

Physico-Chemical Properties of Binary Mixture of 2,3-Dichloroaniline and Carbon Tetrachloride at 300 K

MAHENDRA KUMAR^{1,2,*}, CHANDRESHVAR PRASAD YADAV¹, DHARMENDRA KUMAR PANDEY¹ and DHANANJAY SINGH³

¹Department of Physics, P.P.N. (P.G.) College (Affiliated to Chhatrapati Shahu Ji Maharaj University, Kanpur), Kanpur-208001, India

²Department of Physics, D.B.S. (P.G.) College (Affiliated to Chhatrapati Shahu Ji Maharaj University, Kanpur), Kanpur-208006, India

³Department of Chemistry, P.P.N. (P.G.) College (Affiliated to Chhatrapati Shahu Ji Maharaj University, Kanpur), Kanpur-208001, India

*Corresponding author: E-mail: mahendrakumar.physics@gmail.com

Received: 28 May 2021;

Accepted: 24 July 2021;

Published online: 20 September 2021;

AJC-20511

The physico-chemical properties of binary mixture of 2,3-dichloroaniline (2,3-DCA) and carbon tetrachloride (CCl₄) have been studied in the present work by the measurement of density, viscosity, ultrasonic velocity and estimation of thermo-physical parameters at 300 K. The density, compressibility, free length, relative association and free volume were found to increase while viscosity, ultrasonic velocity, acoustic impedance and internal pressure were received to decay with an increase in mole fraction of solvent in the binary mixture. The thermal relaxation time and Gibb's free energy were found to decay with a slow rate up to 50% mole fraction of the solvent, after that these quantities decay very fast in chosen binary mixture. The ultrasonic absorption was also found to increase slowly up to 50% mole fraction of solvent and after that a fast decay was observed in the present binary mixture. The molecular interaction, structural ordering, stability and related features of the prepared binary mixture were also analyzed based on measured and estimated parameters.

Keywords: Binary mixture, Thermo-physical properties, 2,3-Dichloroaniline, Carbon tetrachloride.

INTRODUCTION

In recent years, behaviour of mixed components rather than single component has much importance because of their wide range of applications in the field of chemicals and industrial processes. For the investigation of thermodynamical properties of mixture, practical aspects are needed for the study of multi-component systems. The ultrasonic parameters provide insight into the structure of binary mixture and intensity of intermolecular interactions among components of binary mixtures as they are related to several thermophysical properties [1-4]. The magnitude of non-linear deviations from ideal values of velocities, densities and viscosities of liquid mixtures with composition is attributed to difference in molecular size, molar mass, temperature, pressure, mole fraction and strength of interaction between unlike molecules. The analysis of molecular interaction on the knowledge of variation in thermodynamic parameters and their excess values with composition provides an insight into the molecular process [5-12]. In modern times, ultrasonic studies are extensively carried out to measure thermo-

dynamic properties of liquids and predict intermolecular interaction between liquid mixtures.

2,3-Dichloroaniline (2,3-DCA) is a polar liquid having associative nature whereas carbon tetrachloride (CCl₄) is a non-polar liquid with zero dipole moment and has a low dielectric constant. The liquid 2,3-DCA is a base substance for synthesis and a vital chemical substance for manufacturing of medicines, herbicides, pesticides, agricultural fungicides, insecticides and dyes, *etc.* [13-20]. Carbon tetrachloride is a colourless, dense, highly toxic, volatile, non-flammable liquid having a characteristic odour. Carbon tetrachloride is useful in refrigeration fluid, fire extinguishers, propellants and dry-cleaning agent. Carbon tetrachloride is a highly potent hepatotoxin that can cause serious damage to liver and can also harmful to central nervous system (CNS) if high adequate concentrations are present [21-25]. The intermolecular interactions for binary mixture of CCl₄ with benzaldehyde [18], acetone [19], benzene and substituted benzenes [20], organic liquids [21], dimethyl carbonate [22], toluene [23], acetylacetone [24], *etc.* have been reported by measurement and estimation of density, viscosity,

ultrasonic velocity and other related parameters [25]. No work on the ultrasonic non-destructive characterization of 2,3-dichloroaniline (2,3-DCA) and its binary mixture with CCl_4 has been found to be reported in literature while they have a prominent role in field of chemical and pharmaceutical industries. In this work, the ultrasonic study of binary mixture 2,3-DCA with CCl_4 to explore their characteristic feature related to molecular interaction at 300 K is carried out.

