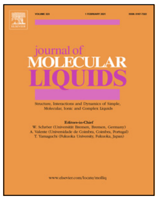
Journal of Molecular Liquids 342 (2021) 117550

Contents lists available at ScienceDirect

Journal of Molecular Liquids

journal homepage: www.elsevier.com/locate/molliq



# Spectroscopic thermodynamic properties of binary liquid mixtures of non-polar and polar solvents (Tetra chloromethane, 2-chloroaniline,2-methylaniline, and 2-methoxyaniline) at various temperatures

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ARTICLE INFO

*Article history:*
Received 21 April 2021
Revised 4 September 2021
Accepted 9 September 2021
Available online 13 September 2021

*Keywords:*
Density
Speed of sound
Tetrachloromethane
Ortho-substituent aniline and FT-IR properties

ABSTRACT

The densities ($\rho$), viscosities ($\eta$) and speeds of sound($u$) are reported for binary mixtures of tetrachloromethane with ortho-substituted aniline (2-chloroaniline, 2-methylaniline, and 2-methoxyaniline) over the entire composition range from 298.15 K to 318.15 K and at atmospheric pressure (0.1 MPa). The excess functions and deviation in viscosity are calculated from the densities, speeds of sound and viscosities at experimental temperatures. FT-IR properties have been carried out and analyzed to study the presence of specific interaction such as the formation of hydrogen bonding of the type (N-H·····Cl) between unlike molecules in the binary liquid mixtures. A good agreement is observed among the excess parameters and FT-IR properties.

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## 1. Introduction

Many studies have shown that thermodynamic data for binary mixtures of tetrachloromethane with Lewis bases can be interpreted in terms of the formation of either 1: 1, or both 1: 1 and 2: 1 complex. Such an interpretation is physically realistic and it has been substantiated in some cases by other types of investigation, including measurements of NMR relaxation rates [1,2], measurements of dielectric properties [3], and determination of stable phases in the solid-state [4,5]. The simplest and most convenient thermodynamic model that has been applied for this purpose assumes that the species present at equilibrium mix ideally. In effect, all of the deviations of the mixture from ideal solution behavior are treated in terms of chemical equilibria with neglect of activity coefficients terms (activity coefficients equal to unity at all compositions, temperatures, and pressures). Accordingly, there has been considerable interest in investigating the accuracy with which this simple "ideal associated solution" model describes the equilibrium properties of associated solutions. The present work endeavors to find the basic strength of the o-substituted group in the aniline molecule that can affect both the sign and magnitude of various thermodynamic functions when mixed with tetrachloromethane. The present work is a continuation of our earlier studies [6–8] of thermodynamic and physicochemical properties of non-aqueous binary liquid mixtures. A survey of the literature reveals that the thermo-physical properties of pure molecules of tetrachloromethane and *ortho*-substituted aniline have been reported extensively because of their importance from both fundamental and industrial points of view. However, the thermo-physical properties of liquid mixtures of tetrachloromethane and *ortho*-substituted aniline have not been explored systematically. The present investigation is the continuation of our earlier research [6] on the thermodynamic properties of binary liquid mixtures. Tetrachloromethane is an effective solvent within the chemical industry and is used to clean machinery and electrical equipment. Chloroaniline is widely used in polymer, rubber, pharmaceutical and dye industries.Methylaniline used as an intermediate for dyes, agrochemicals and other organic products manufacturing.Methoxyaniline used in the manufacture of azo dyes, pharmaceuticals, textile-processing chemicals. We aim to explore the thermo-physical properties such as density, speed of sound, viscosity and their excess properties for the mixed solvents of these binary mixtures to expand the basic needs for scientific research. Modern research machines can take infrared measure-

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https://doi.org/10.1016/j.molliq.2021.117550
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ments across the whole range of interest as frequently as 32 times a second. This can be done whilst simultaneous measurements are made using other techniques. This makes the observations of chemical reactions and processes quicker and more accurate. Infrared spectroscopy is widely used in both research and industry as a simple and reliable technique for measurement, quality control, and dynamic measurement. This is especially useful in measuring the degree of polymerization in polymer manufacture. Infrared spectroscopy has been highly successful for applications in both organic and inorganic chemistry. Infrared spectroscopy has also been successfully utilized in the field of semiconductor microelectronics: for example, infrared spectroscopy can be applied to semiconductors like silicon, gallium arsenide, gallium nitride, zinc selenide, amorphous silicon, silicon nitride, etc.

## 2. Experimental

### 2.1. Materials

O-substituted aniline and tetrachloromethane chemicals were purchased from S.D. Fine Chemical, India. O-substituted aniline was dried and other chemicals were placed over potassium hydroxide pellets for several days and then distilled under nitrogen at reduced pressure [65].Carbon tetrachloride was shaken with potassium hydroxide solution in 50 mass percentalcohols removed from the potassium hydroxide layer and then shaken with water and small portions of concentrated sulfuric acid.The water content was measured using Analab (Micro Aqua Cal 100) Karl Fischer Titrator [7] and Karl Fisher reagent from Merck by conductometric titration with the dual platinum electrode. It can detect water content from less than 10 ppm to 100 %and issummarized in Table 1. All chemicals purity was checked by GC analysis and found to be > 0.99 and the procedure is as follows. The sample solution is injected through an injector usually with a syringe. The injector is heated 150–250 °C which causes the volatile sample solutes to vaporize. The vaporized solutes are transported into the column (30 m × 0.25 mm × 0.25 μm) by the carrier gas (Constant Flow Hydrogen, 1 mL/min is used as carrier gas).The column is maintained in a temperature controlled oven. The various components are separated inside the column by their physical properties and the temperature and composition of the column. The various solutes travel through the column at different rates (35 °C for 7 min to 60 °C at the rate of 5 °C/min for 2 min to 210 °C at the rate of 10 °C). The fastest-moving solute elutes the column first then is followed by the remaining solutes in corresponding order. The detector (FID) measures the quantity of the components that exit the column. To measure a sample with an unknown concentration, a standard sample with aknown concentration is injected into the instrument. The standard sample peak retention time (appearance time) and area are compared to the test sample to calculate the concentration.Table 1 contains information regarding their source and purity and Table 2 shows their physical properties, density, speed of sound, and viscosity.The measured physical values (Table 2) have been compared with NIST literature[9–46].

The density, speed of sound, and viscosity of tetrachloromethanehas been graphically compared with average absolute deviation ($AAD = (100/n)\sum_{i=1}^{n}(F_{0,lit} - F_{0,exp}/F_{0,lit})_i$) and given as Fig. 1S-3S(*Supplementary information*). Density values with good agreement with literature except Rout et al. [16] and Dash et al. [20]. The deviation is very high approximately 0.7% at 318.15 K. These two researchers worked on a pycnometer of bulb capacity 25 mL and another cause is temperature bath and purity of the liquid. In the case of speed of sound, the values are well agreement with Takagi et al. [28]. From Fig. 2S(*Supplementary information*) the deviation is maximum varied up to 0.4% in the case of Nath et al. [17]and Lagemann et al [29] is 0.2% at a higher temperature 318.15 K. With other literature the values are 0.09 [17], −0.04 [18], −0.15 [20] and 0.18% [29] at 303.15 K. In the case of researchers [16] and [20] the values are scattered more. The deviation may be arising due to the equipment, frequency, purity, and temperature control. The viscosity value (Fig. 3S) (*Supplementary information*) of tetrachloromethane reported by Klofutar et al. [26] is 0.899 mPa.s at 298.15 K, and data are higher by 0.16% (maximum) than our values inthe entire temperature ranges. This slight temperature dependence is obvious. Joshi et al. [11] and Aminabhavi et al [12] are very scattered. These may be experimental procedures and time calculations.The viscosities of 2-chloroaniline were in good agreement with Friend et al.[61] and the data comparison was shown in Table 1S in *Supplementary information*. Whereas with Aminabhavi et al. [62] the values were scattered>50%. So, we omitted values in Table 2. This may be a slip of procedure.The experimental density of 2-methoxyaniline was graphically (Fig. 4S *in Supplementary information*) compared with Perkin et al. [63], Thole et al [64], and Friend et al [61]. Perkin et al. [63] values deviate-0.35% from our experimental values. It may be a typographical mistake by Perkin. In the case of 2-chloroaniline viscosity values agreed with 100% with our experimental values. In the case of Thole and Friend, it may experimentally error.

### 2.2. Apparatus and procedure

All the binary liquid mixtures were prepared by weighing the required amounts of pure liquids in an electric balance (ER-120A, Afoset) with a precision of ± 0.1 mg by syringing each component into air-tight stopper bottles to minimize evaporation losses. The uncertainty of the mole fraction was $\pm 1 \times 10^{-4}$. The details of the density, speeds of sound, and viscosity methods and their measurement techniques were described elsewhere [47]. After mixing the sample, the bubble-free homogeneous sample was transferred into the U-tube of the densimeter using a syringe. The density measurements were performed with a Rudolph Research Analytical digital densimeter (DDH-2911 Model), equipped with a builtinsolid-state thermostat and a resident program with an accuracy of temperature of ± 0.03 K. The speed of sound in pure liquids and their mixtures were measured by using a multi-frequency ultrasonic interferometer (M−82 Model, Mittal Enterprise, New Delhi, India) single-crystal variable path, operated at 2 MHz at (303.15–318.15) K by using a digital constant temperature water bath.The viscosities of pure liquids and their mixtures were determined at

**Table 1**
List of chemicals with details of source, CAS number, Purity and water content.

| Name of the chemical | Source | CAS number | purification method | Mass fraction purity by GC* | Water content mass fraction by KF |
|---|---|---|---|---|---|
| carbon tetra chloride | Sigma Aldrich India | 56–23-5 | no purification | 0.995 | 0.0001 |
| 2-chloroaniline | Sigma Aldrich India | 95–51-2 | no purification | 0.996 | 0.00042 |
| 2-methylaniline | Sigma Aldrich India | 95–53-4 | no purification | 0.995 | 0.00042 |
| 2-methoxyaniline | Sigma Aldrich India | 90–04-0 | no purification | 0.995 | 0.00042 |

*Gas Chromatography;

**Table 2**
Densities, viscosity and speeds of sounds data of pure components at different temperatures and 0.1 MPa pressure.

| component | density($\rho$)($kg{\cdot}m^{-3}$) | | speed of sound, (u / $m{\cdot}s^{-1}$) | | viscosity (mPa·s) | | $C_P$($J{\cdot}K^{-1}\cdot mol^{-1}$) |
|---|---|---|---|---|---|---|---|
| | experimental | literature | experimental | literature | experimental | literature | |
| tetra chloromethane | | | | | | | |
| 298.15 | 1584.50 | 1585.1[11]<br>1584.0[12]<br>1584.3[21]<br>1584.3[22]<br>1584.3[24]<br>1584.3[25]<br>1584.4[27] | 921.10 | 921.1[28] | 0.897 | 0.928[11]<br>0.821[12]<br>0.899[26] | 130.96[30] |
| 303.15 | 1574.62 | 1574.9[11]<br>1574.8[12]<br>1574.8[13]<br>1574.8[14]<br>1574.8[15]<br>1584.8[16]<br>1584.8[20]<br>1574.6[21]<br>1574.6[22]<br>1574.6[24]<br>1574.6[25]<br>1575.1[27]<br>1574.8[29] | 905.38 | 908.0[16]<br>906.2[17]<br>905.0[18]<br>904.0[19]<br>908.0[20]<br>907.0[29] | 0.838 | 0.870[11]<br>0.768[12]<br>0.845[13]<br>0.845[14]<br>0.845[15]<br>0.914[16]<br>0.844[23]<br>0.838[26] | 131.85* |
| 308.15 | 1564.80 | 1565.0[11]<br>1565.0[12]<br>1568.4[16]<br>1568.4[20]<br>1564.8[21]<br>1564.8[22]<br>1564.8[24] | 889.72 | 893.0[16]<br>893.0[20] | 0.784 | 0.816[11]<br>0.717[12]<br>0.796[16] | 132.63[30] |
| 313.15 | 1555.10 | 1555.4[11]<br>1562.2[16]<br>1562.2[20]<br>1554.9[21]<br>1554.9[22]<br>1555.0[25]<br>1555.0[29] | 874.20 | 883.0[16]<br>877.6[17]<br>874.0[18]<br>873.5[19]<br>883.0[20]<br>874.2[28]<br>876.0[29] | 0.734 | 0.768[11]<br>0.748[16]<br>0.739[23]<br>0.735[26] | 133.31* |
| 318.15 | 1545.10 | 1556.4[16]<br>1556.4[20]<br>1545.1[22]<br>1564.8[22]<br>1564.8[24] | 858.71 | 875.0[16]<br>875.0[20] | 0.689 | 0.703[16] | 133.89[30] |
| 2-methylaniline [8] | | | | | | | |
| 298.15 | 994.44 | 994.3[34]<br>994.4[35] | 1598.5 | 1602.35[34]<br>1602.35[35] | | 3.725[39] | 217.16* |
| 303.15 | 990.19 | 990.4[31]<br>990.1[33]<br>990.2[32]<br>990.2[36]<br>990.2[37]<br>994.4[38] | 1578.5 | 1577.70[31]<br>1577.82[32]<br>1579.00[33]<br>1577.67[36]<br>1577.67[37] | 3.258 | 3.255[39] | 218.43 |
| 308.15 | 985.99 | 986.0[32]<br>986.1[31]<br>986.1[36]<br>986.1[37]<br>991.9[38] | 1558.2 | 1558.20[31]<br>1558.38[32]<br>1558.18[36]<br>1558.18[37] | 2.824 | 2.820[39] | 219.09 |
| 313.15 | 981.69 | 981.7[33]<br>989.6[38] | 1538.5 | 1539.00[33] | 2.487 | 2.485[39] | 220.52 |
| 318.15 | 977.60 | | 1518.5 | | 2.153 | 2.150[39] | 221.12* |
| 2-Chloroaniline [6] | | | | | | | |
| 298.15 | 1207.53 | 1207.8[60] | 1485.9 | | 4.209 | | 199.76* |
| 303.15 | 1202.69 | 1202.4[40]<br>1202.7[41] | 1469.5 | 1470.00[41]<br>1471.00[43] | 3.825 | 3.8256[40] | 200.55 |
| 308.15 | 1198.02 | 1197.6[42] | 1453.3 | 1455.13[45] | 3.412 | | 201.37 |
| 313.15 | 1193.02 | 1193.0[41] | 1435.2 | 1436.00[41] | 3.060 | | 202.22 |
| 318.15 | 1188.49 | | 1418.9 | | 2.762 | | 203.09 |
| 2-methoxyaniline [7] | | | | | | | |
| 298.15 | 1095.26 | | 1613.5 | | 5.374 | | 189.04* |
| 303.15 | 1091.57 | 1091.7 [9] | 1595.4 | | 4.922 | 4.9236[7] | 190.52 |
| 308.15 | 1087.99 | | 1579.2 | | 4.512 | | 192.05 |
| 313.15 | 1084.32 | | 1566.3 | | 4.185 | | 193.62 |
| 318.15 | 1080.82 | | 1554.8 | | 3.886 | | 195.25 |

