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Study of Intermolecular Interactions in Binary Mixtures of 2-Methoxyaniline with Chlorinated Ethanes at Various Temperatures

B. Mukesh[1] · **T. Sreekanth**[2] · **M. Gowrisankar**[3] · **M. Raveendra**[4]

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Abstract

Densities (ρ), speeds of sound (u), and viscosities (η) are reported for binary mixtures of 2-methoxyaniline with chlorinated ethane components (1,2-dichloroethane, 1,1,2-trichloroethane and 1,1,2,2-tetrachloroethane) over the entire composition range of mole fraction at $T = (303.15–313.15)$ K and at atmospheric pressure (0.1 MPa). The excess properties: excess molar volume, excess isentropic compressibility and deviation in viscosity are calculated from the experimental density, speed of sound and viscosity. Excess properties are correlated using the Redlich–Kister polynomial equation. The excess partial molar volumes and excess partial molar isentropic compressibilities are calculated for all the binary systems throughout the composition range and at infinite dilutions. The results are analyzed in terms of electron donor–acceptor interactions leading to the formation of intermolecular complexes and hydrogen bonding between 2-methoxyaniline and chlorinated ethane molecules. The V^E results are analyzed in light of the Prigogine–Flory–Patterson theory. Analysis of each of the three contributions viz. interactional, free volume and P^* to V^E has shown that the interactional contribution are positive for all studied systems, the free volume and P^* contributions are negative for all the binary mixtures.

Keywords Density · Speed of sound · Viscosity · 2-Methoxyaniline · Chlorinated ethane molecules · PFP theory

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✉ T. Sreekanth
bmkchandra48@gmail.com

[1] Department of Physics, Vignan Institute of Technology and Science, Deshmukhi, Telangana 508284, India

[2] Department of Physics, J.N.T.U.H-College of Engineering, Sulthanpur, Medak, Telangana 502273, India

[3] Department of Chemistry, J.K.C.C. Acharya Nagarjuna University, Guntur, AP 522006, India

[4] Department of Chemistry, Rajiv Gandhi University of Knowledge Technologies, Dr. APJ Abdul Kalam, IIIT, Ongole, AP 523001, India

1 Introduction

Thermodynamic and transport properties of liquid mixtures provide important information for better understanding of the molecular liquid structure and intermolecular interactions of the liquid mixtures. Understanding molecular interactions are essential in diverse fields of protein folding, drug design, sensors, and nanotechnology [1–3].

2-Methoxyaniline is used in the manufacture of dyes for tattooing and coloration of paper. 1,2-dichloroethane is used in the production of polyvinyl chloride (PVC); it is also used as a solvent that is added to leaded gasoline to replace lead whereas 1,1,2-trichloroethane and 1,1,2,2-tetrachloroethane are non flammable solvents, used in fats, waxes, resins, oils, rubber, paints, varnishes, natural resins, and alkaloids.

This work is a part of our program to provide data for the molecular interactions between solvents in binary systems [4, 5]. The aim of the present work is to study the effect of hydrogen bonding intensity and number of chlorine atoms in the binary blends of 2-methoxyaniline with chlorinated ethane molecules on the excess and deviation in viscosity function values. Several researchers investigated density and viscosity of binary mixtures of acetophenone with chloroalkanes and chloroalkenes at 303.15 K [6] dibutyl ether with 1,2-dichloroethane, +1,2-dichloroethane, +1,2-dichloroethane [7]. Excess enthalpies of oxolane and 1,4-dioxane with chloroethanes and chloroethenes [8], pentan-3-one with 1,2-dichloroethane,1,3-dichloropropane, 1,4-dichlorobutane, trichloromethane,1,1,1-trichloroethane, 1,1,2,2-tetrachloroethane binary mixtures at 298.15 K [9]. There is no information accessible indicating itemized physical properties such as density, speed of sound, and viscosity for the binary mixtures of 2-methoxyaniline with chlorinated ethane components. Hence, this research work was undertaken for these binary blends.

In this paper we report densities (ρ), speeds of sound (u) and viscosities (η) of the binary mixtures of 2-methoxyaniline with chlorinated ethane components (including pure liquids) at different temperatures over the entire composition range. By using these data, the excess molar volume (V^{E}), excess isentropic compressibility (κ_S^{E}), deviation in viscosity ($\Delta\eta$), excess Gibbs energy of activation of viscous flow ($G^{*\mathrm{E}}$), excess partial molar volumes ($\overline{V}^{\circ\mathrm{E}}_{\mathrm{m},1}$, $\overline{V}^{\circ\mathrm{E}}_{\mathrm{m},2}$), excess partial isentropic compressibility ($\bar{K}^{\circ\mathrm{E}}_{\mathrm{s,m},1}$, $\bar{K}^{\circ\mathrm{E}}_{\mathrm{s,m},2}$) have been calculated.

2 Experimental

2.1 Chemicals

Chemicals used in the present study were 2-methoxyaniline (Sigma Aldrich), chlorinated ethane (1,2-dichloroethane, 1,1,2-trichloroethane and 1,1,2,2-tetrachloroethane) chemicals were purchased from S.D. Fine Chemicals Ltd. Prior to experimental measurements, liquids were purified as described in the literature [10–12]. The details of the chemicals and purification methods are presented in Table 1.

2.2 Apparatus and Procedure

All the binary liquid mixtures were prepared by weighing appropriate amounts of pure liquids on an Afcoset-ER-120A electric balance using a syringe in a narrow mouth

Table 1 List of chemicals with details of source, CAS number, purity and water content

Name of the chemical	Source	CAS number	Purification method	Purity in mole fraction (after purification)[a]	Water content %[b]	Analysis method[a]
2-Methoxyaniline	Sigma Aldrich India	90-04-0	No purification	0.995	0.004	GC
1,2-Dichloroethane	S.D. Fine Chemicals, India	107-06-2	Distillation	0.995	0.017	GC
1,1,2-Trichloroethane	S.D. Fine Chemicals, India	71-55-06	Distillation	0.995	0.008	GC
1,1,2,2-Tetrachloroethane	S.D. Fine Chemicals, India	79-34-5	Distillation	0.995	0.007	GC

[a] Gas chromatography

[b] Karl–Fischer method

stoppered bottle. The uncertainty of the electronic balance was ± 0.05 mg while the accuracy of the mole fraction is $\pm 1 \times 10^{-4}$.

Densities and speeds of sound of the pure liquids as well as their binary mixtures were measured using an automatic digital vibrating tube density and speed of sound analyzer (Anton Parr, DSA 5000). The instrument was first calibrated by comparing the density and speed of sound of distilled water and dry air with the literature. Dynamic viscosities are measured using an Anton Parr micro-viscometer (Lovis 2000 ME) mounted on the master instrument DSA 5000 M. It can measure viscosity from 0.3 mPa·s to 10,000 mPa·s by the rolling ball technique. The temperature was controlled by a built-in precise Peltier thermostat with an accuracy of 0.02 K. Before measurement, calibration was carried out using standard oils (S3, N26, N100 liquids for 1.59 mm, 1.8 mm and 2.5 mm capillaries, respectively) provided by Anton Paar Co., Austria. The standard uncertainties of temperature, density, speed of sound and viscosity are estimated to be 0.002 K, $\pm 5 \times 10^{-5}$ g·cm^{-3}, ± 0.05 m·s^{-1} and ± 1.5%, respectively.

