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Research Article

Study of acoustic and thermodynamic parameters for binary mixture containing cyclohexane and the substituted benzenes at different temperatures

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Abstract: Ultrasonic velocity, density and viscosity have been measured for binary mixtures of Cyclohexane and the substituted benzene at different temperatures 288K, 298K, 308K, 318K, the frequency being fixed at 6 MHz. These experimental data have been used to study the different thermodynamic parameters and their excess values.

Keywords: Binary mixtures, ultrasonic velocity, internal pressure, adiabatic compressibility, Gibbs' free energy.

INTRODUCTION

Ultrasonic investigation of liquid mixtures consisting of polar and non-polar components, are of considerable importance in understanding the intermolecular interaction between the component molecules and find applications in several industrial and technological processes[1-7]. The variation in ultrasonic velocity and related parameters throw light upon the structural changes associated with the liquid mixtures having strongly as well as weakly interacting components. This has been studied for various binary and ternary mixtures[8-9] with respect to variation in concentration and temperature.

In the present paper various acoustic and the derived thermodynamic parameters along with their excess values are studied for four binary mixtures containing Cyclohexane and the substituted benzene group of liquids at different temperatures.

Mixture - I: Cyclohexane + Benzene

Mixture - II: Cyclohexane + Chlorobenzene

Mixture - III: Cyclohexane + NitrobBenzene

Mixture - IV: Cyclohexane + Pyridine

Cyclohexane belongs to the alicyclic hydrocarbon. It is non-polar, unassociated, inert hydrocarbon and has globular structure. It is highly inert towards an electrophille or nucleophille at ordinary temperature. Due to the above properties of Cyclohexane, dispersive types of interactions are possible between it and other components.

The liquid which are successively used along with Cyclohexane are Benzene, Chlorobenzene, Nitrobenzene and Pyridine.

Benzene is a non-polar solvent. It is a cyclic hydrocarbon with a continuous pi bond. Chlorobenzene is a poor electron donor towards the electron seeking proton of any group. It has low dielectric constant and dipole moment. The chlorine atom being an electron withdrawing atom attracts the π-electron of benzene ring and thus a decrease of electron density of the ring takes place. This makes the benzene ring a relatively poor electron donor towards the Cyclohexane molecules.

Nitrobenzene is a polar solvent with high dielectric constant and dipole moment. Hence intermolecular interaction in this case is large.

Pyridine is a basic heterocyclic organic compound with a lower dielectric constant and dipole moment. Pyridine molecules are spherical in shape and tightly packed.

Experimental technique: The liquid mixtures of fixed concentration (6:4) in mole fraction were prepared by taking analytical reagent grade and spectroscopic reagent grade chemicals with minimum assay of 99.9% and obtained from Merck Ltd (India)[10].

The density, viscosity, and ultrasonic velocity of all liquid mixtures were measured at temperatures 288K, 298K, 308K, 318K and at frequency 6MHz.

Ultrasonic velocity measurements were made using an ultrasonic interferometer (Model M-84, supplied by M/S Mittal Enterprises, New Delhi), at different temperatures and different frequencies with the accuracy of ±0.1m·s−1. The measuring cell of interferometer is a specially designed double-walled vessel with provision for temperature constancy. An electronically operated digital constant temperature bath (Model SSI-03 Spl, supplied by M/S Mittal Enterprises, New Delhi), operating in the temperature range of−10°C to 85°C 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. The specific gravity bottle with the experimental mixture was immersed in the temperature-controlled water bath. The weight of the sample was measured using an electronic digital balance with an accuracy of ±0.1 mg (Model: SHIMADZU AX-200, Kyoto, Japan).

An Oswald viscometer (10 ml) with an accuracy of $\pm$ 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

1. **Adiabatic compressibility:** The ultrasonic velocity in a liquid medium, in terms of Bulk modulus (B) and density of the medium is given by the Newton-Laplace equation[11].

$$U = \sqrt{\frac{B}{\rho}} = \sqrt{\frac{1}{\rho.\beta}}$$

Or, $\beta = \frac{1}{\rho.U^2}.........\left(N^{-1}.m^2\right)$ ... (1)

2. **Intermolecular free length:** Intermolecular free length (L_f), is calculated using the standard expression [12]

$L_f = K_T.\beta^{1/2}............(m)$ ... (2)

Where, 'K_T' $\{=(93.875 + 0.375.T) \times 10^{-8}\}$ is Jacobson's temperature dependent constant and 'β' is the adiabatic compressibility.