Standard uncertainties $u$ are$u(\rho)$ = ±0.8 $kg.m^{-3}$, $u(u)$ = ±0.5 $m.s^{-1}$, $u(\eta)$ = ±0.17 % mPa.s, $u(T)$ = 0.01 K for density and speed of sound, $u(T)$ = 0.01 K for viscosity and $u(p)$ = 1 kPa
*With Polynomial fitting

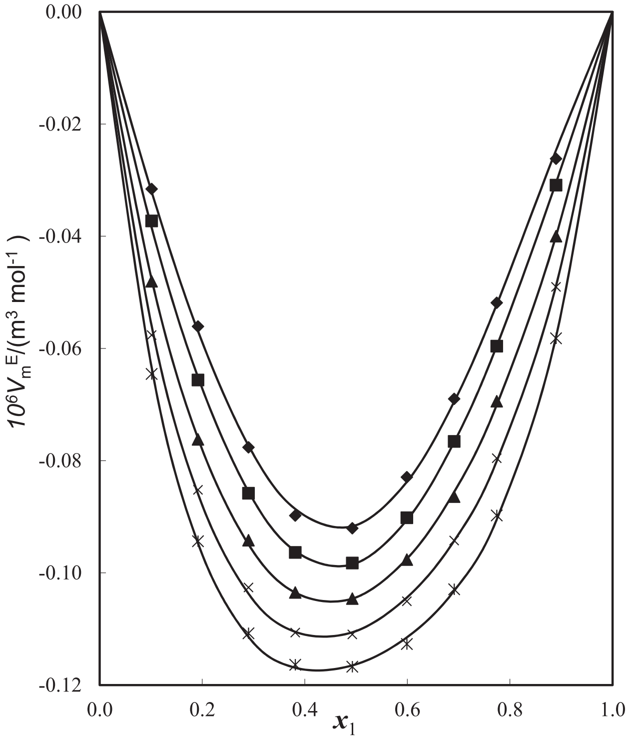

**Fig. 1.** Eexcess molar volume ($V^E$) with mole fraction for the binary mixtures oftetra chloromethane (1) + 2-chloroaniline (2) binary mixtures at temperatures, $T$/K = 298.15,◆; At $T$/K = 303.15,■; $T$/K = 308.15, ▲; $T$/K = 313.15, ×; $T$/K = 318.15,∗. The points represent experimental values and lines represent values calculated from equation (7) using the coefficients given in table 4.

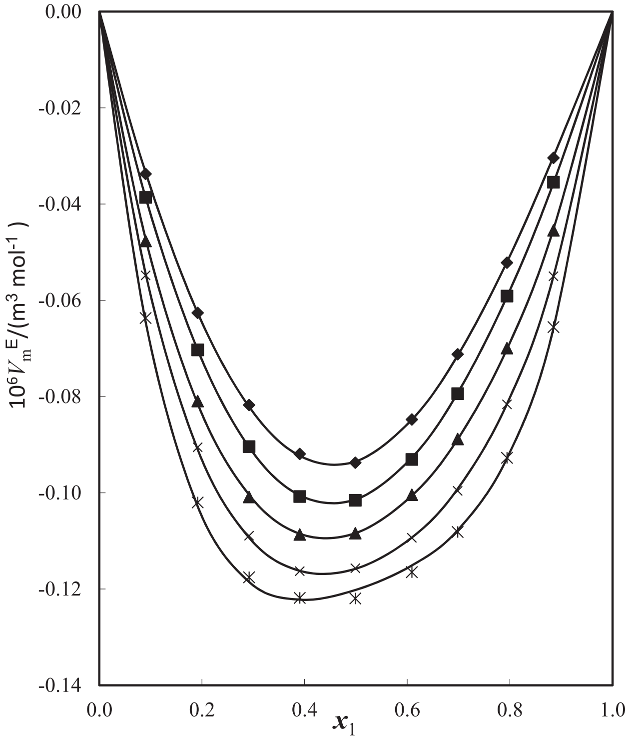

**Fig. 2.** Excess molar volume ($V^E$) with mole fraction for the binary mixtures of tetra chloromethane (1) + 2-methylaniline (2) binary mixtures at temperatures, $T$/K = 298.15,◆; At $T$/K = 303.15,■; $T$/K = 308.15, ▲; $T$/K = 313.15, ×; $T$/K = 318.15,∗. The points represent experimental values and lines represent values calculated from equation (7) using the coefficients given in table 4.

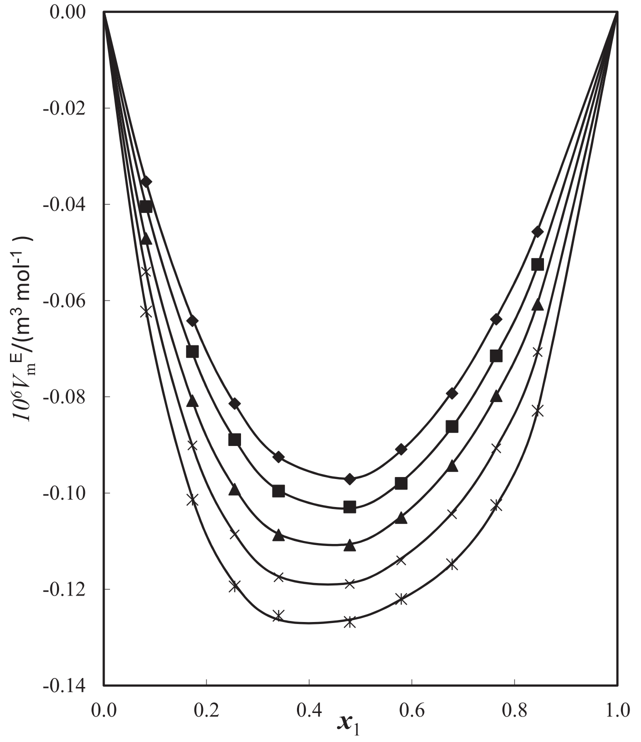

**Fig. 3.** Excess molar volume ($V^E$) with mole fraction for the binary mixtures of tetra chloromethane (1) + 2-methoxyaniline (2) binary mixtures at temperatures, $T$/K = 298.15,◆; At $T$/K = 303.15,■; $T$/K = 308.15, ▲; $T$/K = 313.15, ×; $T$/K = 318.15,∗. The points represent experimental values and lines represent values calculated from equation (7) using the coefficients given in table 4.

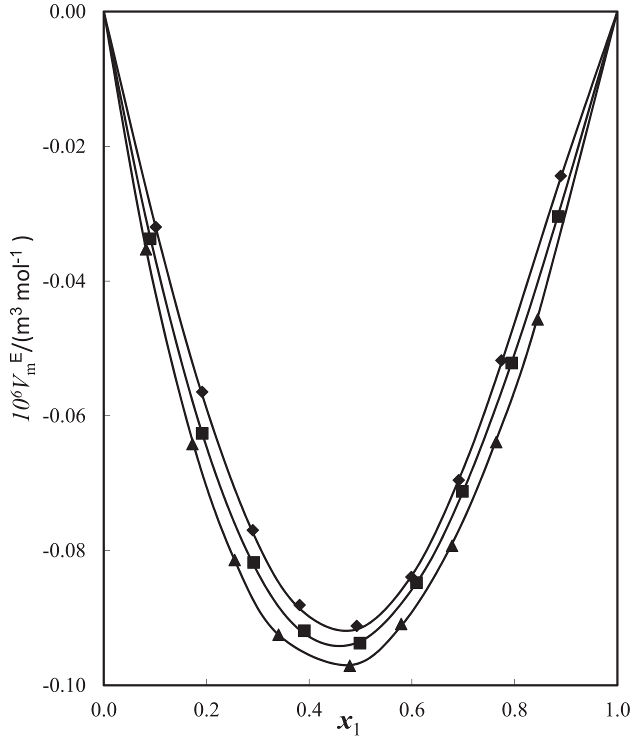

**Fig. 4.** Excess molar volume ($V^E$) with mole fraction for the binary mixtures of tetra chloromethane (1) + 2-chloroaniline (2),◆;tetra chloromethane (1) + 2-methylaniline (2),■;tetra chloromethane (1) + 2-methoxyaniline (2), ▲;binary mixtures at temperatures, $T$/K = 298.15. The points represent experimental values and lines represent values calculated from equation (7) using the coefficients given in table 4.

atmospheric pressure at T = (303.15–318.15) K by using an Ubbelohde viscometer. The viscometer was thoroughly cleaned, perfectly dried and filled with the sample liquid by fitting the viscometer to about 30°from the vertical and also its limbs were closed with Teflon caps to avoid evaporation. The viscometer was kept in a transparent walled bath with the thermal stability of ± 0.1 K for about 20 min to obtain thermal equilibrium.Proper calibration at each temperature was achieved with doubly distilled, de-ionized water {$\rho$ = 997.043 ± 0.002 kg·m$^{-3}$ at 298.15 K, conductivity, $\kappa$ = 5.5μS/m} and with dry air as standards.The uncertainty in density measurement of liquid mixtures was $\pm$0.8 kg.m$^{-3}$ [66,67].The uncertainty in the measured speed of sound is $\pm$0.5 m·s$^{-1}$ [66,67].The experimental uncertainty of viscosity was estimated as $\pm$0.17 %mPa.s [66,67].The temperature of the liquids during the measurements was maintained with in an uncertainty of ± 0.01 K [66,67] in an electronically controlled thermostatic water bath.

## 3. Results and discussion:

The excess thermodynamic / deviation functions were calculated with the following equations

$$V^E/cm^3.mol^{-1} = V \;--\; (x_1V_1 + \; x_2V_2) \tag{1}$$

$$\Delta\eta/\; mPa\cdot s = \eta - (x_1\eta_1 + x_1\eta_1) \tag{2}$$

$$DG^{*E}/\; J.mol^{-1} = RT[\ln\eta V - \;(x_1 ln\eta_1 V_1 + x_2 ln\eta_2 V_2)] \tag{3}$$

where $\eta$, $\eta_1$,$\eta_2$,$V$,$V_1$and$V_2$ are viscosities and molar volumes of the mixture and ~~of~~ the pure components respectively.

The isentropic compressibilities were calculated from the relation [48]

$$\kappa_s = \;(u^2\rho)^{-1} \tag{4}$$

where$\rho$is the density and$u$is the speed of sound of the binary mixture.

Further, the excess isentropic compressibilities $(\kappa_s^E)$ are calculated from the following relations recommended by Benson and Kiyohara[49]

$$k_s^E = k_s - k_s^{id} \tag{5}$$

$$k_s^{id} = \sum_{\mathrm{i}=1}^{2}\varphi_\mathrm{i}\left[\kappa_{\mathrm{s,i}} + \frac{\mathrm{TV_i}\left(\alpha_\mathrm{i}^2\right)}{\mathrm{C_{p,i}}}\right] - \left\{\frac{\mathrm{T}\left(\sum_{\mathrm{i}=1}^{2}\mathrm{x_iV_i}\right)\left(\sum_{\mathrm{i}=1}^{2}\varphi_\mathrm{i}\alpha_\mathrm{i}\right)^2}{\sum_{\mathrm{i}=1}^{2}\mathrm{x_iC_{p,i}}}\right\} \tag{6}$$

where $\varphi_\mathrm{i}$, $C_{p,\mathrm{i}}$, $V_i$, $\kappa_{\mathrm{s,i}}$and $\alpha_i$ are the volume fraction, molar heat capacity, molar volume, isentropic compressibility and coefficient of isobaric thermal expansion of pure components respectively. The $C_{p,\mathrm{i}}$ values are taken from our previous papers [6,7,8] and [30]. Some values are calculated on extrapolating with the help of polynomial fitting.

$$\phi_i = \frac{x_iV_{m,i}}{\sum_{i=1}^{2}x_iV_{m,i}}$$

$$\alpha_p^{id} = \phi_1\alpha_{p,1} + \phi_2\alpha_{p,2}$$

$$V_m^{id} = x_1V_{m,1} + x_2V_{m,2}$$

$$C_p^{id} = x_1C_{p,1} + x_2C_{p,2}$$

The coefficient of thermal expansion, $\alpha_p$, can be calculated as

$$\alpha_p = -\frac{1}{\rho}\left(\frac{\Delta\rho}{\Delta T}\right)_p$$

$\alpha_p$ values have been obtained from a linear dependence of $\rho$ with *T*.