The purity of all these solvents were confirmed by comparison of the measured densities, speeds of sound and viscosities of the pure liquids with literature values [4, 5, 13–20] and these are listed in the Table 2.

3 Results and Discussion

The experimental densities and viscosities for all the binary systems at various compositions are used to calculate the excess thermodynamic functions using following equations:

$$V^{\mathrm{E}} = \frac{x_1 M_1 + x_2 M_2}{\rho} - \left(\frac{x_1 M_1}{\rho_1} + \frac{x_2 M_2}{\rho_2} \right) \tag{1}$$

$$\Delta\eta = \eta - \left(x_1 \eta_1 + x_2 \eta_2 \right) \tag{2}$$

$$G^{*\mathrm{E}} = RT\left(\ln \eta V - \left(x_1 \ln \eta_1 V_1 + x_2 \ln \eta_2 V_2 \right) \right) \tag{3}$$

where ρ, η, V and $G^{*\mathrm{E}}$ are density, viscosity molar volume and excess Gibbs energy of activation of viscous flow of the binary mixture. x_1, M_1, ρ_1, η_1, V_1 and x_2, M_2, ρ_2, η_2 and V_2 are the mole fraction, molar mass, density, viscosity and molar volume of pure components 1 and 2, respectively. R is the gas constant and T is the absolute temperature.

The experimental data are used to compute isentropic compressibility (κ_S) by using the following relation:

$$\kappa_S = \left(u^2 \rho \right)^{-1} \tag{4}$$

The method used for calculating κ_s^{E} (Benson–Kiyohara approach) was outlined previously [21].

V^{E}, κ_S^{E} and $\Delta\eta$ values were fitted with a Redlich–Kister [22] polynomial equation,

$$Y^{\mathrm{E}} = x_1 x_2 \sum_{i=0}^{j} A_i \left(1 - 2x_1 \right)^i \tag{5}$$

where Y^{E} is V^{E}, κ_S^{E}, or $\Delta\eta$. Values of the coefficients A_i have been determined by using the method of least-squares. The densities, speeds of sound and viscosities for mixtures of

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

Component	Density $(\rho)\times 10^3$/kg·m^{-3}		Speed of sound (u/m·s^{-1})		Viscosity (mPa·s)	
	Experimental	Literature	Experimental	Literature	Experimental	Literature
2-Methoxyaniline						
303.15 K	1.09175 [4, 5]	1.0917 [13]	1597.4 [4, 5]		4.923 [4, 5]	4.9236 [13, 14]
308.15 K	1.08799 [4, 5]		1583.2 [4, 5]		4.571 [4, 5]	
313.15 K	1.08432 [4, 5]		1569.9 [4, 5]		4.217 [4, 5]	
1,2-Dichloroethane						
303.15 K	1.23860	1.23879 [15] 1.2386 [16] 1.2387 [17] 1.23802 [19]	1175.2	1175.0 [15] 1176.0 [18] 1174.0 [17] 1175.0 [16] 1174.0 [19]	0.724	0.7237 [17] 0.7236 [16]
308.15 K	1.23092	1.2305 [17] 1.2305 [16]	1165.3	1164.8 [17] 1165.7 [16]	0.682	0.6737 [17] 0.6736 [16]
313.15 K	1.22342	1.2237 [17] 1.2236 [16] 1.22342 [19]	1155.2	1153.5 [17] 1155.0 [16] 1139.0 [19]	0.642	0.6419 [17] 0.6420 [16]
1,1,2-Trichloroethane						
303.15 K	1.42453	1.4250 [17] 1.4249 [16]	1129.5	1129.2 [17] 1130.0 [16]	1.019	1.0192 [17, 20] 1.0193 [16]
308.15 K	1.41683	1.4161 [17] 1.4160 [16]	1116.8	1115.9[17 1116.0 [16]	0.953	0.9483[17 0.9483 [16]
313.15 K	1.40916	1.4096 [17] 1.4097 [16]	1104.3	1096.1 [17] 1097.0 [16]	0.887	0.8888 [17] 0.8888 [16]
1,1,2,2-Tetrachloroethane						
303.15 K	1.57931	1.57963 [15] 1.5780 [16] 1.5796 [17] 1.57860 [19]	1135.2	1128.0 [15] 1132.0 [18] 1134.7 [17] 1135.0 [16] 1137.0 [19]	1.456	1.4562 [17] 1.4562 [16]
308.15 K	1.57118	1.5706 [17] 1.5706 [16]	1115.2	1115.5 [17] 1115.7 [16]	1.359	1.3544 [17] 1.3543 [16]
313.15 K	1.56333	1.5637 [17] 1.5638 [16] 1.56317 [19]	1095.3	1095.5 [17] 1095.3 [16] 1104.0 [19]	1.264	1.2613 [17] 1.2613 [16]

various mole fractions of 2-methoxyaniline in chlorinated ethane molecules are presented in Tables 3, 4 along with excess molar volumes, excess isentropic compressibilities, viscosity deviations and excess Gibbs energies of activation of viscous flow at various temperatures. Figures 1, 2, 3, 4 show the behaviors of V^{E}, κ_S^{E}, $\Delta\eta$ and $G^{*\mathrm{E}}$ as a function of the molar fraction of 2-methoxyaniline, respectively.

The standard deviations $\sigma(Y^{\mathrm{E}})$ have been calculated by using the formula

Table 3 Density (ρ), excess molar volumes (V^E), speed of sound (u), excess isentropic compressibility (κ_S^E), viscosity (η), deviation in viscosity ($\Delta\eta$) and excess Gibbs energy of activation of viscous flow (G^{*E}) of binary liquid mixtures of 2-methoxyaniline with chlorinated ethane components at $T=(303.15$ to $318.15)$ K and 0.1 MPa pressure