EXPERIMENTAL

Experimental procedure: The binary mixture can be prepared by taking constituents either by volume percentage or by weight percentage or by both volume and weight percentage [26]. To prepare binary mixture in present study, 2,3-dichloroaniline (solute, 98% purity, S.D. Fine Chem. Ltd., India) and carbon tetrachloride (solvent, 99% purity, Molychem Chemicals Pvt. Ltd., India) were taken as constituent liquids. Basic specifications of used chemicals are listed in Table-1. No further purification was carried out in the purchased chemicals. Six samples were prepared by mixing constituents in volume by volume ratio with variation in mole fraction (ϕ : 0 to 1) at 300 K. The pressure was measured with digital Barometer and found to be 100.5 ± 0.1 kPa. The volume/mole fraction of components in the prepared binary mixtures is listed in Table-2. The uncertainties in volume/mole fraction for present binary mixtures were ± 0.1 and $\pm 1 \times 10^{-4}$, respectively. No crystallization was observed in the pure and binary samples at 300 K. The prepared homogeneous binary mixtures were kept in borosil bottle with air tight caps to achieve good thermal equilibrium.

The densities of prepared mixtures were measured using density bottle method. The calibration of this method was performed using distilled water. The uncertainty in density measurements was found to be of the order of $\pm 1 \times 10^{-3}$ g cm^{-3} . The temperature of the samples was controlled by using a water bath equipped with a thermostat of accuracy ± 0.1 °C. The Poiseuille method was used for the viscosity measurement of a binary mixture in the present experiment [27-30]. The experimental viscosity was determined by taking the average of five outflow time readings. For temperature constancy in the viscosity

measurement unit, a water bath thermostat is used with an accuracy of ± 0.1 °C. The uncertainty in present viscosity measurement was $\pm 2.1\%$.

The ultrasonic velocity of the liquid mixtures has been measured using an ultrasonic interferometer (M/s Mittal Enterprises, Model F-81) working at 2 MHz frequency with an accuracy of ± 0.1 m s^{-1} . The reliability of measurement was checked by obtaining sound velocity data for distilled water at 300 K. The sound velocity in distilled water at 300 K and at 1 atm was found to be 1502.5 ± 0.6 m s^{-1} . The reported ultrasonic velocity in distilled water at 25 °C and 28 °C is 1496.65 ± 0.1 m s^{-1} and 1504.29 ± 0.1 m s^{-1} , respectively [31]. The uncertainty in ultrasonic velocity measurements is ± 0.6 m s^{-1} . The required properties are measured for freshly prepared binary mixture and needed precautions to reduce the possible experimental error are taken for all measurements. Since both measured and literature velocities were found in good agreement, thus used velocity measurement unit in the present study provides accurate value with high precision.

Thermo-physical parameters: The experimental values of density (ρ), viscosity (η) and ultrasonic velocity (U) of binary mixtures have been used for the evaluation of compressibility (β), acoustic impedance (Z), free length (L_f), excess molar volume (V^{Ex}), free volume (V_f), internal pressure (π_i), relative association (R_A), thermal relaxation time (τ), Gibbs free energy (ΔG) and ultrasonic absorption (α/f^2) to explore the nature and strength of intermolecular interactions. The related formulations are given as follows [32-34]:

$$\beta = \frac{1}{\rho U^2} \quad (1)$$

$$Z = \rho U \quad (2)$$

$$L_f = K_T \beta^{1/2} \quad (3)$$

$$V^{\text{Ex}} = \left(\frac{x_1 M_1 + x_2 M_2}{\rho_{\text{mix}}} \right) + \left(\frac{x_1 M_1}{\rho_1} + \frac{x_2 M_2}{\rho_2} \right) \quad (4)$$

TABLE-1
CHEMICALS USED WITH THEIR NAME/FORMULA, ABBREVIATION, CAS NUMBER, SOURCE, MOLAR MASS, MOLE FRACTION PURITY, PURIFICATION METHOD, ANALYSIS METHOD AND WATER CONTENT (%)

Chemical name/formula	Abbreviation	CAS number	Source	Molar mass (g mol^{-1})	Mole fraction purity* (%)	Analysis Method	Water content** (%)
2,3-Dichloroaniline ($\text{C}_6\text{H}_5\text{NCl}_2$)	2,3-DCA	608-27-5	S.D. Fine Chem. Ltd., Mumbai, India	162.01	98	GC [#]	Not detected
Carbon tetrachloride	CCl_4	56-23-5	Molychem Chem. Pvt. Ltd Mumbai, India	153.80	99	GC [#]	0.02

*Purity, **Water content and analysis method [#]GC (gas chromatography) as stated by the supplier.

