

Studies of Associated Solutions: Evaluation of Thermodynamic Parameters of Blends of 2-Methylaniline and Substituted Ethanols at Various Temperatures

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Abstract Densities (ρ), speeds of sound (u), and viscosities (η) are reported for binary mixtures of 2-methylaniline with substituted ethanols (2-phenylethanol, 2-chloroethanol and 2-aminoethanol) over the entire composition range of mole fraction at $T = (303.15\text{--}318.15)$ K and at atmospheric pressure, 0.1 MPa. The excess molar volume, excess isentropic compressibility and deviation in viscosity are calculated from the corresponding experimental densities, speeds of sound and viscosities. The excess properties are correlated using the Redlich–Kister polynomial smoothing equation. Excess partial molar volumes and excess partial molar isentropic compressibilities were calculated for all the binary systems throughout the composition range and also at infinite dilution. The variations in these properties with composition, for all the binary mixtures, suggest that loss of dipolar association, difference in size and shape of the component molecules, dipole–dipole interaction and hydrogen bonding between molecules of 2-methylaniline with 2-phenylethanol, 2-chloroethanol and 2-aminoethanol are involved.

Keywords Density · Speed of sound · Viscosity · 2-Methylaniline · Substituted ethanols · PFP theory

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1 Introduction

The study of molecular interactions in mixed solvent systems is of great significance owing to the practical applications of these systems in various technologies, as they provide a wide choice of solutions with appropriate properties. The excess thermodynamic properties of binary liquid mixtures have been very useful for obtaining information on the intermolecular interactions in the systems. The negative or positive deviations from the ideal value depend on the type and extent of the interactions between unlike molecules, as well as on the composition and the temperature [1].

Knowledge of the physico-chemical properties of non-aqueous binary liquid mixtures has relevance in theoretical and applied areas of research and such results are frequently used in the design process (flow, mass transfer or heat transfer calculations) in many chemical and industrial processes. The excess properties derived from these physical property data reflect the physico-chemical behavior of the liquid mixtures with respect to the solution structure and intermolecular interactions between the component molecules of the mixture [2].

This work is part of our program to give information/data for the characterization of molecular interactions between solvents in binary systems [3]. The liquids were chosen for the present study on the basis of their industrial importance. 2-Methylaniline is an important compound used in the manufacturing of dyes and of rubber vulcanization accelerators. It is also used in the fabrication of hypnotic and anesthetic pharmaceuticals and pesticides. On the other hand, 2-chloroethanol is a polar, bi-functional compound, consisting of both a hydroxyl group as a proton donor and halogen atom as a proton acceptor. It is a versatile solvent used in many industrial areas and is also a mutagenic chemical. 2-Aminoethanol is a widely used agent in the carbon dioxide and hydrogen sulfide removal processes. 2-Phenylethanol has found usage in artificial essences and as a base solvent for some flavor compounds.

In the present study, our focus is on the study of liquid mixtures of substituted ethanols with 2-methylaniline because there have been a few studies on these mixtures [4, 5]. It is expected that there will be a significant degree of H-bonding in these binary mixtures because 2-methylaniline and substituted ethanols both have a proton donor and a proton acceptor group [6]. To understand the possible associations between 2-methylaniline and substituted ethanols through $-\text{OH}\cdots\text{NH}$ and $-\text{NH}\cdots\text{OH}$ bonds, we report the densities, speeds of sound and viscosities for three binary systems (2-methylaniline with 2-phenylethanol, with 2-chloroethanol, and with 2-aminoethanol) at $T = (303.15\text{--}318.15)$ K and under 0.1 MPa pressure. The experimental data has been used to compute the excess volumes (V^E), excess isentropic compressibilities (κ_S^E) and deviations in viscosity ($\Delta\eta$). The results are used to qualitatively discuss specific interactions between unlike molecules.

2 Experimental Methods

2.1 Materials

Chemicals used in the present study are 2-methylaniline (Sigma–Aldrich), 2-phenylethanol (Sigma–Aldrich), 2-chloroethanol and 2-aminoethanol. These chemicals were purchased from S.D. Fine Chemicals Ltd. 2-chloroethanol and 2-aminoethanol chemicals were further purified by standard methods [7, 8] like distillation and fractional distillation under reduced

pressure, and only the middle fractions were collected. Before use, the chemicals were stored over 0.4 nm molecular sieves for about 72 h to remove water and gas. The purity of the liquid samples was checked by gas chromatography. The water contents were determined by the Karl–Fischer method. 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 the pure liquids on an Afcoset-ER-120A electric balance, using a syringe, in a narrow mouth stoppered bottle. The resolution of electronic balance was ± 0.01 mg while the accuracy of the mole fraction was $\pm 1 \times 10^{-4}$.

Densities and speeds of sound were measured with a digital oscillating Density and Sound Analyzer (DSA 5000 M, Anton Parr, Austria) with a reproducibility of $\pm 1 \times 10^{-6}$ g·cm⁻³ for the density and $\pm 1 \times 10^{-2}$ m·s⁻¹ for the speed of sound. The speed of sound was measured using a propagation time technique at the frequency 3 MHz. The densimeter was calibrated randomly with dry air at atmospheric pressure and doubly-distilled, freshly degassed and deionized water ($\rho = 997.075$ kg·m⁻³ at 298.15 K) supplied by Anton-Paar as described elsewhere. After each measurement, distilled water and anhydrous ethanol were used to clean the vibrating tube. The combined expanded uncertainties associated with the measurements for temperature, density and speed of sound are estimated to be within 0.01 K, 0.8×10^{-3} g·cm⁻³ and 0.5 m·s⁻¹, respectively, at the 95% confidence level.

The viscosities of the pure liquids and their mixtures were determined at atmospheric pressure at $T = (303.15\text{--}318.15)$ K by using an Ubbelohde viscometer, which was calibrated with benzene, carbon tetrachloride, acetonitrile and doubly distilled water. The Ubbelohde viscometer bulb capacity was 15 mL and the capillary tube had a length of about 90 mm with 0.5 mm internal diameter. The viscometer was thoroughly cleaned and perfectly dried, filled with the sample liquid by fitting the viscometer to about 30° from the vertical, and its limbs were closed with Teflon caps to avoid evaporation. The viscometer was kept in a transparent walled bath with a thermal stability of ± 0.01 K for about 20 min to obtain thermal equilibrium before making the measurement. An electronic digital stopwatch with an uncertainty of ± 0.01 s was used for flow time measurements. The experimental uncertainty of viscosity of pure liquids was estimated at 0.17 mPa·s and the uncertainty of temperature ± 0.01 K. The combined expanded uncertainty of viscosity was estimated as 0.2 mPa·s. The purities of all these solvents were compared with the measured densities, speeds of sound and viscosities of the pure liquids with literature [9–28] and these comparisons are listed in Table 2 and graphically compared with the average absolute deviation (AAD) with literature values in Supplementary Material as Fig. 1S–9S. Density, speed of sound and viscosity values of 2-methylaniline were taken from our previous papers [29, 30].