x_1	Density ρ/g·cm^{-3}			V^E/cm^3·mol^{-1}		
	303.15 K	308.15 K	313.15 K	303.15 K	308.15 K	313.15 K
2-Methoxyaniline (1) + 1,2-dichloroethane (2)						
0.0000	1.23860	1.23092	1.22342	0.0000	0.0000	0.0000
0.0875	1.22132	1.21421	1.20726	−0.0173	−0.0199	−0.0240
0.1658	1.20683	1.20018	1.19362	−0.0317	−0.0366	−0.0395
0.2536	1.19152	1.18532	1.17918	−0.0438	−0.0485	−0.0520
0.3656	1.17333	1.16764	1.16199	−0.0549	−0.0587	−0.0625
0.4856	1.15529	1.15010	1.14491	−0.0581	−0.0620	−0.0657
0.5902	1.14066	1.13586	1.13105	−0.0544	−0.0583	−0.0623
0.6816	1.12863	1.12415	1.11965	−0.0462	−0.0508	−0.0554
0.7704	1.11757	1.11338	1.10913	−0.0355	−0.0403	−0.0432
0.8841	1.10426	1.10039	1.09645	−0.0195	−0.0225	−0.0257
1.0000	1.09157	1.08799	1.08432	0.0000	0.0000	0.0000
2-Methoxyaniline (1) + 1,1,2-trichloroethane (2)						
0.0000	1.42423	1.41665	1.40932	0.0000	0.0000	0.0000
0.0884	1.39002	1.38315	1.37641	−0.0399	−0.0562	−0.0673
0.1905	1.35204	1.34572	1.33946	−0.0923	−0.1115	−0.1246
0.2923	1.31544	1.30963	1.30382	−0.1305	−0.1536	−0.1688
0.4076	1.27552	1.27015	1.26481	−0.1629	−0.1823	−0.1994
0.5076	1.24201	1.23703	1.23206	−0.1703	−0.1896	−0.2074
0.5945	1.21373	1.20906	1.20441	−0.1634	−0.1822	−0.2024
0.7016	1.17988	1.17559	1.17129	−0.1362	−0.1560	−0.1768
0.8104	1.14663	1.14270	1.13869	−0.0913	−0.1115	−0.1292
0.9023	1.11949	1.11577	1.11198	−0.0478	−0.0620	−0.0758
1.0000	1.09157	1.08799	1.08432	0.0000	0.0000	0.0000
2-Methoxyaniline (1) + 1,1,2,2-tetrachloroethane (2)						
0.0000	1.57931	1.57118	1.56333	0.0000	0.0000	0.0000
0.0911	1.53342	1.52593	1.51866	−0.0714	−0.0811	−0.0896
0.1962	1.48086	1.47393	1.46719	−0.1409	−0.1509	−0.1616
0.2806	1.43888	1.43237	1.42601	−0.1815	−0.1905	−0.2000
0.3816	1.38898	1.38291	1.37697	−0.2148	−0.2205	−0.2280
0.4976	1.33204	1.32653	1.32108	−0.2256	−0.2330	−0.2395
0.6023	1.28109	1.27606	1.27104	−0.2156	−0.2245	−0.2323
0.7116	1.22835	1.22379	1.21922	−0.1847	−0.1950	−0.2056
0.8104	1.18110	1.17693	1.17273	−0.1384	−0.1488	−0.1609
0.9212	1.12857	1.12480	1.12095	−0.0635	−0.0724	−0.0811
1.0000	1.09157	1.08799	1.08432	0.0000	0.0000	0.0000
Speed of sound (u/m·s^{-1})				κ_S^E/TPa^{-1}		
2-Methoxyaniline (1) + 1,2-dichloroethane(2)						
0.0000	1175.2	1165.3	1155.2	0.000	0.000	0.000
0.0875	1217.8	1208.8	1200.3	−5.560	−7.111	−9.323

Table 3 (continued)

Speed of sound (u/m·s^{-1})				κ_S^E/TPa^{-1}		
0.1658	1255.8	1247.8	1239.0	−9.727	−12.53	−14.76
0.2536	1298.6	1290.6	1281.6	−13.81	−16.84	−19.26
0.3656	1353.9	1344.4	1334.9	−18.41	−20.72	−22.97
0.4856	1410.3	1399.8	1389.8	−20.50	−22.37	−24.48
0.5902	1456.2	1445.9	1435.3	−19.94	−22.09	−23.86
0.6816	1493.0	1482.8	1473.4	−17.62	−19.78	−22.27
0.7704	1525.0	1516.1	1507.6	−13.56	−16.40	−19.26
0.8841	1562.4	1553.9	1544.8	−7.223	−10.049	−12.385
1.0000	1597.4	1583.2	1569.9	0.000	0.000	0.000
2-Methoxyaniline (1) + 1,1,2-trichloroethane(2)						
0.0000	1129.5	1116.8	1104.3	0.000	0.000	0.000
0.0884	1171.5	1160.1	1148.8	−6.188	−7.942	−9.733
0.1905	1220.8	1210.2	1199.1	−11.85	−14.64	−16.93
0.2923	1271.6	1260.5	1249.4	−16.61	−19.27	−21.70
0.4076	1329.7	1317.7	1305.7	−20.22	−22.28	−24.22
0.5076	1379.3	1366.9	1354.9	−21.15	−22.99	−24.94
0.5945	1421.9	1409.5	1398.0	−20.65	−22.48	−24.67
0.7016	1471.6	1460.9	1450.2	−17.55	−20.20	−22.62
0.8104	1519.4	1509.8	1500.7	−12.38	−15.23	−18.14
0.9023	1558.6	1547.8	1539.3	−7.097	−9.049	−11.82
1.0000	1597.4	1583.2	1569.9	0.000	0.000	0.000
2-Methoxyaniline (1) + 1,1,2,2-tetrachloroethane(2)						
0.0000	1135.2	1115.2	1095.3	0.000	0.000	0.000
0.0911	1176.2	1158.8	1139.8	−7.243	−9.803	−11.13
0.1962	1223.7	1206.3	1188.9	−13.16	−15.68	−18.32
0.2806	1263.4	1246.1	1228.4	−17.16	−19.72	−22.00
0.3816	1311.3	1293.8	1276.3	−20.30	−22.39	−24.55
0.4976	1367.1	1349.7	1332.8	−21.84	−23.68	−25.71
0.6023	1417.5	1401.3	1385.5	−21.24	−23.29	−25.46
0.7116	1468.2	1454.7	1440.7	−17.89	−20.66	−23.06
0.8104	1513.8	1501.9	1489.7	−13.45	−16.27	−18.75
0.9212	1564.7	1552.0	1542.0	−6.991	−8.429	−10.92
1.0000	1597.4	1583.2	1569.9	0.000	0.000	0.000

Standard uncertainty u are: $u(x) = \pm 1\times10^{-4}$, $u(\rho) = \pm 5\times10^{-5}$ g·cm^{-3}, $u(u) = \pm 0.05$ m·s^{-1}

$$\sigma\left(Y^{\mathrm{E}}\right) = \left(\frac{\sum\left(Y^{\mathrm{E}}_{\mathrm{exp}} - Y^{\mathrm{E}}_{\mathrm{cal}}\right)^2}{m-n}\right)^{\frac{1}{2}} \tag{6}$$