The density, speed of sound, and viscosity for mixtures of various mole fractions of 2-methylcyclohexanone are presented in Table 3 along with excess/deviation functions.

Excess/deviation functions ($V^E$, $\Delta\eta$ and $\kappa_s^E$) values are fitted to a Redlich-Kister polynomial equation [50]

$$Y^E = x_1x_2\sum_{i=0}^{j}A_i(2x_1 - 1)^i \tag{7}$$

Where $Y^E$ is the $V^E$, $\Delta\eta$ and $\kappa_s^E$.Values of the coefficients $A_i$ are determined by using the method of least-squares. The standard deviations $\sigma$ ($Y^E$) were calculated by the formula as follows

$$\sigma(Y^E) = [\;\Sigma(Y^E_{exp} - \;Y^E_{cal})^2/(n-j)]^{1/2} \tag{8}$$

where $n$ is the total number of experimental points and$j$is the number of $A_i$ coefficients considered ($i$ + 1 in the present study). The coefficients, $A_{i,}$ and corresponding standard deviation values ($\sigma$) are presented in Table 4.

The donor–acceptor effects are measured by UV or in some cases FTIR spectroscopy. According to Mulliken's classical theoretical study on donor–acceptor complexes the common types of donors can be divided into $n$ (lone pair) and $\pi$ (bonding $\pi$-orbital) and the acceptors as σ (vacant orbitals) $\pi^*$ (antibonding $\pi$-orbital) and $\sigma^*$ (antibonding σ-orbital).

Table 3 shows that the values of excess functions ($V^E$ and $\kappa_s^E$) are negative and deviation in viscosity ($\Delta\eta$), excess Gibbs energy of activation ($\Delta G^{*E}$) of flow is positive for all the binary systems over the whole composition range and at all experimental temperatures.

Negative values of excess molar volume, excess isentropic compressibility,and positive values of deviation in viscosity ($\Delta\eta$), excess Gibbs energy of activation ($\Delta G^{*E}$) of flow values are arises due to

(i) $n$-σinteraction between *ortho*-substituted aniline and tetrachloromethane componentsin the binary liquid mixtures

(ii) Formation of charge-transfer complexes of the type C-Cl (acceptor) and base (donor)

(iii) Formation of hydrogen bonding between chlorine atom of tetrachloromethane and Nitrogen atom of amines groups of *ortho*-substituted Anilinein the binary liquid mixtures.

The data included in Table 3(Figs. 1-4) shows that the values of excess molar volume is negative overthe entire composition range in all the mixtures at all thetemperatures. The negative values $V^E$are indicative of important interactions between unlike molecules. These interactions may be explained as follows. When a non-polar molecule such as tetrachloromethane is situated in anelectric field set by the presence of a polar molecule, the *ortho*-substituted aniline, a dipole is induced. The force between the permanentdipole of the *ortho*-substituted aniline and the induced dipole of tetrachloromethane is always attractive. This leads to anegative deviation in excess volume. The negative values $V^E$fall in the following order: 2-methoxyaniline > 2-methyl aniline > 2-chloroaniline.The mixtures of tetrachloromethane with 2-methylaniline aniline show excess molar volumes values morenegative than with 2-chloroaniline and tetrachloromethane with 2-methoxy aniline shows greater values than with 2-methylaniline, which could be attributed to the complex formation between2-methoxy aniline and tetrachloromethane by moremesomeric effect. Further, the data suggest that the rise in temperature results in an increase in negative values of excess molar volume

**Table 3**

Density ($\rho$), excess molar volumes ($V^E$), speed of sound ($u$), excess isentropic compressibility ($\kappa_s^E$), viscosity (η), deviation in viscosity ($\Delta\eta$) and excess Gibbs energy of activation of viscous flow ($G^{*E}$) as a function of mole fraction, $x_1$ oftetra chloromethane of binary liquid mixtures at $T$ = (303.15 to 318.15) K and 0.1 MPa pressure.

| $x_1$ | Density(ρ)Kg·m$^{-3}$ | $V^E 10^6$m$^3$·mol$^{-1}$ | $u$ m.s$^{-1}$ | $\kappa_s^E 10^{10}$m$^2$.N$^{-1}$ | viscosity (η / mPa·s) | $\Delta\eta$ / mPa·s | G*$^E$J·mol$^{-1}$ |
|---|---|---|---|---|---|---|---|
| tetra chloromethane (1) + 2-chloroaniline (2) | | | | | | | |
| 298.15 K | | | | | | | |
| 0.0000 | 1207.53 | 0.0000 | 1485.9 | 0.0000 | 4.209 | 0.000 | 0.000 |
| 0.1011 | 1243.22 | −0.0320 | 1407.4 | −0.1209 | 3.882 | 0.008 | 0.187 |
| 0.1916 | 1275.64 | −0.0565 | 1343.8 | −0.2209 | 3.589 | 0.015 | 0.339 |
| 0.2901 | 1311.42 | −0.0770 | 1279.8 | −0.3130 | 3.268 | 0.020 | 0.484 |
| 0.3811 | 1344.91 | −0.0881 | 1224.3 | −0.3752 | 2.970 | 0.023 | 0.596 |
| 0.4923 | 1386.40 | −0.0912 | 1160.5 | −0.4150 | 2.604 | 0.025 | 0.696 |
| 0.5989 | 1426.77 | −0.0839 | 1103.0 | −0.4095 | 2.250 | 0.025 | 0.743 |
| 0.6911 | 1462.16 | −0.0696 | 1056.4 | −0.3707 | 1.942 | 0.022 | 0.731 |
| 0.7741 | 1494.42 | −0.0518 | 1016.7 | −0.3054 | 1.664 | 0.019 | 0.666 |
| 0.8897 | 1540.07 | −0.0244 | 965.5 | −0.1720 | 1.273 | 0.011 | 0.445 |
| 1.0000 | 1584.50 | 0.0000 | 921.1 | 0.0000 | 0.897 | 0.000 | 0.000 |
| 303.15 K | | | | | | | |
| 0.0000 | 1202.69[6] | 0.0000 | 1469.5 [6] | 0.0000 | 3.825 | 0.000 | 0.000 |
| 0.1011 | 1238.05 | −0.0383 | 1391.6 | −0.1342 | 3.532 | 0.009 | 0.186 |
| 0.1916 | 1270.12 | −0.0651 | 1327.6 | −0.2398 | 3.269 | 0.016 | 0.337 |
| 0.2901 | 1305.47 | −0.0857 | 1262.8 | −0.3327 | 2.981 | 0.023 | 0.481 |
| 0.3811 | 1338.53 | −0.0958 | 1206.8 | −0.3942 | 2.713 | 0.026 | 0.592 |
| 0.4923 | 1379.48 | −0.0986 | 1142.9 | −0.4339 | 2.382 | 0.028 | 0.690 |
| 0.5989 | 1419.30 | −0.0907 | 1085.8 | −0.4295 | 2.063 | 0.027 | 0.736 |
| 0.6911 | 1454.21 | −0.0770 | 1039.7 | −0.3915 | 1.785 | 0.024 | 0.724 |
| 0.7741 | 1486.02 | −0.0599 | 1000.5 | −0.3260 | 1.533 | 0.020 | 0.658 |
| 0.8897 | 1530.97 | −0.0307 | 949.8 | −0.1876 | 1.179 | 0.012 | 0.438 |
| 1.0000 | 1574.62 | 0.0000 | 905.4 | 0.0000 | 0.838 | 0.000 | 0.000 |
| 308.15 K | | | | | | | |
| 0.0000 | 1198.02[6] | 0.0000 | 1453.3[6] | 0.0000 | 3.413 | 0.000 | 0.000 |
| 0.1011 | 1233.06 | −0.0463 | 1376.1 | −0.1504 | 3.158 | 0.011 | 0.182 |
| 0.1916 | 1264.77 | −0.0752 | 1311.5 | −0.2608 | 2.927 | 0.018 | 0.328 |
| 0.2901 | 1299.67 | −0.0949 | 1245.8 | −0.3536 | 2.675 | 0.025 | 0.468 |
| 0.3811 | 1332.29 | −0.1039 | 1189.3 | −0.4142 | 2.439 | 0.028 | 0.575 |
| 0.4923 | 1372.67 | −0.1050 | 1125.2 | −0.4522 | 2.149 | 0.030 | 0.670 |
| 0.5989 | 1411.94 | −0.0974 | 1068.6 | −0.4502 | 1.868 | 0.030 | 0.712 |
| 0.6911 | 1446.37 | −0.0853 | 1023.1 | −0.4146 | 1.622 | 0.026 | 0.698 |
| 0.7741 | 1477.72 | −0.0689 | 984.5 | −0.3507 | 1.400 | 0.022 | 0.634 |
| 0.8897 | 1521.97 | −0.0388 | 934.3 | −0.2077 | 1.086 | 0.012 | 0.419 |
| 1.0000 | 1564.80 | 0.0000 | 889.7 | 0.0000 | 0.784 | 0.000 | 0.000 |
| 313.15 K | | | | | | | |
| 0.0000 | 1193.02[6] | 0.0000 | 1435.3 [6] | 0.0000 | 3.060 | 0.000 | 0.000 |
| 0.1011 | 1227.78 | −0.0553 | 1358.9 | −0.1657 | 2.837 | 0.012 | 0.178 |
| 0.1916 | 1259.15 | −0.0855 | 1294.1 | −0.2814 | 2.634 | 0.020 | 0.321 |
| 0.2901 | 1293.63 | −0.1040 | 1227.9 | −0.3746 | 2.411 | 0.026 | 0.457 |
| 0.3811 | 1325.84 | −0.1115 | 1171.2 | −0.4347 | 2.203 | 0.029 | 0.560 |
| 0.4923 | 1365.70 | −0.1111 | 1107.4 | −0.4739 | 1.947 | 0.032 | 0.652 |
| 0.5989 | 1404.48 | −0.1046 | 1051.3 | −0.4728 | 1.699 | 0.032 | 0.693 |
| 0.6911 | 1438.47 | −0.0936 | 1006.4 | −0.4385 | 1.482 | 0.029 | 0.680 |
| 0.7741 | 1469.41 | −0.0784 | 968.5 | −0.3769 | 1.284 | 0.025 | 0.616 |
| 0.8897 | 1513.02 | −0.0470 | 918.8 | −0.2275 | 1.005 | 0.014 | 0.408 |
| 1.0000 | 1555.10 | 0.0000 | 874.2 | 0.0000 | 0.734 | 0.000 | 0.000 |
| 318.15 K | | | | | | | |
| 0.0000 | 1188.49[6] | 0.0000 | 1418.9[6] | 0.0000 | 2.762 | 0.000 | 0.000 |
| 0.1011 | 1222.90 | −0.0631 | 1343.0 | −0.1823 | 2.566 | 0.014 | 0.176 |
| 0.1916 | 1253.88 | −0.0950 | 1277.4 | −0.3019 | 2.387 | 0.022 | 0.316 |
| 0.2901 | 1287.88 | −0.1124 | 1210.5 | −0.3962 | 2.189 | 0.028 | 0.449 |
| 0.3811 | 1319.61 | −0.1178 | 1153.4 | −0.4555 | 2.004 | 0.032 | 0.550 |
| 0.4923 | 1358.89 | −0.1176 | 1089.6 | −0.4945 | 1.776 | 0.035 | 0.639 |
| 0.5989 | 1397.10 | −0.1125 | 1034.1 | −0.4969 | 1.554 | 0.034 | 0.677 |
| 0.6911 | 1430.56 | −0.1019 | 989.8 | −0.4650 | 1.361 | 0.032 | 0.665 |
| 0.7741 | 1461.01 | −0.0874 | 952.4 | −0.4035 | 1.185 | 0.028 | 0.604 |
| 0.8897 | 1503.88 | −0.0552 | 903.3 | −0.2492 | 0.934 | 0.016 | 0.399 |
| 1.0000 | 1545.10 | 0.0000 | 858.7 | 0.0000 | 0.689 | 0.000 | 0.000 |
| tetra chloromethane (1) + 2-methylaniline (2) | | | | | | | |
| 298.15 K | | | | | | | |
| 0.0000 | 994.44 | 0.0000 | 1598.5 | 0.0000 | 3.718 | 0.000 | 0.000 |
| 0.0901 | 1043.10 | −0.0337 | 1517.8 | −0.0696 | 3.473 | 0.009 | 0.149 |
| 0.1916 | 1098.92 | −0.0626 | 1432.5 | −0.1346 | 3.194 | 0.017 | 0.299 |
| 0.2917 | 1155.03 | −0.0818 | 1353.6 | −0.1825 | 2.917 | 0.022 | 0.427 |
| 0.3903 | 1211.35 | −0.0919 | 1280.5 | −0.2117 | 2.642 | 0.025 | 0.530 |
| 0.4988 | 1274.56 | −0.0938 | 1205.3 | −0.2239 | 2.337 | 0.026 | 0.608 |
| 0.6089 | 1340.04 | −0.0848 | 1134.2 | −0.2132 | 2.026 | 0.026 | 0.642 |
| 0.6981 | 1394.12 | −0.0712 | 1080.3 | −0.1877 | 1.772 | 0.023 | 0.625 |
| 0.7941 | 1453.40 | −0.0522 | 1025.9 | −0.1444 | 1.497 | 0.019 | 0.545 |
| 0.8847 | 1510.40 | −0.0304 | 977.8 | −0.0885 | 1.235 | 0.013 | 0.387 |