3 Results and Discussion

The experimental densities and sound velocities for all the binary systems at various compositions were used to calculate the excess thermodynamic functions, using the following equations:

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	Water content mass fraction ^a	Analysis method
2-Methylamine	Sigma Aldrich India	95-53-4	No purification	0.995	0.0004	GC
2-Phenylethanol	Sigma Aldrich India	60-12-8	No purification	0.995	0.0004	GC
2-Chloroethanol	S.D. Fine Chemicals, India	107-07-3	Distillation under reduced pressure	0.995	0.0004	GC
2-Aminoethanol	S.D. Fine Chemicals, India	141-43-5	Fractional distillation	0.995	0.0004	GC

GC: gas chromatography

^aKarl–Fischer method

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

Component	Density ($\rho/\text{g}\cdot\text{cm}^{-3}$)		Speed of sound ($u/\text{m}\cdot\text{s}^{-1}$)		Viscosity ($\eta/\text{mPa}\cdot\text{s}$)	
	Experimental	Literature	Experimental	Literature	Experimental	Literature
2-Methylaniline						
303.15 K	0.9902	0.9902 [9]	1578.5	1579.0 [11]	3.258	3.255 [28]
308.15 K	0.9860	0.9860 [10]	1558.2	1558.4 [10]	2.824	2.820 [28]
313.15 K	0.9817	0.9817 [11]	1538.5	1539.0 [11]	2.487	2.485 [28]
318.15 K	0.9776	–	1518.5	–	2.153	2.150 [28]
2-Phenylethanol						
303.15 K	1.0123	1.0123 [12]	1509.6	1510.5 [12]	9.027	
		1.0124 [14]		1510.5 [14, 21]		
		1.0124 [21]				
308.15 K	1.0085	1.0086 [12]	1491.6	1494.8 [12, 14]	7.653	7.518 [15]
		1.0086 [13, 15]		1494.7 [21]		7.438 [14]
		1.0086 [14]				7.190 [16]
		1.0086 [16]				7.190 [17]
313.15 K	1.0047	1.0047 [12]	1474.8	1479.1 [12]	6.335	
		1.0048 [14, 21]		1479.2 [14, 21]		
318.15 K	1.0009	1.0010 [12]	1457.7	1463.6 [12]	4.984	4.980 [16]
		1.0010 [13, 14]		1463.3 [14, 21]		5.232 [15]
		1.0010 [15, 16]				
		1.0093 [21]				
2-Chloroethanol						
303.15 K	1.1921	1.1925 [20]	1342.8		2.583	2.669 [20]
		1.1921 [24]				2.580 [24]
308.15 K	1.1852	1.1871 [20]	1328.9	1329.9 [19]	2.234	2.261 [24]
		1.1849 [18, 19]				2.347 [20]
		1.1866 [24]				
313.15 K	1.1780	1.1738 [18]	1315.3	–	1.882	–
318.15 K	1.1709	1.1736 [19]	1301.2	1300.2 [19]	1.564	–
2-Aminoethanol						
303.15 K	1.0087	1.0087 [23]	1704.9	1703.7 [23]	14.424	14.423 [22]
		1.0081 [22]				14.956 [23]
308.15 K	1.0045	1.0041 [22]	1689.9	1686.7 [25]	12.015	–
		1.0047 [25]				
		1.0045 [26]				
313.15 K	1.0004	1.0008 [23]	1675.0	1671.2 [23]	9.778	9.562 [22]

Table 2 continued

Component	Density ($\rho/\text{g}\cdot\text{cm}^{-3}$)		Speed of sound ($u/\text{m}\cdot\text{s}^{-1}$)		Viscosity ($\eta/\text{mPa}\cdot\text{s}$)	
	Experimental	Literature	Experimental	Literature	Experimental	Literature
318.15 K	0.9960	1.0004 [22]	1659.4	1653.5 [25]	7.558	9.839 [23]
		0.9965 [26]				–
		0.9960 [22, 27]				

The standard uncertainties u are: $u(\rho) = 0.5 \times 10^3 \text{ g}\cdot\text{cm}^{-3}$, $u(u) = 0.2 \text{ m}\cdot\text{s}^{-1}$, $u(\eta) = 0.17 \text{ mPa}\cdot\text{s}$, $u(T) = 0.01 \text{ K}$ for density and speed of sound, $u(T) = 0.01 \text{ K}$ for viscosity and $u(p) = 1 \text{ kPa}$

$$V^E(\text{m}^3 \cdot \text{mol}^{-1}) = [x_1M_1 + x_2M_2]/\rho - [x_1M_1/\rho_1 + x_2M_2/\rho_2] \tag{1}$$

Deviation in viscosity were be calculated by following equation,

$$\Delta\eta(\text{mPa} \cdot \text{s}) = \eta - (x_1\eta_1 + x_2\eta_2) \tag{2}$$

where ρ , η and V are the density, viscosity and molar volume of the binary mixture. Also 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.

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

$$\kappa_S = (u^2 \rho)^{-1} \tag{3}$$

The method used for calculating κ_S^E (Benson–Kiyohara approach) was outlined previously [31]:

$$\kappa_S^E = \kappa_S - \kappa_S^{\text{id}} \tag{4}$$

where κ_S is the isentropic compression and κ_S^{id} is the isentropic compression of the ideal mixture,

$$\kappa_S^{\text{id}} = \sum_{i=1}^2 \phi_i [\kappa_{S,i} + TV_i(\alpha_i^2)/C_{p,i}] - \left\{ T \left(\sum_{i=1}^2 x_i V_i \right) \left(\sum_{i=1}^2 \phi_i \alpha_i \right)^2 / \sum_{i=1}^2 x_i C_{p,i} \right\} \tag{5}$$

where ϕ_i is the volume fraction of component i , and κ_S , V_m , α_p and C_p are the isentropic compressibility, molar volume, coefficient of isobaric thermal expansion and molar heat capacity, respectively, and R is the gas constant and T is the absolute temperature. The values of ϕ_i , V_m^{id} , α_p^{id} and C_p^{id} were calculated using the following relations:

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

$$V_m^{\text{id}} = x_1 V_{m,1} + x_2 V_{m,2}$$

and

$$C_p^{\text{id}} = x_1 C_{p,1} + x_2 C_{p,2}$$

Values of C_p were obtained from the literature [32]. α_p was calculated from measured densities by the relation,

$$\alpha_p = \frac{1}{V_m} \left(\frac{\partial V_m}{\partial T} \right)_p = -\frac{1}{\rho} \left(\frac{\partial \rho}{\partial T} \right)_p = -\left(\frac{\partial \ln \rho}{\partial T} \right)_p \quad (6)$$

The V^E , κ_S^E and $\Delta\eta$ values were fitted with a Redlich–Kister [33] polynomial equation,

$$Y^E = x_1 x_2 \sum_{i=0}^j A_i (1 - 2x_1)^i \quad (7)$$

where Y^E represents V^E , κ_S^E and $\Delta\eta$. Values of the coefficients A_i have been determined by using the method of least squares. The standard deviations $\sigma(Y^E)$ have been calculated by using the formula

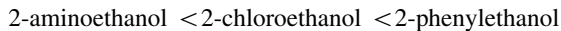
$$\sigma(Y^E) = \left[\sum_m \left(Y_{\text{exp}}^E - Y_{\text{calc}}^E \right)^2 / (m - n)^{1/2} \right] \quad (8)$$

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 6.

