefraction Vs Molar volume

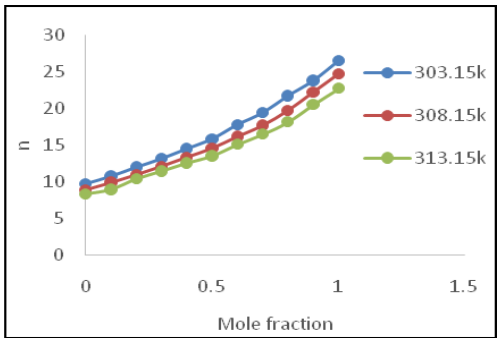

Figure 12. Mole fraction Vs LJP Repulsive term

Results and Discussion

The experimental values of density, velocity and viscosity at three different temperatures for the binary mixture were used to calculate the acoustical and thermos dynamical parameters and the relevant data are presented in Tables 1 to 4. The variation of these parameters are displayed graphically in Figures 1-12.

Density is a parameter giving information about solvent-solvent interactions. From the Table 1and Figure1 it is observed that in all the three temperatures the density and velocity increases with increasing mole fraction and decreases with higher temperatures. The increase in density indicates the presence of solvent-solvent interactions in the binary mixture which may bring a bonding between them [11]. Figure3 Shows that ultrasonic velocity increases with mole fraction and decreases with temperature. The structural changes occurring in the mixture with the increase in mole fraction leads to increase in velocity which may result in the increase in intermolecular forces. From Table1 and Figure 2, it is found that the viscosity of all liquid mixtures were found to be increased with increasing concentration of Anisole. Due to the increase of Anisole molecules in solution, the medium became denser. Further, the increase in the number of particles in solution is responsible for increasing the cohesive force between the liquid layers. Thereby, the co-efficient of viscosity increases in all systems.

The variation of adiabatic compressibility (β_a), Intermolecular free length (L_f) and acoustic impedance (Z) with the increase in mole fraction are presented in Table 2. Based on the model for sound propagation proposed by Eyring and Kincaid [11], ultrasonic velocity should increase, if the intermolecular free length decreases and vice versa. A reduction in adiabatic compressibility is an indication that component molecules are held close to each

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other. The intermolecular free length (L_f) is the distance covered by a sound wave between the surface of the neighbouring molecules and it depends upon the intermolecular attractive and repulsive forces. The decrease in the value of adiabatic compressibility (β_a) and Intermolecular free length (Lf) with concentration further strengthens the strong molecular association between the unlike molecules through hydrogen bonding.

The increase in the value of L_f with temperature implies that the mean distance between the molecules increases thereby decreasing the potential energy of interaction between them, leading to the decrease in the values of velocity and density. Molecular association increases the acoustic impedance (Z). It is the opposition exerted by the medium to displacement of the mediums particles by sound energy. A continuous decrease in (β_a), L_f and increase of Z with concentration are the clear evidence for the existence of strong interactions like dipole-dipole and dipole induced dipole interaction.

The relaxation time decreases with increase in concentration, is due to the structural relaxation process and in such a situation, it is suggested that the molecule get rearranged due to co-operative process. The Gibb's free energy increase with increasing temperature may be due to the dissociation of intermediate compound between binary liquids. It is observed that free energy increase favours the dissociation of products from reaction.

Internal pressure in a liquid system is a measure of intermolecular cohesive forces [16]. From the Table 3, it is observed that as the mole fraction of Anisole increases, free volume decreases whereas internal pressure increases. This suggests the close packing of the molecule inside the shield, which may be brought about by the increasing magnitude of interactions. [17,18].

It can be observed that the molar volume V_m decreases with increase in concentration of Anisole. This is because of the fact, that molecular weight is directly proportional to the molar volume. Moreover, molar volume increase with rise in temperature in our study, which probably from the fact that thermal energy facilitates an increase in the molecular separation in the liquid mixtures and thus leads to an increase in molar volume.

Table 4 and Figure 11 shows that the available volume V_a decreases with the increase in molefraction of Anisole. Available volume is a direct measure of compactness and strength of bonding between the molecules of the liquid mixture. [14]. The decrease in V_a is due to the net packing of molecules inside the shell which may be formed by complexation between unlike molecules through hydrogen bonding in the binary mixture.

Lennard-Jones potential $\varphi(r)$ is given by the relation [17].

$$\varphi(r) = -Ar^{-6} + Dr^{-n}$$

where r and n are intermolecular distance and Lennard-Jones potential repulsive term exponent respectively. A and D are constants. The first term arises from

attractive forces while the second term arises from repulsive forces. Larger the value of n smaller is the second term. Thus large value of n indicates the dominance of attractive forces over repulsive forces. The values of n increase with the increase in mole fraction of Anisole as shown in Table 4 and Figure 12 for a fixed frequency. The increase in n indicates the increasing dominance of attractive force over repulsive forces in the binary liquid mixture. Further, the values of n decrease with the increase in temperature for a particular concentration which indicates the increase in repulsive forces due to reduction in molecular interaction in the binary mixture [18].

Conclusion

The ultrasonic method was found to be a powerful tool in characterizing the physico-chemical behavior of liquid mixtures. It is concluded from the experimental and calculated parameters that the density, viscosity, velocity increases with increase in mole fraction of Anisole. This is due to the dipole induced dipole interaction between the binary mixture. The temperature variation indicates that the strength of intermolecular interaction decreases with rise in temperature.

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