TABLE-2
VOLUME/MOLE FRACTION OF COMPONENT IN BINARY SAMPLES OF 2,3-DCA WITH CCl_4

Components		$D_1\text{C}_0$	$D_{0.8}\text{C}_{0.2}$	$D_{0.6}\text{C}_{0.4}$	$D_{0.4}\text{C}_{0.6}$	$D_{0.2}\text{C}_{0.8}$	$D_0\text{C}_1$
2,3-DCA	Volume fraction	1	0.8	0.6	0.4	0.2	0
	Mole fraction	1	0.7715	0.5587	0.3601	0.1743	0
CCl_4	Volume fraction	0	0.2	0.4	0.6	0.8	1
	Mole fraction	0	0.2285	0.4413	0.6399	0.8257	1

$$V_f = \left(\frac{M_{\text{eff}} U}{\eta K} \right)^{3/2} \quad (5)$$

$$\pi_i = bRT \left(\frac{K\eta}{U} \right)^{1/2} \left(\frac{\rho^{3/2}}{M^{7/6}} \right) \quad (6)$$

$$R_A = \frac{\rho}{\rho_0} \left(\frac{U}{U_0} \right)^{3/2} \quad (7)$$

$$\tau = \frac{4\beta\eta}{3} \quad (8)$$

$$\Delta G = -KT \log \left(\frac{h}{KT\tau} \right) \quad (9)$$

$$\left(\frac{\alpha}{f^2} \right) = \frac{8\pi^2\eta}{3\rho U^3} \quad (10)$$

where, K_T is temperature-dependent constant and its value is 198×10^8 . The quantity 'b' is cubic packing factor assumed to be 2 for all liquids. T is absolute temperature and R is constant having value $1.0974 \times 10^5 \text{ cm}^{-1}$. Term h is Planck's constant. ρ_0 and U_0 are density and ultrasonic velocity of pure solvent while K is temperature-independent constant and its value is 4.28×10^9 . The effective molecular weight (M_{eff}) of binary

mixture is given as $\sum_{i=1}^n x_i M_i$. where, x_i and M_i are mole fraction and molecular weight of individual component in mixture. The ρ_{mix} is density of binary mixture while quantities (x_1, M_1, ρ_1) and (x_2, M_2, ρ_2) are the mole fraction, molar mass and density of pure components, 2,3-dichloroaniline (2,3-DCA) and carbon tetrachloride (CCl_4), respectively.

RESULTS AND DISCUSSION

The measured density, viscosity, ultrasonic velocity for individual liquids 2,3-dichloroaniline (2,3-DCA) and carbon

tetrachloride (CCl_4) were found very close to the literature values (Table-3) [14,18,19,21,24,25]. Experimental value of density, viscosity and ultrasonic velocity of binary mixture 2,3-DCA with CCl_4 at 300 K and atmospheric pressure 100.5 kPa are presented in Table-4. The acoustical and thermodynamic parameters of the binary mixtures were calculated with the help of measured quantities and formulation (eqns. 1-10) are given in Table-5.

The measured density was found to increase while the viscosity and ultrasonic velocity were expected to decrease with an increase in the mole fraction of solvent in the binary mixtures (Fig. 1). The same feature of density, viscosity and ultrasonic velocity has been also reported for binary mixture of CCl_4 with acetone, benzene, dimethyl carbonate [19,20,22, 25]. An increase in density with an increase in mole fraction of CCl_4 indicates that solute-solvent interactions resulted in the formation of bonding. The decrease in viscosity is due to weak inter- and intramolecular forces between constituents with an increase in the mole fraction of solvent in the binary