The observed V^E values can be discussed in terms of several effects, which are (i) physical, (ii) chemical and (iii) geometrical contributions [34]. (i) The physical interactions involve expansion due to mutual breaking of OH...O and N–H...N bonds present in the self-associated substituted ethanol and the amine; (ii) the chemical or specific interactions involves contraction due to complex formation between unlike molecules, which include hydrogen bond (HNH...OH and OH...N); and (iii) structural contributions resulting from geometrical fitting of one component into other, due to the difference in the free volumes between components, resulting in negative contributions to V^E .

The excess volume data reported in Tables 3, 4, and 5 at $T = 303.15$ K are graphically represented in Fig. 1 and in Fig. 10S at 313.15 K. The excess volume has negative values for all the studied systems over the whole composition range and at all experimental temperatures. The negative value of V^E may be due to the predominance of chemical or specific interactions and structural contributions dominating over physical contributions. Hence, the OH...N bond is stronger than the OH...O and N–H...N bonds in the mixtures [35]. The existence of strong O–H...N bond was also confirmed through NMR, IR and UV studies [36]. The same trend is observed for the system water + 2-aminoethanol [37].

The values of V^E for the binary mixtures of 2-methylaniline with the substituted ethanols are in the following order:



The more negative excess volume in the system 2-methylaniline + 2-aminoethanol reveals that more efficient packing and/or attractive interactions occur between these two components when mixed together. Consequently, its structure and smaller size lead to easier interstitial accommodation with 2-methylaniline molecules compared to 2-methylaniline with 2-chloroethanol or with 2-phenylethanol. Similar results have been reported earlier [38, 39]. Hence the above order is justified.

Table 3 Density (ρ), excess molar volumes (V^E), speed of sound (u), excess isentropic compressibility (κ_S^E), viscosity (η) and deviation in viscosity ($\Delta\eta$) of binary liquid mixtures of 2-methylaniline with 2-phenylethanol at $T = (303.15\text{--}318.15)$ K and 0.1 MPa pressure

x_1	Density (ρ) ($\text{g}\cdot\text{cm}^{-3}$)	V^E ($\text{cm}^3\cdot\text{mol}^{-1}$)	u ($\text{m}\cdot\text{s}^{-1}$)	κ_S^E (TPa^{-1})	Viscosity (η) ($\text{mPa}\cdot\text{s}$)	$\Delta\eta$ ($\text{mPa}\cdot\text{s}$)
$T = 303.15$ K						
0.0000	1.0123	0.0000	1509.6	0.000	9.027	0.000
0.0857	1.0108	− 0.0218	1518.7	− 2.365	8.539	0.007
0.1652	1.0094	− 0.0439	1526.3	− 3.965	8.086	0.012
0.2724	1.0074	− 0.0703	1535.9	− 5.579	7.473	0.017
0.3731	1.0054	− 0.0883	1543.8	− 6.347	6.895	0.021
0.4711	1.0034	− 0.0963	1551.0	− 6.635	6.332	0.023
0.6147	1.0001	− 0.0893	1560.2	− 6.142	5.503	0.022
0.7123	0.9977	− 0.0722	1565.8	− 5.226	4.937	0.020
0.8105	0.9952	− 0.0503	1570.7	− 3.879	4.366	0.015
0.8904	0.9931	− 0.0299	1574.2	− 2.400	3.900	0.009
1.0000	0.9902 [29]	0.0000	1578.5 [29]	0.000	3.258 [30]	0.000
$T = 308.15$ K						
0.0000	1.0085	0.0000	1491.6	0.000	7.653	0.000
0.0857	1.0069	− 0.0248	1500.8	− 2.616	7.247	0.008
0.1652	1.0055	− 0.0489	1508.4	− 4.387	6.868	0.013
0.2724	1.0035	− 0.0757	1517.7	− 6.033	6.356	0.019
0.3731	1.0015	− 0.0938	1525.4	− 6.803	5.874	0.022
0.4711	0.9994	− 0.1008	1532.1	− 6.994	5.402	0.024
0.6147	0.9961	− 0.0943	1541.3	− 6.597	4.708	0.024
0.7123	0.9937	− 0.0782	1546.7	− 5.715	4.234	0.021
0.8105	0.9911	− 0.0548	1551.4	− 4.303	3.755	0.016
0.8904	0.9890	− 0.0324	1554.9	− 2.889	3.364	0.010
1.0000	0.9860 [29]	0.0000	1558.2 [29]	0.000	2.824 [30]	0.000
$T = 313.15$ K						
0.0000	1.0047	0.0000	1474.8	0.000	6.335	0.000
0.0857	1.0032	− 0.0278	1484.4	− 3.101	6.014	0.009
0.1652	1.0017	− 0.0529	1491.7	− 4.839	5.714	0.014
0.2724	0.9996	− 0.0797	1500.5	− 6.452	5.307	0.020
0.3731	0.9975	− 0.0978	1507.8	− 7.189	4.923	0.023
0.4711	0.9954	− 0.1053	1514.2	− 7.346	4.548	0.025
0.6147	0.9920	− 0.0993	1523.1	− 7.020	3.994	0.025
0.7123	0.9896	− 0.0832	1528.2	− 6.135	3.616	0.022
0.8105	0.9870	− 0.0593	1532.9	− 4.827	3.234	0.017
0.8904	0.9848	− 0.0369	1536.1	− 3.307	2.921	0.012
1.0000	0.9817 [29]	0.0000	1538.5 [29]	0.000	2.487 [30]	0.000
$T = 318.15$ K						
0.0000	1.0009	0.0000	1457.7	0.000	4.984	0.000
0.0857	0.9994	− 0.0323	1467.4	− 3.399	4.751	0.010
0.1652	0.9979	− 0.0574	1474.7	− 5.398	4.532	0.016
0.2724	0.9958	− 0.0847	1483.0	− 6.885	4.234	0.021

Table 3 continued

x_1	Density (ρ) ($\text{g}\cdot\text{cm}^{-3}$)	V^E ($\text{cm}^3\cdot\text{mol}^{-1}$)	u ($\text{m}\cdot\text{s}^{-1}$)	κ_S^E (TPa^{-1})	Viscosity (η / $\text{mPa}\cdot\text{s}$)	$\Delta\eta$ ($\text{mPa}\cdot\text{s}$)
0.3731	0.9937	- 0.1018	1489.7	- 7.478	3.952	0.025
0.4711	0.9915	- 0.1093	1495.8	- 7.612	3.676	0.026
0.6147	0.9881	- 0.1043	1504.3	- 7.280	3.269	0.026
0.7123	0.9856	- 0.0891	1509.6	- 6.640	2.991	0.023
0.8105	0.9830	- 0.0652	1514.2	- 5.427	2.708	0.019
0.8904	0.9808	- 0.0413	1517.0	- 3.772	2.476	0.013
1.0000	0.9776 [29]	0.0000	1518.5 [29]	0.000	2.153 [30]	0.000

Standard uncertainty $u(x) = 1 \times 10^{-4}$. Combined expanded uncertainties $U_c(\rho) = 0.8 \times 10^{-3} \text{ g}\cdot\text{cm}^{-3}$, $U_c(u) = 0.5 \text{ m}\cdot\text{s}^{-1}$, and $U(\eta) = 0.2 \text{ mPa}\cdot\text{s}$

Further, inspection of Tables 3, 4, 5 and Fig. 1 indicates that as the temperature increases from 303.15 to 318.15 K, the values of V^E for all these binary systems decrease. An increase in temperature increases the kinetic energy and therefore the breaking up of associate species present in the pure liquids, releasing more free dipoles of unlike molecules into the mixture, which results in interactions with each other and the formation of greater numbers of H-bonds between 2-methylaniline and the substituted ethanol's monomer. This has already been described in the literature for 1-alkanol + hexane systems [40], alcohol + triethylene glycol systems [41] and 1-hexanol + ether systems [42].

