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Densities, Viscosities and Speeds of Sound of Binary Mixtures of 2-Chloroaniline with *o*-Chlorotoluene, *m*-Chlorotoluene and *p*-Chlorotoluene at Different Temperatures

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Abstract Densities (ρ), speeds of sound (u) and viscosities (η) are reported for binary mixtures of 2-chloroaniline (CA) with chlorotoluenes [*o*-chlorotoluene (*o*-CT), *m*-chlorotoluene (*m*-CT), and *p*-chlorotoluene (*p*-CT)] over the entire range of mole fraction at 303.15, 308.15, 313.15 and 318.15 K and atmospheric pressure. By using this data, the excess molar volumes, excess isentropic compressibilities, and deviation in viscosity for the binary systems were calculated and fitted to the Redlich–Kister equation to determine the fitting parameters and the root-mean-square deviations. The excess molar volumes, excess isentropic compressibilities, deviations in viscosity and excess Gibbs energy of activation of viscous flow have been analyzed in terms of charge-transfer complexes, and dipole–dipole interactions between unlike molecules in the mixtures. The viscosity data have been correlated using three equations: the Grunberg–Nissan, Katti–Chaudhri and Hind et al.

Keywords Viscosity · Density · Charge-transfer complexes · 2-Chloroaniline · Chlorotoluenes

1 Introduction

The thermodynamic and transport properties of liquid mixtures have attracted much attention from the point of view of both theoretical and engineering applications. Many engineering applications require data on the density, speed of sound and viscosity of liquid

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mixtures. They also provide information about the nature and molecular interactions between liquid mixture components.

The liquids were chosen in the present study on the basis of their industrial importance. 2-Chloroaniline is used as parent substance in the production of antioxidants, agricultural, pharmaceutical and rubber chemicals. It is also used in manufacture of intermediates for synthetic dyes, and organic pigments, especially for red color. Chlorotoluenes are used as intermediates in the pesticide, pharmaceutical and dye industries. A fundamental understanding of the mixture behavior of 2-chloroaniline with chlorotoluenes is therefore important from the technical and engineering standpoint.

The present investigation is a continuation of our earlier research [1–6] on thermodynamic properties of binary liquid mixtures. In this paper we report measurements of densities, speeds of sound and viscosities for three binary systems, 2-chloroaniline + *o*-chlorotoluene, 2-chloroaniline + *m*-chlorotoluene and 2-chloroaniline + *p*-chlorotoluene at 303.15, 308.15, 313.15 and 318.15 K and atmospheric pressure. The aim of this work is to investigate the influence on both the sign and magnitude of excess/deviation properties by the introduction of a chlorogroup into the toluene molecule in mixtures with 2-chloroaniline.

Several researchers investigated density, speed of sound, and viscosity of binary mixtures of dimethylformamide with chlorotoluenes [7], tetrahydrofuran with chlorotoluenes [8], dimethylsulfoxide with chlorotoluenes [9], and benzyl alcohol with chlorotoluenes [10]. However, no attempt has been made to measure excess/deviation properties of binary mixtures of 2-chloroaniline with chlorotoluenes. We report here excess volume (V^{E}), excess isentropic compressibility (κ_S^{E}), deviation in viscosity ($\Delta\eta$) and excess Gibbs energy of activation of viscous flow ($G^{*\mathrm{E}}$) for the above said binary systems. The variations of these properties with composition of the binary mixtures are discussed in terms of molecular interactions between components and structural effects.

2 Experimental

The mass fraction purity of the liquids obtained from Merck, and S.D. Fine Chemicals Ltd., India are as follows: 2-chloroaniline (0.995), *o*-chlorotoluene (0.995), *m*-chlorotoluene (0.995), and *p*-chlorotoluene (0.996). Prior to experimental measurements, all the liquids were used after double distillations and partially degassed with a vacuum pump under an inert atmosphere. The purity of these solvents was ascertained by comparing the measured density, speed of sound, and viscosity of the pure components with available literature values [11–16] as shown in Table 1. The binary mixtures of 2-chloroaniline with *o*-chlorotoluene, *m*-chlorotoluene, and *p*-chlorotoluene were prepared in glass bottles with air-tight stoppers, and adequate precautions were taken to minimize losses through evaporation. The weighing of solutions was made using Afoset ER-120A electronic balance with a precision of ±0.1 mg. The uncertainty in solution composition expressed in mole fraction was found to be less than 1×10^{-4}. After mixing, 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 built-in solid-state thermostat and a resident program with giving temperature control to ± 0.03 K. The uncertainty in the density measurements was found to be less than $\pm 4 \times 10^{-5}$ g·cm^{-3}. Proper calibration at each temperature was achieved with doubly distilled, deionized water and air as standards. A

Table 1 Comparison of experimental and literature values of density (ρ), and viscosity (η) data of pure components at $T = 303.15$ K

Pure components	Density (ρ) (g·cm^{-3})		Viscosity (η) (mPa·s)		Speed of sound (u) (m·s^{-1})	Literature	Cp (J·K^{-1}·mol^{-1})
	Experimental	Literature	Experimental	Literature	Experimental		
2-Chloroaniline	1.20269	1.20270 [11]	3.8256	3.8256 [11]	1469.6	1471 [12]	208.0 [15]
o-Chlorotoluene	1.07284	1.07283 [13]	0.886	0.884 [16]	1285.0	1284 [16]	179.59 [14]
m-Chlorotoluene	1.06283	1.06281 [13]	0.743	0.746 [13]	1281.1	1280 [16]	173.07 [14]
p-chlorotoluene	1.05954	1.05951 [13]	0.782	0.782 [13]	1172.6	1271 [16]	172.74 [14]

multi–frequency ultrasonic interferometer (M-82 Model, Mittal Enterprise, New Delhi, India) operated at 2 MHz was used to measure the ultrasonic velocities in the binary liquid mixtures; the temperature was controlled by a digital, constant temperature water bath. The uncertainty in the measurement of speed sound is ±0.2 %.

The viscosities of the pure liquids and their mixtures were determined at atmospheric pressure, at 303.15, 308.15, 313.15 and 318.15 K, using an Ubbelohde viscometer that was calibrated with benzene and doubly distilled water. The Ubbelohde viscometer bulb has a capacity of 15 mL and the capillary tube with a length of about 90 mm with 0.5 mm internal diameter. The viscometer was thoroughly cleaned and dried, and was filled with the sample liquid 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. An electronic digital stopwatch with an uncertainty ±0.01 s was used for flow time measurements. The viscosity values of pure liquids and mixtures are calculated using the relation:

$$\eta = (at - b/t)\rho \tag{1}$$

where, a and b are the characteristic constants of the viscometer, ρ is the density, and t represents the flow time. The uncertainty of viscosity thus estimated was found to be ±0.005 mPa·s.

3 Calculations

From these experimental values of densities (ρ) viscosity (η) and sound speed (u) data were used to calculate excess molar volumes (V^{E}), excess isentropic compressibility (κ_S^{E}) deviation in viscosity ($\Delta\eta$) and excess Gibbs energy of activation of viscous flow ($G^{*\mathrm{E}}$) for binary mixtures of 2-chloroaniline with chlorotoluene at 303.15, 308.15, 313.15, 318.15 K are shown in Table 2 and also excess properties are graphically represented in Figs. 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, and 16, respectively V^{E}, $\Delta\eta$, $G^{*\mathrm{E}}$ and κ_S^{E} are calculated from the experimental measurements by Eqs. 2–9:

$$V^{\mathrm{E}} = \frac{x_1M_1 + x_2M_2}{\rho} - \left(\frac{x_1M_1}{\rho_1} + \frac{x_2M_2}{\rho_2}\right) \tag{2}$$

$$\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2) \tag{3}$$

$$G^{*\mathrm{E}} = RT(\ln \eta V - (x_1 \ln \eta_1 V_1 + x_2 \ln \eta_2 V_2)) \tag{4}$$

where for each equation, ρ, V, and η are the density, the molar volume and the dynamic viscosity of the mixtures and x_i, V_i, M_i, and η_i ($i = 1, 2$) are the mole fraction, molar volume, the molar mass and the dynamic viscosity of the components 2-chloroaniline (1) with chlorotoluene (2), respectively. R is the gas constant and T the absolute temperature

$$\kappa_S^{\mathrm{E}} = \kappa_S - \kappa_S^{\mathrm{id}} \tag{5}$$

$$\kappa_S = \frac{1}{u^2\rho} \tag{6}$$

where κ_S^{id} is the ideal value of the isentropic compressibility and was calculated from the following equation [17]:

Table 2 Mole fraction of 2-chloroaniline (x_1), density (ρ), excess volumes (V^E),viscosity (η), excess viscosity ($\Delta\eta$), excess Gibbs energy of activation of viscous flow (G^{*E}), Grunberg–Nissan interaction parameters (d_{12}), Katti–Chaudhri interaction parameters (W_{vis}/RT), and Hind interaction parameters (H_{12}), Tamura and Kurata (T_{12}) and Heric–Brewer (Δ_{12}) for binary mixtures at 303.15, 308.15, 313.15, and 318.15 K

x_1	ρ (g·cm^{-3})	V^E (cm^3·mol^{-1})	η (mPa·s)	$\Delta\eta$ (mPa·s)	G^{*E} (kJ·mol^{-1})	W_{vis}/RT	d_{12}	H_{12}	T_{12}	Δ_{12}
2-Chloroaniline + *o*-chlorotoluene										
303.15 K										
0.0000	1.07284	0.0000	0.8860	0.000	0.000					
0.1100	1.08631	−0.0519	1.0729	0.032	0.761	0.312	0.311	1.659	1.710	3.006
0.2176	1.09970	−0.0931	1.2843	0.066	1.322	0.312	0.311	1.647	1.706	1.673
0.3228	1.11300	−0.1236	1.5208	0.100	1.697	0.312	0.311	1.637	1.704	1.229
0.4258	1.12619	−0.1412	1.7823	0.131	1.897	0.312	0.311	1.629	1.704	1.007
0.5266	1.13927	−0.1473	2.0685	0.154	1.933	0.312	0.311	1.623	1.705	0.874
0.6253	1.15220	−0.1396	2.3786	0.167	1.817	0.312	0.311	1.619	1.709	0.785
0.7219	1.16502	−0.1205	2.7112	0.164	1.556	0.311	0.311	1.616	1.715	0.722
0.8165	1.17769	−0.0892	3.0646	0.140	1.162	0.311	0.311	1.616	1.722	0.674
0.9092	1.19026	−0.0500	3.4369	0.087	0.640	0.311	0.311	1.619	1.731	0.637
1.0000	1.20269	0.0000	3.8256	0.000	0.000					
308.15 K										
0.0000	1.06822	0.0000	0.8080	0.000	0.000					
0.1100	1.08183	−0.0691	0.9808	0.034	0.893	0.360	0.361	1.530	1.577	3.055
0.2176	1.09521	−0.1103	1.1755	0.070	1.554	0.361	0.361	1.525	1.579	1.723
0.3228	1.10848	−0.1374	1.3920	0.106	1.997	0.361	0.361	1.523	1.584	1.279
0.4258	1.12164	−0.1529	1.6298	0.138	2.233	0.361	0.361	1.523	1.590	1.057
0.5266	1.13469	−0.1563	1.8877	0.162	2.277	0.361	0.361	1.525	1.599	0.924
0.6253	1.14765	−0.1513	2.1643	0.175	2.139	0.361	0.361	1.530	1.609	0.835
0.7219	1.16050	−0.1357	2.4576	0.172	1.831	0.361	0.361	1.536	1.622	0.771
0.8165	1.17321	−0.1078	2.7651	0.146	1.365	0.360	0.361	1.545	1.636	0.724
0.9092	1.18576	−0.0659	3.0843	0.091	0.751	0.359	0.361	1.557	1.653	0.687