Table 4 Viscosity (η), deviation in viscosity ($\Delta\eta$) and excess Gibbs energy of activation of viscous flow (G^{*E}) of binary liquid mixtures of 2-methoxyaniline with chlorinated ethane components at $T = (303.15$ to $318.15)$ K and 0.1 MPa pressure

x_1	Viscosity (η/mPa·s)			$\Delta\eta$/mPa·s		
	303.15 K	308.15 K	313.15 K	303.15 K	308.15 K	313.15 K
2-Methoxyaniline (1) + 1,2-dichloroethane(2)						
0.0000	0.724	0.682	0.642	0.000	0.000	0.000
0.0875	1.101	1.037	0.974	0.010	0.014	0.019
0.1658	1.438	1.353	1.270	0.017	0.026	0.035
0.2536	1.814	1.704	1.596	0.025	0.036	0.047
0.3656	2.291	2.148	2.004	0.032	0.044	0.055
0.4856	2.797	2.617	2.436	0.034	0.047	0.058
0.5902	3.235	3.022	2.808	0.033	0.045	0.056
0.6816	3.613	3.372	3.130	0.028	0.040	0.051
0.7704	3.980	3.711	3.439	0.022	0.033	0.043
0.8841	4.448	4.139	3.829	0.012	0.018	0.026
1.0000	4.923	4.571	4.217	0.000	0.000	0.000
2-Methoxyaniline (1) + 1,1,2-trichloroethane(2)						
0.0000	1.019	0.953	0.887	0.000	0.000	0.000
0.0884	1.376	1.291	1.204	0.012	0.018	0.023
0.1905	1.786	1.676	1.566	0.023	0.034	0.045
0.2923	2.192	2.055	1.916	0.032	0.045	0.056
0.4076	2.647	2.478	2.307	0.037	0.051	0.063
0.5076	3.038	2.841	2.641	0.038	0.051	0.064
0.5945	3.376	3.154	2.929	0.037	0.050	0.062
0.7016	3.789	3.535	3.279	0.031	0.044	0.056
0.8104	4.204	3.918	3.629	0.022	0.033	0.043
0.9023	4.554	4.236	3.918	0.012	0.019	0.027
1.0000	4.923	4.571	4.217	0.000	0.000	0.000
2-Methoxyaniline (1) + 1,1,2,2-tetrachloroethane(2)						
0.0000	1.456	1.359	1.264	0.000	0.000	0.000
0.0911	1.786	1.673	1.562	0.015	0.022	0.029
0.1962	2.165	2.029	1.895	0.029	0.040	0.051
0.2806	2.464	2.309	2.154	0.035	0.048	0.061
0.3816	2.820	2.639	2.458	0.041	0.054	0.067
0.4976	3.224	3.013	2.802	0.043	0.056	0.069
0.6023	3.584	3.347	3.111	0.040	0.054	0.068
0.7116	3.957	3.692	3.427	0.035	0.048	0.062
0.8104	4.291	4.001	3.709	0.026	0.039	0.052
0.9212	4.660	4.337	4.012	0.011	0.019	0.027
1.0000	4.923	4.571	4.217	0.000	0.000	0.000

G^{*E}/J·mol^{-1}

2-Methoxyaniline (1) + 1,2-dichloroethane(2)				2-Methoxyaniline (1) + 1,1,2-trichloroethane(2)				2-Methoxyaniline (1) + 1,1,2-trichloroethane(2)			
x_1	303.15 K	308.15 K	313.15 K	x_1	303.15 K	308.15 K	313.15 K	x_1	303.15 K	308.15 K	313.15 K
0.0000	0.000	0.000	0.000	0.0000	0.000	0.000	0.000	0.0000	0.000	0.000	0.000
0.0875	6.389	6.508	6.599	0.0884	4.041	4.186	4.339	0.0911	2.315	2.454	2.608
0.1658	9.374	9.563	9.719	0.1905	6.539	6.766	7.020	0.1962	3.896	4.088	4.296

Table 4 (continued)

G^{*E}/J·mol^{-1}

2-Methoxyaniline (1) + 1,2-dichloroethane(2)				2-Methoxyaniline (1) + 1,1,2-trichloroethane(2)				2-Methoxyaniline (1) + 1,1,2-trichloroethane(2)			
x_1	303.15 K	308.15 K	313.15 K	x_1	303.15 K	308.15 K	313.15 K	x_1	303.15 K	308.15 K	313.15 K
0.2536	11.04	11.25	11.41	0.2923	7.661	7.899	8.14	0.2806	4.553	4.759	4.97
0.3656	11.57	11.77	11.91	0.4076	7.851	8.070	8.29	0.3816	4.842	5.038	5.23
0.4856	10.82	11.00	11.14	0.5076	7.360	7.558	7.763	0.4976	4.657	4.832	5.011
0.5902	9.435	9.590	9.712	0.5945	6.575	6.755	6.933	0.6023	4.122	4.280	4.451
0.6816	7.791	7.925	8.036	0.7016	5.238	5.386	5.540	0.7116	3.279	3.413	3.563
0.7704	5.902	6.017	6.104	0.8104	3.549	3.657	3.769	0.8104	2.306	2.419	2.540
0.8841	3.140	3.198	3.258	0.9023	1.916	1.974	2.046	0.9212	1.016	1.074	1.142
1.0000	0.000	0.000	0.000	1.0000	0.000	0.000	0.000	1.0000	0.000	0.000	0.000

Standard uncertainty u are: $u\ (\eta) = 1.5\%$, $u\ (T) = 0.01$ K for density and speed of sound, $u\ (T) = 0.02$ K, for viscosity and $u(p) = 1$ kPa

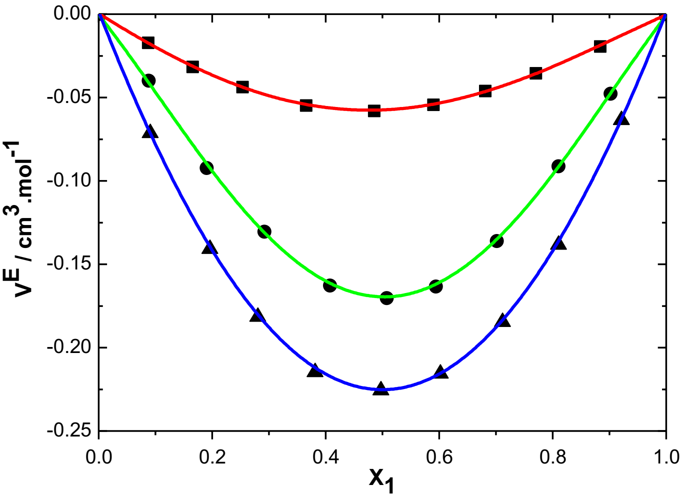

Fig. 1 Variation of excess molar volume (V^E) with mole fraction (x_1) of 2-methoxyaniline for the binary liquid mixtures of 2-methoxyaniline with 1,1,2,2-tetrachloroethane (▲);1,1,2-trichloroethane (●) and 1,2-dichloroethane (■) at 303.15 K

where m is the total number of experimental points and n is the number of parameters. The coefficients, A_i and corresponding standard deviation values (σ) are presented in Table 5.