Excess isentropic compressibility (κ_S^E) data for the mixtures of 2-methylaniline with substituted ethanols are graphically depicted in Fig. 2 at 303.15 K and Fig. 11S at 313.15 K and the data are given in Tables 3, 4, 5, and they show that the κ_S^E values are negative over the entire mole fraction range at $T = (303.15\text{--}318.15)$ K and become more negative with increasing temperature for all three binary mixtures. Thus, the mixtures are less compressible than the pure components, i.e., a greater resistance to compression (enhanced rigidity) is observed. The three systems show both enhanced rigidity ($\kappa_S^E < 0$) and contraction ($V^E < 0$) over the entire composition range. In other words, the volume decreases (more compact packing of molecules), and simultaneously the whole system becomes more rigid (less compressible) [43]. As shown in Fig. 2, in the mixtures of 2-methylaniline and 2-phenylethanol, the trend of κ_S^E is identical to that of the excess molar volumes. The same trend can be observed for other two systems. Interpretation of the κ_S^E data is generally not simple because the $\kappa_S^E < 0$ values are affected by both the molecular packing and the patterns of molecular aggregation induced by the molecular interactions. However, in these three binary systems it seems that the interpretation of negative κ_S^E values will be the same as for the negative V^E values.

The values of κ_S^E for the binary mixtures of 2-methylaniline with substituted ethanols are in the following order:



An examination of the curves in Fig. 3 at 303.15 K and Fig. 12S at 313.15 K indicates that the negative values for binary mixture containing 2-methylaniline and 2-phenylethanol are smaller than those of the other binary systems, because the phenyl group ($-\text{C}_6\text{H}_5$) has steric hindrance with the $-\text{NH}_2$ group. 2-Methylaniline has a free NH_2 group and easily interacts with the neighboring molecules [44–46]. Hence the above order is justified. The

Table 4 Density (ρ), excess molar volumes (V^E), speed of sound (u), excess isentropic compressibility (κ_S^E), viscosity (η), and deviation in viscosity ($\Delta\eta$) of binary liquid mixtures of 2-methylaniline with 2-chloroethanol at $T = (303.15\text{--}318.15)$ K and 0.1 MPa pressure

x_1	Density (ρ) ($\text{g}\cdot\text{cm}^{-3}$)	V^E ($\text{cm}^3\cdot\text{mol}^{-1}$)	u ($\text{m}\cdot\text{s}^{-1}$)	κ_S^E (TPa^{-1})	Viscosity (η) ($\text{mPa}\cdot\text{s}$)	$\Delta\eta$ ($\text{mPa}\cdot\text{s}$)
$T = 303.15$ K						
0.0000	1.1921	0.0000	1342.8	0.000	2.583	0.000
0.1141	1.1582	− 0.0358	1382.5	− 3.203	2.670	0.010
0.2148	1.1315	− 0.0623	1414.4	− 5.181	2.744	0.016
0.3201	1.1065	− 0.0864	1444.6	− 6.378	2.820	0.021
0.4123	1.0865	− 0.0973	1468.9	− 6.941	2.884	0.023
0.5121	1.0667	− 0.1019	1492.8	− 6.994	2.953	0.024
0.6225	1.0467	− 0.0938	1516.7	− 6.460	3.026	0.023
0.7112	1.0319	− 0.0795	1534.2	− 5.714	3.084	0.021
0.8184	1.0153	− 0.0545	1553.0	− 4.233	3.151	0.016
0.9017	1.0034	− 0.0322	1565.7	− 2.579	3.202	0.010
1.0000	0.9902 [29]	0.0000	1578.5 [29]	0.000	3.258 [30]	0.000
$T = 308.15$ K						
0.0000	1.1852	0.0000	1328.9	0.000	2.234	0.000
0.1141	1.1518	− 0.0396	1368.0	− 3.610	2.312	0.011
0.2148	1.1255	− 0.0681	1399.1	− 5.600	2.378	0.017
0.3201	1.1008	− 0.0907	1428.5	− 6.847	2.445	0.022
0.4123	1.0811	− 0.1022	1451.9	− 7.256	2.502	0.025
0.5121	1.0616	− 0.1055	1475.4	− 7.377	2.562	0.026
0.6225	1.0418	− 0.0983	1498.9	− 6.976	2.626	0.025
0.7112	1.0272	− 0.0842	1515.8	− 6.202	2.676	0.022
0.8184	1.0109	− 0.0599	1534.1	− 4.714	2.734	0.017
0.9017	0.9990	− 0.0362	1546.4	− 2.977	2.777	0.011
1.0000	0.9860 [29]	0.0000	1558.2 [29]	0.000	2.824 [30]	0.000
$T = 313.15$ K						
0.0000	1.1780	0.0000	1315.3	0.000	1.882	0.000
0.1141	1.1452	− 0.0443	1353.9	− 3.984	1.963	0.012
0.2148	1.1194	− 0.0739	1384.1	− 5.961	2.031	0.019
0.3201	1.0950	− 0.0958	1412.8	− 7.209	2.099	0.023
0.4123	1.0756	− 0.1066	1435.6	− 7.656	2.157	0.026
0.5121	1.0563	− 0.1102	1458.3	− 7.710	2.219	0.027
0.6225	1.0369	− 0.1035	1481.2	− 7.350	2.285	0.027
0.7112	1.0224	− 0.0897	1497.8	− 6.570	2.337	0.025
0.8184	1.0063	− 0.0642	1515.6	− 5.094	2.396	0.019
0.9017	0.9946	− 0.0400	1527.5	− 3.292	2.440	0.012
1.0000	0.9817 [29]	0.0000	1538.5 [29]	0.000	2.487 [30]	0.000
$T = 318.15$ K						
0.0000	1.1709	0.0000	1301.2	0.000	1.564	0.000
0.1141	1.1387	− 0.0490	1339.2	− 4.365	1.645	0.014
0.2148	1.1133	− 0.0783	1368.9	− 6.564	1.711	0.020
0.3201	1.0893	− 0.1004	1396.6	− 7.629	1.777	0.025

Table 4 continued

x_1	Density (ρ) ($\text{g}\cdot\text{cm}^{-3}$)	V^E ($\text{cm}^3\cdot\text{mol}^{-1}$)	u ($\text{m}\cdot\text{s}^{-1}$)	κ_S^E (TPa^{-1})	Viscosity (η / $\text{mPa}\cdot\text{s}$)	$\Delta\eta$ ($\text{mPa}\cdot\text{s}$)
0.4123	1.0702	- 0.1110	1418.7	- 7.992	1.834	0.027
0.5121	1.0512	- 0.1145	1440.8	- 8.045	1.893	0.028
0.6225	1.0320	- 0.1080	1463.0	- 7.637	1.958	0.028
0.7112	1.0178	- 0.0950	1479.3	- 6.976	2.008	0.025
0.8184	1.0019	- 0.0707	1496.8	- 5.534	2.067	0.021
0.9017	0.9904	- 0.0450	1508.3	- 3.637	2.108	0.013
1.0000	0.9776 [29]	0.0000	1518.5 [29]	0.000	2.153 [30]	0.000

Standard uncertainty $u(x) = 1 \times 10^{-4}$, Combined expanded uncertainties $U_c(\rho) = 0.8 \times 10^{-3} \text{ g}\cdot\text{cm}^{-3}$, $U_c(u) = 0.5 \text{ m}\cdot\text{s}^{-1}$, and $u(\eta) = 0.2 \text{ mPa}\cdot\text{s}$

negative values of κ_S^E increase with increasing temperature, which suggests that specific interactions increase due to the enhanced thermal energy. Comelli et al. [47] have also reported similar behavior for κ_S^E .