3. **Free Volume:** Suryanarayana *et al.*[13,14] obtained a relation for free volume in terms of ultrasonic velocity (U) and the viscosity of the liquid (η) as,

$$V_f = \left(\frac{M_{eff}.U}{K.\eta}\right)^{3/2}.........\left(m^3.mol^{-1}\right) \quad \quad \text{... (3)}$$

Where 'M_{eff}' is the effective mass of the mixture, 'K' is a dimensionless constant independent of temperature and liquid. Its value is 4.281×10^9.

4. **Internal Pressure:** Internal pressure can be calculated by using the relation[15,16]

$$\pi_i = bRT\left(\frac{K\eta}{U}\right)^{\frac{1}{2}}\left(\frac{\rho^{\frac{2}{3}}}{M^{\frac{7}{6}}}\right)............(N.m^{-2}) \quad \quad \text{... (4)}$$

Where, 'b' stands for the cubic packing factor, which is assumed to be '2' for all liquids and solutions. 'K' is a dimensionless constant independent of temperature and nature of liquids. Its value is 4.281×10^9, R is the gas constant, T is the absolute temperature, η is the viscosity, U is the ultrasonic velocity, ρ is the density and M is the effective molecular weight.

5. **Viscous Relaxation Time (τ):** The relaxation time can be calculated using the relation[17,]

$$\tau = \frac{4}{3}.(\beta.\eta)............(s) \quad \quad \text{... (5)}$$

Where, 'β' is the adiabatic compressibility and 'η' is the viscosity of the mixture.

6. **Acoustic impedance (Z):** The specific acoustic impendence is given by[18]

$$Z = \rho.U.........(Kg.m^2.s^{-1}) \quad \quad \text{... (6)}$$

7. **Gibb's free energy:** The variation of 'τ' with temperature can be expressed in the form of Eyring salt process theory[17].

$$\frac{1}{\tau} = \frac{KT}{h}exp\left(\frac{-\Delta G}{KT}\right)$$

The above equation can be rearranged as

$$\Delta G = 2.30.KT\log\left(\frac{KT\tau}{h}\right) \;\dots \text{(k.J.mol}^{-1}) \qquad \dots \textbf{(7)}$$

Where, 'τ' is the viscous relaxation time, 'T' is the absolute temperature, 'K' is the Boltzmann's constant and 'h' is the Plank's constant.

8. **Molar volume:** Molar volume can be calculated by using the relation[19]

$$V_m = \frac{M_{eff}}{\rho}\dots\dots\dots \text{(m}^3 \text{ mol}^{-1}) \qquad \dots\textbf{(8)}$$

Where, M_{eff} is the effective molecular weight and ρ is the density of the solution.

9. **Available volume:** Available volume is the direct measure of compactness and strength of binding between the molecules of liquid or liquid mixture. Schaffs *et al.*[20] shown that the available volume can be obtained by the relation
    Another parameter which can be calculate from ultrasonic velocity is the available volume and is given by

$$V_a = V_m.\left(1 - \frac{U}{U_\infty}\right)\dots \text{(m}^3 \text{ mol}^{-1}) \qquad \dots \textbf{(9)}$$

Where, $V_m = M/\rho$, is the molar volume, U = velocity, and U_∞ = Schaff's limiting value taken as 1600 m/s for liquids.

10. **Rao's constant:** Rao's constant is also known as molar sound velocity and it is an additive property. It has been found to be invariant with temperature and pressure for un-associated organic and inorganic liquid. R can be evaluated by an equation given by Bagchi *et al*[21]

$$R = \left(\frac{M_{eff}}{\rho}\right).U^{\frac{1}{3}} = V_m.U^{\frac{1}{3}}\dots \text{(m}^3.\text{mol}^{-1}.\text{(m/s)}^{1/3}) \;\dots\textbf{(10)}$$