Table 2 continued

x_1	ρ (g·cm^{-3})	V^E (cm^3·mol^{-1})	η (mPa·s)	$\Delta\eta$ (mPa·s)	G^{*E} (kJ·mol^{-1})	W_{vis}/RT	d_{12}	H_{12}	T_{12}	Δ_{12}
1.0000	1.19802	0.0000	3.4122	0.000	0.000					
313.15 K										
0.0000	1.06334	0.0000	0.7280	0.006	0.000					
0.1100	1.07701	−0.0774	0.8896	0.037	1.089	0.433	0.434	1.409	1.453	3.128
0.2176	1.09041	−0.1225	1.0700	0.075	1.866	0.426	0.427	1.408	1.458	1.789
0.3228	1.10364	−0.1479	1.2713	0.114	2.414	0.430	0.430	1.415	1.470	1.348
0.4258	1.11677	−0.1611	1.4898	0.148	2.690	0.428	0.428	1.421	1.482	1.124
0.5266	1.12982	−0.1659	1.7257	0.175	2.748	0.429	0.429	1.432	1.498	0.992
0.6253	1.14277	−0.1612	1.9757	0.189	2.583	0.429	0.429	1.445	1.516	0.903
0.7219	1.15564	−0.1472	2.2376	0.185	2.215	0.429	0.43	1.461	1.536	0.840
0.8165	1.16837	−0.1221	2.5073	0.156	1.645	0.427	0.429	1.477	1.556	0.792
0.9092	1.18086	−0.0746	2.7831	0.097	0.907	0.427	0.43	1.498	1.581	0.756
1.0000	1.19302	0.0000	3.0602	0.006	0.000					
318.15 K										
0.0000	1.05896	0.0000	0.6420	0.000	0.000					
0.1100	1.07272	−0.0903	0.7935	0.040	1.314	0.522	0.524	1.285	1.326	3.219
0.2176	1.08611	−0.1366	0.9642	0.082	2.290	0.523	0.524	1.294	1.340	1.886
0.3228	1.09930	−0.1587	1.1532	0.125	2.944	0.524	0.524	1.306	1.357	1.442
0.4258	1.11240	−0.1713	1.3585	0.163	3.294	0.524	0.524	1.321	1.377	1.220
0.5266	1.12542	−0.1745	1.5777	0.193	3.358	0.524	0.524	1.339	1.400	1.087
0.6253	1.13837	−0.1700	1.8077	0.209	3.155	0.524	0.524	1.361	1.424	0.998
0.7219	1.15124	−0.1576	2.0451	0.204	2.700	0.523	0.524	1.385	1.452	0.935
0.8165	1.16396	−0.1314	2.2860	0.173	2.012	0.522	0.524	1.411	1.481	0.887
0.9092	1.17646	−0.0861	2.5263	0.107	1.105	0.521	0.524	1.440	1.511	0.850
1.0000	1.18849	0.0000	2.7621	0.000	0.000					

Table 2 continued

x_1	ρ (g·cm^{-3})	V^E (cm^3·mol^{-1})	η (mPa·s)	$\Delta\eta$ (mPa·s)	G^{*E} (kJ·mol^{-1})	W_{vis}/RT	d_{12}	H_{12}	T_{12}	Δ_{12}
2-Chloroaniline + *m*-chlorotoluene										
303.15 K										
0.0000	1.06283	0.0000	0.7430	0.000	0.000					
0.1012	1.07611	−0.0581	0.9087	0.032	0.886	0.391	0.39	1.480	1.535	5.873
0.2056	1.08996	−0.0992	1.1091	0.068	1.591	0.391	0.39	1.465	1.529	3.213
0.3125	1.10437	−0.1292	1.3483	0.108	2.093	0.391	0.39	1.451	1.525	2.356
0.4056	1.11712	−0.1447	1.5867	0.142	2.349	0.391	0.39	1.441	1.524	2.003
0.5026	1.13060	−0.1517	1.8666	0.173	2.435	0.391	0.39	1.433	1.525	1.805
0.6025	1.14470	−0.1482	2.1896	0.195	2.332	0.391	0.39	1.427	1.529	1.717
0.7145	1.16076	−0.1312	2.5946	0.198	1.986	0.391	0.39	1.424	1.537	1.750
0.8180	1.17581	−0.0996	3.0086	0.170	1.448	0.391	0.39	1.425	1.549	2.002
0.9100	1.18937	−0.0592	3.4081	0.107	0.796	0.390	0.39	1.430	1.562	2.914
1.0000	1.20269	0.0000	3.8256	0.000	0.000					
308.15 K										
0.0000	1.05835	0.0000	0.7010	0.000	0.000					
0.1012	1.07171	−0.0703	0.8581	0.035	1.063	0.462	0.462	1.412	1.463	5.945
0.2056	1.08557	−0.1152	1.0466	0.076	1.911	0.462	0.462	1.408	1.467	3.285
0.3125	1.09996	−0.1439	1.2694	0.120	2.515	0.463	0.462	1.408	1.475	2.428
0.4056	1.11268	−0.1585	1.4889	0.157	2.823	0.463	0.462	1.410	1.484	2.075
0.5026	1.12612	−0.1635	1.7431	0.190	2.927	0.463	0.462	1.415	1.497	1.877
0.6025	1.14020	−0.1599	2.0317	0.213	2.804	0.463	0.462	1.425	1.513	1.789
0.7145	1.15628	−0.1456	2.3863	0.215	2.386	0.462	0.462	1.439	1.536	1.821
0.8180	1.17131	−0.1140	2.7402	0.182	1.740	0.462	0.462	1.457	1.561	2.074
0.9100	1.18485	−0.0728	3.0733	0.114	0.955	0.461	0.462	1.477	1.586	2.986
1.0000	1.19802	0.0000	3.4122	0.000	0.000					

Table 2 continued

x_1	ρ (g·cm^{-3})	V^E (cm^3·mol^{-1})	η (mPa·s)	$\Delta\eta$ (mPa·s)	G^{*E} (kJ·mol^{-1})	W_{vis}/RT	d_{12}	H_{12}	T_{12}	Δ_{12}
313.15 K										
0.0000	1.05314	0.0000	0.6630	0.000	0.000					
0.1012	1.06662	−0.0828	0.8120	0.038	1.230	0.05	0.527	1.347	1.395	6.010
0.2056	1.08050	−0.1286	0.9900	0.082	2.223	0.05	0.529	1.354	1.409	3.352
0.3125	1.09489	−0.1563	1.1993	0.130	2.952	0.05	0.534	1.366	1.428	2.500
0.4056	1.10761	−0.1696	1.4019	0.169	3.307	0.05	0.533	1.378	1.445	2.146
0.5026	1.12107	−0.1736	1.6341	0.204	3.433	0.05	0.533	1.394	1.467	1.949
0.6025	1.13519	−0.1719	1.8932	0.227	3.287	0.05	0.533	1.414	1.492	1.860
0.7145	1.15132	−0.1596	2.2045	0.227	2.793	0.05	0.533	1.442	1.525	1.892
0.8180	1.16641	−0.1297	2.5075	0.191	2.033	0.05	0.532	1.471	1.558	2.144
0.9100	1.17990	−0.0805	2.7856	0.119	1.118	0.05	0.533	1.503	1.593	3.057
1.0000	1.19302	0.0000	3.0602	0.000	0.000					
318.15 K										
0.0000	1.04856	0.0000	0.6240	0.000	0.000					
0.1012	1.06213	−0.0943	0.7685	0.043	1.503	0.633	0.635	1.298	1.344	6.118
0.2056	1.07606	−0.1451	0.9398	0.093	2.704	0.634	0.635	1.314	1.366	3.457
0.3125	1.09041	−0.1687	1.1384	0.145	3.561	0.635	0.635	1.335	1.392	2.601
0.4056	1.10311	−0.1792	1.3294	0.189	3.998	0.635	0.635	1.358	1.419	2.247
0.5026	1.11658	−0.1839	1.5445	0.227	4.146	0.635	0.635	1.385	1.450	2.050
0.6025	1.13071	−0.1822	1.7801	0.251	3.971	0.635	0.635	1.417	1.486	1.961
0.7145	1.14683	−0.1679	2.0559	0.250	3.379	0.634	0.635	1.458	1.53	1.994
0.8180	1.16196	−0.1406	2.3156	0.209	2.462	0.633	0.635	1.501	1.573	2.246
0.9100	1.17548	−0.0924	2.5448	0.129	1.351	0.631	0.635	1.542	1.614	3.159
1.0000	1.18849	0.0000	2.7621	0.000	0.000					