A correlation between the signs of $\Delta\eta$ and V^E has been observed for a number of binary solvent systems [48, 49], i.e., $\Delta\eta$ is positive when V^E is negative and vice versa. In general, for systems where dispersion and dipolar interactions are operating, the $\Delta\eta$ values are found to be negative, whereas donor–acceptor and hydrogen-bonding interactions lead to the formation of complex species between unlike molecules and thereby result in positive $\Delta\eta$ values [50]. The viscosity deviation data of the liquid mixtures are graphically given in Fig. 3 and viscosity deviation data are given in Tables 3, 4, 5. The positive values of viscosity deviation for the binary systems investigated suggest that the hetero-molecular complexes between unlike molecules are relatively more numerous than those present in the pure components [51]. The effect of temperature increase is to disrupt hetero and homo association of the molecules resulting in an increase in fluidity of the liquids, giving higher $\Delta\eta$ values at higher temperatures. The viscosity deviation values are found to be opposite to the sign of excess molar volumes for all binary mixtures, which is in agreement with the view proposed by Brocos et al. [52, 53].

As Tables 3, 4 and 5 show, the excess properties obtained are negative over the entire composition range and absolute values of V^E , κ_S^E , and $\Delta\eta$ increase (become more negative) with increasing temperature from $T = (303.15\text{--}318.15)$ K. This effect can be attributed to the fact that the attractive interactions between like molecules become weaker with increasing temperature. Also, increasing temperature expands the volume of the mixture so that more space occurs between larger molecules that will become available for the smaller molecules to fill upon mixing. This packing effect will decrease the mixture volume.

4 Partial Molar Properties

The interpretations of excess partial molar properties ($\bar{V}_{m,1}^E$, $\bar{V}_{m,2}^E$, $\bar{K}_{s,m,1}^E$ and $\bar{K}_{s,m,2}^E$) and excess partial molar properties at infinite dilution ($\bar{V}_{m,1}^{\circ E}$, $\bar{V}_{m,2}^{\circ E}$, $\bar{K}_{s,m,1}^{\circ E}$ and $\bar{K}_{s,m,2}^{\circ E}$) of components 2 have previously been described [54].

Table 5 Density (ρ), excess molar volumes (V^E), speed of sound (u), excess isentropic compressibility (κ_S^E), viscosity (η) and deviation in viscosity ($\Delta\eta$) of binary liquid mixtures of 2-methylaniline with 2-aminoethanol at $T = (303.15\text{--}318.15)$ K and 0.1 MPa pressure

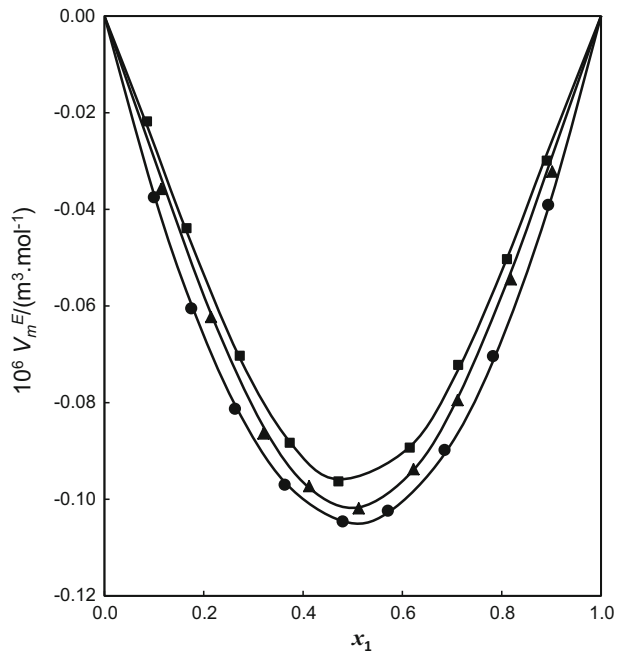
x_1	Density (ρ) ($\text{g}\cdot\text{cm}^{-3}$)	V^E ($\text{cm}^3\cdot\text{mol}^{-1}$)	U ($\text{m}\cdot\text{s}^{-1}$)	κ_S^E (TPa^{-1})	Viscosity (η) ($\text{mPa}\cdot\text{s}$)	$\Delta\eta$ ($\text{mPa}\cdot\text{s}$)
$T = 303.15$ K						
0.0000	1.0087	0.0000	1704.9	0.000	14.424	0.000
0.0997	1.0062	-0.0375	1688.8	-3.246	13.321	0.010
0.1749	1.0045	-0.0605	1677.4	-4.908	12.486	0.015
0.2629	1.0026	-0.0813	1664.9	-6.264	11.508	0.020
0.3629	1.0006	-0.0970	1651.5	-7.074	10.395	0.023
0.4797	0.9984	-0.1046	1637.2	-7.413	9.093	0.025
0.5707	0.9968	-0.1024	1626.7	-7.158	8.076	0.025
0.6849	0.9949	-0.0898	1614.1	-6.371	6.799	0.023
0.7826	0.9934	-0.0704	1603.6	-5.238	5.704	0.019
0.8935	0.9917	-0.0391	1591.4	-3.165	4.459	0.012
1.0000	0.9902 [29]	0.0000	1578.5 [29]	0.000	3.258 [30]	0.000
$T = 308.15$ K						
0.0000	1.0045	0.0000	1689.9	0.000	12.015	0.000
0.0997	1.0021	-0.0411	1673.4	-3.663	11.109	0.010
0.1749	1.0003	-0.0652	1661.2	-5.362	10.424	0.016
0.2629	0.9985	-0.0863	1647.8	-6.640	9.620	0.021
0.3629	0.9965	-0.1020	1633.8	-7.525	8.704	0.024
0.4797	0.9943	-0.1096	1618.7	-7.792	7.633	0.027
0.5707	0.9927	-0.1077	1607.9	-7.615	6.796	0.027
0.6849	0.9908	-0.0958	1594.9	-6.799	5.745	0.025
0.7826	0.9893	-0.0765	1584.2	-5.636	4.842	0.020
0.8935	0.9876	-0.0437	1571.8	-3.529	3.815	0.012
1.0000	0.9860 [29]	0.0000	1558.2 [29]	0.000	2.824 [30]	0.000
$T = 313.15$ K						
0.0000	1.000	0.0000	1675	0.000	9.778	0.000
0.0997	0.998	-0.0458	1658.1	-4.048	9.063	0.012
0.1749	0.996	-0.0681	1645.5	-5.927	8.521	0.018
0.2629	0.994	-0.0899	1631.4	-7.232	7.884	0.023
0.3629	0.992	-0.1075	1616.4	-7.930	7.159	0.026
0.4797	0.990	-0.1158	1600.5	-8.100	6.309	0.028
0.5707	0.989	-0.1142	1589.3	-7.905	5.646	0.028
0.6849	0.987	-0.1016	1576.3	-7.248	4.812	0.027
0.7826	0.985	-0.0819	1565.5	-6.140	4.095	0.023
0.8935	0.983	-0.0477	1552.6	-3.850	3.278	0.014
1.0000	0.9817 [29]	0.0000	1538.5 [29]	0.000	2.487 [30]	0.000
$T = 318.15$ K						
0.0000	0.9960	0.0000	1659.4	0.000	7.558	0.000
0.0997	0.9937	-0.0495	1642.0	-4.441	7.032	0.013
0.1749	0.9921	-0.0758	1628.7	-6.337	6.632	0.019
0.2629	0.9902	-0.0972	1613.9	-7.649	6.161	0.024