**Table 3** (*continued*)

| $x_1$ | Density(ρ)Kg·m$^{-3}$ | $V^E 10^6$m$^3$·mol$^{-1}$ | $u$ m.s$^{-1}$ | $\kappa_S^E 10^{10}$m$^2$.N$^{-1}$ | viscosity (η / mPa·s) | $\Delta\eta$ / mPa·s | G*$^E$J·mol$^{-1}$ |
|---|---|---|---|---|---|---|---|
| 1.0000 | 1584.50 | 0.0000 | 921.1 | 0.0000 | 0.897 | 0.000 | 0.000 |
| 303.15 K | | | | | | | |
| 0.0000 | 990.19[8] | 0.0000 | 1578.5[8] | 0.0000 | 3.258 | 0.000 | 0.000 |
| 0.0901 | 1038.52 | −0.0386 | 1498.6 | −0.0780 | 3.049 | 0.009 | 0.141 |
| 0.1916 | 1093.92 | −0.0703 | 1413.4 | −0.1467 | 2.812 | 0.018 | 0.285 |
| 0.2917 | 1149.57 | −0.0904 | 1334.6 | −0.1964 | 2.575 | 0.023 | 0.406 |
| 0.3903 | 1205.40 | −0.1008 | 1261.8 | −0.2269 | 2.340 | 0.027 | 0.502 |
| 0.4988 | 1268.02 | −0.1016 | 1187.1 | −0.2402 | 2.079 | 0.028 | 0.576 |
| 0.6089 | 1332.88 | −0.0931 | 1116.7 | −0.2308 | 1.812 | 0.028 | 0.606 |
| 0.6981 | 1386.42 | −0.0794 | 1063.4 | −0.2057 | 1.593 | 0.024 | 0.587 |
| 0.7941 | 1445.07 | −0.0591 | 1009.6 | −0.1614 | 1.356 | 0.020 | 0.509 |
| 0.8847 | 1501.43 | −0.0355 | 961.9 | −0.1011 | 1.130 | 0.013 | 0.359 |
| 1.0000 | 1574.62 | 0.0000 | 905.4 | 0.0000 | 0.838 | 0.000 | 0.000 |
| 308.15 K | | | | | | | |
| 0.0000 | 985.99[8] | 0.0000 | 1558.2 [8] | 0.0000 | 2.824 | 0.000 | 0.000 |
| 0.0901 | 1034.03 | −0.0477 | 1479.4 | −0.0884 | 2.650 | 0.010 | 0.133 |
| 0.1916 | 1089.00 | −0.0810 | 1394.5 | −0.1620 | 2.452 | 0.019 | 0.267 |
| 0.2917 | 1144.18 | −0.1009 | 1315.6 | −0.2119 | 2.254 | 0.025 | 0.380 |
| 0.3903 | 1199.49 | −0.1087 | 1243.0 | −0.2426 | 2.056 | 0.028 | 0.469 |
| 0.4988 | 1261.52 | −0.1084 | 1168.7 | −0.2556 | 1.836 | 0.030 | 0.535 |
| 0.6089 | 1325.76 | −0.1004 | 1099.1 | −0.2484 | 1.611 | 0.029 | 0.562 |
| 0.6981 | 1378.79 | −0.0888 | 1046.5 | −0.2251 | 1.426 | 0.026 | 0.542 |
| 0.7941 | 1436.85 | −0.0699 | 993.4 | −0.1814 | 1.224 | 0.020 | 0.466 |
| 0.8847 | 1492.59 | −0.0455 | 946.2 | −0.1184 | 1.032 | 0.013 | 0.326 |
| 1.0000 | 1564.80 | 0.0000 | 889.7 | 0.0000 | 0.784 | 0.000 | 0.000 |
| 313.15 K | | | | | | | |
| 0.0000 | 981.69[8] | 0.0000 | 1538.5 [6] | 0.0000 | 2.487 | 0.000 | 0.000 |
| 0.0901 | 1029.43 | −0.0548 | 1460.6 | −0.0987 | 2.340 | 0.011 | 0.127 |
| 0.1916 | 1083.99 | −0.0905 | 1375.9 | −0.1772 | 2.172 | 0.021 | 0.256 |
| 0.2917 | 1138.70 | −0.1089 | 1296.9 | −0.2273 | 2.003 | 0.027 | 0.363 |
| 0.3903 | 1193.53 | −0.1163 | 1224.4 | −0.2577 | 1.834 | 0.031 | 0.447 |
| 0.4988 | 1255.00 | −0.1157 | 1150.6 | −0.2717 | 1.646 | 0.033 | 0.510 |
| 0.6089 | 1318.66 | −0.1093 | 1081.8 | −0.2675 | 1.452 | 0.032 | 0.534 |
| 0.6981 | 1371.20 | −0.0996 | 1029.9 | −0.2466 | 1.292 | 0.029 | 0.513 |
| 0.7941 | 1428.69 | −0.0816 | 977.4 | −0.2026 | 1.118 | 0.023 | 0.441 |
| 0.8847 | 1483.82 | −0.0550 | 930.6 | −0.1352 | 0.951 | 0.015 | 0.308 |
| 1.0000 | 1555.10 | 0.0000 | 874.2 | 0.0000 | 0.734 | 0.000 | 0.000 |
| 318.15 K | | | | | | | |
| 0.0000 | 977.60[8] | 0.0000 | 1518.5[8] | 0.0000 | 2.153 | 0.000 | 0.000 |
| 0.0901 | 1025.03 | −0.0637 | 1441.6 | −0.1106 | 2.033 | 0.012 | 0.119 |
| 0.1916 | 1079.15 | −0.1020 | 1356.8 | −0.1923 | 1.895 | 0.023 | 0.239 |
| 0.2917 | 1133.34 | −0.1175 | 1277.8 | −0.2430 | 1.755 | 0.029 | 0.338 |
| 0.3903 | 1187.62 | −0.1219 | 1205.4 | −0.2723 | 1.616 | 0.034 | 0.417 |
| 0.4988 | 1248.49 | −0.1220 | 1132.1 | −0.2868 | 1.459 | 0.036 | 0.474 |
| 0.6089 | 1311.50 | −0.1165 | 1064.1 | −0.2850 | 1.296 | 0.034 | 0.492 |
| 0.6981 | 1363.49 | −0.1081 | 1012.9 | −0.2659 | 1.162 | 0.031 | 0.472 |
| 0.7941 | 1420.37 | −0.0927 | 961.2 | −0.2239 | 1.016 | 0.026 | 0.406 |
| 0.8847 | 1474.85 | −0.0656 | 914.9 | −0.1528 | 0.874 | 0.016 | 0.281 |
| 1.0000 | 1545.10 | 0.0000 | 858.7 | 0.0000 | 0.689 | 0.000 | 0.000 |
| tetra chloromethane (1) + 2-methoxyaniline (2) | | | | | | | |
| 298.15 K | | | | | | | |
| 0.0000 | 1095.26 | 0.0000 | 1613.5 | 0.0000 | 5.374 | 0.000 | 0.000 |
| 0.0821 | 1130.69 | −0.0353 | 1544.8 | −0.0705 | 5.011 | 0.005 | 0.204 |
| 0.1726 | 1170.60 | −0.0642 | 1469.9 | −0.1285 | 4.611 | 0.010 | 0.394 |
| 0.2547 | 1207.63 | −0.0814 | 1403.8 | −0.1658 | 4.247 | 0.013 | 0.546 |
| 0.3403 | 1247.08 | −0.0925 | 1337.5 | −0.1912 | 3.866 | 0.016 | 0.687 |
| 0.4788 | 1312.87 | −0.0971 | 1236.5 | −0.2070 | 3.249 | 0.018 | 0.877 |
| 0.5789 | 1361.99 | −0.0909 | 1168.4 | −0.2008 | 2.800 | 0.018 | 0.964 |
| 0.6781 | 1412.03 | −0.0793 | 1104.8 | −0.1803 | 2.355 | 0.017 | 0.980 |
| 0.7641 | 1456.55 | −0.0639 | 1052.5 | −0.1505 | 1.967 | 0.014 | 0.907 |
| 0.8447 | 1499.29 | −0.0457 | 1005.8 | −0.1115 | 1.603 | 0.010 | 0.735 |
| 1.0000 | 1584.50 | 0.0000 | 921.1 | 0.0000 | 0.897 | 0.000 | 0.000 |
| 303.15 K | | | | | | | |
| 0.0000 | 1091.57[7] | 0.0000 | 1595.4[7] | 0.0000 | 4.922 | 0.000 | 0.000 |
| 0.0821 | 1126.70 | −0.0405 | 1526.6 | −0.0801 | 4.596 | 0.009 | 0.207 |
| 0.1726 | 1166.22 | −0.0706 | 1450.4 | −0.1417 | 4.234 | 0.017 | 0.398 |
| 0.2547 | 1202.85 | −0.0889 | 1383.1 | −0.1788 | 3.904 | 0.022 | 0.551 |
| 0.3403 | 1241.86 | −0.0996 | 1316.0 | −0.2027 | 3.559 | 0.026 | 0.694 |
| 0.4788 | 1306.85 | −0.1029 | 1214.9 | −0.2171 | 2.996 | 0.029 | 0.885 |
| 0.5789 | 1355.36 | −0.0980 | 1147.7 | −0.2119 | 2.587 | 0.029 | 0.973 |
| 0.6781 | 1404.75 | −0.0862 | 1085.4 | −0.1937 | 2.179 | 0.027 | 0.988 |
| 0.7641 | 1448.68 | −0.0715 | 1034.4 | −0.1656 | 1.824 | 0.022 | 0.914 |
| 0.8447 | 1490.80 | −0.0525 | 988.7 | −0.1261 | 1.489 | 0.017 | 0.741 |
| 1.0000 | 1574.62 | 0.0000 | 905.4 | 0.0000 | 0.838 | 0.000 | 0.000 |
| 308.15 K | | | | | | | |

(continued on next page)