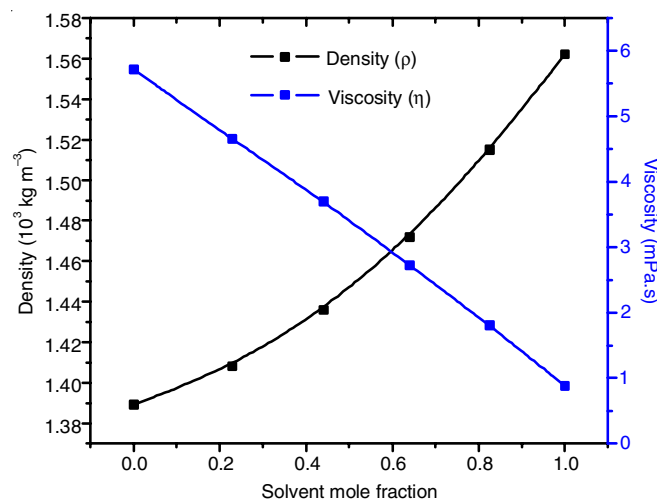


Fig. 1. Density and viscosity vs. solvent mole fraction of 2,3-DCA + CCl_4

TABLE-3
MEASURED AND LITERATURE DENSITY, VISCOSITY AND ULTRASONIC VELOCITY OF PURE 2,3-DCA AND CCl_4 AT 300 K

Components	Temp. (K)	Density (10^3 kg m^{-3})		Viscosity (mPa.s)		Velocity (m s^{-1})	
		Present	Lit.	Present	Lit.	Present	Lit.
2,3-DCA	300	1.389	–	5.717	–	1460.7	–
	317	–	1.378 [14]	–	3.74 [14]	–	1458.0 [14]
CCl_4	296	–	1.596 [21]	–	–	–	929.5 [21]
	300	1.562	–	0.879	–	916	–
	303	–	1.559 [25]	–	–	–	906 [19,25]
	308	–	1.544 [18]	–	0.796 [24]	–	–

TABLE-4
EXPERIMENTAL VALUE OF DENSITY, VISCOSITY AND ULTRASONIC VELOCITY AS A FUNCTION OF SOLVENT MOLE FRACTION OF BINARY MIXTURE OF 2,3-DCA WITH CCl_4 AT 300 K AND ATMOSPHERIC PRESSURE 100.5 kPa

Properties	D_1C_0	$D_{0.8}C_{0.2}$	$D_{0.6}C_{0.4}$	$D_{0.4}C_{0.6}$	$D_{0.2}C_{0.8}$	D_0C_1
Solvent mole fraction	0	0.2285	0.4413	0.6399	0.8257	1
Density (10^3 kg m^{-3})	1.389	1.408	1.436	1.472	1.515	1.562
Viscosity (mPa.s)	5.717	4.652	3.694	2.719	1.805	0.879
Ultrasonic velocity (m s^{-1})	1460.7	1348	1207	1088	988	916

Standard uncertainties u (solvent mole fraction) = $\pm 1 \times 10^{-4}$, $u(\rho)$ = $\pm 1 \times 10^{-3} \text{ g cm}^{-3}$, $u(\eta)$ = $\pm 2.1\%$, $u(U)$ = $\pm 0.6 \text{ m s}^{-1}$, $u(T)$ = $\pm 0.1 \text{ }^\circ\text{C}$ and $u(p)$ = $\pm 0.1 \text{ kPa}$

TABLE-5
CALCULATED VALUE OF MOLAR MASS (M_w), COMPRESSIBILITY (β), ACOUSTIC IMPEDANCE (Z), FREE LENGTH (L_f), EXCESS MOLAR VOLUME (V^{Ex}), FREE VOLUME (V_f), INTERNAL PRESSURE (π_i), RELATIVE ASSOCIATION (R_A), THERMAL RELAXATION TIME (τ), GIBBS FREE ENERGY (ΔG) AND ULTRASONIC ABSORPTION (α/f^2) AS A FUNCTION OF SOLVENT MOLE FRACTION OF BINARY MIXTURE OF 2,3-DCA WITH CCl_4 AT 300 K AND ATMOSPHERIC PRESSURE 100.5 kPa