Table 5 continued

x_1	Density (ρ) ($\text{g}\cdot\text{cm}^{-3}$)	V^E ($\text{cm}^3\cdot\text{mol}^{-1}$)	U ($\text{m}\cdot\text{s}^{-1}$)	k_s^E (TPa^{-1})	Viscosity (η)/ mPa·s)	$\Delta\eta$ (mPa·s)
0.3629	0.9881	-0.1119	1598.5	-8.416	5.624	0.028
0.4797	0.9859	-0.1188	1582.2	-8.608	4.994	0.029
0.5707	0.9844	-0.1173	1570.9	-8.549	4.503	0.029
0.6849	0.9825	-0.1066	1557.4	-7.810	3.884	0.028
0.7826	0.9809	-0.0880	1546.2	-6.609	3.352	0.024
0.8935	0.9792	-0.0528	1533.1	-4.200	2.744	0.016
1.0000	0.9776 [29]	0.0000	1518.5 [29]	0.000	2.153 [30]	0.000

Standard uncertainty $u(x) = 1 \times 10^{-4}$, Combined expanded uncertainties $U_c(\rho) = 0.8 \times 10^{-3} \text{ g}\cdot\text{cm}^{-3}$, $U_c(u) = 0.5 \text{ m}\cdot\text{s}^{-1}$, and $u(\eta) = 0.2 \text{ mPa}\cdot\text{s}$

Fig. 1 Variation of excess molar volume (V^E) with mole fraction (x_1) of 2-methylaniline for the binary liquid mixtures of 2-methylaniline with 2-aminoethanol (filled triangle), 2-chloroethanol (filled circle), and 2-phenylethanol (filled square) at 303.15 K



In general, negative values of excess partial molar volume of component 1 and excess partial molar volumes of component 2 ($\bar{V}_{m,1}^E$, $\bar{V}_{m,2}^E$, $\bar{K}_{s,m,1}^E$ and $\bar{K}_{s,m,2}^E$) indicate the presence of significant solute–solvent interactions between unlike molecules, whereas a positive excess partial molar volume of component 1 and excess partial molar volume component 2 data indicate the presence of solute–solute/solvent–solvent interactions between like molecules in the mixtures [55, 56].

A close perusal of Supplementary Table 1S and Fig. 4 indicates that the values of $\bar{V}_{m,1}^E$ and $\bar{V}_{m,2}^E$ are negative for all the binary mixtures over the whole composition range.

Fig. 2 Excess isentropic compressibility (κ_s^E) with mole fraction (x_1) of 2-methoxyaniline in the binary liquid mixtures of 2-methylaniline with 2-aminoethanol (filled triangle), 2-chloroethanol (filled circle), and 2-phenylethanol (filled square) at 303.15 K

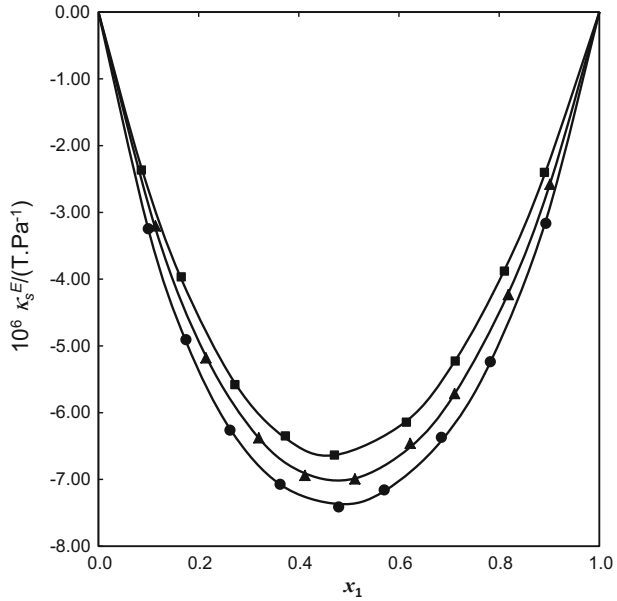
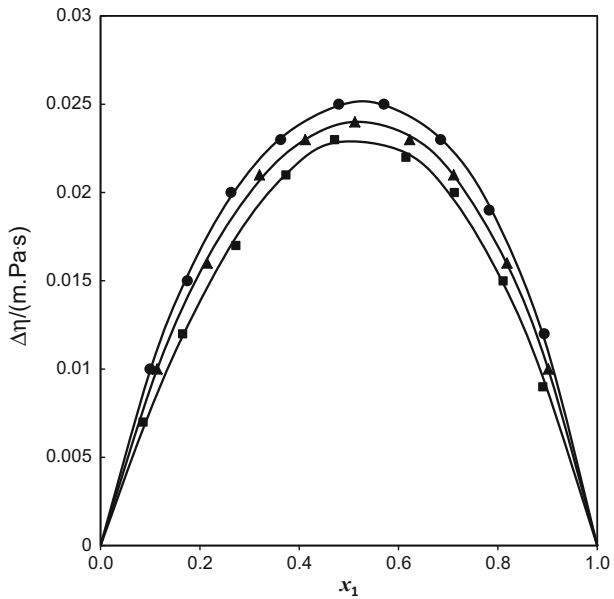