**Table 3** (*continued*)

| $x_1$ | Density(ρ)$Kg{\cdot}m^{-3}$ | $V^E 10^6 m^3{\cdot}mol^{-1}$ | $u$ $m.s^{-1}$ | $\kappa_s^E 10^{10} m^2.N^{-1}$ | viscosity (η / mPa·s) | $\Delta\eta$ / mPa·s | $G^{*E} J{\cdot}mol^{-1}$ |
|---|---|---|---|---|---|---|---|
| 0.0000 | 1087.99[7] | 0.0000 | 1579.2[7] | 0.0000 | 4.512 | 0.000 | 0.000 |
| 0.0821 | 1122.83 | −0.0471 | 1511.1 | −0.0908 | 4.218 | 0.012 | 0.208 |
| 0.1726 | 1161.97 | −0.0808 | 1434.5 | −0.1560 | 3.892 | 0.023 | 0.402 |
| 0.2547 | 1198.20 | −0.0992 | 1366.5 | −0.1924 | 3.593 | 0.031 | 0.557 |
| 0.3403 | 1236.75 | −0.1087 | 1298.8 | −0.2142 | 3.280 | 0.037 | 0.701 |
| 0.4788 | 1300.94 | −0.1108 | 1197.5 | −0.2265 | 2.768 | 0.041 | 0.894 |
| 0.5789 | 1348.82 | −0.1051 | 1130.6 | −0.2223 | 2.394 | 0.040 | 0.983 |
| 0.6781 | 1397.56 | −0.0943 | 1068.9 | −0.2071 | 2.020 | 0.036 | 0.998 |
| 0.7641 | 1440.88 | −0.0798 | 1018.4 | −0.1812 | 1.694 | 0.030 | 0.922 |
| 0.8447 | 1482.39 | −0.0608 | 973.0 | −0.1418 | 1.385 | 0.022 | 0.746 |
| 1.0000 | 1564.80 | 0.0000 | 889.7 | 0.0000 | 0.784 | 0.000 | 0.000 |
| 313.15 K | | | | | | | |
| 0.0000 | 1084.32[7] | 0.0000 | 1566.3[7] | 0.0000 | 4.185 | 0.000 | 0.000 |
| 0.0821 | 1118.88 | −0.0540 | 1497.6 | −0.1014 | 3.917 | 0.016 | 0.213 |
| 0.1726 | 1157.64 | −0.0901 | 1419.3 | −0.1704 | 3.619 | 0.030 | 0.411 |
| 0.2547 | 1193.48 | −0.1086 | 1349.9 | −0.2062 | 3.346 | 0.039 | 0.569 |
| 0.3403 | 1231.59 | −0.1175 | 1281.0 | −0.2260 | 3.057 | 0.047 | 0.716 |
| 0.4788 | 1295.01 | −0.1189 | 1179.1 | −0.2365 | 2.585 | 0.052 | 0.914 |
| 0.5789 | 1342.30 | −0.1140 | 1112.6 | −0.2336 | 2.239 | 0.052 | 1.005 |
| 0.6781 | 1390.42 | −0.1044 | 1051.6 | −0.2211 | 1.892 | 0.047 | 1.022 |
| 0.7641 | 1433.16 | −0.0907 | 1001.8 | −0.1976 | 1.587 | 0.039 | 0.945 |
| 0.8447 | 1474.07 | −0.0707 | 957.0 | −0.1580 | 1.299 | 0.029 | 0.766 |
| 1.0000 | 1555.10 | 0.0000 | 874.2 | 0.0000 | 0.734 | 0.000 | 0.000 |
| 318.15 K | | | | | | | |
| 0.0000 | 1080.82[7] | 0.0000 | 1554.8[7] | 0.0000 | 3.886 | 0.000 | 0.000 |
| 0.0821 | 1114.84 | −0.0623 | 1486.0 | −0.1124 | 3.643 | 0.020 | 0.218 |
| 0.1726 | 1153.21 | −0.1014 | 1406.5 | −0.1852 | 3.371 | 0.037 | 0.421 |
| 0.2547 | 1188.64 | −0.1194 | 1335.7 | −0.2206 | 3.121 | 0.049 | 0.582 |
| 0.3403 | 1226.27 | −0.1255 | 1265.8 | −0.2384 | 2.856 | 0.058 | 0.733 |
| 0.4788 | 1288.90 | −0.1268 | 1163.1 | −0.2475 | 2.420 | 0.065 | 0.936 |
| 0.5789 | 1335.57 | −0.1220 | 1096.6 | −0.2459 | 2.099 | 0.064 | 1.030 |
| 0.6781 | 1383.06 | −0.1148 | 1035.8 | −0.2366 | 1.776 | 0.058 | 1.047 |
| 0.7641 | 1425.21 | −0.1025 | 986.3 | −0.2153 | 1.491 | 0.048 | 0.969 |
| 0.8447 | 1465.52 | −0.0829 | 941.8 | −0.1754 | 1.221 | 0.035 | 0.785 |
| 1.0000 | 1545.10 | 0.0000 | 858.7 | 0.0000 | 0.689 | 0.000 | 0.000 |

From Fig. 5, negative values of $\kappa_s^E$for a given system are attributed to the complex formation through N····Cl specific interactions between tetrachloromethane and *ortho*-substitutedaniline.There is, however, a possibility of involving in the formation of charge transfer interaction between the n-donor nitrogen atom of the *ortho*-substituted aniline and σ-acceptor chlorine atom of tetrachloromethane in the binary liquid mixtures [51,52].There is evidence for the Stevenson and Coppinger [53], Biaselle,Miller [54] and Oswal et al[55] observed a charge transfer band UV spectrum in solutions of alkylamine with tetrachloromethane

The negative values of$\kappa_s^E$fall in the following order: Tetrachloromethane + 2-methoxy aniline > tetrachloromethane + 2-methylaniline > tetrachloromethane + 2-chloroaniline

The trends in experimental deviation in viscosities (Fig. 6) and excess Gibbs energy of activation of flow (Fig. 7) may be explained qualitatively in terms of molecular interactions between like / unlike molecules and the difference in size and shape of the unlike components. The positive values of deviation in viscosities and excess Gibbs energy of activation of flow indicate thepresence of specific interactions in the binary mixtures, attributed to the formation of donor–acceptor complexes through (C-Cl·····$H_2N$) hydrogen bonding between tetrachloromethane and *ortho*-substituted aniline molecules and also the mutual loss of dipolar association of the *ortho*-substituted aniline components[56].

### 3.1. Partial molar properties

The interpretations of excess partial molar properties ($\bar{V}^{E}_{m,1}$,$\bar{V}^{E}_{m,2}$,$\bar{K}^{E}_{s,m,1}$and$\bar{K}^{E}_{s,m,2}$) and excess partial molar properties at infinite dilution ($\bar{V}^{\circ E}_{m,1}$, $\bar{V}^{\circ E}_{m,2}\bar{K}^{\circ E}_{s,m,1}$and$\bar{K}^{\circ E}_{s,m,2}$) of components 2 have previously been described [47].

Tables 5 and 6 show that the values of,$\bar{V}^{\circ E}_{m,1}$, $\bar{V}^{\circ E}_{m,2}$, $\bar{K}^{\circ E}_{s,m,1}$ and $\bar{K}^{\circ E}_{s,m,2}$ are negative over the whole composition ranges at experimental temperatures. Negative values may be attributed to the hydrogen bonding between unlike moleculesare dominant over the breaking up of self-associated structures of the components of the mixtures.

### 3.2. FT-IR properties

The infrared spectrum, which gives important information about the functional groups, can be appreciably influenced by the surrounding condensed medium [57]. Hence, FT-IR plays a significant role in the study of intermolecular hydrogen bonds present in the molecular structure of complexes in the binary mixture [58,59,60].

The dilute solution of primary amines gives two sharp bands due to asymmetric (high frequency) and symmetric (low frequency) stretching vibrations between 3200 and 3500 $cm^{-1}$.In aromatic amines, lower νN-H absorption values are expected due to negative inductive (-*I*) and positive electrometric (+*E*) effects and nitrogen atom is partially rehybridized towards $SP^2$ due to conjugation.

In the present paper, infrared spectra have been recorded for the pure liquids and binary mixtures at room temperature, and the variations of the frequencies of these mixtures are given in Table 7 and Figs. 8-14. FT-IR absorption bands of pure compounds are tetrachloromethane, 2-chloroaniline, 2-methylaniline, 2-methoxyaniline exhibited intense bands at 789.79 $cm^{-1}$(C-Cl stretching), 3370.86 $cm^{-1}$(–$NH_2$ stretching), 3363.56 $cm^{-1}$(–$NH_2$

**Table 4**
Coefficients $A_i$ of equation (7) along with standard deviations $\sigma$ of binary mixture properties.

| T/K | $A_0$ | $A_1$ | $A_2$ | $\sigma$ |
|---|---|---|---|---|
| tetra chloromethane (1) + 2-chloroaniline (2) | | | | |
| $V_m^E 10^6$ $m^3.mol^{-1}$ | | | | |
| 298.15 | −0.366 | 0.066 | 0.104 | 0.0009 |
| 303.15 | −0.393 | 0.071 | 0.043 | 0.0005 |
| 308.15 | −0.419 | 0.074 | −0.054 | 0.0008 |
| 313.15 | −0.444 | 0.076 | −0.155 | 0.0013 |
| 318.15 | −0.469 | 0.079 | −0.253 | 0.0005 |
| $\kappa_s^E 10^{10}$ $m^2.N^{-1}$ | | | | |
| 298.15 | −1.6625 | −0.2609 | 0.1932 | 0.0003 |
| 303.15 | −1.7388 | −0.2718 | 0.0645 | 0.0003 |
| 308.15 | −1.8148 | −0.2843 | −0.1160 | 0.0004 |
| 313.15 | −1.8984 | −0.3067 | −0.2771 | 0.0003 |
| 318.15 | −1.9840 | −0.3381 | −0.4581 | 0.0003 |
| $\Delta\eta$ / mPa·s | | | | |
| 298.15 | 0.101 | 0.011 | 0.003 | 0.0003 |
| 303.15 | 0.112 | 0.008 | −0.004 | 0.0004 |
| 308.15 | 0.121 | 0.005 | 0.000 | 0.0004 |
| 313.15 | 0.128 | 0.013 | 0.019 | 0.0004 |
| 318.15 | 0.138 | 0.015 | 0.034 | 0.0005 |
| $G^{*E}$ $J\cdot mol^{-1}$ | | | | |
| 298.15 | 2.797 | 1.442 | 0.785 | 0.0072 |
| 303.15 | 2.774 | 1.411 | 0.759 | 0.0068 |
| 308.15 | 2.691 | 1.334 | 0.702 | 0.0061 |
| 313.15 | 2.620 | 1.290 | 0.693 | 0.0059 |
| 318.15 | 2.566 | 1.251 | 0.692 | 0.0059 |
| tetra chloromethane (1) + 2-methylaniline (2) | | | | |
| $V_m^E 10^6$ $m^3.mol^{-1}$ | | | | |
| 298.15 | −0.374 | 0.072 | 0.0373 | 0.0009 |
| 303.15 | −0.406 | 0.076 | −0.0043 | 0.0005 |
| 308.15 | −0.434 | 0.077 | −0.1181 | 0.0011 |
| 313.15 | −0.463 | 0.067 | −0.2172 | 0.0014 |
| 318.15 | −0.481 | 0.069 | −0.3693 | 0.0030 |
| $\kappa_s^E 10^{10}$ $m^2.N^{-1}$ | | | | |
| 298.15 | −0.8956 | −0.0094 | 0.0557 | 0.0002 |
| 303.15 | −0.9607 | −0.0316 | −0.0179 | 0.0003 |
| 308.15 | −1.0230 | −0.0569 | −0.1534 | 0.0002 |
| 313.15 | −1.0888 | −0.0898 | −0.2848 | 0.0003 |
| 318.15 | −1.1485 | −0.1170 | −0.4378 | 0.0003 |
| $\Delta\eta$ / mPa·s | | | | |
| 298.15 | 0.105 | 0.007 | 0.022 | 0.0003 |
| 303.15 | 0.113 | 0.007 | 0.012 | 0.0005 |
| 308.15 | 0.120 | 0.003 | 0.008 | 0.0003 |
| 313.15 | 0.132 | 0.007 | 0.016 | 0.0002 |
| 318.15 | 0.142 | 0.008 | 0.021 | 0.0005 |
| $G^{*E}$ $J\cdot mol^{-1}$ | | | | |
| 298.15 | 2.430 | 1.172 | 0.627 | 0.0053 |
| 303.15 | 2.301 | 1.068 | 0.540 | 0.0045 |
| 308.15 | 2.142 | 0.945 | 0.454 | 0.0033 |
| 313.15 | 2.040 | 0.878 | 0.418 | 0.0033 |
| 318.15 | 1.892 | 0.781 | 0.364 | 0.0031 |
| tetra chloromethane (1) + 2-methoxyaniline (2) | | | | |
| $V_m^E 10^6$ $m^3.mol^{-1}$ | | | | |
| 298.15 | −0.385 | 0.075 | −0.0264 | 0.00003 |
| 303.15 | −0.410 | 0.073 | −0.0890 | 0.00003 |
| 308.15 | −0.439 | 0.084 | −0.1684 | 0.00003 |
| 313.15 | −0.472 | 0.077 | −0.2553 | 0.00002 |
| 318.15 | −0.502 | 0.069 | −0.3716 | 0.00033 |
| $\kappa_s^E 10^{10}$ $m^2.N^{-1}$ | | | | |
| 298.15 | −1.145 | −0.098 | −0.149 | 0.0015 |
| 303.15 | −1.207 | −0.120 | −0.329 | 0.0031 |
| 308.15 | −1.277 | −0.143 | −0.485 | 0.0045 |
| 313.15 | −1.331 | −0.176 | −0.675 | 0.0063 |
| 318.15 | −1.388 | −0.206 | −0.862 | 0.0075 |
| $\Delta\eta$ / mPa·s | | | | |
| 298.15 | 0.073 | 0.009 | 0.000 | 0.0003 |
| 303.15 | 0.118 | 0.006 | 0.011 | 0.0002 |
| 308.15 | 0.164 | 0.005 | 0.004 | 0.0003 |
| 313.15 | 0.211 | 0.009 | 0.007 | 0.0003 |
| 318.15 | 0.260 | 0.008 | 0.009 | 0.0002 |
| $G^{*E}$ $J\cdot mol^{-1}$ | | | | |
| 298.15 | 3.600 | 2.071 | 1.206 | 0.0104 |
| 303.15 | 3.631 | 2.084 | 1.222 | 0.0102 |
| 308.15 | 3.671 | 2.097 | 1.207 | 0.0101 |
| 313.15 | 3.752 | 2.160 | 1.256 | 0.0108 |
| 318.15 | 3.843 | 2.216 | 1.291 | 0.0111 |