The excess parameters of liquid mixtures depend on

i. Donor–acceptor complex formation
ii. Complex formation through new hydrogen bonds between C–Cl·····H–N
iii. The number of chlorine atoms

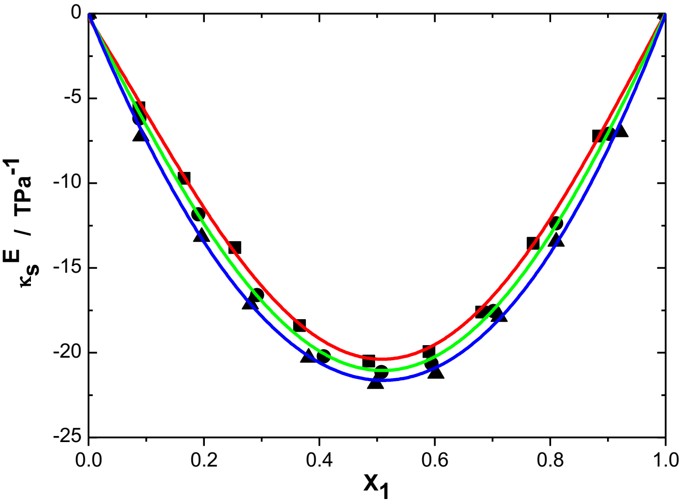

Fig. 2 Excess isentropic compressibility (κ_S^E) with mole fraction (x_1) of 2-methoxyanilinein the binary liquid mixtures of 2-methoxyaniline with 1,1,2,2-tetrachloroethane (▲);1,1,2-trichloroethane (●) and 1,2-dichloroethane (■) at 303.15 K

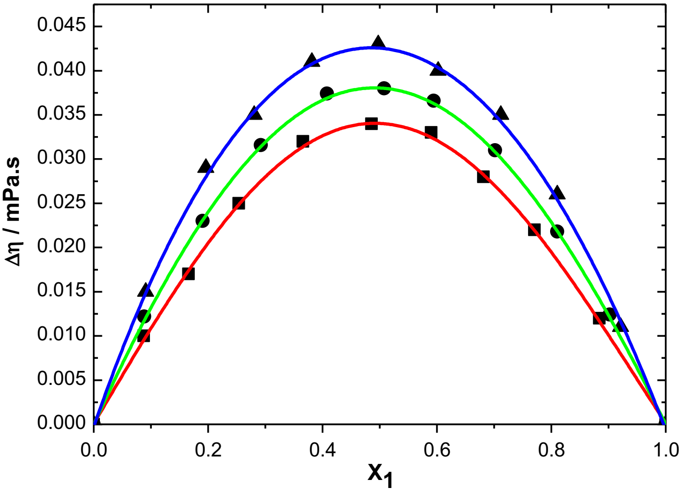

Fig. 3 Variation of deviation in viscosity (Δη) with mole fraction (x_1) of 2-methoxyanilinein the binary liquid mixtures of 2-methoxyaniline with 1,1,2,2-tetrachloroethane (▲); 1,1,2-trichloroethane (●) and 1,2-dichloroethane (■) at 303.15 K

The negative values of excess molar volume, along with negative of excess isentropic compressibility and positive values of deviation in viscosity and excess Gibbs energy of activation of viscous flow for all binary systems indicate that specific interactions are dominant over non-specific interactions between the component molecules in liquid mixtures.

According to these investigations, negative values of excess molar volume and excess isentropic compressibility of 2-methoxyaniline with chlorinated ethane molecules indicate

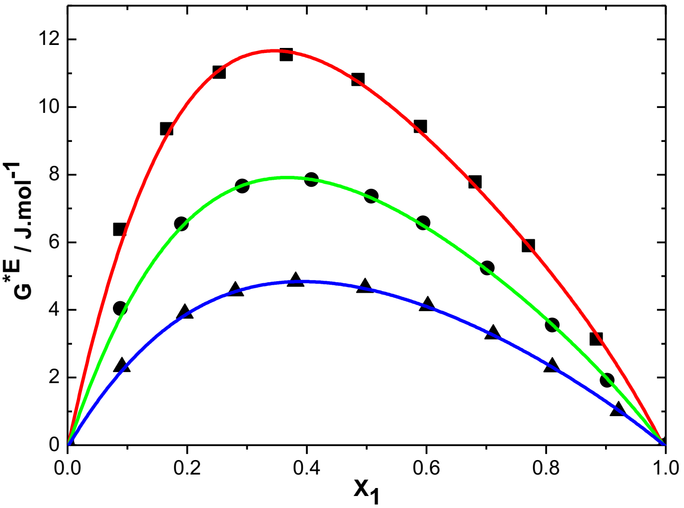

Fig. 4 Excess Gibbs energy (G^{*E}) of activation of viscous flow with mole fraction (x_1) of 2-methoxyanilinein the binary liquid mixtures of 2-methoxyaniline with 1,1,2,2-tetrachloroethane (▲); 1,1,2-trichloroethane (●) and 1,2-dichloroethane (■) at 303.15 K

that the formation of transition complex with new hydrogen bonds between component molecules in liquid mixtures (C–Cl·····H–N) [23].

The κ_S^E values become more negative with increase in the temperature of the binary systems of 2-methoxyaniline with chlorinated alkane, because of excessive thermal dissociation of self-association than cross-association in liquid mixtures and more interstitial accommodation of one component into another [24]. Finally, concluded that negative values of excess isentropic compressibility (κ_S^E) over the entire range of composition of 2-methoxyaniline with chlorinated alkanes confirm that strong intermolecular hydrogen bonding interaction between unlike molecules of 2-methoxyaniline and chlorinated alkane dominate over the structural breaking effect of chlorinated alkane on addition of 2-methoxyaniline (Table 6).

The magnitude of excess functions follows the sequence:

1,2-dichloroethane > 1,1,2-trichloroethane > 1,1,2,2-tetrachloroethane

This order suggests that the increase of strength of interaction between unlike molecules as the number of chlorine atoms increases from 1,2-dichloroethane to 1,1,2,2-tetrachloroethane results in a decrease of excess molar volume, and excess isentropic compressibility. Hence, here above order is observed.

Generally, positive values of deviation viscosity ($\Delta\eta$) indicate the predominance of hydrogen bonding interaction between unlike molecules over the dissociation effects of the mixing components [25].

The data presented in Figs. 3, 4 reveal that $\Delta\eta$ and G^{*E} are positive for all the binary systems over the entire composition range, which indicates that the hydrogen bonding interaction between the nitrogen atom of 2-methoxyaniline and acidic hydrogen atom of chlorinated ethane molecules [25, 26].

Table 5 Coefficients of Redlich–Kister equation and standard deviation (σ) values for liquid mixtures of 2-methoxyaniline with chlorinated ethane components at $T = (303.15–318.15)$ K