11. **Wada's constant:-**

    Molar compressibility is also known as Wada's constant, which is dependent on adiabatic compressibility and density, is given by the relation[22]

$$W = \left(\frac{M_{eff}}{\rho}\right).\beta^{-\frac{1}{7}} = V_m.\beta^{-\frac{1}{7}}\dots \text{(m}^3.\text{mol}^{-1}.\text{(Kg}^{-1}.\text{ms}^2)^{1/7}) \qquad \dots \textbf{(11)}$$

12. **Surface tension:-**
    Surface tension can be calculated by using the relation[23]

$$S = 6.3 \text{X} 10^{-4}.\rho.U^{\frac{3}{2}}\dots\dots\dots.. \text{(N.m}^{-1}) \qquad \dots \textbf{(12)}$$

RESULT AND DISCUSSION

The experimental values of density, viscosity and velocity are presented in table-1. Calculated values of acoustic and thermodynamic parameters are presented in table-2 to table-5 and excess values of the parameters are shown in table-6 to table-8. Variations of some parameters with temperature are shown in **Fig.1 To Fig.-8.**

Table – 1: Values of Density (ρ), Viscosity (η) and velocity (U) in binary mixtures for different temperatures at 6 MHz frequency.

Binary mixture	Density (ρ) Kg.m^{-3}				Viscosity (η x 10^{-3}) (N.s.m^{-2})				Velocity (U) m.s^{-2}			
	288 K	298 K	308 K	318 K	288 K	298 K	308 K	318 K	288 K	298 K	308 K	318 K
M - I: Ben + C.H	835.26	830.94	824.87	818.23	0.912	0.680	0.536	0.459	1280.4	1247.7	1220.1	1165.1
M- II: C.Ben + C.H	998.17	991.77	984.93	975.25	1.087	0.840	0.651	0.549	1276.4	1242	1206	1161.4
M- III: N.Ben C.H	869.94	864.22	859.31	856.69	1.023	0.933	0.835	0.646	1274.4	1241.8	1205.9	1156.1
M - IV: Pyr. + C.H	887.56	882.59	876.93	869.56	1.044	0.829	0.689	0.571	1330.2	1290.5	1265.4	1224.6

Table –2: Calculated values of adiabatic compressibility (β), free length (L$_f$) and free volume (V$_f$) in binary mixtures for different temperatures at 6 MHz frequency.

Binary mixture	Adiabatic compressibility (β) (10^{-10}N^{-1}.m^2)				Free length (L$_f$) (10^{-10} m)				Free volume (V$_f$) (10^{-7} m^3.mol^{-1})			
	288 K	298 K	308 K	318 K	288 K	298 K	308 K	318 K	288 K	298 K	308 K	318 K
M - I: Ben + C.H	7.303	7.731	8.144	9.003	0.530	0.551	0.577	0.609	1.357	2.029	2.799	3.305
M - II: C.Ben + C.H	6.149	6.537	6.981	7.602	0.486	0.506	0.534	0.560	1.463	2.066	2.899	3.540
M-III: N.Ben + C.H	7.078	7.504	8.002	8.733	0.521	0.542	0.572	0.600	1.749	1.931	2.183	3.013
M - IV: Pyr. + C.H	6.367	6.803	7.122	7.669	0.495	0.517	0.539	0.562	1.186	1.602	2.053	2.591

Table – 3: Calculated values of viscous relaxation time (τ), Gibb's free energy (ΔG) and acoustic impedance (Z) in binary mixtures for different temperatures at 6 MHz frequency.

Binary mixture	Viscous relaxation time (τ) (x 10^{-12} s)				Gibb's free energy (ΔG) (x 10^{-20} k.J.mol^{-1})				Acoustic impedance (Z) (x 10^6 Kg.m^2.s^{-1})			
	288 K	298 K	308 K	318 K	288 K	298 K	308 K	318 K	288 K	298 K	308 K	318 K
M - I: Ben + C.H	0.888	0.701	0.582	0.550	0.665	0.605	0.561	0.568	1.069	1.037	1.006	1.069
M - II: C.Ben + C.H	0.891	0.732	0.606	0.556	0.667	0.623	0.577	0.573	1.274	1.232	1.188	1.133
M - III: N.Ben + C.H	0.966	0.934	0.891	0.752	0.699	0.723	0.742	0.705	1.109	1.073	1.036	1.099
M - IV: Pyr. + C.H	0.886	0.752	0.654	0.584	0.665	0.634	0.610	0.594	1.181	1.139	1.110	1.181

Table – 4: Calculated values of internal pressure, available volume (V_a) and molar volume (V_m) in binary mixtures for different temperatures at 6 MHz frequency.