Table 2 continued

x_1	ρ (g·cm^{-3})	V^E (cm^3·mol^{-1})	η (mPa·s)	$\Delta\eta$ (mPa·s)	G^{*E} (kJ·mol^{-1})	W_{vis}/RT	d_{12}	H_{12}	T_{12}	Δ_{12}
2-Chloroaniline + *p*-chlorotoluene										
303.15 K										
0.0000	1.05954	0.0000	0.782	0.000	0.000					
0.1012	1.07314	−0.0655	0.9617	0.043	1.150	0.508	0.507	1.598	1.658	2.246
0.2102	1.08795	−0.1125	1.1753	0.083	1.838	0.445	0.444	1.561	1.629	1.281
0.3125	1.10200	−0.1367	1.405	0.121	2.242	0.419	0.418	1.540	1.616	0.981
0.4056	1.11505	−0.1536	1.6441	0.155	2.476	0.413	0.411	1.531	1.616	0.845
0.5142	1.13050	−0.1607	1.9625	0.193	2.591	0.417	0.415	1.534	1.629	0.758
0.6125	1.14473	−0.1556	2.2873	0.219	2.518	0.426	0.425	1.548	1.651	0.712
0.6952	1.15687	−0.1430	2.5858	0.228	2.301	0.436	0.435	1.567	1.678	0.688
0.7859	1.17036	−0.1195	2.936	0.213	1.875	0.448	0.447	1.597	1.714	0.671
0.8859	1.18541	−0.0776	3.3438	0.152	1.158	0.460	0.460	1.638	1.762	0.659
1.0000	1.20269	0.0000	3.8256	0.000	0.000					
308.15 K										
0.0000	1.05394	0.0000	0.7280	0.000	0.000					
0.1012	1.06773	−0.0798	0.8921	0.041	1.188	0.516	0.517	1.479	1.534	2.255
0.2102	1.08263	−0.1288	1.0975	0.090	2.171	0.517	0.517	1.484	1.547	1.353
0.3125	1.09677	−0.1530	1.3182	0.138	2.813	0.518	0.517	1.492	1.562	1.079
0.4056	1.10986	−0.1665	1.5429	0.181	3.157	0.518	0.517	1.502	1.579	0.950
0.5142	1.12539	−0.1704	1.8329	0.222	3.272	0.518	0.517	1.519	1.604	0.859
0.6125	1.13972	−0.1663	2.1198	0.245	3.108	0.518	0.517	1.539	1.630	0.804
0.6952	1.15199	−0.1576	2.3773	0.246	2.772	0.517	0.517	1.559	1.654	0.770
0.7859	1.16558	−0.1337	2.6739	0.223	2.199	0.517	0.517	1.584	1.684	0.740
0.8859	1.18076	−0.0928	3.0141	0.153	1.318	0.515	0.517	1.616	1.721	0.715
1.0000	1.19802	0.0000	3.4122	0.000	0.000					

Table 2 continued

x_1	ρ (g·cm^{-3})	V^E (cm^3·mol^{-1})	η (mPa·s)	$\Delta\eta$ (mPa·s)	G^{*E} (kJ·mol^{-1})	W_{vis}/RT	d_{12}	H_{12}	T_{12}	Δ_{12}
313.15 K										
0.0000	1.05034	0.0000	0.6820	0.000	0.000					
0.1012	1.06410	−0.0897	0.8377	0.044	1.379	0.590	0.591	1.404	1.456	2.329
0.2102	1.07889	−0.1410	1.0314	0.096	2.522	0.591	0.591	1.418	1.476	1.428
0.3125	1.09291	−0.1667	1.2379	0.148	3.267	0.591	0.591	1.435	1.499	1.154
0.4056	1.10586	−0.1779	1.4458	0.192	3.668	0.592	0.591	1.455	1.524	1.025
0.5142	1.12125	−0.1835	1.7106	0.235	3.800	0.592	0.591	1.482	1.557	0.933
0.6125	1.13545	−0.1803	1.9681	0.258	3.609	0.592	0.591	1.512	1.590	0.878
0.6952	1.14758	−0.1692	2.1949	0.258	3.220	0.591	0.591	1.540	1.622	0.844
0.7859	1.16109	−0.1502	2.4511	0.232	2.553	0.590	0.591	1.574	1.658	0.815
0.8859	1.17606	−0.1042	2.7372	0.159	1.531	0.589	0.591	1.616	1.702	0.790
1.0000	1.19302	0.0000	3.0602	0.000	0.000					
318.15 K										
0.0000	1.04568	0.0000	0.6320	0.000	0.000					
0.1012	1.05954	−0.1008	0.7836	0.050	1.685	0.721	0.723	1.345	1.395	2.461
0.2102	1.07435	−0.1548	0.9716	0.110	3.082	0.722	0.723	1.371	1.426	1.560
0.3125	1.08839	−0.1812	1.1704	0.168	3.992	0.723	0.723	1.401	1.461	1.286
0.4056	1.10132	−0.1900	1.3684	0.219	4.483	0.723	0.723	1.432	1.496	1.157
0.5142	1.11671	−0.1928	1.6162	0.267	4.645	0.723	0.723	1.475	1.542	1.065
0.6125	1.13095	−0.1916	1.8516	0.292	4.412	0.723	0.723	1.518	1.587	1.010
0.6952	1.14312	−0.1835	2.0537	0.292	3.936	0.722	0.723	1.557	1.628	0.976
0.7859	1.15664	−0.1637	2.2747	0.261	3.121	0.721	0.723	1.604	1.674	0.947
0.8859	1.17161	−0.1145	2.5112	0.177	1.871	0.720	0.723	1.658	1.727	0.921
1.0000	1.18849	0.0000	2.7621	0.000	0.000					

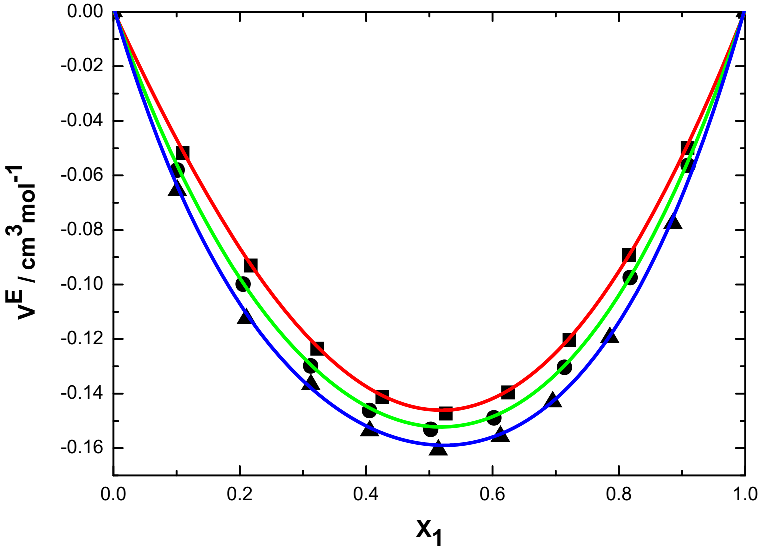

Fig. 1 Curves of excess molar volume (V^E) versus mole fraction for the binary mixtures of 2-chloroaniline + *o*-chlorotoluene (*square*), *m*-chlorotoluene (*circle*), *p*-chlorotoluene (*triangle*) at 303.15 K

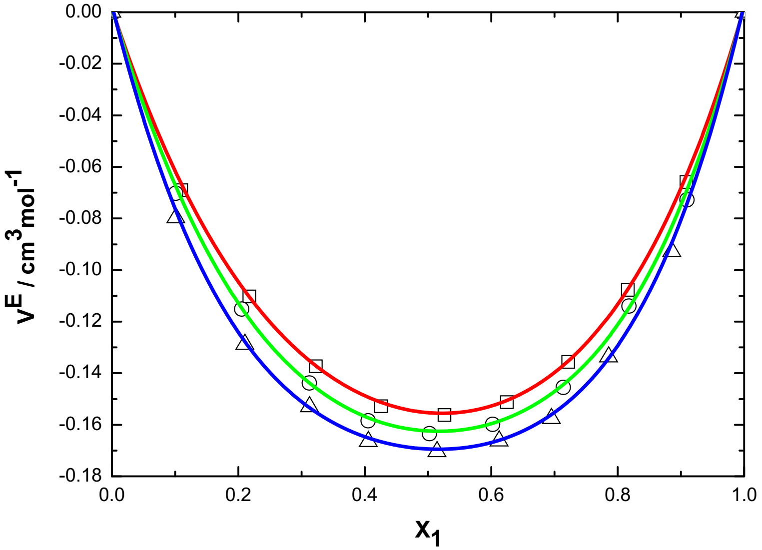

Fig. 2 Curves of excess molar volume (V^E) versus mole fraction for the binary mixtures of 2-chloroaniline + *o*-chlorotoluene (*square*), *m*-chlorotoluene (*circle*), *p*-chlorotoluene (*triangle*) at 308.15 K

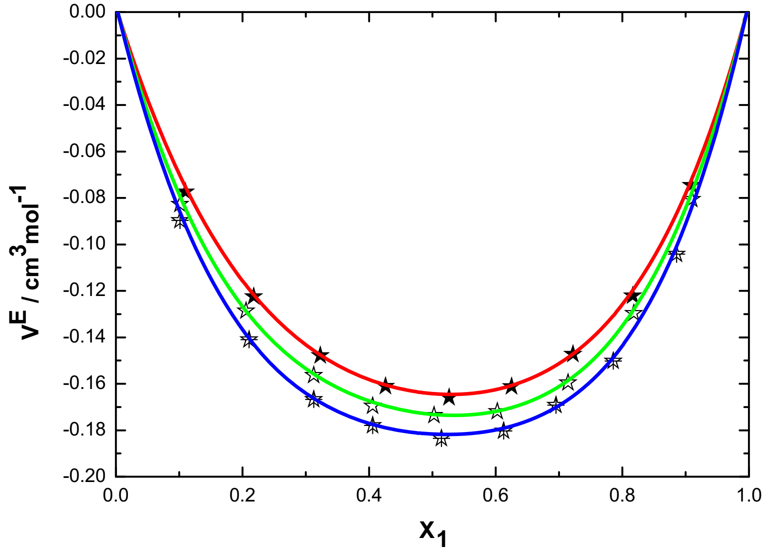

Fig. 3 Curves of excess molar volume (V^E) versus mole fraction for the binary mixtures of 2-chloroaniline + *o*-chlorotoluene (*black star*), *m*-chlorotoluene (*white star*), *p*-chlorotoluene (*crossed star*) at 313.15 K