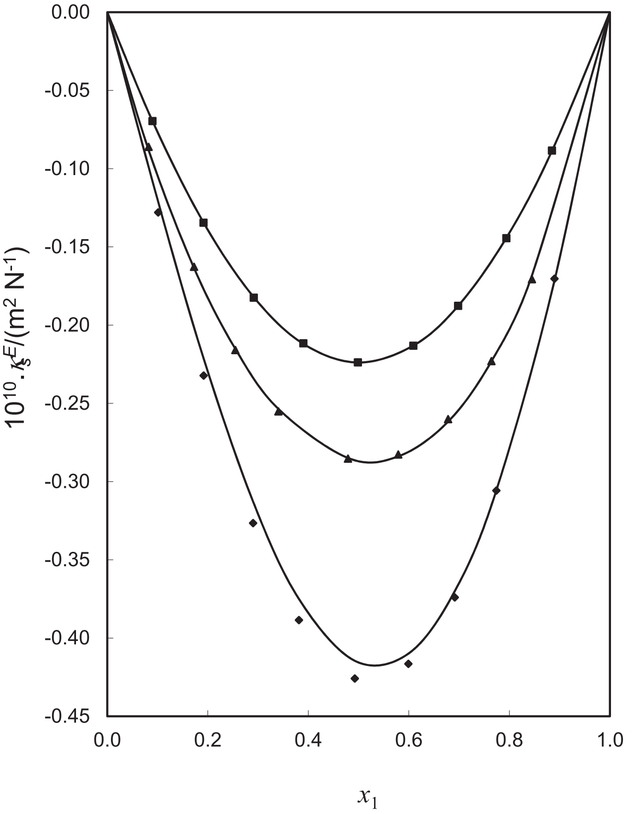

**Fig. 5.** Excess isentropic compressibilities ($\kappa_s^E$) with mole fraction $x_1$of tetra chloromethane (1) + 2-chloroaniline (2),◆; tetra chloromethane (1) + 2-methylaniline (2),■;tetra chloromethane (1) + 2-methoxyaniline (2), ▲;binary mixtures at temperatures, *T*/K = 298.15. The points represent experimental values and lines represent values calculated from equation (7) using the coefficients given in table 4.

stretching), and 3369.12 cm$^{-1}$(–NH$_2$ stretching), respectively,From Table 7, it is clear that the values of C-Cl . . .·. . .· H$_2$N stretching frequency for the equimolar mixtures of tetrachloromethane + 2-chloroaniline (3384.02 cm$^{-1}$), tetrachloromethane + 2-methylaniline (3379.74 cm$^{-1}$) and tetrachloromethane + 2-methoxyaniline (3374.00 cm$^{-1}$) are observed. The difference in the stretching frequency represents the bond strength and leading to specific interactions through hydrogen bonding between the unlike molecules promoting attractive interactions leading to volume contraction which supports the excess molar volume behaviour. Based on the magnitude of the shift in the stretching frequencies of C-Cl ····· H$_2$N band in the studied all binaries, the strength of molecular interaction between tetrachloromethane and ortho – mono substituted aniline follows the order: (tetrachloromethane + 2-methoxyaniline) > (tetrachloromethane + 2-chloroaniline) > (tetrachloromethane + 2-methylaniline) the absorption band shifted to alower frequency. The frequency shifts are caused by the strong intermolecular interaction like hydrogen bonding between the unlike molecules in the binary mixtures. The C-Cl stretching vibration of aromatic amines is observed as bands in the region 760–830 cm$^{-1}$. Further, the values of ν(C-Cl) bands for the equimolar mixtures of tetrachloromethane + 2-chloroaniline, tetrachloromethane + 2-methylaniline, and tetrachloromethane + 2-methoxyaniline were in the order of 790.38, 789.92, and 789.88 cm$^{-1}$ respectively and their corresponding Δν values of ν(C-Cl) absorption band of tetrachloromethane (789.79 cm$^{-1}$)were 0.59, 0.13 and 0.09 cm$^{-1}$respectively. The frequency shifts are caused by the strong intermolecular interaction like hydrogen bonding between unlike mole-

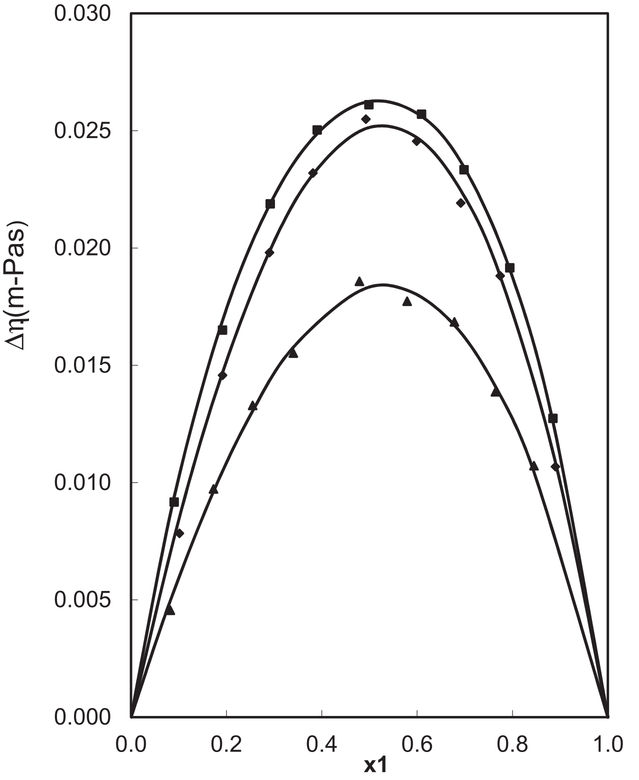

**Fig. 6.** Deviation in viscosity, Δ*η*vs. mole fraction, $x_1$of tetra chloromethane (1) + 2-chloroaniline (2),◆; tetra chloromethane (1) + 2-methylaniline (2),■;tetra chloromethane (1) + 2-methoxyaniline (2), ▲;binary mixtures at temperatures, *T*/K = 298.15. The points represent experimental values and lines represent values calculated from equation (7) using the coefficients given in table 4.

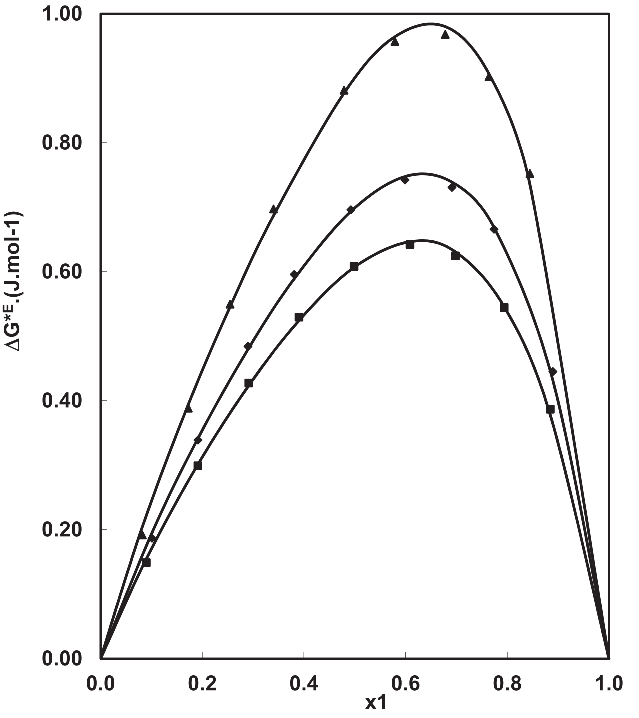

**Fig. 7.** Excess Gibbs free energies, $\Delta G^{*E}$*vs.* mole fraction, $x_1$of tetra chloromethane (1) + 2-chloroaniline (2),◆; tetra chloromethane (1) + 2-methylaniline (2),■;tetra chloromethane (1) + 2-methoxyaniline (2), ▲;binary mixtures at temperatures, *T*/K = 298.15. The points represent experimental values and lines represent values calculated from equation (7) using the coefficients given in table 4.

**Table 5**
The values of $\bar{V}^{\circ}_{m,1}$,$V^{*}_{m,1}$,$\bar{V}^{\circ E}_{m,1}$,$\bar{V}^{\circ}_{m,2}$, $V^{*}_{m,2}$and$\bar{V}^{\circ E}_{m,2}$of the components for tetrachloro methane with substituted aniline of binary mixtures at T= (303.15–318.15) K

| T/K | $\bar{V}^{\circ}_{m,1}$ | $V^{*}_{m,1}$ | $\cdot\bar{V}^{\circ E}_{m,1}$ | $V^{*}_{m,2}$ | $V^{*}_{m,2}$ | $\bar{V}^{\circ E}_{m,2}$ |
|---|---|---|---|---|---|---|
| ($10^6$ $m^3.mol^{-1}$) | | | | | | |
| tetra chloromethane (1) + 2-chloroaniline (2) | | | | | | |
| 298.15 | 96.75 | 97.08 | −0.328 | 105.45 | 105.65 | −0.196 |
| 303.15 | 97.27 | 97.69 | −0.421 | 105.79 | 106.07 | −0.278 |
| 308.15 | 97.75 | 98.30 | −0.547 | 106.08 | 106.48 | −0.399 |
| 313.15 | 98.24 | 98.91 | −0.676 | 106.41 | 106.93 | −0.523 |
| 318.15 | 96.92 | 99.55 | −2.631 | 106.06 | 107.34 | −1.280 |
| tetra chloromethane (1) + 2-methylaniline (2) | | | | | | |
| 298.15 | 96.67 | 97.08 | −0.408 | 107.48 | 107.75 | −0.264 |
| 303.15 | 97.20 | 97.69 | −0.486 | 107.88 | 108.21 | −0.334 |
| 308.15 | 97.67 | 98.30 | −0.629 | 108.20 | 108.67 | −0.475 |
| 313.15 | 98.17 | 98.91 | −0.747 | 108.54 | 109.15 | −0.613 |
| 318.15 | 98.63 | 99.55 | −0.919 | 108.82 | 109.61 | −0.781 |
| tetra chloromethane (1) + 2-methoxyaniline (2) | | | | | | |
| 298.15 | 96.59 | 97.08 | −0.487 | 112.12 | 112.46 | −0.337 |
| 303.15 | 97.11 | 97.69 | −0.573 | 112.39 | 112.82 | −0.426 |
| 308.15 | 97.61 | 98.30 | −0.692 | 112.67 | 113.19 | −0.524 |
| 313.15 | 98.11 | 98.91 | −0.804 | 112.92 | 113.57 | −0.650 |
| 318.15 | 98.61 | 99.55 | −0.943 | 113.16 | 113.97 | −0.805 |

**Table 6**
The values of$\bar{K}^{\circ}_{s,m,1}$, $K^{*}_{s,m,1}$, $\bar{K}^{\circ E}_{s,m,1}$, $\bar{K}^{\circ}_{s,m,2}$, $K^{*}_{s,m,2}$ and $\bar{K}^{\circ E}_{s,m,2}$of the components for tetrachloro methane with substituted aniline of binary mixtures at $T$ = (303.15–318.15) K

| T/K | $\bar{K}^{\circ}_{s,m,1}$ | $K^{*}_{s,m,1}$ | $\bar{K}^{\circ E}_{s,m,1}$ | $\cdot\bar{K}^{\circ}_{s,m,2}$ | $K^{*}_{s,m,2}$ | $\bar{K}^{\circ E}_{s,m,2}$ |
|---|---|---|---|---|---|---|
| $10^{10}$ $m^2.N^{-1}$ | | | | | | |
| tetra chloromethane (1) + 2-chloroaniline (2) | | | | | | |
| 298.15 | 6.013 | 7.221 | −1.208 | 2.232 | 3.963 | −1.730 |
| 303.15 | 6.166 | 7.568 | −1.402 | 2.137 | 4.084 | −1.946 |
| 308.15 | 6.290 | 7.936 | −1.647 | 1.993 | 4.208 | −2.215 |
| 313.15 | 6.454 | 8.323 | −1.869 | 1.869 | 4.351 | −2.482 |
| 318.15 | 6.634 | 8.738 | −2.104 | 1.706 | 4.486 | −2.780 |
| tetra chloromethane (1) + 2-methylaniline (2) | | | | | | |
| 298.15 | 6.391 | 7.221 | −0.831 | 3.391 | 4.240 | −0.849 |
| 303.15 | 6.621 | 7.568 | −0.947 | 3.376 | 4.386 | −1.010 |
| 308.15 | 6.817 | 7.936 | −1.120 | 3.306 | 4.539 | −1.233 |
| 313.15 | 7.039 | 8.323 | −1.284 | 3.234 | 4.697 | −1.463 |
| 318.15 | 7.269 | 8.738 | −1.469 | 3.159 | 4.862 | −1.703 |
| tetra chloromethane (1) + 2-methoxyanilineaniline (2) | | | | | | |
| 298.15 | 6.025 | 7.221 | −1.196 | 2.570 | 3.962 | −1.392 |
| 303.15 | 6.153 | 7.568 | −1.416 | 2.395 | 4.050 | −1.655 |
| 308.15 | 6.317 | 7.936 | −1.619 | 2.245 | 4.151 | −1.905 |
| 313.15 | 6.492 | 8.323 | −1.831 | 2.087 | 4.269 | −2.183 |
| 318.15 | 6.694 | 8.738 | −2.044 | 1.907 | 4.363 | −2.456 |

**Table 7**
Experimental FT-IR frequencies with wave numbers ($cm^{-1}$) and shifting of bands of 2-methylcyclohexanone (1) + mono substituted aniline (2) of binary mixtures at $T$ = 298.15 K

| compound | Band | experimental ν ($cm^{-1}$) |
|---|---|---|
| 2-methylcyclohexanone | OH | 3403.83 |
| 2-chloroaniline | $NH_2$ | 3370.86 |
| 2-methylaniline | $NH_2$ | 3363.56 |
| 2-methoxyaniline | $NH_2$ | 3369.12 |
| 2-methylcyclohexanone + 2-chloroaniline | OH . . .∴. . . $NH_2$ | 3369.29 |
| 2-methylcyclohexanone + 2-methylaniline | OH . . .∴. . . $NH_2$ | 3374.50 |
| 2-methylcyclohexanone + 2-methoxyaniline | OH . . .∴. . . $NH_2$ | 3371.75 |
| Shift in wave numbers with respect to mono-substituted aniline | | |
| 2-methylcyclohexanone + 2-chloroaniline | (5) – (2) | −1.57 |
| 2-methylcyclohexanone + 2-methylaniline | (6) - (3) | 10.94 |
| 2-methylcyclohexanone + 2-methoxyaniline | (7) – (4) | 2.64 |