Binary mixture	Internal pressure(Π_i) (x 10^6 N.m^{-2})				Available volume (V_a) (m$^3\cdot$mol^{-1})				Molar volume (V_m) (m$^3\cdot$mol^{-1})			
	288 K	298 K	308 K	318 K	288 K	298 K	308 K	318 K	288 K	298 K	308 K	318 K
M - I: Ben + C.H	443.3	399.7	369.3	443.3	0.019	0.021	0.023	0.027	0.096	0.097	0.098	0.098
M-II: C.Ben +C.H	418.0	383.9	352.8	338.5	0.020	0.023	0.025	0.028	0.101	0.102	0.103	0.104
M-III: N.Ben +C.H	345.2	344.0	340.1	314.5	0.025	0.028	0.031	0.035	0.124	0.124	0.125	0.125
M - IV: Pyr. + C.H	480.3	448.0	424.4	480.3	0.015	0.018	0.019	0.022	0.091	0.092	0.093	0.093

Table – 5: Calculated values of Surface tension (S) in binary mixtures for different temperatures at 6 MHz frequency.

Binary mixture	Surface tension (S) $(N.m^{-1})$			
	288 K	298 K	308 K	318 K
M - I: Ben + C.H	24109.1	23071.5	22147.2	20500.3
M - II: C.Ben + C.H	28676.4	27348.5	25987.6	24318.1
M - III: N.Ben + C.H	24933.7	23825.4	22670.4	21215.7
M - IV: Pyr. + C.H	27127.7	25777.2	24868.4	23476.4

Table – 6: Excess values of adiabatic compressibility, free length and free volume in binary mixtures for different temperatures at 6 MHz frequency.

Binary mixture	Excess Adiabatic comp. (β^E) $(10^{-10}N^{-1}.m^2)$				Excess Free length (L^E) $(10^{-10}$ m)				Excess Free volume (V_f^E) $(10^{-7}$ $m^3.mol^{-1})$			
	288 K	298 K	308 K	318 K	288 K	298 K	308 K	318 K	288 K	298 K	308 K	318 K
M - I: Ben + C.H	0.52	0.350	0.138	0.324	0.020	0.013	0.006	0.012	0.298	0.551	0.828	0.894
M - II: C.Ben + C.H	-0.05	-0.163	-0.259	-0.209	-0.000	-0.004	-0.007	-0.005	-0.032	-0.025	0.205	0.230
M - III: N.Ben + C.H	1.824	1.836	1.895	2.148	0.079	0.079	0.082	0.089	0.635	0.396	0.077	0.343
M - IV: Pyr. + C.H	0.47	0.461	0.247	0.258	0.022	0.021	0.014	0.014	0.400	0.512	0.546	0.674

Table – 7: Excess values of internal pressure, Acoustic impedance and Gibb's free energy in Binary mixtures for different temperatures at 6 MHz frequency.

Binary mixture	Excess internal pressure(Π_i^E) (x 10^6 N.m^{-2})				Excess acoustic impedance (Z^E) (x 10^6 Kg.m^2.s^{-1})				Excess Gibb's free energy (ΔG^E) (x 10^{-20} k.J.mol^{-1})			
	288 K	298 K	308 K	318 K	288 K	298 K	308 K	318 K	288 K	298 K	308 K	318 K
M - I: Ben + C.H	-41.910	-47.807	-50.595	-46.238	-0.048	-0.031	-0.016	-0.025	-0.050	-0.077	-0.097	-0.087
M - II: C.Ben + C.H	7.36	5.790	-3.040	-1.909	0.000	0.008	0.012	0.015	-0.001	-0.001	-0.030	-0.026
M - III: N.Ben + C.H	-104.14	-71.63	-45.75	-51.09	-0.379	-0.359	-0.349	-0.34	-0.011	0.046	0.102	0.081
M - IV: Pyr. + C.H	-100.57	-90.739	-77.068	-73.627	-0.084	-0.081	-0.061	-0.059	-0.106	-0.105	-0.098	-0.101

Table – 8: Excess values of viscous relaxation time (τ^E) and surface tension (S^E) in Binary mixtures for different temperatures at 6 MHz frequency.