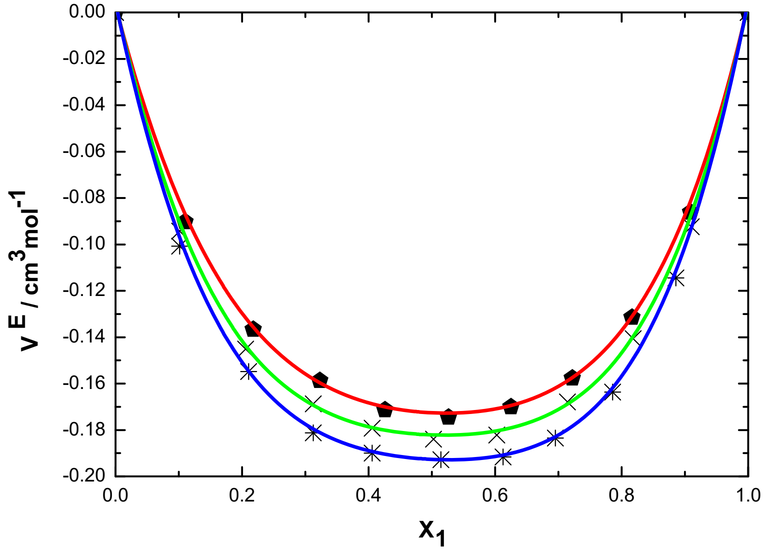

Fig. 4 Curves of excess molar volume (V^E) versus mole fraction for the binary mixtures of 2-chloroaniline + *o*-chlorotoluene (*pentagon*), *m*-chlorotoluene (*cross symbol*), *p*-chlorotoluene (*cross with plus symbol*) at 318.15 K

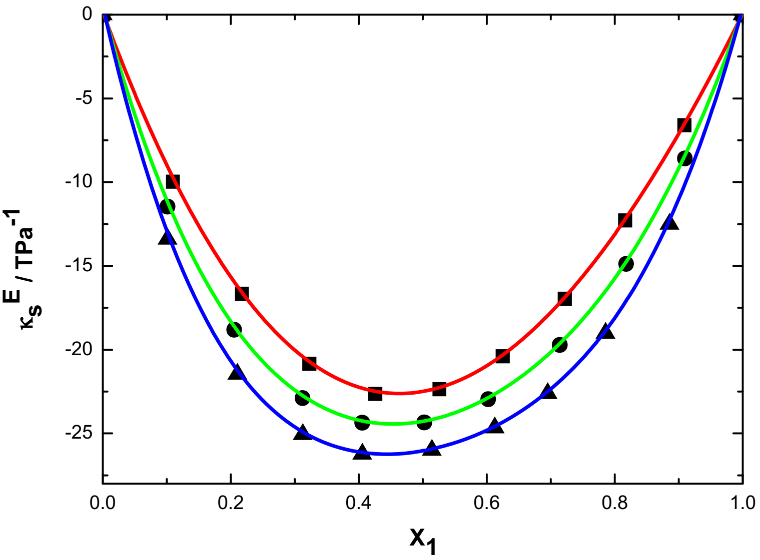

Fig. 5 Curves of excess isentropic compressibility (κ_S^E) versus mole fraction for the binary mixtures of 2-chloroaniline + *o*-chlorotoluene (*square*), *m*-chlorotoluene (*circle*), *p*-chlorotoluene (*triangle*) at 303.15 K

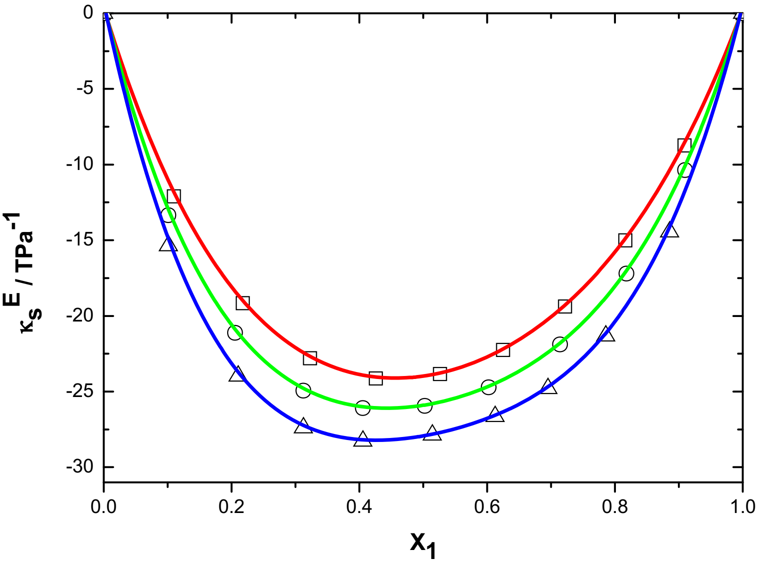

Fig. 6 Curves of excess isentropic compressibility (κ_S^E) versus mole fraction for the binary mixtures of 2-chloroaniline + *o*-chlorotoluene (*square*), *m*-chlorotoluene (*circle*), *p*-chlorotoluene (*triangle*) at 308.15 K

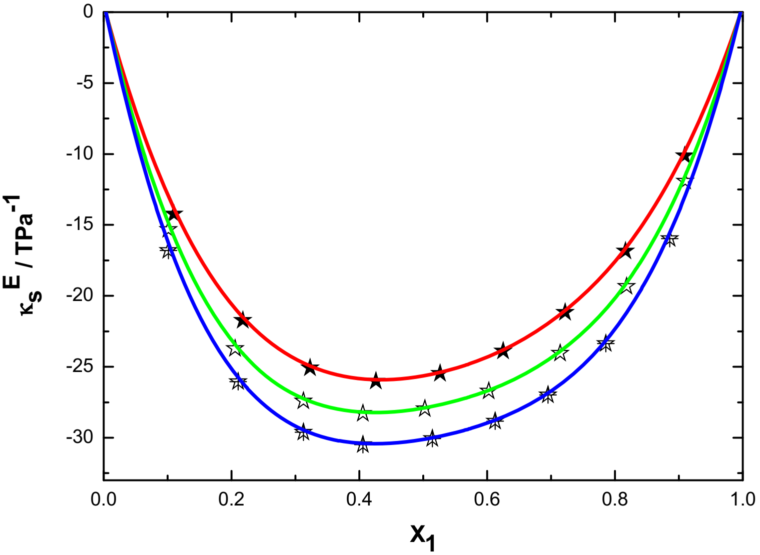

Fig. 7 Curves of excess isentropic compressibility (κ_S^E) versus mole fraction for the binary mixtures of 2-chloroaniline + *o*-chlorotoluene (*black star*), *m*-chlorotoluene (*white star*), *p*-chlorotoluene (*crossed star*) at 313.15 K

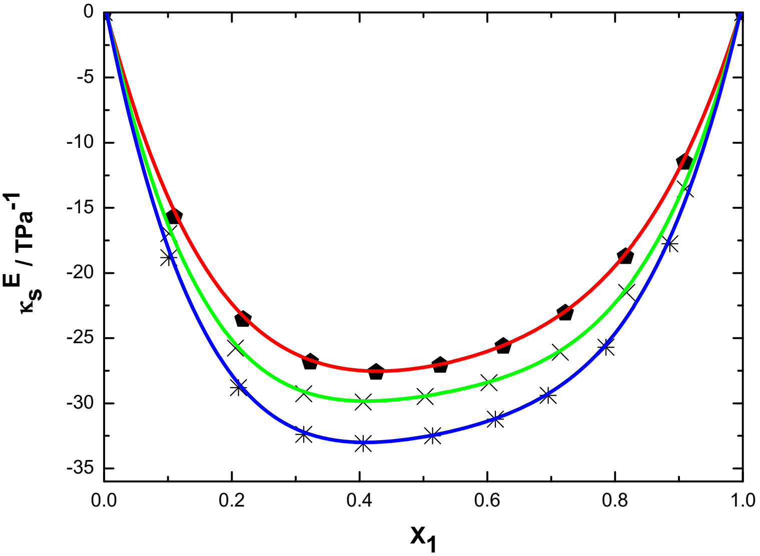

Fig. 8 Curves of excess isentropic compressibility (κ_S^E) versus mole fraction for the binary mixtures of 2-chloroaniline + *o*-chlorotoluene (*pentagon*), *m*-chlorotoluene (*cross symbol*), *p*-chlorotoluene (*cross with plus symbol*) at 318.15 K

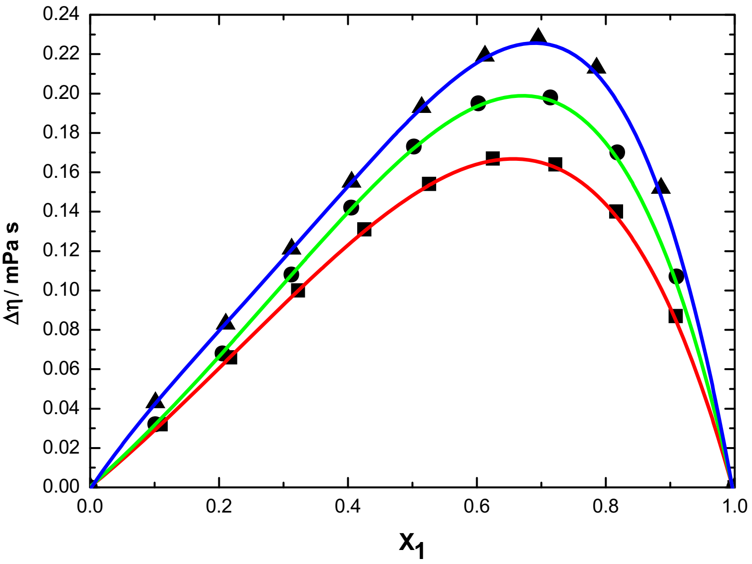

Fig. 9 Curves of deviation in viscosity ($\Delta\eta$) versus mole fraction for the binary mixtures of 2-chloroaniline + *o*-chlorotoluene (*square*), *m*-chlorotoluene (*circle*), *p*-chlorotoluene (*triangle*) at 303.15 K