Parameters	D_1C_0	$D_{0.8}C_{0.2}$	$D_{0.6}C_{0.4}$	$D_{0.4}C_{0.6}$	$D_{0.2}C_{0.8}$	D_0C_1
Solvent mole fraction	0	0.2285	0.4413	0.6399	0.8257	1
Molar mass ($g\ mol^{-1}$)	162.01	160.14	158.39	156.76	155.23	153.80
Compressibility ($10^{-10}\ N^{-1}\ m^2$)	3.374	3.908	4.780	5.738	6.761	7.630
Acoustic impedance ($10^6\ kg\ m^{-2}\ s^{-1}$)	2.029	1.897	1.733	1.601	1.496	1.430
Free length ($10^{-10}\ m$)	0.363	0.391	0.432	0.474	0.514	0.547
Excess molar volume ($10^6\ m^3\ mol^{-1}$)	0	1.2463	1.6792	1.4838	0.8318	0
Free volume ($10^{-10}\ m^3\ mol^{-1}$)	300.80	356.99	420.38	561.03	884.77	2291.90
Internal pressure ($N\ m^{-2}$)	8.869	8.519	8.234	7.656	6.748	5.046
Relative association	1	1.041	1.102	1.169	1.243	1.314
Thermal Relax. Time ($10^{-12}\ s$)	2.572	2.424	2.355	2.081	1.627	0.894
Gibbs free energy ($10^{-21}\ joule\ mol^{-1}$)	4.996	4.890	4.838	4.615	4.173	3.096
Ultrasonic absorption ($10^{-17}\ Np\ s^{-2}\ cm^{-1}$)	34.721	35.464	38.468	37.709	32.475	19.249

Standard uncertainties $u(\text{mole fraction}) = \pm 1 \times 10^{-4}$, $u(M_w) = \pm 0.001\ g\ mol^{-1}$, $u(\beta) = \pm 0.001 \times 10^{-10}\ N^{-1}\ m^2$, $u(Z) = \pm 0.001 \times 10^6\ kg\ m^{-2}\ s^{-1}$, $u(L_f) = \pm 0.001 \times 10^{-10}\ m$, $u(V^{Ex}) = \pm 0.0001 \times 10^6\ m^3\ mol^{-1}$, $u(V_f) = \pm 0.01 \times 10^{-10}\ m^3\ mol^{-1}$, $u(\pi_i) = \pm 1 \times 10^{-3}\ N\ m^{-2}$, $u(R_A) = \pm 1 \times 10^{-3}$, $u(\tau) = \pm 0.001 \times 10^{-12}\ s$, $u(\Delta G) = \pm 0.001 \times 10^{-21}\ joule\ mol^{-1}$, $u(\alpha/f^2) = \pm 0.001 \times 10^{-17}\ Np\ s^{-2}\ cm^{-1}$, $u(T) = \pm 0.1\ ^\circ C$ and $u(p) = \pm 0.1\ kPa$

mixtures. A decrease in the ultrasonic velocity also support that physical forces among the molecules of binary mixture reduce with the solvent mole fraction.

The compressibility was found to increase while acoustic impedance decrease with an increase in mole fraction of solvent in the binary mixtures (Fig. 2). Similar results have been reported in the literature for binary mixture of CCl_4 with benzene [20]. The intermolecular bonding strength is inversely proportional to compressibility of mixture. An enhancement in the compressibility with an increase in mole fraction of solvent reveals the decrease of intermolecular bonding in present binary system. Since weak molecular interaction is responsible for the low hindrance in acoustic waves in liquids. A decrease in the acoustic impedance confirms the existence of dispersive forces between polar and non-polar liquids in present binary system.

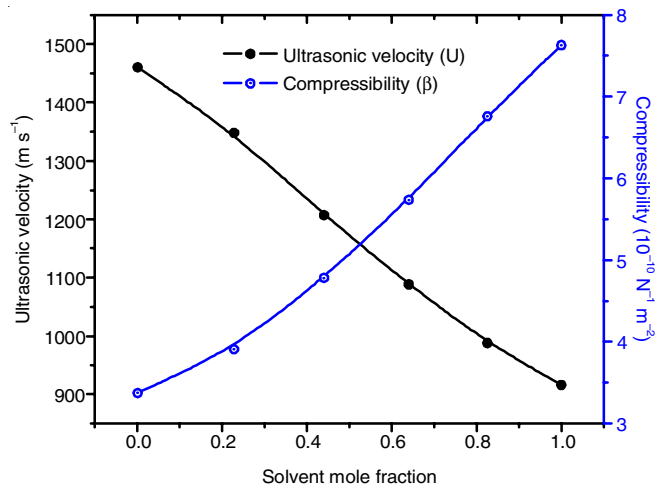


Fig. 2. Ultrasonic velocity and compressibility vs. solvent mole fraction of 2,3-DCA + CCl_4

The free length defines average space among atoms of a molecular system. The strong atomic bonding either caused by dipole-dipole interaction or ionic interaction or hydrogen

bonding, which decrease the low atomic separation and compressibility. Therefore, free length of molecular system or binary mixture is governed by adiabatic compressibility. The calculated free length for present binary mixtures is shown in Fig. 3. The variation of free length with mole fraction is found to resemble same characteristics as adiabatic compressibility (Fig. 2). Similar results have been reported in literature for the binary mixture of CCl_4 with acetone and toluene [35,36].