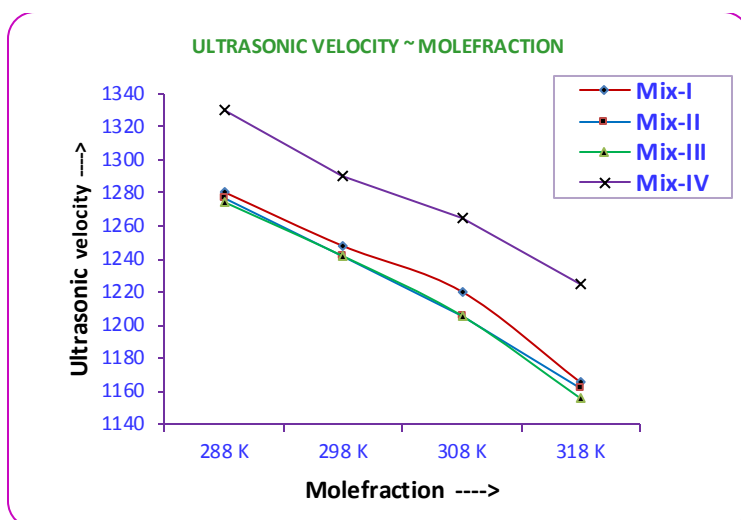| Binary mixture	Excess vis. relaxation time (τ^E) (x 10^{-12} s)				Excess surface tension (S^E) (N.m^{-1})			
	288 K	298 K	308 K	318 K	-1570.9	-998.7	-490.0	-760.7
M - I: Ben + C.H	-0.121	-0.145	-0.149	-0.121	-187.7	-28.2	99.5	49.0
M - II: C.Ben + C.H	-0.014	-0.022	-0.049	-0.037	-10447	-9905.4	-9529.7	-8959.1
M - III: N.Ben + C.H	-0.030	0.097	0.188	0.116	-5229.3	-3744.7	-2141.4	-672.5
M - IV: Pyr. + C.H	-0.272	-0.219	-0.172	-0.153	-1570.9	-998.7	-490.0	-760.7

Fig.1: Variation of ultrasonic velocity with temp.
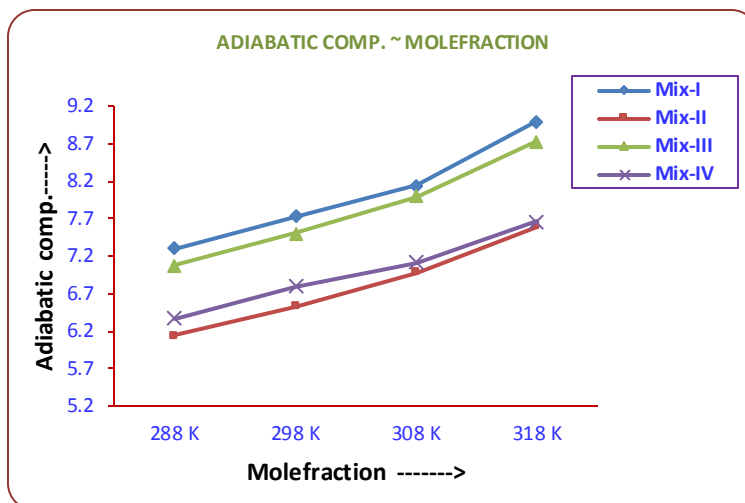

Fig. 2: Variation of adiabatic compressibility with temp.
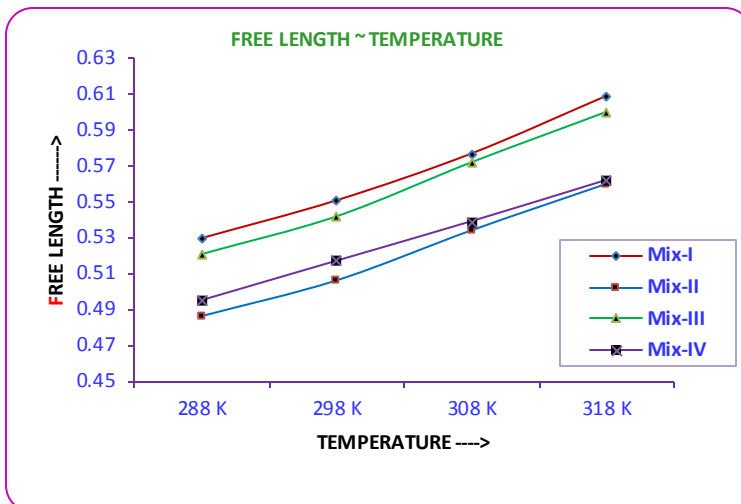