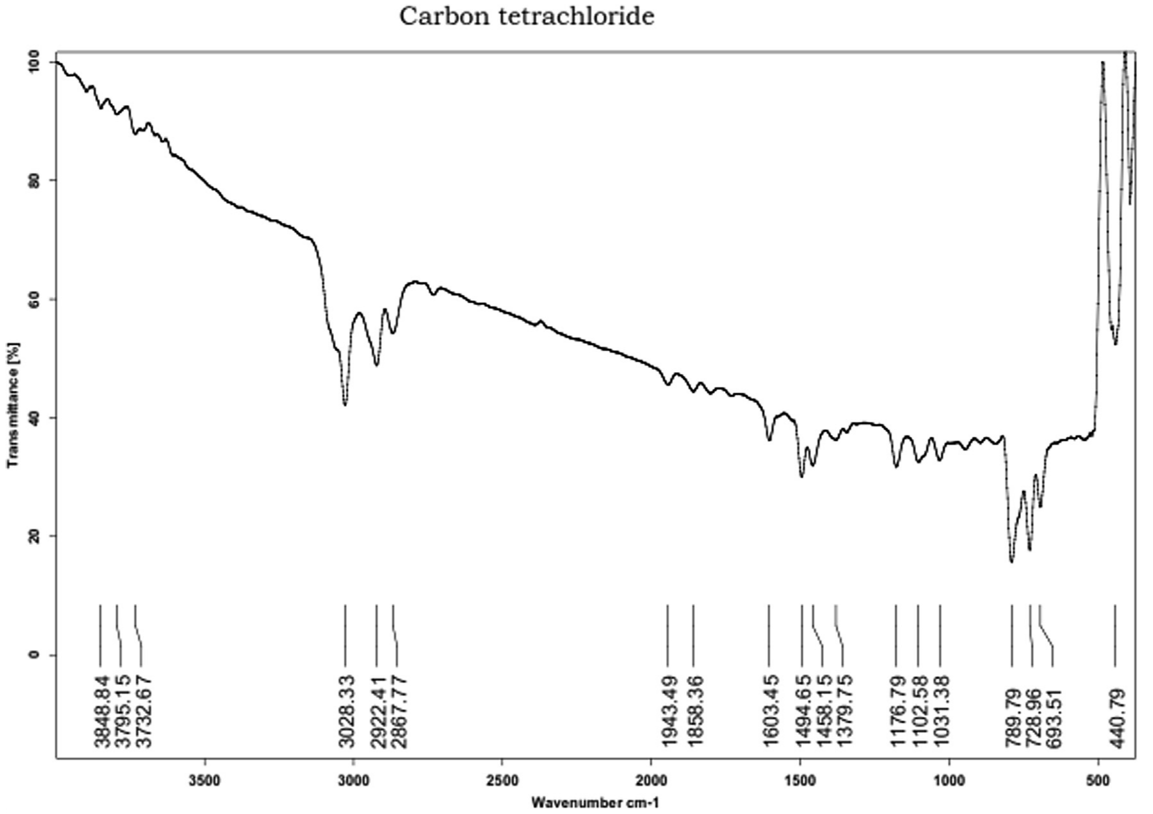

**Fig. 8.** Experimental Fourier Transform Infrared spectra of pure tetrachloromethane.

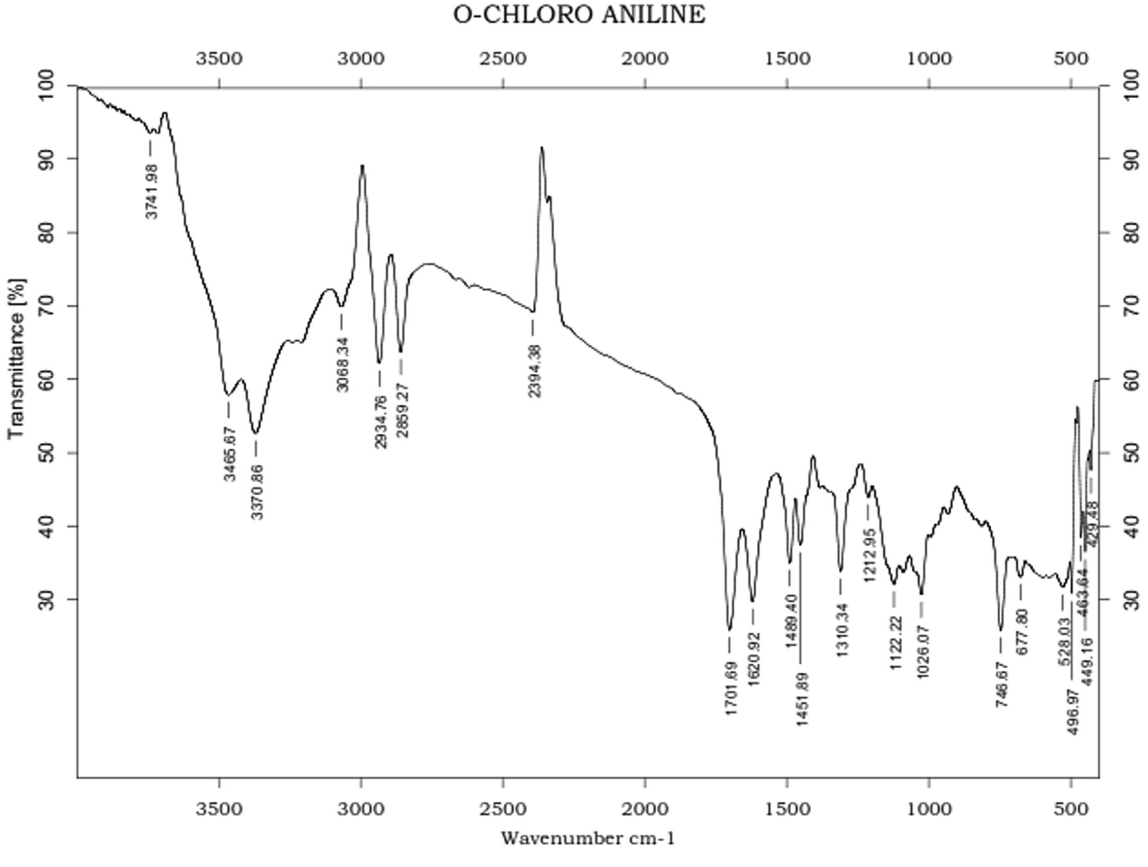

**Fig. 9.** Experimental Fourier Transform Infrared spectra of pure 2-chloroaniline.

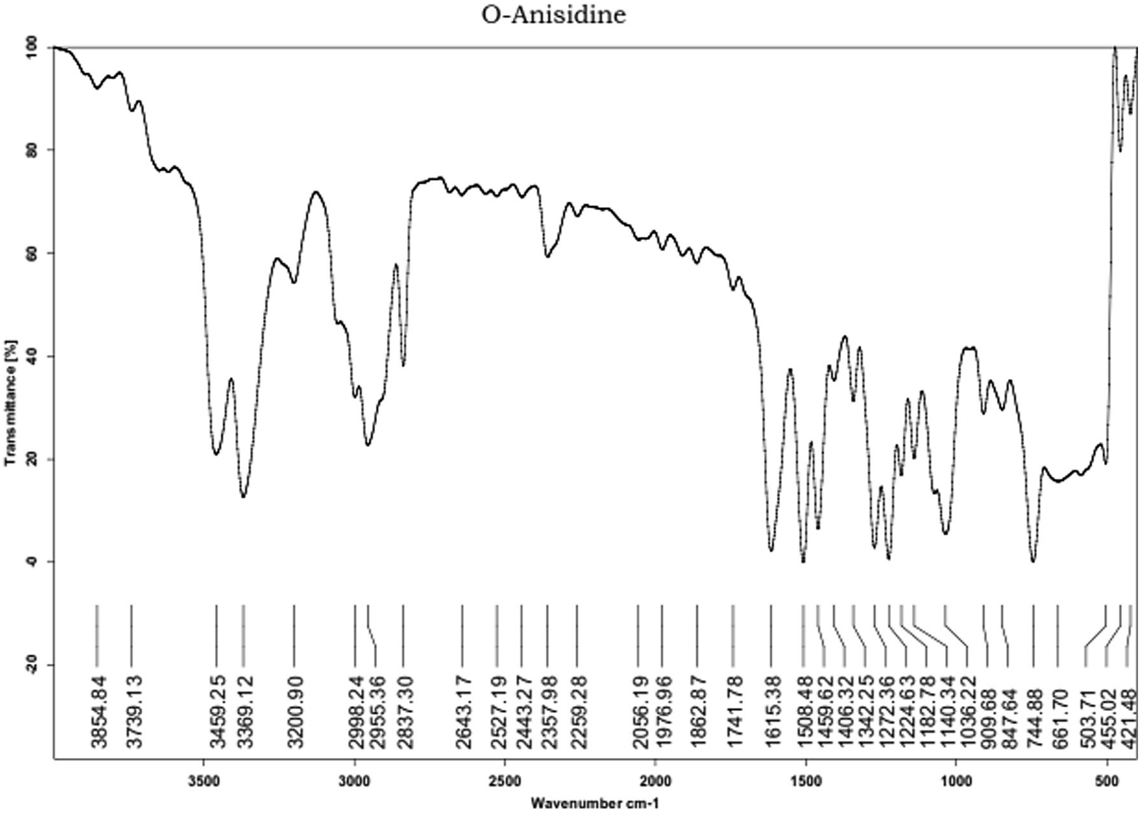

**Fig. 10.** Experimental Fourier Transform Infrared spectra of pure 2-methoxyaniline.

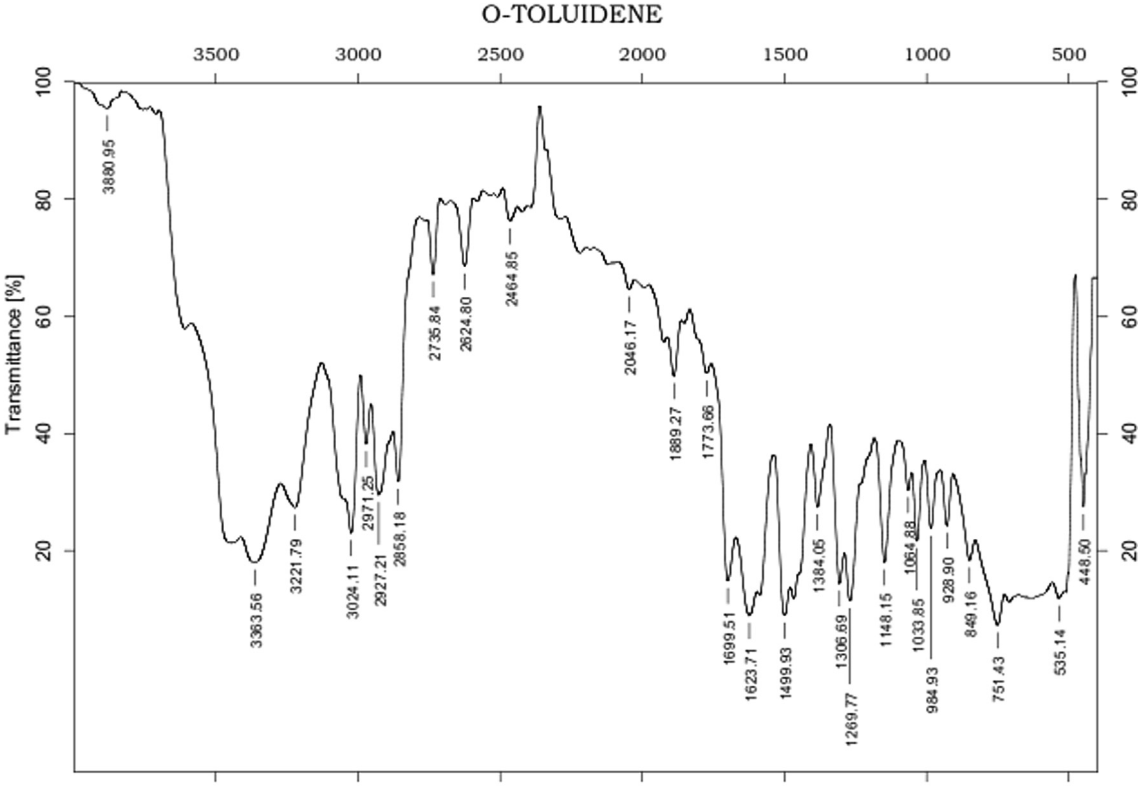

**Fig. 11.** Experimental Fourier Transform Infrared spectra of pure 2-methyl aniline.

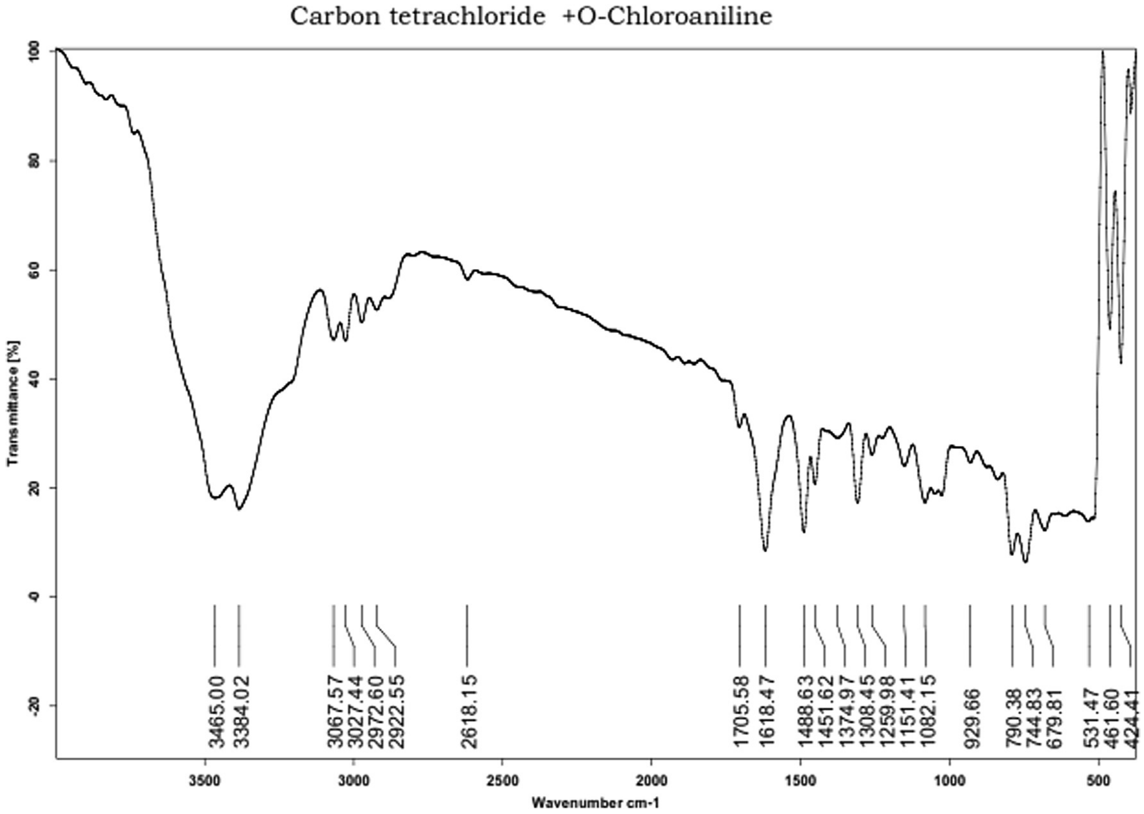

**Fig. 12.** Experimental Fourier Transform Infrared spectra of binary liquid mixtures (tetrachloromethane + 2-chloro aniline) in the ratio 1:1.

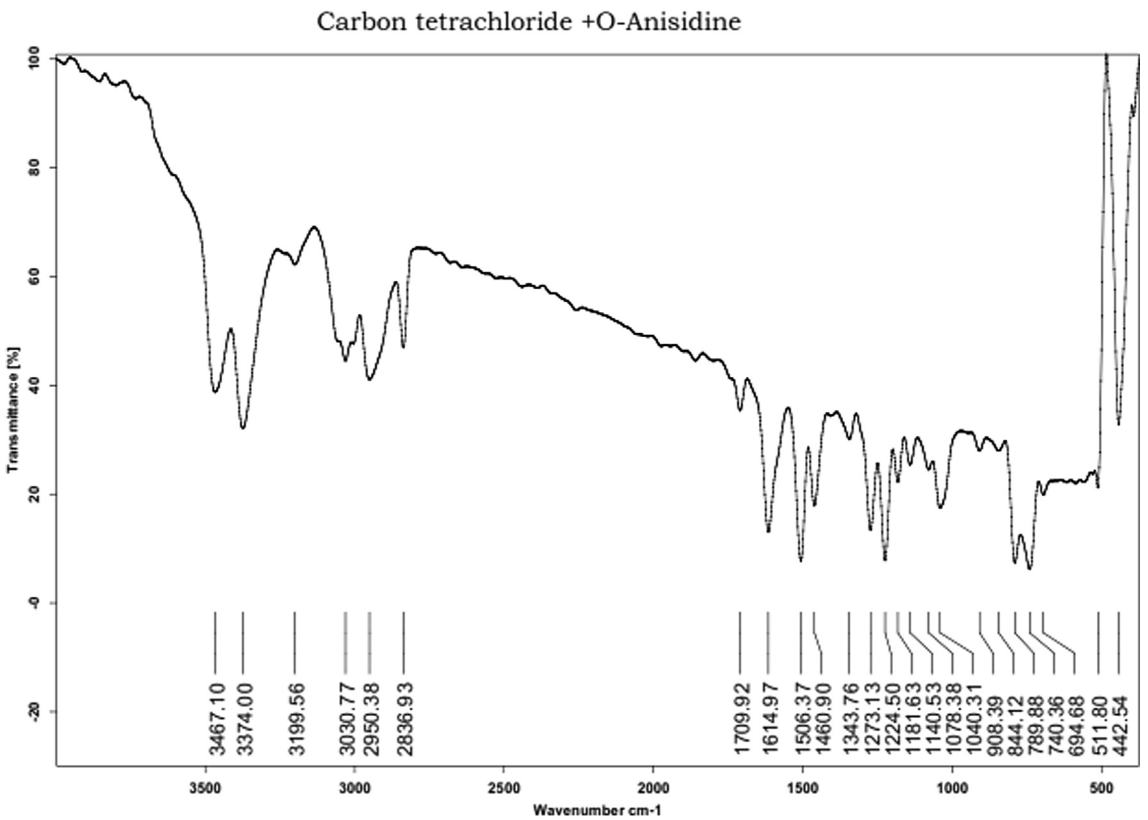

**Fig. 13.** Experimental Fourier Transform Infrared spectra of binary liquid mixtures (tetrachloromethane + 2-methoxy aniline) in the ratio 1:1.

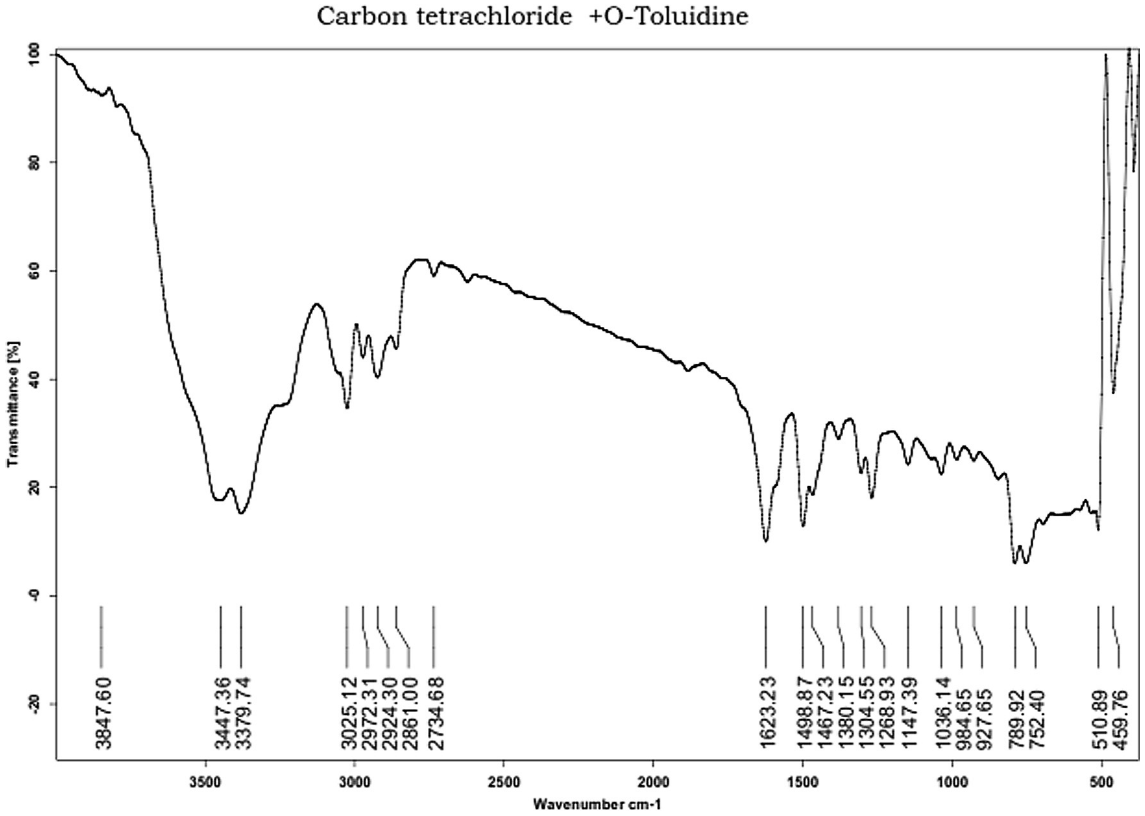

**Fig. 14.** Experimental Fourier Transform Infrared spectra of binary liquid mixtures (tetrachloromethane + 2-methyl aniline) in the ratio 1:1.

cules in the binary liquid mixtures. These spectra are shown in Figs. 12–14.

The N-H bending vibrations of aromatic amines are observed as strong bands in the region 1650–1580 cm$^{-1}$. FT-IR(Figs. 9-11) absorption bands of pure compounds are 2-chloroaniline, 2-methylaniline, 2-methoxyaniline exhibited bands at 1620.92 cm$^{-1}$, 1623.71 cm$^{-1}$, and 1615.38 cm$^{-1}$(N-H bending vibrations), respectively. From Figs. 12- 14,it is clear that the values of N-H bending vibrations frequency for the equimolar mixtures of tetrachloromethane + 2-chloroaniline (1618.47 cm$^{-1}$), Tetra chloromethane + 2-methylaniline (1623.23 cm$^{-1}$) and tetrachloromethane + 2-methoxyaniline (1614.97 cm$^{-1}$) are observed.Their corresponding $\Delta\nu$ values of $\nu$(N-H) absorption bands were −2.45, −0.48, and −0.41 cm$^{-1}$respectively.The frequency shifts are caused by the strong intermolecular interaction like hydrogen bonding between unlike molecules in the binary liquid mixtures,