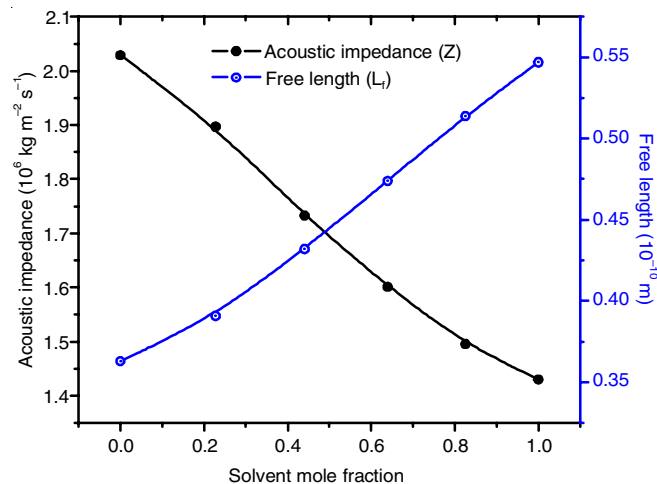


Fig. 3. Acoustic impedance and free length vs. solvent mole fraction of 2,3-DCA + CCl_4

The excess molar volume of mixture is influenced by three types of interactions viz. as physical, chemical and structural interaction between the components of liquid mixtures due to differences in size and shape of the component molecules of mixtures. The values of the excess molar volume are positive for all mole fractions of present binary mixture. The maximum value of excess molar volume occurs at 50% solvent mole fraction in chosen binary mixture (Fig. 4). Similar results have been reported for the binary mixture of dimethyl carbonate with methanol, chloroform, carbon tetrachloride, cyclohexane

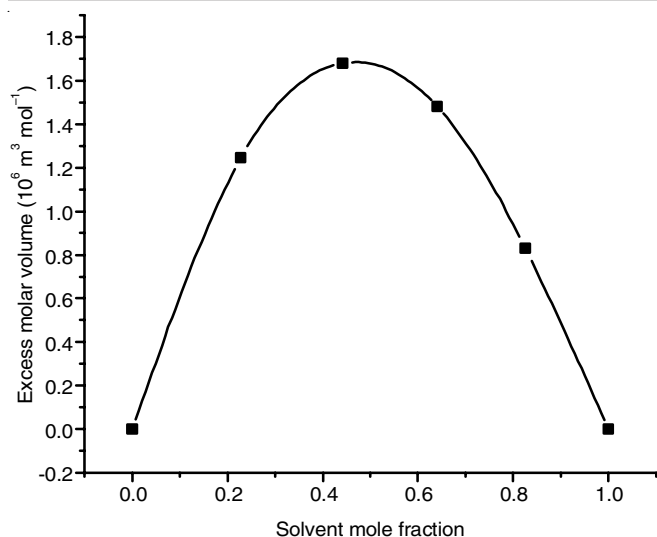


Fig. 4. Excess molar volume vs. solvent mole fraction of 2,3-DCA + CCl_4

and dichloromethane by Aminabhavi & Banerjee [22]. An enhancement in excess molar volume is due to the dominant dispersive forces between unlike molecules due to the effective interaction among molecules caused by hydrogen bonding, charge transfer and dipole-dipole interaction.

The free volume of 2,3-DCA with CCl_4 was found to increase with an increase in solvent mole fraction (Fig. 5). A rapid increase in the free volume was also observed at 50% solvent mole fraction. The enhancement in free volume shows that strength of interaction decreases with an increase in solvent mole fraction. Thus, weak forces exist in present binary system whose strength decrease rapidly after 50% solvent mole fraction due to the reduction of hydrogen bonding between solute and solvent molecules influenced by excess cohesive forces among solvent molecules.