Binary mixtures	Functions	a_0	a_1	a_2	σ
303.15 K					
2-Methoxyaniline + 1,2-dichloroethane	V^E/m^3·mol^{-1}	− 0.231	0.024	0.045	0.001
	$\Delta\eta$/mPa•s	0.136	− 0.005	− 0.027	0.001
	κ_S^E/TPa^{-1}	− 81.54	− 1.841	19.26	0.328
2-Methoxyaniline + 1,1,2-trichloroethane	V^E/m^3·mol^{-1}	− 0.684	− 0.015	0.245	0.001
	$\Delta\eta$/mPa•s	0.153	− 0.006	− 0.012	0.001
	κ_S^E/TPa^{-1}	− 84.16	− 2.748	9.842	0.233
2-Methoxyaniline + 1,1,2,2-tetra chloroethane	V^E/m^3·mol^{-1}	− 0.909	− 0.006	0.052	0.001
	$\Delta\eta$/mPa•s	0.173	− 0.014	− 0.005	0.001
	κ_S^E/TPa^{-1}	− 85.86	− 4.268	− 6.286	0.385
308.15 K					
2-Methoxyaniline + 1,2-dichloroethane	V^E/m^3·mol^{-1}	− 0.248	0.025	0.018	0.001
	$\Delta\eta$/mPa•s	0.189	− 0.003	− 0.019	0.001
	κ_S^E/TPa^{-1}	− 89.68	− 4.564	− 6.282	0.172
2-Methoxyaniline + 1,1,2-trichloroethane	V^E/m^3·mol^{-1}	− 0.758	− 0.002	0.088	0.001
	$\Delta\eta$/mPa•s	0.208	− 0.005	0.015	0.001
	κ_S^E/TPa^{-1}	− 92.34	− 3.134	− 12.56	0.087
2-Methoxyaniline + 1,1,2,2-tetra chloroethane	V^E/m^3·mol^{-1}	− 0.932	− 0.009	− 0.082	0.001
	$\Delta\eta$/mPa•s	0.226	− 0.003	0.059	0.001
	κ_S^E/TPa^{-1}	− 93.55	− 1.054	− 31.91	0.468
313.15 K					
2-Methoxyaniline + 1,2-dichloroethane	V^E/m^3·mol^{-1}	− 0.261	0.029	− 0.019	0.001
	$\Delta\eta$/mPa•s	0.236	0.002	0.023	0.001
	κ_S^E/TPa^{-1}	− 96.95	− 5.015	− 33.26	0.209
2-Methoxyaniline + 1,1,2-trichloroethane	V^E/m^3·mol^{-1}	− 0.828	− 0.022	− 0.021	0.001
	$\Delta\eta$/mPa•s	0.259	0.002	0.057	0.001
	κ_S^E/TPa^{-1}	− 99.47	− 7.502	− 41.18	0.231
2-Methoxyaniline + 1,1,2,2-tetra chloroethane	V^E/m^3·mol^{-1}	− 0.958	− 0.159	− 0.205	0.001
	$\Delta\eta$/mPa•s	0.279	0.009	0.123	0.001
	κ_S^E/TPa^{-1}	− 101.01	− 6.467	− 57.09	0.468

3.1 Partial Molar Properties

The interpretations of excess partial molar properties ($\overline{V}^{E}_{m,1}$, $\overline{V}^{E}_{m,2}$, $\overline{K}^{E}_{s,m,1}$ and $\overline{K}^{E}_{s,m,2}$) and excess partial molar properties at infinite dilution($\overline{V}^{\circ E}_{m,1}$, $\overline{V}^{\circ E}_{m,2}$ $\overline{K}^{\circ E}_{s,m,1}$ and $\overline{K}^{\circ E}_{s,m,2}$) of components 2 have previously been described [27].

A close perusal of Table 6 indicates that the values of $\overline{V}^{E}_{m,1}$, $\overline{V}^{E}_{m,2}$ $\overline{K}^{E}_{s,m,1}$ and $\overline{K}^{E}_{s,m,2}$ are negative for all the binary mixtures over the whole composition range. Negative values may be ascribed due to complex formation through dipole–dipole interactions between component molecules.

From the Table 7, it can be seen that the values of $\overline{V}^{\circ E}_{m,1}$, $\overline{V}^{\circ E}_{m,2}$ $\overline{K}^{\circ E}_{s,m,1}$ and $\overline{K}^{\circ E}_{s,m,2}$ are negative for all the binary mixtures over the whole composition range. The negative values

Table 6 The values $\bar{V}^{E}_{m,1}$,$\bar{V}^{E}_{m,2}$,$\bar{K}^{\mathrm{E}}_{\mathrm{s,m,1}}$,$\bar{K}^{\mathrm{E}}_{\mathrm{s,m,2}}$ of liquid mixtures of 2-methoxyaniline with chlorinated ethane components at T = (303.15–313.15) K

x_1	303.15 K		308.15 K		313.15 K		303.15 K		308.15 K		313.15 K	
	$\bar{V}^{\mathrm{E}}_{m,1}$	$\bar{V}^{\mathrm{E}}_{m,2}$	$\bar{V}^{\mathrm{E}}_{m,1}$	$\bar{V}^{\mathrm{E}}_{m,2}$	$\bar{V}^{\mathrm{E}}_{m,1}$	$\bar{V}^{\mathrm{E}}_{m,2}$	$\bar{K}^{\mathrm{E}}_{\mathrm{s,m,1}}$	$\bar{K}^{\mathrm{E}}_{\mathrm{s,m,2}}$	$\bar{K}^{\mathrm{E}}_{\mathrm{s,m,1}}$	$\bar{K}^{\mathrm{E}}_{\mathrm{s,m,2}}$	$\bar{K}^{\mathrm{E}}_{\mathrm{s,m,1}}$	$\bar{K}^{\mathrm{E}}_{\mathrm{s,m,2}}$
	$\mathrm{cm^3{\cdot}mol^{-1}}$						$\mathrm{TPa^{-1}}$					
2-Methoxyaniline (1) + 1,2-dichloroethane(2)												
0.0000	−0.162	0.000	−0.206	0.000	−0.256	0.000	−74.72	0.000	−114.5	0.000	−151.9	0.000
0.0875	−0.159	0.000	−0.185	−0.001	−0.213	−0.002	−67.74	−0.358	−86.20	−1.262	−114.4	−2.129
0.1658	−0.148	−0.002	−0.164	−0.004	−0.180	−0.007	−58.73	−1.687	−65.98	−4.172	−87.75	−6.529
0.2536	−0.129	−0.007	−0.139	−0.011	−0.147	−0.015	−47.11	−4.795	−48.07	−8.895	−64.28	−12.72
0.3656	−0.099	−0.021	−0.105	−0.026	−0.110	−0.031	−32.12	−11.55	−31.10	−16.45	−42.13	−20.84
0.4856	−0.067	−0.048	−0.072	−0.052	−0.076	−0.054	−18.12	−21.91	−18.51	−25.70	−25.64	−28.80
0.5902	−0.042	−0.079	−0.047	−0.082	−0.051	−0.082	−8.971	−32.52	−11.00	−34.39	−15.65	−35.53
0.6816	−0.025	−0.111	−0.028	−0.114	−0.032	−0.114	−3.675	−41.70	−6.404	−42.40	−9.361	−42.50
0.7704	−0.012	−0.143	−0.015	−0.150	−0.018	−0.154	−0.844	−49.08	−3.280	−50.66	−4.935	−52.00
0.8841	−0.003	−0.182	−0.004	−0.200	−0.005	−0.219	0.202	−53.56	−0.846	−62.32	−1.320	−71.75
1.0000	0.000	−0.210	0.000	−0.253	0.000	−0.308	0.000	−48.16	0.000	−76.21	0.000	−106.6
2-Methoxyaniline (1) + 1,1,2-trichloroethane (2)												
0.0000	−0.454	0.000	−0.686	0.000	−0.864	0.000	−88.96	0.000	−124.2	0.000	−169.8	0.000
0.0884	−0.468	0.001	−0.601	−0.004	−0.704	−0.008	−76.54	−0.596	−93.02	−1.398	−126.6	−2.484
0.1905	−0.427	−0.005	−0.494	−0.020	−0.545	−0.034	−61.07	−3.129	−66.09	−5.702	−88.92	−9.202
0.2923	−0.350	−0.033	−0.385	−0.056	−0.410	−0.077	−45.87	−7.984	−46.61	−11.85	−61.45	−17.12
0.4076	−0.247	−0.098	−0.270	−0.120	−0.284	−0.144	−30.35	−16.34	−30.94	−20.21	−39.44	−25.48
0.5076	−0.161	−0.182	−0.183	−0.197	−0.195	−0.219	−19.22	−25.72	−21.17	−28.43	−25.99	−31.94
0.5945	−0.099	−0.269	−0.120	−0.276	−0.132	−0.295	−11.66	−34.99	−14.66	−36.41	−17.36	−37.81
0.7016	−0.043	−0.377	−0.061	−0.385	−0.072	−0.406	−5.152	−46.88	−8.427	−47.90	−9.548	−47.76
0.8104	−0.011	−0.461	−0.022	−0.497	−0.029	−0.539	−1.494	−58.05	−3.736	−62.51	−4.063	−65.21
0.9023	−0.001	−0.483	−0.005	−0.584	−0.008	−0.668	−0.242	−65.29	−1.095	−78.39	−1.157	−90.32
1.0000	0.000	−0.424	0.000	−0.656	0.000	−0.826	0.000	−69.07	0.000	−100.4	0.000	−133.1