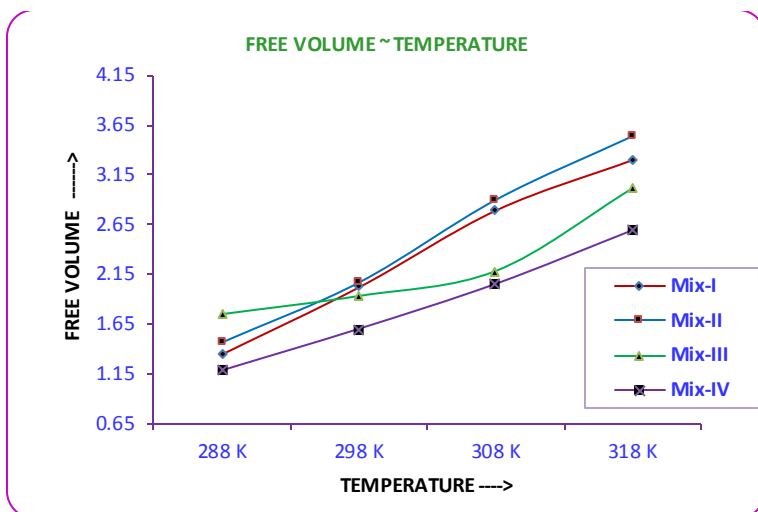
Fig.3: Variation of free length with temp.

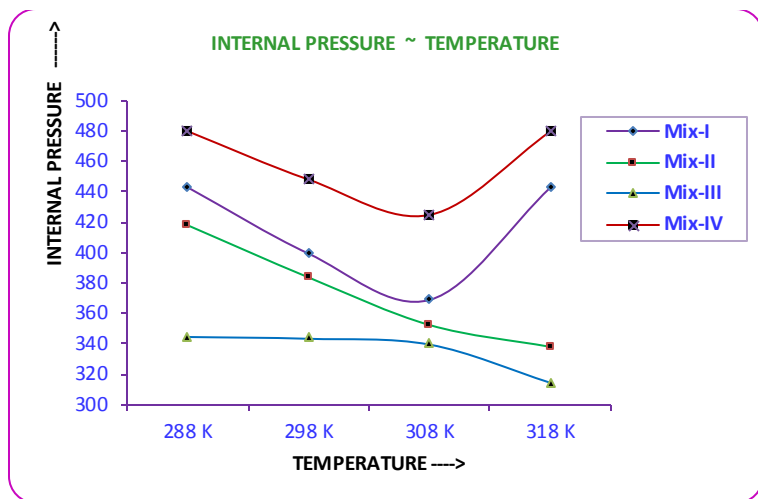**Fig.4:** Variation of free volume with temp.
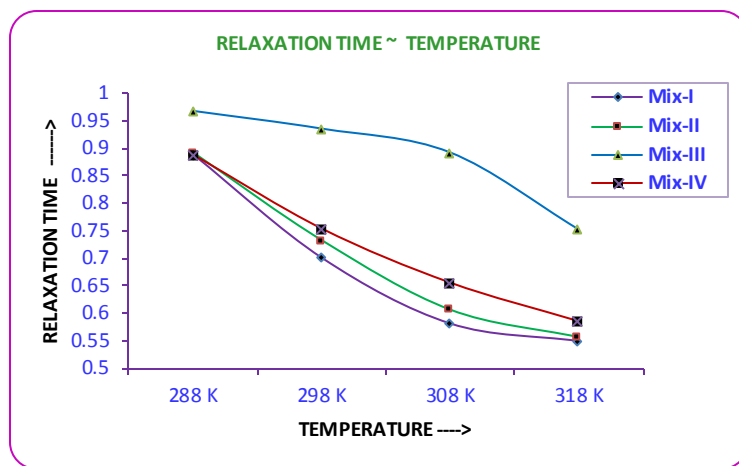