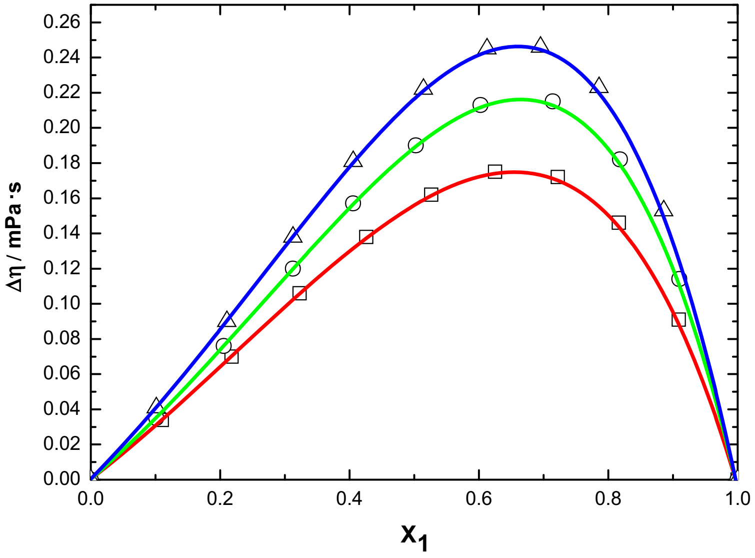

Fig. 10 Curves of deviation in viscosity ($\Delta\eta$) versus mole fraction for the binary mixtures of 2-chloroaniline + *o*-chlorotoluene (*square*), *m*-chlorotoluene (*circle*), *p*-chlorotoluene (*triangle*) at 308.15 K

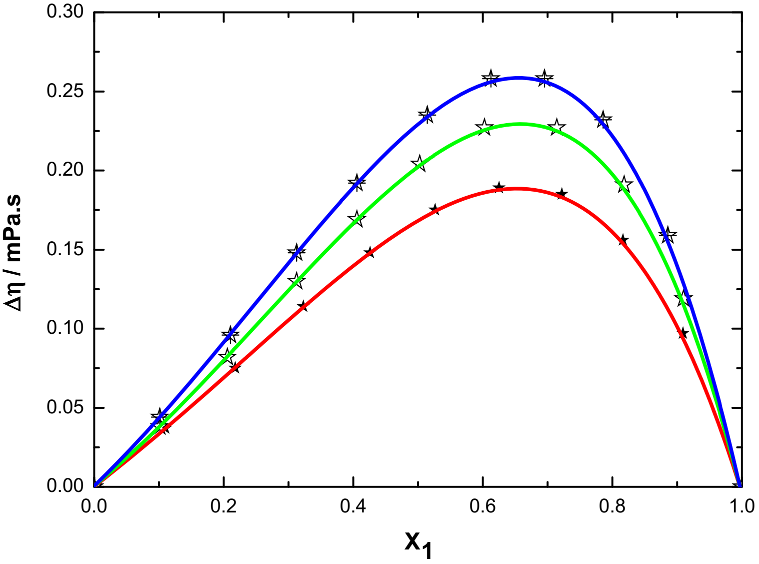

Fig. 11 Curves of deviation in viscosity ($\Delta\eta$) versus mole fraction for the binary mixtures of 2-chloroaniline + *o*-chlorotoluene (*black star*), *m*-chlorotoluene (*white star*), *p*-chlorotoluene (*crossed star*) at 313.15 K

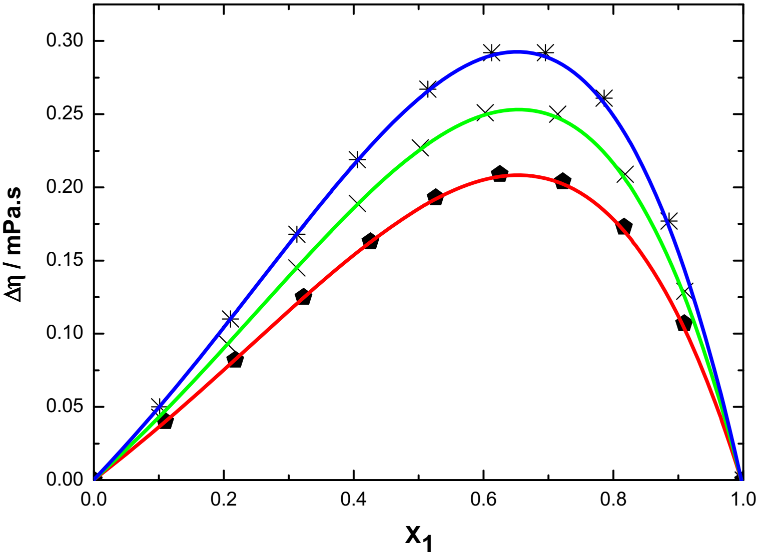

Fig. 12 Curves of deviation in viscosity ($\Delta\eta$) versus mole fraction for the binary mixtures of 2-chloroaniline + *o*-chlorotoluene (*pentagon*), *m*-chlorotoluene (*cross symbol*), *p*-chlorotoluene (*cross with plus symbol*) at 318.15 K

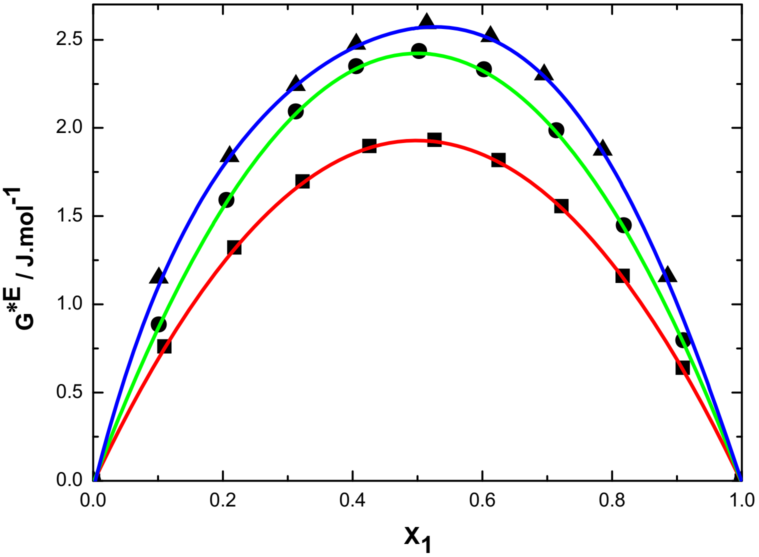

Fig. 13 Excess Gibbs energy of activation of viscous flow (G^{*E}) with mole fraction of 2-chloroaniline in the binary liquid mixtures of 2-chloroaniline with *o*-chlorotoluene (*square*), *m*-chlorotoluene (*circle*), *p*-chlorotoluene (*triangle*) at 303.15 K

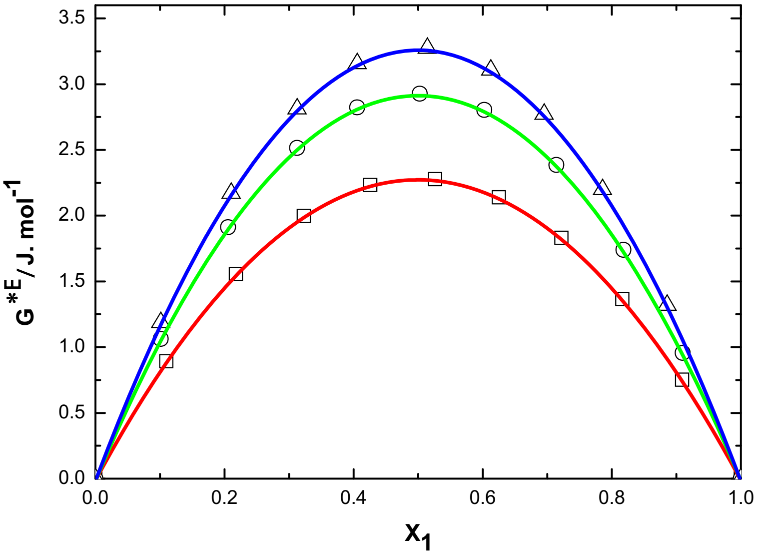

Fig. 14 Excess Gibbs energy of activation of viscous flow (G^{*E}) with mole fraction of 2-chloroaniline in the binary liquid mixtures of 2-chloroaniline with *o*-chlorotoluene (*square*), *m*-chlorotoluene (*circle*), *p*-chlorotoluene (*triangle*) at 308.15 K

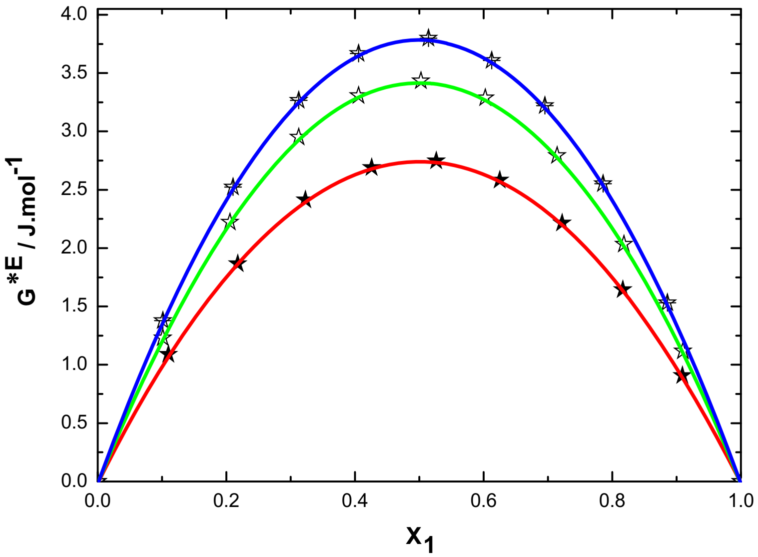

Fig. 15 Excess Gibbs energy of activation of viscous flow (G^{*E}) with mole fraction (x_1) of 2-chloroaniline + *o*-chlorotoluene (*black star*), *m*-chlorotoluene (*white star*), *p*-chlorotoluene (*crossed star*) at 313.15 K