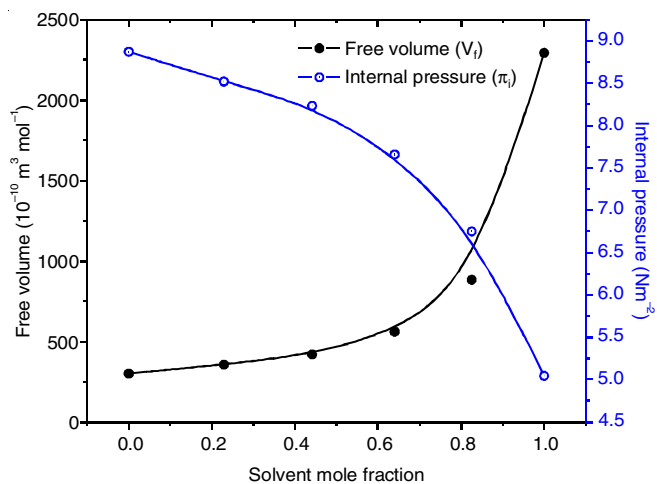


Fig. 5. Free volume and internal pressure vs. solvent mole fraction of 2,3-DCA + CCl_4

From Fig. 5, it is clear that internal pressure decrease with an increase in the mole fraction of solvent in present binary mixture and variation trend is observed opposite to free volume. Internal pressure gives an idea about the solubility of mixture and is a measure of resultant attractive and repulsive forces

between the interacting components in mixture. The suspension of solutes influenced by the internal forces in solvent and their interactions with solvent arise through hydrogen bonding, charge transfer and columbic/van der Waal's interaction [29]. Therefore, variation of internal pressure with mole fraction justifies the same results as free volume.

The relative association was found to increase with increase in mole fraction of solvent in the binary mixture (Fig. 6). The association of atoms in binary mixture depends upon the free length of solvent and density of solute. The strength of association is governed by nature of molecular interaction, which can be defined on basis of compressibility. This indicates that association of solute with solvent molecules is high whose strength decreases with solvent mole fraction.

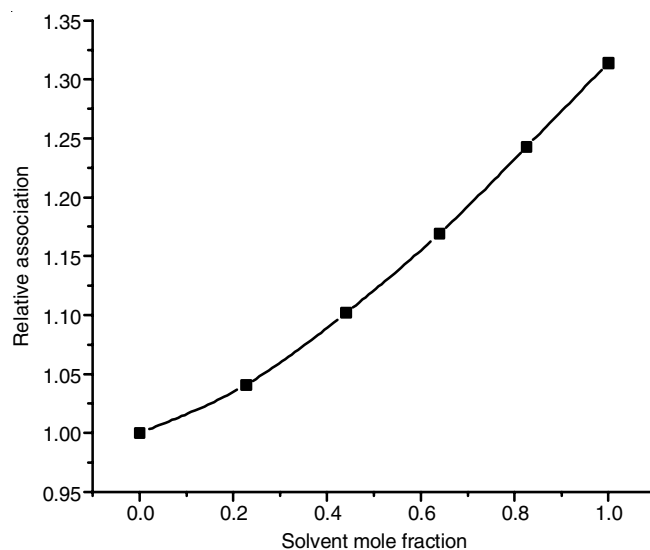


Fig. 6. Relative association vs. solvent mole fraction of 2,3-DCA + CCl_4 at 300 K

The thermal relaxation time and Gibbs free energy were found to decrease with an increase in the mole fraction of solvent in present binary mixture (Fig. 7). Both of the parameters were found to decrease with a slow rate up to 50% mole fraction of

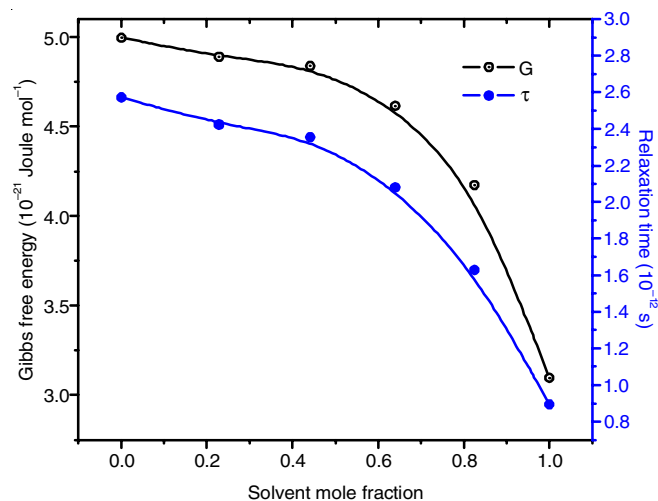


Fig. 7. Gibbs free energy and relaxation time vs. solvent mole fraction of 2,3-DCA + CCl_4

solvent, however decrease rapidly after it. A similar trend of results has also been reported for the binary mixtures of methylmethacrylate with 2-methoxy ethanol, 2-ethoxy ethanol and 2-butoxy ethanol [33]. The thermal relaxation time is governed by the adiabatic compressibility and viscosity of binary mixture. The compressibility is found to enhance with the mole fraction of solvent while viscosity is found to decrease in the binary system. Therefore, both of these parameters jointly influence solvent mole fraction dependent variation of thermal relaxation time in present binary mixture. Hence, re-establishment time confirmed that effective molecular interaction exists among constituent molecules up to 50% solvent mole fraction. The pico-second order of thermal relaxation time is due to the structural relaxation process, which confirms that molecules are rearranged in binary mixture with hydrogen bonding and dipole interaction through accommodating process [37,38].