Fig.5: Variation of internal pressure with temp.
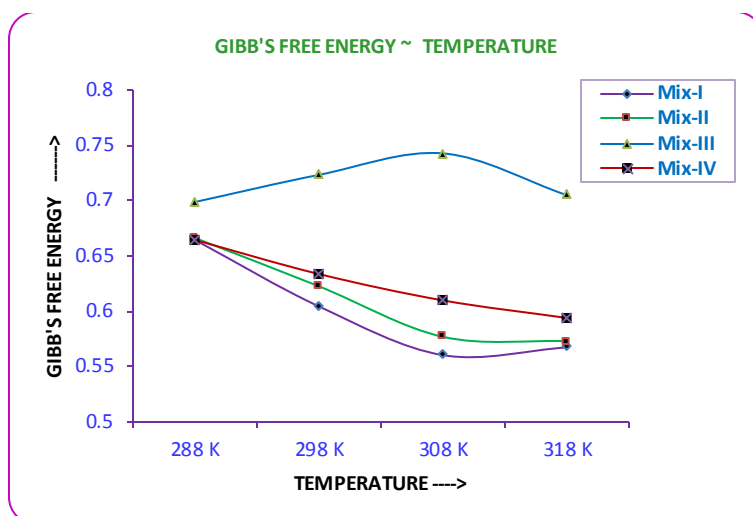

Fig.6: Variation of relaxation time with temp.

GIBB'S FREE ENERGY ~ TEMPERATURE

Fig.7: Variation of Gibb's free energy with temp.

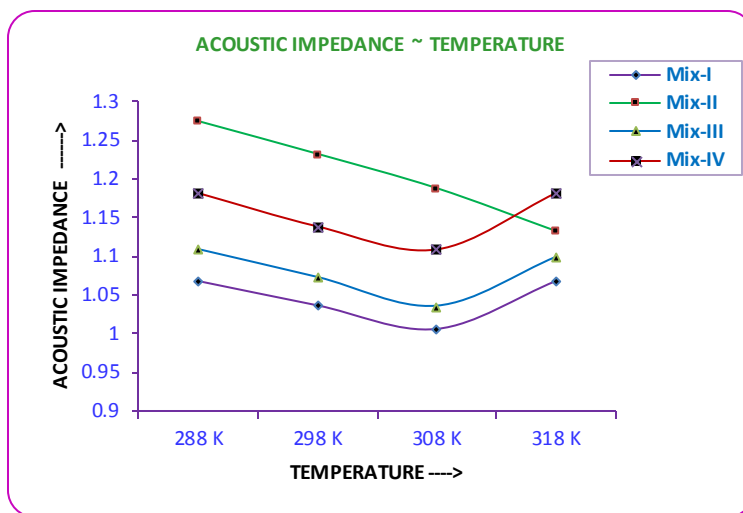**ACOUSTIC IMPEDANCE ~ TEMPERATURE**

Fig.8: Variation of acoustic impedance with temp.Aa

Intermolecular free length is the distance between the surfaces of the neighboring molecules in the mixture. Variation in free length indicates variation in molecular forces in the mixture which depends on the experimental density as well as temperature. At any temperature (say 318 K) free length is minimum for mixture of chlorobenzene + cyclohexane and maximum for the mixture of benzene + cyclohexane, this is because the density of the former mixture is maximum and that for the latter is minimum. However interaction in both the mixtures is weak. In the chlorobenzene + cyclohexane mixture, the benzene ring in chlorobenzene is a poor electron donor towards the cyclohexane molecules. Further since benzene and cyclohexane both are non-polar, the interaction between them is also weak.

Ultrasonic velocity in a solution depends on free length. As free length increases with temperature, ultrasonic velocity decreases with increase in temperature.

Adiabatic compressibility and free volume show a reverse trend i.e they increase as temperature increases. At a particular temperature (say 318 K) free volume is minimum for mixture of pyridine + cyclohexane and increases from nitrobenzene + cyclohexane to benzene + cyclohexane to chlorobenzene + cyclohexane. Free volume is more in case of chlorobenzene + cyclohexane and benzene + cyclohexane

mixtures the intermolecular interactions in both the cases being small. It is minimum for pyridine + cyclohexane as pyridine molecules are spherical in shape and are closely packed. Further a dipole – induced dipole interaction with cyclohexane may cause a decrease in volume. In the mixture nitrobenzene + cyclohexane, nitrobenzene also has a larger dipole moment, but the interaction with cyclohexane may be small due to steric hindrance.