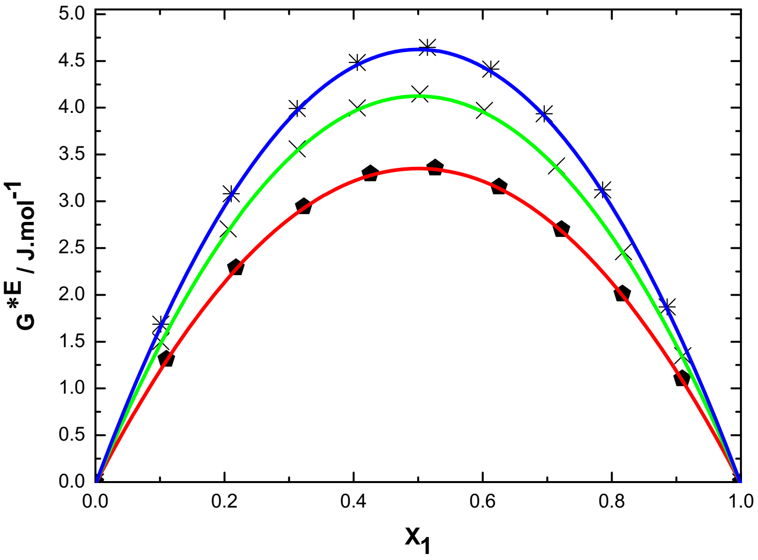

Fig. 16 Excess Gibbs energy of activation of viscous flow (G^{*E}) with mole fraction of 2-chloroaniline + *o*-chlorotoluene (*pentagon*), *m*-chlorotoluene (*cross symbol*), *p*-chlorotoluene (*cross with plus symbol*) at 318.15 K

$$\kappa_S^{\mathrm{id}} = \sum_{i=1}^{2} \varphi_i \left[\kappa_{S,i} + T \frac{V_i\left(\alpha_i^2\right)}{C_{\mathrm{p},i}} \right] - \left\{ T \frac{\left(\sum\limits_{i=1}^{2} x_i V_i\right)\left(\sum\limits_{i=1}^{2} \varphi_i \alpha_i\right)^2}{\sum\limits_{i=1}^{2} x_i C_{\mathrm{p},i}} \right\} \tag{7}$$

where φ_i is ideal state volume fraction of component i in mixture and is defined by relation:

$$\varphi_i = \frac{x_i V_i^{\mathrm{o}}}{\sum x_i V_i^{\mathrm{o}}} \tag{8}$$

T is temperature and κ_{Si}, V_i^{o}, α_i^{o} and C_{pi} are isentropic compressibility, molar volume, coefficent of isobaric thermal expansion and molar heat capacity, respectively, for pure component I; α_i^{o} was calculated from measured densities by relation:

$$\alpha_i^{\mathrm{o}} = \left(\frac{\rho_{T_1}}{\rho_{T_1}} - 1 \right) \Big/ (T_2 - T_1). \tag{9}$$

3.1 Theory

The observed V^{E} values are the resultant of physical and chemical forces and they may be recognized as: (i) the breaking of liquid order on mixing with the second component; (ii) non-specific physical interactions and unfavorable interactions between unlike molecules; (iii) specific interactions appearing in the mixture between dissimilar molecules by hydrogen bond formation; and (iv) specific dipole–dipole interactions in the mixture between solvent and co-solvent molecules. The first two factors contribute for the expansion of volume and the latter two factors contribute to the reduction of the volume. From the V^{E} curves shown in Figs. 1, 2, 3, and 4, it is clear that the volume reducing factors are dominant over the expansion factors in the present systems. The negative excess molar volumes show that the volume reduction factors play an important role between unlike molecules. This indicates the formation of hydrogen bond complexes. Thus, the observed negative V^{E} indicate the predominance of formation of (–NH····π) bonds over the rupture of bonds present in pure 2-chloroaniline molecules. 2-Chloroaniline and chlorotoluenes are polar solvents; their high dipole moments favor dipole–dipole interactions. Hence, there will be dipole–dipole interactions between unlike molecules of the three systems, contributing to the reduction in the volume.

An examination of curves in Figs. 1, 2, 3, and 4 indicates that the excess molar volumes for all of the binary systems are negative over the entire composition range at 303.15, 308.15, 313.15 and 318.15 K.

The algebraic values of V^{E} for the binary mixtures of 2-chloroaniline with chlorotoluenes fall in the following order: *o*-chlorotoluene < *m*-chlorotoluene < *p*-chlorotoluene.

The above order suggests that the dipole moments of the pure solvents are influencing the V^{E} data of the binary liquid mixtures. The dipole moments are 2-chloroaniline (1.78 D), *o*-chlorotoluene (1.56 D), *m*-chlorotoluene (1.82 D) and *p*-chlorotoluene (2.21 D). This type of behavior was observed earlier [8, 9]. The more negative V^{E} data of *p*-chlorotoluene when compared with other chlorotoluenes are due to its high dipole moment, which leads to stronger dipole–dipole interactions. Further, introduction of a chloro atom into the toluene molecule influences the magnitude of V^{E} to a considerable extent. This type of behavior was reported earlier [18].

An examination of plots in Figs. 5, 6, 7, and 8 reveal that κ_S^{E} is negative over the entire composition range for the binary mixtures of 2-chloroaniline with chlorotoluenes. This may be attributed to the relative strength of the following effects which influences the free spaces between component molecules [19, 20].

(a) Loss of dipolar association and difference in size and shape of component molecules leads to decrease in speed of sound and increase in isentropic compressibility.

(b) Dipole–dipole interactions, electron donor–acceptor interactions or charge-transfer complexes between unlike molecules lead to an increase in speed of sound and decrease in isentropic compressibility. If the strength of the interaction between the components increases, the magnitudes of the parameters become greater. In the present investigation, there is a possibility of electron donor–acceptor type or charge transfer interactions between the nitrogen atom of the amino group of 2-chloroaniline molecule, which acts as donor and the π-electron of the benzene ring of aromatic hydrocarbons, which act as acceptors (chlorotoluenes due to +M effect) resulting in negative κ_S^{E} values [21].

The algebraic values of κ_S^{E} for the binary mixtures of 2-chloroaniline with chlorotoluene are in the following order: *o*-chlorotoluene < *m*-chlorotoluene < *p*-chlorotoluene.

Viscosities of the binary mixtures of 2-chloroaniline with the chlorotoluenes are given in Figs. 9, 10, 11, and 12, which indicate that the deviations in viscosity ($\Delta\eta$) of all the binary mixtures are positive over the entire composition range at 303.15, 308.15, 313.15 and 318.15 K. Fort and Moore [22] observed that the positive deviations in viscosity indicate the specific interactions involving the formation of hetero-molecular complexes. The deviation in viscosity variation gives a qualitative estimation of the strength of the intermolecular interactions. The deviation in viscosity [23] may be generally explained by considering the following factors:

(i) The difference in size and shape of the component molecules and the loss of dipolar association in pure component may contribute to a decrease in viscosity.

(ii) Specific interactions between unlike components such as dipole–dipole interactions and charge transfer complexes may cause increases in viscosity in mixtures compared to the pure components. The former effect produces negative deviations in viscosity and the latter effect produces positive deviations in viscosity.

The positive values of viscosity deviation for the binary systems investigated suggest that the viscosities of associates formed between unlike molecules are relatively greater than those of the pure components. The deviations in viscosity are found to be opposite in sign to the excess molar volumes for all binary mixtures, which is in agreement with the view proposed by Brocos et al. [24, 25].

According to Reed and Taylor [26] and Palepu et al. [27], the excess Gibbs energy of activation of viscous flow ($G^{*\mathrm{E}}$) may be considered as a reliable criterion to detect or exclude the presence of interaction between unlike molecules. According to these authors, the magnitude of the positive $G^{*\mathrm{E}}$ values is an excellent indicator of the strength of specific interactions. The results presented in Figs. 13, 14, 15, and 16 indicate that excess Gibbs energies of activation of viscous flow are positive for three binary mixtures over the entire composition range at all investigated temperatures. The positive values of excess Gibbs energy of activation of viscous flow for the binary systems investigated suggest that the

Table 3 Mole fraction of 2-chloroaniline (x_1), viscosity (η), Theoretical values of Grunberg and Nissan (Eq. 13), Katti and Chaudhri (Eq. 14) and Hind et al. (Eq. 15) and Heric-Brewer (Eq. 16) parameters including standard deviation (SD) for binary mixtures at 303.15, 308.15, 313.15, and 318.15 K

x_1	η (mPa·s)	Equation 13	Equation 14	Equation 15	Equation 16
2-Chloroaniline + *o*-chlorotoluene					
303.15 K					
0.0000	0.886	0.886	0.886	0.886	0.886
0.1100	1.0729	1.073	1.072	1.067	1.073
0.2176	1.2843	1.284	1.283	1.278	1.285
0.3228	1.5208	1.521	1.518	1.517	1.521
0.4258	1.7823	1.782	1.777	1.783	1.782
0.5266	2.0685	2.069	2.063	2.072	2.069
0.6253	2.3786	2.379	2.376	2.384	2.378
0.7219	2.7112	2.711	2.711	2.716	2.711
0.8165	3.0646	3.065	3.071	3.069	3.064
0.9092	3.4369	3.437	3.445	3.439	3.437
1.0000	3.8256	3.826	3.826	3.826	3.826
SD		0.001	0.004	0.004	0.001
308.15 K					
0.0000	0.8080	0.808	0.808	0.808	0.808
0.1100	0.9808	0.981	0.981	0.981	0.981
0.2176	1.1755	1.175	1.175	1.178	1.176
0.3228	1.3920	1.392	1.392	1.396	1.392
0.4258	1.6298	1.630	1.630	1.635	1.630
0.5266	1.8877	1.888	1.888	1.891	1.888
0.6253	2.1643	2.164	2.164	2.166	2.164
0.7219	2.4576	2.458	2.458	2.456	2.457
0.8165	2.7651	2.765	2.765	2.761	2.765
0.9092	3.0843	3.084	3.084	3.080	3.084
1.0000	3.4122	3.412	3.412	3.412	3.412
SD		0.001	0.001	0.002	0.003
313.15 K					
0.0000	0.7280	0.728	0.728	0.728	0.728
0.1100	0.8896	0.889	0.889	0.896	0.889
0.2176	1.0700	1.070	1.070	1.081	1.071
0.3228	1.2713	1.271	1.271	1.283	1.271
0.4258	1.4898	1.490	1.490	1.499	1.490
0.5266	1.7257	1.726	1.726	1.730	1.726
0.6253	1.9757	1.976	1.976	1.974	1.976
0.7219	2.2376	2.237	2.238	2.23	2.237
0.8165	2.5073	2.508	2.508	2.496	2.507
0.9092	2.7831	2.783	2.783	2.774	2.783
1.0000	3.0602	3.060	3.060	3.060	3.060
SD		0.001	0.001	0.001	0.008
318.15 K					
0.0000	0.6420	0.642	0.642	0.642	0.642