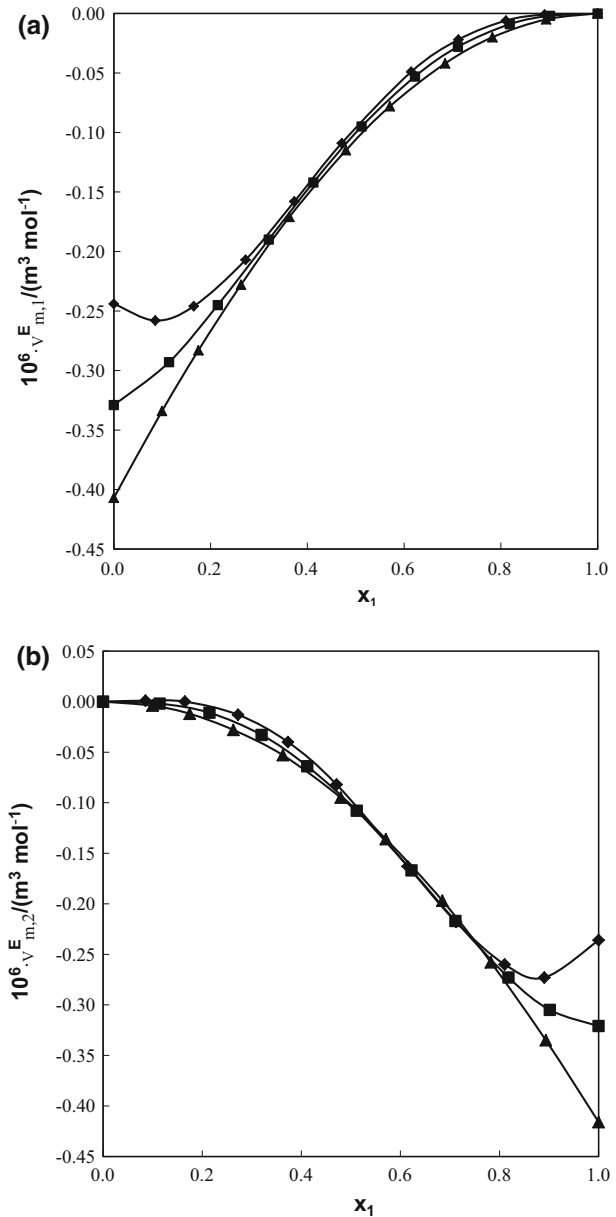
Fig. 3 Variation of the deviation in viscosity ($\Delta\eta$) from a linear mixing rule with mole fraction (x_1) of 2-methoxyaniline in the binary liquid mixtures of 2-methoxyaniline with 2-aminoethanol (filled triangle), 2-chloroethanol (filled circle), and 2-phenylethanol (filled square) at 303.15 K



Negative values may be attributed to hydrogen bonded complex formation between the components of the mixtures (Table 6).

From the Supplementary Table 2S and Fig. 5 it may be inferred that the values of $\overline{K}_{s,m,1}^E$ and $\overline{K}_{s,m,2}^E$ are negative for all the binary mixtures over the whole composition range. The

Fig. 4 Variation of excess partial molar volumes, $\bar{V}_{m,1}^E$ and $\bar{V}_{m,2}^E$, of 2-methoxyaniline with 2-aminoethanol (filled triangle), 2-chloroethanol (filled circle), and 2-phenylethanol (filled square) at 303.15 K



negative values indicate donor–acceptor interactions between the components of the mixtures.

From Table 7, it can be seen that the values of $\bar{V}_{m,1}^E$ and $\bar{V}_{m,2}^E$ are negative for all the binary mixtures over the whole composition range. The negative $\bar{V}_{m,1}^E$ and $\bar{V}_{m,2}^E$ values indicate strong specific interactions through a hetero association complex formation between 2-methoxyaniline and a substituted ethanol molecule.

Table 6 Coefficients of Redlich–Kister equation and standard deviation (σ) values for liquid mixtures of 2-methylaniline binary mixtures with substituted ethanols at $T = (303.15\text{--}18.15)$ K

T/K	A_0	A_1	A_2	σ
2-methylaniline + 2-phenylethanol				
V^E ($\text{cm}^3\cdot\text{mol}^{-1}$)				
303.15	– 0.385	– 0.004	0.145	0.001
308.15	– 0.407	0.001	0.126	0.001
313.15	– 0.423	– 0.005	0.088	0.001
318.15	– 0.438	– 0.006	0.035	0.001
κ_S^E (TPa^{-1})				
303.15	– 26.45	3.061	– 1.303	0.034
308.15	– 28.05	2.400	– 5.011	0.044
313.15	– 29.19	2.693	– 10.89	0.073
318.15	– 30.33	2.156	– 16.34	0.001
$\Delta\eta$ (mPa·s)				
303.15	0.092	0.006	– 0.001	0.001
308.15	0.097	0.005	0.008	0.001
313.15	0.099	0.00	0.026	0.001
318.15	0.104	0.007	0.039	0.001
2-methylaniline + 2-chloroethanol				
V^E ($\text{cm}^3\cdot\text{mol}^{-1}$)				
303.15	– 0.404	– 0.004	0.079	0.001
308.15	– 0.420	– 0.006	0.036	0.001
313.15	– 0.439	– 0.002	– 0.005	0.001
318.15	– 0.454	– 0.008	– 0.061	0.001
κ_S^E (TPa^{-1})				
303.15	– 28.03	1.973	– 3.99	0.039
308.15	– 29.55	1.715	– 8.25	0.028
313.15	– 31.03	1.902	– 11.80	0.039
318.15	– 32.43	2.284	– 16.21	0.031
$\Delta\eta$ (mPa·s)				
303.15	0.096	0.008	0.016	0.001
308.15	0.102	0.009	0.020	0.001
313.15	0.109	0.011	0.029	0.001
318.15	0.112	0.008	0.049	0.001
2-methylaniline + 2-aminoethanol				
V^E ($\text{cm}^3\cdot\text{mol}^{-1}$)				
303.15	– 0.419	0.005	0.007	0.001
308.15	– 0.439	– 0.001	– 0.031	0.001
313.15	– 0.459	– 0.004	– 0.064	0.001
318.15	– 0.476	– 0.004	– 0.122	0.001
κ_S^E (TPa^{-1})				
303.15	– 29.38	1.907	– 8.309	0.042
308.15	– 30.83	2.155	– 12.39	0.070
313.15	– 32.34	2.706	– 16.58	0.001
318.15	– 34.36	2.671	– 19.38	0.077

Table 6 continued

T/K	A_0	A_1	A_2	σ
$\Delta\eta$ (mPa·s)				
303.15	0.099	0.009	0.029	0.001
308.15	0.109	0.009	0.016	0.001
313.15	0.113	0.011	0.044	0.001
318.15	0.117	0.014	0.062	0.001

Fig. 5 Variation of excess partial molar isentropic compressibilities, $\bar{K}_{s,m,1}^E$ and $\bar{K}_{s,m,2}^E$, of 2-methoxyaniline with 2-aminoethanol (filled triangle), 2-chloroethanol (filled circle), and 2-phenylethanol (filled square) at 303.15 K