When the concentrations of the liquids change or the molecules are closely packed the effective free volume sometimes changes due to the transmission of collision effect through the molecules. The change in adiabatic compressibility thus does not follow the same pattern as that of free volume. It is minimum for chlorobenzene + cyclohexane mixture and increases successively from pyridine + cyclohexane to nitrobenzene + cyclohexane to benzene + cyclohexane. This may be due to the effect of density on adiabatic compressibility.

In all the binary mixtures, internal pressure decreases as temperature increases indicating weakening of interaction. At a particular temperature (say 318 k), internal pressure is the minimum for nitrobenzene + cyclohexane mixture and is maximum for pyridine + cyclohexane mixture. Pyridine molecules are spherical in shape, closely packed and have a finite dipole moment (2.2 D). Hence the intramolecular and intermolecular interaction is maximum in the case of pyridine mixture. Nitrobenze has a high dipole moment, but the complex structure of nitrobenzene molecules leads to less intermolecular interaction due to steric hindrance. Same structure also gives a less force of cohesion.

Acoustic impedance decreases as temperature increases for all the binary mixtures. It is the minimum for benzene + cyclohexane mixture amongst all the binary mixtures at all temperatures. This is so because both benzene and cyclohexane are non-polar and only dispersive forces act between them.

Molecular volume increases with rise in temperature. This is because thermal energy facilitates increase in molecular separation. Molar volume should increase with molecular weight, which is evident in our observation. Available volume changes in the same way as molar volume. Available volume is a direct measure of the compactness and strength of bonding between the molecules of the liquid mixture.

Viscous relaxation time and the Gibb's free energy both decrease as temperature increases. As temperature increases excitation energy increases and hence relaxation time decreases. Relaxation time is the time taken for the excitation energy to appear as translational energy. Further since kinetic energy of the molecules increases, longer time is taken for rearrangement of molecules and this suggests a decrease in Gibb's free energy.

Surface tension is minimum for benzene + cyclohexane mixture and maximum for chlorobenzene + cyclohexane mixture. This is because surface tension depends on density and velocity ($S \sim \rho.U^{3/2}$)

Excess values of thermodynamic parameters play a major role in understanding the nature of molecular interaction. Excess free volume is the result of contribution from several opposing effects. This may be divided in to three types; physical, chemical and structural.

Physical effect, which arises due to dipole-induced dipole interaction resulting in disruption of the bonds, gives a positive value of excess free volume. Chemical or specific intermolecular interaction (formation of new bond) results in a volume decrease giving negative value of excess free volume. Structural contribution gives mostly negative value to excess free volume.

Excess free volume which is positive in all the binary mixtures is supported by negative excess viscosity and negative excess velocity. Negative excess viscosity predicts easier flow of liquid mixture.

Excess internal pressure is negative in all the mixtures indicating weak intermolecular interaction. In chlorobenzene + cyclohexane mixture, it however changes from positive to negative as temperature increases. This is consistent with the change in the excess free volume and excess surface tension, which change from negative to positive.

Sati et al. attribute negative values of excess Gibb's free energy to weak interaction (dispersion forces) between molecules. Excess Gibb's free energy is found to be negative in all the binary mixtures except in the mixture nitrobenzene + cyclohexane. For this mixture it is negative at low temperatures and becomes positive at higher temperature. Low temperature effect may be due to steric hindrance. Positive excess values indicate charge transfer interaction.

It is seen that the excess density is negative for the mixtures of benzene + cyclohexane, nitrobenzene + cyclohexane and pyridine + cyclohexane being the maximum negative for nitrobenzene + cyclohexane mixture. This indicates that, the density decreases when the mixtures are formed. The excess value of density is positive for the mixture of chlorobenzene + cyclohexane.

CONCLUSION

Study of the thermodynamic parameters and their excess values in the binary mixtures indicate the nature of interaction between Cyclohexane and the benzene group of liquids. Although Cyclohexane is non-polar the intermolecular interaction is evident through the excess values of the thermodynamic parameters.

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