Table 3 continued

x_1	η (mPa·s)	Equation 13	Equation 14	Equation 15	Equation 16
0.1100	0.7935	0.793	0.793	0.806	0.794
0.2176	0.9642	0.964	0.964	0.983	0.964
0.3228	1.1532	1.153	1.153	1.172	1.153
0.4258	1.3585	1.358	1.358	1.372	1.359
0.5266	1.5777	1.578	1.578	1.582	1.578
0.6253	1.8077	1.808	1.808	1.802	1.808
0.7219	2.0451	2.045	2.045	2.031	2.045
0.8165	2.2860	2.286	2.286	2.267	2.286
0.9092	2.5263	2.526	2.527	2.511	2.526
1.0000	2.7621	2.762	2.762	2.762	2.762
SD		0.001	0.001	0.001	0.014
2-Chloroaniline + *m*-chlorotoluene					
303.15 K					
0.0000	0.7430	0.743	0.743	0.743	0.743
0.1012	0.9087	0.909	0.908	0.902	0.909
0.2056	1.1091	1.109	1.108	1.102	1.109
0.3125	1.3483	1.348	1.347	1.344	1.349
0.4056	1.5867	1.587	1.580	1.587	1.587
0.5026	1.8666	1.867	1.857	1.871	1.867
0.6025	2.1896	2.190	2.181	2.197	2.189
0.7145	2.5946	2.595	2.597	2.602	2.594
0.8180	3.0086	3.009	3.022	3.014	3.008
0.9100	3.4081	3.408	3.421	3.410	3.408
1.0000	3.8256	3.826	3.826	3.826	3.826
SD		0.001	0.008	0.005	0.001
308.15 K					
0.0000	0.7010	0.701	0.701	0.701	0.701
0.1012	0.8581	0.858	0.859	0.861	0.858
0.2056	1.0466	1.047	1.047	1.053	1.047
0.3125	1.2694	1.269	1.269	1.278	1.270
0.4056	1.4889	1.489	1.491	1.498	1.489
0.5026	1.7431	1.743	1.747	1.749	1.743
0.6025	2.0317	2.032	2.036	2.033	2.032
0.7145	2.3863	2.386	2.384	2.382	2.386
0.8180	2.7402	2.740	2.734	2.731	2.740
0.9100	3.0733	3.073	3.069	3.065	3.073
1.0000	3.4122	3.412	3.412	3.412	3.412
SD		0.001	0.003	0.006	0.001
313.15 K					
0.0000	0.6630	0.663	0.663	0.663	0.663
0.1012	0.8120	0.812	0.813	0.823	0.812
0.2056	0.9900	0.990	0.991	1.008	0.991
0.3125	1.1993	1.199	1.198	1.217	1.199

Table 3 continued

x_1	η (mPa·s)	Equation 13	Equation 14	Equation 15	Equation 16
0.4056	1.4019	1.402	1.404	1.416	1.402
0.5026	1.6341	1.633	1.637	1.641	1.634
0.6025	1.8932	1.893	1.896	1.890	1.892
0.7145	2.2045	2.204	2.202	2.191	2.204
0.8180	2.5075	2.507	2.502	2.489	2.507
0.9100	2.7856	2.785	2.782	2.770	2.785
1.0000	3.0602	3.060	3.060	3.060	3.060
SD		0.001	0.003	0.002	0.001
318.15 K					
0.0000	0.6240	0.624	0.624	0.624	0.624
0.1012	0.7685	0.768	0.769	0.787	0.769
0.2056	0.9398	0.940	0.940	0.968	0.940
0.3125	1.1384	1.138	1.138	1.167	1.139
0.4056	1.3294	1.329	1.331	1.350	1.330
0.5026	1.5445	1.544	1.548	1.552	1.545
0.6025	1.7801	1.780	1.783	1.772	1.780
0.7145	2.0559	2.056	2.054	2.032	2.056
0.8180	2.3156	2.316	2.311	2.286	2.315
0.9100	2.5448	2.545	2.542	2.522	2.545
1.0000	2.7621	2.762	2.762	2.762	2.762
SD		0.001	0.002	0.021	0.001
2-Chloroaniline + *p*-chlorotoluene					
303.15 K					
0.0000	0.7820	0.782	0.782	0.782	0.782
0.1012	0.9617	0.961	0.956	0.536	0.956
0.2102	1.1753	1.175	1.177	0.411	1.175
0.3125	1.4050	1.405	1.415	0.425	1.412
0.4056	1.6441	1.644	1.654	0.549	1.656
0.5142	1.9625	1.962	1.978	0.826	1.975
0.6125	2.2873	2.286	2.301	1.201	2.296
0.6952	2.5858	2.589	2.585	1.608	2.588
0.7859	2.9360	2.933	2.927	2.150	2.932
0.8859	3.3438	3.347	3.331	2.863	3.337
1.0000	3.8256	3.826	3.826	3.826	3.826
SD		0.007	0.009	0.011	0.007
308.15 K					
0.0000	0.7280	0.728	0.728	0.728	0.728
0.1012	0.8921	0.892	0.892	0.469	0.892
0.2102	1.0975	1.097	1.095	0.323	1.098
0.3125	1.3182	1.318	1.315	0.313	1.318
0.4056	1.5429	1.543	1.542	0.410	1.543
0.5142	1.8329	1.833	1.828	0.650	1.833
0.6125	2.1198	2.120	2.115	0.987	2.120

Table 3 continued

x_1	η (mPa·s)	Equation 13	Equation 14	Equation 15	Equation 16
0.6952	2.3773	2.377	2.380	1.357	2.377
0.7859	2.6739	2.674	2.681	1.856	2.673
0.8859	3.0141	3.014	3.022	2.516	3.014
1.0000	3.4122	3.412	3.412	3.412	3.412
SD		0.001	0.005	0.013	0.001
313.15 K					
0.0000	0.6820	0.682	0.682	0.682	0.682
0.1012	0.8377	0.838	0.838	0.411	0.838
0.2102	1.0314	1.031	1.029	0.249	1.032
0.3125	1.2379	1.238	1.235	0.217	1.238
0.4056	1.4458	1.446	1.444	0.291	1.446
0.5142	1.7106	1.711	1.706	0.501	1.711
0.6125	1.9681	1.968	1.963	0.804	1.968
0.6952	2.1949	2.195	2.197	1.144	2.195
0.7859	2.4511	2.451	2.458	1.605	2.451
0.8859	2.7372	2.737	2.745	2.221	2.737
1.0000	3.0602	3.060	3.060	3.060	3.060
SD		0.001	0.004	0.019	0.001
318.15 K					
0.0000	0.6320	0.632	0.632	0.632	0.632
0.1012	0.7836	0.784	0.784	0.350	0.784
0.2102	0.9716	0.972	0.970	0.171	0.972
0.3125	1.1704	1.170	1.167	0.122	1.171
0.4056	1.3684	1.368	1.367	0.176	1.368
0.5142	1.6162	1.616	1.612	0.360	1.616
0.6125	1.8516	1.852	1.847	0.638	1.851
0.6952	2.0537	2.054	2.056	0.953	2.053
0.7859	2.2747	2.275	2.281	1.385	2.274
0.8859	2.5112	2.511	2.518	1.966	2.511
1.0000	2.7621	2.762	2.762	2.762	2.762
SD		0.001	0.004	0.027	0.001

specific interaction between 2-chloroaniline and chlorotoluene molecules takes place through dipole–dipole interaction. Thus, the values of deviation in viscosity and excess Gibbs energy of activation of viscous flow are dependent on the position of the –Cl group in the toluene molecule, indicating a different extent of molecular interactions in chlorotoluene.

The experimental values of density (ρ) and speed of sound (u) were used to calculate the values of acoustic impedance, Z, intermolecular free length (L_f). The values of Δu, ΔZ, and L_f^E were evaluated with help of the following relation:

$$Y^E = Y - (x_1Y_1 + x_2Y_2) \tag{10}$$

where Y^E is Δu, ΔZ, and L_f^E.

Table 4 Mole fraction of 2-chloroaniline (x_1), density (ρ), speed of sound (u), deviation in ultrasonic speed (Δu), isentropic compressibility (κ_s), intermolecular free length (L_f), acoustic impedance (Z), excess isentropic compressibility (κ_S^E), excess intermolecular free length (L_f^E), deviation in acoustic impedance (ΔZ) of the binary mixtures at 303.15, 308.15, 313.15 and 318.15 K respectively

x_1	ρ (kg·m^{-3})	u (m·s^{-1})	κ_S (TPa^{-1})	Z × 10^{-3} (kg·m^2·s^{-1})	L_f × 10^{-9} (m)	ΔZ (kg·m^{-2}·s^{-1})	Δu (m·s^{-1})	κ_S^E (TPa^{-1})	L_f^E × 10^{-9} (m)
2-Chloroaniline (1) + *o*-chlorotoluene (2)									
303.15 K									
0.0000	1072.84	1285.0	564.5	1378.6	176.40	0.000	0.000	0.000	0.000
0.1100	1086.31	1309.8	536.6	1422.9	167.68	1.505	4.526	−9.985	−2.559
0.2176	1099.70	1332.8	511.9	1465.6	159.98	2.444	7.617	−16.66	−4.221
0.3228	1113.00	1354.4	489.8	1507.4	153.06	3.318	9.814	−20.85	−5.236
0.4258	1126.19	1374.4	470.1	1547.8	146.89	3.687	10.83	−22.64	−5.628
0.5266	1139.27	1392.9	452.4	1586.9	141.37	3.582	10.75	−22.37	−5.498
0.6253	1152.20	1410.2	436.4	1624.9	136.38	3.148	9.826	−20.39	−4.956
0.7219	1165.02	1426.4	421.9	1661.7	131.84	2.464	8.142	−16.96	−4.073
0.8165	1177.69	1441.5	408.6	1697.7	127.70	1.596	5.834	−12.28	−2.913
0.9092	1190.26	1455.9	396.4	1732.9	123.86	0.807	3.119	−6.603	−1.548
1.0000	1202.69	1469.6	385.0	1767.4	120.32	0.000	0.000	0.000	0.000
308.15 K									
0.0000	1068.22	1266.0	584.1	1352.4	183.99	0.000	0.000	0.000	0.000
0.1100	1081.83	1292.8	553.1	1398.6	174.22	3.442	6.173	−12.11	−3.218
0.2176	1095.21	1316.1	527.1	1441.4	166.05	4.446	9.330	−19.16	−4.987
0.3228	1108.48	1337.1	504.6	1482.1	158.96	4.239	10.59	−22.80	−5.819
0.4258	1121.64	1356.6	484.4	1521.7	152.59	3.783	10.89	−24.14	−6.063
0.5266	1134.69	1375.5	465.8	1560.8	146.73	3.689	10.86	−23.85	−5.926
0.6253	1147.65	1393.8	448.5	1599.6	141.28	4.207	10.71	−22.26	−5.504
0.7219	1160.50	1411.5	432.5	1638.1	136.24	5.097	10.31	−19.39	−4.801
0.8165	1173.21	1428.0	418.0	1675.4	131.66	5.628	9.095	−15.02	−3.746