Table 6 (continued)

x_1	303.15 K		308.15 K		313.15 K		303.15 K		308.15 K		313.15 K	
	$\bar{V}^{\mathrm{E}}_{m,1}$	$\bar{V}^{\mathrm{E}}_{m,2}$	$\bar{V}^{\mathrm{E}}_{m,1}$	$\bar{V}^{\mathrm{E}}_{m,2}$	$\bar{V}^{\mathrm{E}}_{m,1}$	$\bar{V}^{\mathrm{E}}_{m,2}$	$\bar{K}^{\mathrm{E}}_{\mathrm{s,m,1}}$	$\bar{K}^{\mathrm{E}}_{\mathrm{s,m,2}}$	$\bar{K}^{\mathrm{E}}_{\mathrm{s,m,1}}$	$\bar{K}^{\mathrm{E}}_{\mathrm{s,m,2}}$	$\bar{K}^{\mathrm{E}}_{\mathrm{s,m,1}}$	$\bar{K}^{\mathrm{E}}_{\mathrm{s,m,2}}$
	$\mathrm{cm^3 \cdot mol^{-1}}$						$\mathrm{TPa^{-1}}$					
2-Methoxyaniline (1) + 1,1,2,2-tetrachloroethane (2)												
0.0000	−0.863	0.000	−1.019	0.000	−1.169	0.000	−111.9	0.000	−146.6	0.000	−190.9	0.000
0.0911	−0.731	−0.006	−0.807	−0.010	−0.881	−0.015	−86.64	−1.178	−101.4	−2.053	−138.5	−3.139
0.1962	−0.583	−0.031	−0.609	−0.044	−0.635	−0.057	−63.68	−4.993	−67.22	−7.647	−94.44	−11.12
0.2806	−0.471	−0.066	−0.479	−0.084	−0.487	−0.101	−49.08	−9.546	−49.60	−13.10	−68.85	−18.06
0.3816	−0.349	−0.129	−0.350	−0.145	−0.353	−0.162	−35.17	−16.41	−35.95	−19.77	−46.71	−25.15
0.4976	−0.228	−0.226	−0.233	−0.233	−0.238	−0.242	−22.92	−25.99	−25.92	−27.58	−29.31	−31.49
0.6023	−0.140	−0.336	−0.149	−0.332	−0.158	−0.330	−14.50	−36.26	−19.05	−36.01	−18.45	−37.70
0.7116	−0.071	−0.469	−0.082	−0.460	−0.091	−0.454	−7.867	−48.97	−12.39	−48.92	−10.25	−49.49
0.8104	−0.030	−0.599	−0.037	−0.607	−0.044	−0.615	−3.557	−62.73	−6.595	−67.64	−4.833	−70.95
0.9212	−0.005	−0.748	−0.007	−0.819	−0.009	−0.881	−0.657	−81.57	−1.441	−101.9	−0.950	−116.7
1.0000	0.000	−0.851	0.000	−1.008	0.000	−1.147	0.000	−97.83	0.000	−138.6	0.000	−170.1

Table 7 The values of $\overline{V}^{\circ}_{\mathrm{m},1}$,$V^{*}_{\mathrm{m},1}$,$\overline{V}^{\circ\mathrm{E}}_{\mathrm{m},1}$,$\overline{V}^{\circ}_{\mathrm{m},2}$, $V^{*}_{\mathrm{m},2}$,$\overline{V}^{\circ\mathrm{E}}_{\mathrm{m},2}$$\overline{K}^{\circ}_{\mathrm{s,m},1}$, $K^{*}_{\mathrm{s,m},1}$, $\overline{K}^{\circ\mathrm{E}}_{\mathrm{s,m},1}$, $\overline{K}^{\circ}_{\mathrm{s,m},2}$, $K^{*}_{\mathrm{s,m},2}$ and $\overline{K}^{\circ\mathrm{E}}_{\mathrm{s,m},2}$ of the components for 2-methoxyaniline with chlorinated ethane components of binary mixtures at T = (303.15–313.15) K

T/K	$\overline{V}^{\circ}_{\mathrm{m},1}$ (cm^3·mol^{-1})	$V^{*}_{\mathrm{m},1}$	$\overline{V}^{\circ\mathrm{E}}_{\mathrm{m},1}$	$\overline{V}^{\circ}_{\mathrm{m},2}$	$V^{*}_{\mathrm{m},2}$	$\overline{V}^{\circ\mathrm{E}}_{\mathrm{m},2}$	$\overline{K}^{\circ}_{\mathrm{s,m},1}$	$K^{*}_{\mathrm{s,m},1}$ TPa^{-1}	$\overline{K}^{\circ\mathrm{E}}_{\mathrm{s,m},1}$	$\overline{K}^{\circ}_{\mathrm{s,m},2}$	$K^{*}_{\mathrm{s,m},2}$	$\overline{K}^{\circ\mathrm{E}}_{\mathrm{s,m},2}$	
2-Methoxyaniline (1) + 1,2-dichloroethane (2)													
303.15	112.66	112.82	−0.162	79.69	79.90	−0.210		−70.67	4.050	−74.72	−43.49	4.671	−48.16
308.15	112.99	113.19	−0.204	80.14	80.40	−0.255		−110.32	4.151	−114.47	−71.40	4.810	−76.21
313.15	113.32	113.57	−0.251	80.58	80.89	−0.308		−147.62	4.250	−151.87	−101.62	4.954	−106.58
2-Methoxyaniline (1) + 1,1,2-trichloroethane (2)													
303.15	112.37	112.82	−0.454	93.24	93.66	−0.424		−84.91	4.050	−88.96	−63.91	5.155	−69.07
308.15	112.52	113.19	−0.673	93.50	94.17	−0.668		−120.04	4.151	−124.19	−95.05	5.329	−100.38
313.15	112.70	113.57	−0.871	93.83	94.66	−0.827		−165.57	4.250	−169.82	−127.59	5.508	−133.10
2-Methoxyaniline (1) + 1,1,2,2-tetrachloroethane (2)													
303.15	111.96	112.82	−0.863	105.40	106.25	−0.851		−107.82	4.050	−111.87	−92.60	5.221	−97.83
308.15	112.17	113.19	−1.022	105.79	106.80	−1.005		−142.49	4.151	−146.64	−133.17	5.466	−138.63
313.15	112.39	113.94	−1.179	106.19	107.33	−1.148		−186.66	4.250	−190.91	−164.39	5.723	−170.12

indicate complex formation through electron acceptor–donor interactions between component molecules.