Gibb's free energy is the sum of Helmholtz energy and normal work done at constant temperature and pressure. On mixing two equilibrium state liquids to form a binary system, maximum work is done by the decrease in Helmholtz energy. Therefore, Gibb's free energy of a binary system measures the maximum work done contributed to form binary system from constituent liquids in equilibrium condition through relaxation process. Therefore, Gibb's free energy directly co-related with thermal relaxation time.

The ultrasonic absorption for present binary mixture is shown in Fig. 8. The ultrasonic absorption is a function of density, viscosity and ultrasonic velocity of binary mixture. It increases up to 50% solvent mole fraction followed by a rapid decrease with an increase in mole fraction of CCl_4 in the binary mixtures. A decrease in the ultrasonic absorption with a mole fraction is due to the existence of weak intermolecular forces between molecules of constituent liquids. The fast decrease in the ultrasonic absorption corresponds to the structural arrangement of molecules towards solvent. Therefore, a binary mixture of 2,3-DCA and CCl_4 in equal ratio consist of less corrosiveness, good stability and high structural arrangement due to

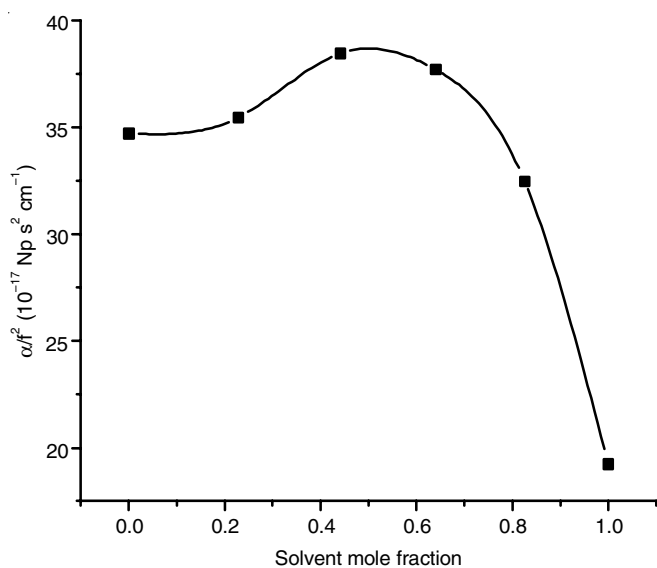


Fig. 8. Ultrasonic absorption vs. solvent mole fraction of 2,3-DCA + CCl_4

rich hydrogen bonding network. Hence, present binary system with equal ratio of constituents may be useful in formation of herbicides and pesticides.

Conclusion

In this work, the experimental determination of density, viscosity, ultrasonic velocity and theoretical approach for the estimation of thermophysical parameters of binary mixture 2,3-dichloroaniline (2,3-DCA) with carbon tetrachloride (CCl_4) at 300 K were conducted. The analysis of estimated parameters justifies the intermolecular physical forces existed between the constituents liquids. The results of acoustic and thermophysical parameters of present binary mixture revealed that weak molecular interactions were present in the binary mixture. The ultrasonic absorption is also received to be mainly governed by compressibility and viscosity of binary mixtures, which provides direct information about their re-structuring time, and nature of molecular interaction. All the properties measured and calculated in this work explores that binary mixture with 50% 2,3-DCA and 50% CCl_4 shall form a low corrosive fluid mixture which is applicable in the synthesis of herbicides, pesticides and fertilizers.

ACKNOWLEDGEMENTS

The authors express their gratitude to Prof. Raja Ram Yadav, Department of Physics, University of Allahabad, Prayagraj and Prof. Devraj Singh, Prof. Rajendra Singh, Institute of Physical Sciences for Study and Research, V.B.S. Purvanchal University, Jaunpur, India for the useful discussion.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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