The dilute solution of aromatic amines givesonesharp band due to N-H wagging vibrations between 910 and 662 cm$^{-1}$. FT-IR (Figs. 9-11)absorption bands of pure compounds are 2-chloroaniline, 2-methylaniline, 2-methoxyaniline exhibited bands at 746.67 cm$^{-1}$, 751.43 cm$^{-1}$, and 744.88 cm$^{-1}$ (N-H wagging vibrations), respectively. From Figs. 12-14, it is clear that the values of N-H wagging vibrations frequency for the equimolar mixtures of tetrachloromethane + 2-chloroaniline (744.83 cm$^{-1}$), tetrachloromethane + 2-methylaniline (752.40 cm$^{-1}$) and tetrachloromethane + 2-methoxyaniline (740.36 cm$^{-1}$) are observed.Their corresponding $\Delta\nu$ values of $\nu$ (N-H wagging vibrations) absorption bands were −1.84, 0.97, and −4.52 cm$^{-1}$ respectively.The frequency shifts are caused by the strong intermolecular interaction like hydrogen bonding between unlike molecules in the binary liquid mixtures

The C-N stretching vibration of aromatic amines is observed as strong bands in the region 1335–1250 cm$^{-1}$.FT-IR (Figs. 9-11) absorption bands of pure compounds are 2-chloroaniline, 2-methylaniline, 2-methoxyaniline exhibited bands at 1310.34 cm$^{-1}$, 1269.77 cm$^{-1}$, and 1272.36 cm$^{-1}$(C-N stretching vibrations), respectively. From Figs. 12-14,it is clear that the values of C-N stretching vibrations frequency for the equimolar mixtures of tetrachloromethane + 2-chloroaniline (1308.45 cm$^{-1}$), tetrachloromethane + 2-methylaniline (1268.93 cm$^{-1}$) and tetrachloromethane + 2-methoxyaniline (1273.13 cm$^{-1}$) are observed and their corresponding $\Delta\nu$ values of $\nu$(C-N) absorption bands were −1.89, −0.84 and 0.77 cm$^{-1}$respectively.The frequency shifts are caused by the strong intermolecular interaction like hydrogen bonding between unlike molecules in the binary liquid mixtures.

## 4. Conclusions

Densities, speeds of sound and viscosities of binary mixtures of tetrachloromethane with o-substituted aniline (2-chloroaniline, 2-methylaniline, and 2-methoxyaniline) have been measured at different temperatures and derived parameters along with their excess values calculated. The results are analyzed in terms of the specific interactions through the hetero molecular association between the components of the mixtures, resulting in the formation of complexes. FT-IR studies have been carried out and analyzed to study the presence of specific interaction such as the formation of hydrogen bonding of the type N-H·····Cl) between unlike molecules in the binary liquid mixtures.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.molliq.2021.117550.

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