3.2 Prigogine–Flory–Patterson Theory

Quantitative estimations of different contributions to V^{E} can be obtained by using the well-known Prigogine–Flory–Patterson theory (PFP). This theory has been described in detail by Patterson and co-workers [28–30] and has been widely used to analyze excess thermodynamic properties for different kinds of mixtures, including polar compounds, by several authors. Their approximate expression for V^{E} in terms of three contributions is

$$\begin{aligned}\frac{V^{\mathrm{E}}}{x_1V_1^*+x_2V_2^*} &= \frac{(\tilde{V}^{1/3}-1)\tilde{V}^{2/3}\Psi_1\theta_2\chi_{12}}{\left[\left(\frac{4}{3}\right)\tilde{V}^{-\frac{1}{3}}-1\right]P_1^*}\ \text{(int.contribution)}\\ &+\frac{-\left(\tilde{V}_1-\tilde{V}_2\right)[(14/9)\tilde{V}^{-1/3}-1]\Psi_1\Psi_2}{\left[\left(\frac{4}{3}\right)\tilde{V}^{-\frac{1}{3}}-1\right]\tilde{V}}\ \text{(fv.contribution)}\\ &+\frac{\left(\tilde{V}_1-\tilde{V}_2\right)\left(P_{1\_}^*P_2^*\right)\Psi_1\Psi_2}{P_2^*\Psi_1+P_1^*\Psi_2}\ (P^*\text{contribution})\end{aligned} \tag{7}$$

The reduced volume of pure component $\tilde{V}_i$ was calculated from the isobaric thermal expansively α_i by the equation:

$$\tilde{V}_i=\left(\frac{1+\left(\frac{4}{3}\right)\alpha_iT}{1+\alpha_iT}\right) \tag{8}$$

The $\tilde{\mathrm{V}}$ of mixture is approximated in Eq. 8 by

$$\tilde{V}=\Psi_1\tilde{V}_1+\Psi_2\tilde{V}_2 \tag{9}$$

The molecular contact energy fraction of components Ψ_1 is given by.

$$\Psi_1=1-\Psi_2=\frac{\phi_1P_1^*}{\phi_1P_1^*+\phi_2P_2^*} \tag{10}$$

The characteristic volume is $V_i^*=V_i^*/\tilde{V}_i$ and the characteristic pressure is given by

$$P_i^*=\frac{T\tilde{V}_i^2\alpha_i}{\kappa_{Ti}} \tag{11}$$

where κ_{Ti} is the isothermal compressibility of pure component *i.*

The hard-core volume fractions of the components 1 and 2 (ϕ_1 and ϕ_2) are defined by

$$\phi_1=1-\phi_2=\frac{x_1V_1^*}{x_1V_1^*+x_2V_2^*} \tag{12}$$

Table 8 PFP interaction parameter, χ_{12} and calculated values of the three contributions from the PFP theory with experimental excess molar volumes at $x_1 = 0.5$ at 303.15 K

Binary mixtures	$\chi_{12}(10^{6})$	Calculated contributions			V^E ($x = 0.5$) $m^3 \cdot mol^{-1}$		
		Interactional (10^{-8})	Free volume	P^* effect	exp	PFP	$\delta/m^3.mol^{-1}$
2-Methoxyaniline + 1,2-dichloroethane	−21.30	1.005	−0.1726	−0.0922	−0.0577	−0.0578	0.0001
2-Methoxyaniline + 1,1,2-trichloroethane	−6.138	1.020	−0.0681	−0.1036	−0.1711	−0.1707	−0.0004
2-Methoxyaniline + 1,1,2,2-tetrachloroethane	−9.482	1.048	−0.0521	−0.0748	−0.2272	−0.2265	−0.0007

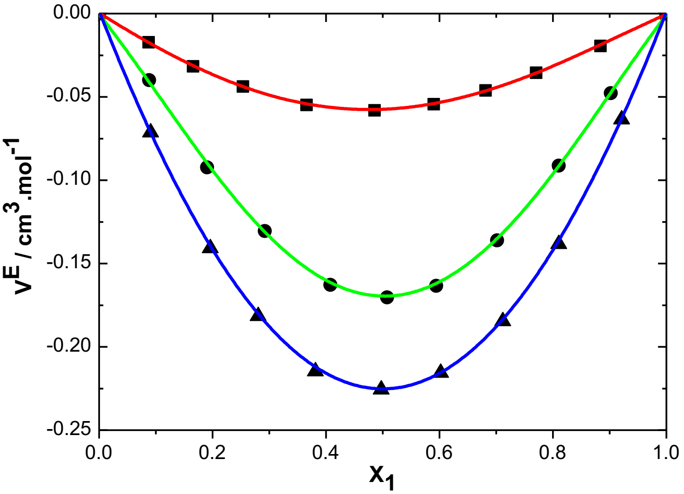

Fig. 5 Excess molar volumes with mole fraction (x_1) of 2-methoxyaniline in the binary liquid mixtures of 2-methoxyaniline with 1,1,2,2-tetrachloroethane (▲); 1,1,2-trichloroethane (●) and 1,2-dichloroethane (■) at 303.15 K calculated with the PFP theory using evaluated parameters

The κ_T values calculated from the following expression,

$$\kappa_T = \kappa_S + \frac{TV\alpha^2}{C_P}. \tag{13}$$

The χ_{12} parameter required for the calculation of V^E using the Flory–Patterson theory was derived by fitting the V^E expression to the experimental equimolar value of V^E for each system investigated. The calculated equimolar values of the three contributions together with the χ_{12} parameter for each system are listed in Table 8. The comparison of experimental V^E values with those calculated from PFP is shown graphically in Fig. 5. Table 8 shows that the interactional contribution was negative for all systems, the free volume and P^* contributions are negative for all the mixtures.

4 Conclusions

This paper reports experimental data of densities, speeds of sound and viscosities of binary blends of 2-methoxyaniline with chlorinated ethane binary mixtures over the entire composition range at $T = (303.15–313.15)$ K with 5 K intervals. From the experimental data, various physicochemical parameters, V^E, κ_s^E and $\Delta\eta$ of the mixtures, the excess partial molar properties and excess partial molar properties at infinite dilution of components were calculated. The present study excess properties become more negative with increase in temperatures for all the studied liquid mixtures. Finally, it can be concluded that the results were analyzed in terms of intermolecular complexes through hydrogen bonding between 2-methoxyaniline and chlorinated ethane molecules.

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