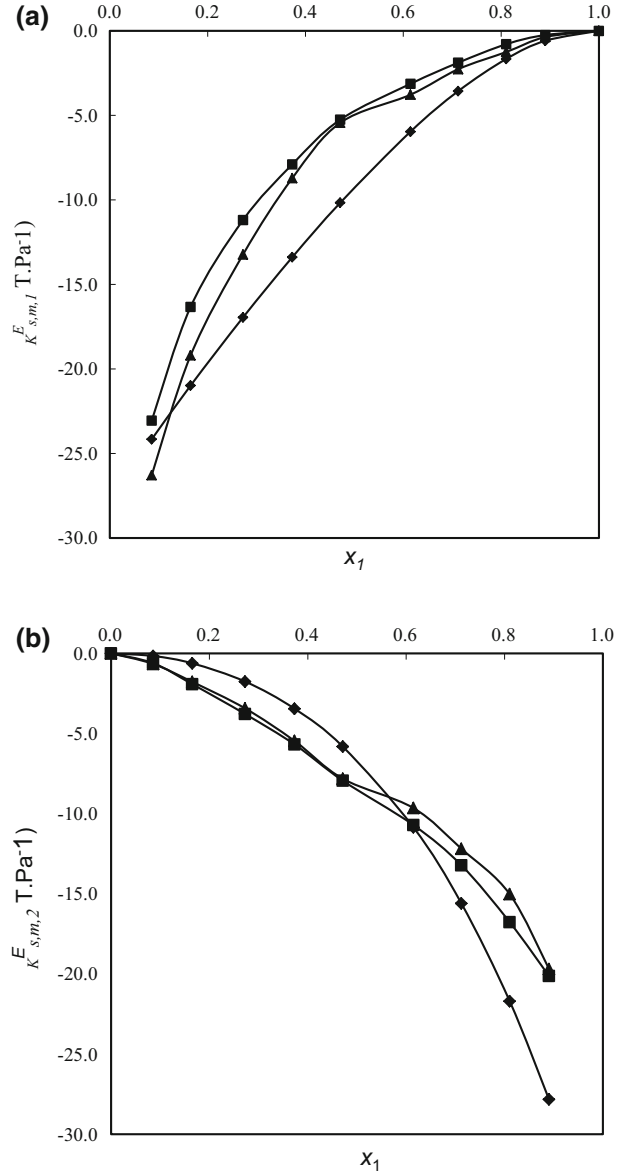


Table 7 The values of $V_{m,1}^\circ$, $V_{m,1}^*$, $V_{m,1}^{\circ E}$, $V_{m,2}^\circ$, $V_{m,2}^*$ and $V_{m,2}^{\circ E}$ of the components for 2-methylaniline binary mixtures with substituted ethanols at $T = (303.15\text{--}18.15)$ K

T/K ($\text{cm}^3 \cdot \text{mol}^{-1}$)	$\bar{V}_{m,1}^\circ$ ($\text{cm}^3 \cdot \text{mol}^{-1}$)	$V_{m,1}^*$ ($\text{cm}^3 \cdot \text{mol}^{-1}$)	$\bar{V}_{m,1}^{\circ E}$ ($\text{cm}^3 \cdot \text{mol}^{-1}$)	$\bar{V}_{m,2}^\circ$ ($\text{cm}^3 \cdot \text{mol}^{-1}$)	$V_{m,2}^*$ ($\text{cm}^3 \cdot \text{mol}^{-1}$)	$\bar{V}_{m,2}^{\circ E}$ ($\text{cm}^3 \cdot \text{mol}^{-1}$)
2-methylaniline(1) + 2-phenylethanol(2)						
303.15	107.99	108.23	− 0.244	120.43	120.67	− 0.236
308.15	108.41	108.69	− 0.280	120.85	121.13	− 0.282
313.15	108.83	109.17	− 0.339	121.25	121.58	− 0.330
318.15	109.22	109.63	− 0.410	121.65	122.05	− 0.398
2-methylaniline(1) + 2-chloroethanol(2)						
303.15	107.90	108.23	− 0.329	67.22	67.54	− 0.321
308.15	108.30	108.69	− 0.390	67.56	67.94	− 0.378
313.15	108.72	109.17	− 0.445	67.91	68.36	− 0.441
318.15	109.10	109.63	− 0.523	68.26	68.77	− 0.507
2-methylaniline(1) + 2-aminoethanol(2)						
303.15	107.82	108.23	− 0.407	60.14	60.55	− 0.416
308.15	108.22	108.69	− 0.471	60.34	60.81	− 0.469
313.15	108.64	109.17	− 0.528	60.54	61.06	− 0.520
318.15	109.02	109.63	− 0.602	60.73	61.32	− 0.594

Table 8 The values of $\bar{K}_{s,m,1}^\circ$, $K_{s,m,1}^*$, $\bar{K}_{s,m,1}^{\circ E}$, $\bar{K}_{s,m,2}^\circ$, $K_{s,m,2}^*$ and $\bar{K}_{s,m,2}^{\circ E}$ of the components for 2-methylaniline binary mixtures with substituted ethanols at $T = (303.15\text{--}18.15)$ K

T/K (TPa^{-1})	$\bar{K}_{s,m,1}^\circ$ (TPa^{-1})	$K_{s,m,1}^*$ (TPa^{-1})	$\bar{K}_{s,m,1}^{\circ E}$ (TPa^{-1})	$\bar{K}_{s,m,2}^\circ$ (TPa^{-1})	$K_{s,m,2}^*$ (TPa^{-1})	$\bar{K}_{s,m,2}^{\circ E}$ (TPa^{-1})
2-methylaniline(1) + 2-phenylethanol(2)						
303.15	− 23.40	4.387	− 27.79	− 32.98	5.231	− 38.21
308.15	− 30.12	4.540	− 34.66	− 38.67	5.399	− 44.07
313.15	− 38.04	4.698	− 42.74	− 47.73	5.564	− 53.30
318.15	− 46.51	4.863	− 51.37	− 55.41	5.738	− 61.15
2-methylaniline(1) + 2-chloroethanol(2)						
303.15	− 29.13	4.387	− 33.52	− 21.89	3.142	− 25.04
308.15	− 35.67	4.540	− 40.21	− 26.72	3.246	− 29.97
313.15	− 41.22	4.698	− 45.92	− 31.61	3.354	− 34.96
318.15	− 47.64	4.863	− 52.50	− 37.59	3.469	− 41.06
2-methylaniline(1) + 2-aminoethanol(2)						
303.15	− 35.05	4.387	− 39.43	− 24.54	2.065	− 26.61
308.15	− 40.52	4.540	− 45.06	− 28.78	2.120	− 30.90
313.15	− 45.88	4.698	− 50.58	− 33.37	2.175	− 35.54
318.15	− 51.15	4.863	− 56.02	− 36.98	2.236	− 39.22

It is seen from Table 8 that the values of $\bar{K}_{s,m,1}^{\circ E}$ and $\bar{K}_{s,m,2}^{\circ E}$ are negative for all the binary systems at each investigated temperature. The negative values may be due to the strong chemical and specific interactions through charge transfer complex formation between the components of the binary mixtures.

5 Conclusion

This paper reports experimental data of densities, speeds of sound and viscosities of binary blends of 2-methylaniline with substituted ethanol (2-phenylethanol, 2-chloroethanol and 2-aminoethanol) binary mixtures over the entire composition range at $T = (303.15\text{--}318.15)$ K with 5 K interval. From the experimental data, various physicochemical parameters, viz., V_m^E , κ_S^E and $\Delta\eta$ of the mixtures, the excess partial molar properties ($\bar{V}_{m,1}^E$, $\bar{V}_{m,2}^E$, $\bar{K}_{s,m,1}^E$ and $\bar{K}_{s,m,2}^E$) and excess partial molar properties at infinite dilution ($\bar{V}_{m,1}^{\circ E}$, $\bar{V}_{m,2}^{\circ E}$, $\bar{K}_{s,m,1}^{\circ E}$ and $\bar{K}_{s,m,2}^{\circ E}$) of the components have been calculated.

References

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