Table 4 continued

x_1	ρ (kg·m^{-3})	u (m·s^{-1})	κ_S (TPa^{-1})	Z × 10^{-3} (kg·m^2·s^{-1})	L_f × 10^{-9} (m)	ΔZ (kg·m^{-2}·s^{-1})	Δu (m·s^{-1})	κ_S^E (TPa^{-1})	L_f^E × 10^{-9} (m)
0.9092	1185.76	1442.4	405.4	1710.3	127.69	4.537	6.090	−8.740	−2.204
1.0000	1198.02	1453.3	395.2	1741.1	124.49	0.000	0.000	0.000	0.000
313.15 K									
0.0000	1063.34	1248.0	603.8	1327.0	191.71	0.000	0.000	0.000	0.000
0.1100	1077.01	1276.3	570.0	1374.6	180.96	5.205	7.745	−14.22	−3.868
0.2176	1090.41	1299.8	542.8	1417.3	172.35	6.441	11.05	−21.72	−5.759
0.3228	1103.64	1320.3	519.8	1457.1	165.05	5.685	11.81	−25.08	−6.484
0.4258	1116.77	1339.2	499.2	1495.6	158.51	4.566	11.53	−26.00	−6.582
0.5266	1129.82	1357.8	480.1	1534.0	152.43	4.161	11.20	−25.46	−6.361
0.6253	1142.77	1376.3	462.0	1572.8	146.68	4.875	11.21	−23.88	−5.943
0.7219	1155.64	1394.5	445.0	1611.5	141.29	6.396	11.33	−21.16	−5.298
0.8165	1168.37	1411.5	429.6	1649.1	136.40	7.595	10.63	−16.83	−4.273
0.9092	1180.86	1425.8	416.6	1683.6	132.26	6.406	7.586	−10.11	−2.616
1.0000	1193.02	1435.2	406.9	1712.2	129.20	0.000	0.000	0.000	0.000
318.15 K									
0.0000	1058.96	12270	627.2	1299.3	201.34	0.000	0.000	0.000	0.000
0.1100	1072.72	1256.3	590.7	1347.7	189.60	5.736	8.186	−15.67	−4.353
0.2176	1086.11	1280.1	561.9	1390.3	180.36	6.793	11.35	−23.55	−6.366
0.3228	1099.30	1300.7	537.7	1429.9	172.59	5.605	11.77	−26.83	−7.062
0.4258	1112.40	1320.0	516.0	1468.3	165.62	4.193	11.24	−27.61	−7.110
0.5266	1125.42	1339.0	495.6	1507.0	159.08	3.828	10.96	−27.07	−6.883
0.6253	1138.37	1358.4	476.1	1546.3	152.82	4.992	11.37	−25.62	−6.508
0.7219	1151.24	1377.6	457.7	1586.0	146.92	7.248	12.08	−23.07	−5.920
0.8165	1163.96	1395.6	441.1	1624.4	141.59	9.093	11.92	−18.72	−4.891
0.9092	1176.46	1410.3	427.4	1659.1	137.19	7.929	8.805	−11.50	−3.067

Table 4 continued

x_1	ρ (kg·m^{-3})	u (m·s^{-1})	κ_S (TPa^{-1})	Z × 10^{-3} (kg·m^2·s^{-1})	L_f × 10^{-9} (m)	ΔZ (kg·m^{-2}·s^{-1})	Δu (m·s^{-1})	κ_S^E (TPa^{-1})	L_f^E × 10^{-9} (m)
1.0000	1188.49	1418.9	417.9	1686.3	134.15	0.000	0.000	0.000	0.000
2-Chloroaniline (1) + *m*-chlorotoluene (2)									
303.15 K									
0.0000	1062.83	1281.1	573.2	1361.6	179.15	0.000	0.000	0.000	0.000
0.1012	1076.11	1306.2	544.6	1405.6	170.21	2.954	6.033	−11.47	−2.992
0.2056	1089.96	1329.3	519.2	1448.9	162.25	3.864	9.468	−18.81	−4.804
0.3125	1104.37	1351.0	496.1	1492.0	155.04	3.542	10.96	−22.89	−5.722
0.4056	1117.12	1368.8	477.7	1529.1	149.30	2.952	11.29	−24.37	−5.991
0.5026	1130.60	1387.0	459.7	1568.2	143.67	2.606	11.20	−24.35	−5.908
0.6025	1144.70	1405.6	442.1	1609.0	138.18	2.862	10.93	−22.95	−5.523
0.7145	1160.76	1426.1	423.6	1655.4	132.37	3.816	10.36	−19.72	−4.740
0.8180	1175.81	1444.2	407.7	1698.1	127.43	4.546	8.941	−14.88	−3.598
0.9100	1189.37	1458.5	395.2	1734.7	123.51	3.814	5.920	−8.590	−2.099
1.0000	1202.69	1469.6	385.0	1767.4	120.32	0.000	0.000	0.000	0.000
308.15 K									
0.0000	1058.35	1261.8	593.4	1335.4	186.94	0.000	0.000	0.000	0.000
0.1012	1071.71	1288.5	562.0	1380.9	177.04	4.391	7.291	−13.34	−3.574
0.2056	1085.57	1311.9	535.2	1424.2	168.59	5.366	10.75	−21.12	−5.510
0.3125	1099.96	1333.3	511.4	1466.6	161.09	4.383	11.66	−24.93	−6.331
0.4056	1112.68	1351.0	492.4	1503.2	155.12	3.215	11.48	−26.1	−6.493
0.5026	1126.12	1369.3	473.6	1542.0	149.19	2.675	11.23	−25.95	−6.364
0.6025	1140.2	1388.6	454.8	1583.2	143.28	3.413	11.38	−24.72	−6.031
0.7145	1156.28	1410.5	434.7	1630.9	136.94	5.615	11.83	−21.89	−5.38
0.8180	1171.31	1429.8	417.6	1674.7	131.56	7.452	11.31	−17.21	−4.301
0.9100	1184.85	1444.2	404.6	1711.1	127.47	6.572	8.115	−10.36	−2.643

Table 4 continued

x_1	ρ (kg·m^{-3})	u (m·s^{-1})	κ_S (TPa^{-1})	Z × 10^{-3} (kg·m^2·s^{-1})	L_f × 10^{-9} (m)	ΔZ (kg·m^{-2}·s^{-1})	Δu (m·s^{-1})	κ_S^E (TPa^{-1})	L_f^E × 10^{-9} (m)
1.0000	1198.02	1453.3	395.2	1741.1	124.49	0.000	0.000	0.000	0.000
313.15 K									
0.0000	1053.14	1247.0	610.6	1313.3	193.88	0.000	0.000	0.000	0.000
0.1012	1066.62	1275.0	576.7	1359.9	183.12	6.259	8.918	−15.32	−4.210
0.2056	1080.50	1298.6	548.8	1403.2	174.24	7.865	12.92	−23.71	−6.336
0.3125	1094.89	1319.5	524.5	1444.7	166.55	6.767	13.68	−27.40	−7.112
0.4056	1107.61	1336.6	505.4	1480.4	160.46	5.308	13.22	−28.29	−7.180
0.5026	1121.07	1354.4	486.2	1518.4	154.39	4.610	12.82	−27.95	−6.985
0.6025	1135.19	1373.5	466.9	1559.2	148.26	5.540	13.09	−26.71	−6.651
0.7145	1151.32	1395.5	446.0	1606.6	141.62	8.303	13.98	−24.03	−6.051
0.8180	1166.41	1414.7	428.3	1650.1	136.01	10.52	13.75	−19.32	−4.967
0.9100	1179.90	1428.4	415.4	1685.3	131.89	9.020	10.10	−11.91	−3.132
1.0000	1193.02	1435.2	406.9	1712.2	129.20	0.000	0.000	0.000	0.000
318.15 K									
0.0000	1048.56	1234.5	625.7	1294.4	200.88	0.000	0.000	0.000	0.000
0.1012	1062.13	1263.4	589.8	1341.8	189.35	7.742	10.19	−16.98	−4.770
0.2056	1076.06	1286.9	561.1	1384.8	180.12	9.771	14.50	−25.76	−7.035
0.3125	1090.41	1307.2	536.7	1425.4	172.28	8.465	15.06	−29.26	−7.746
0.4056	1103.11	1323.7	517.3	1460.2	166.08	6.758	14.38	−29.90	−7.732
0.5026	1116.58	1341.1	497.9	1497.5	159.84	6.055	13.94	−29.48	−7.505
0.6025	1130.71	1360.1	478.0	1537.9	153.46	7.366	14.54	−28.41	−7.221
0.7145	1146.83	1382.4	456.3	1585.4	146.47	10.89	16.12	−26.08	−6.733
0.8180	1161.96	1401.6	438.0	1628.7	140.62	13.65	16.31	−21.48	−5.685
0.9100	1175.48	1414.5	425.2	1662.7	136.49	11.62	12.18	−13.54	−3.673
1.0000	1188.49	1418.9	417.9	1686.3	134.15	0.000	0.000	0.000	0.000

Table 4 continued

x_1	ρ (kg·m^{-3})	u (m·s^{-1})	κ_S (TPa^{-1})	Z × 10^{-3} (kg·m^2·s^{-1})	L_f × 10^{-9} (m)	ΔZ (kg·m^{-2}·s^{-1})	Δu (m·s^{-1})	κ_S^E (TPa^{-1})	L_f^E × 10^{-9} (m)
2-Chloroaniline (1) + p-chlorotoluene (2)									
303.15 K									
0.0000	1059.54	1272.6	582.8	1348.4	182.12	0.000	0.000	0.000	0.000
0.1012	1073.14	1300.0	551.4	1395.1	172.31	4.284	7.446	−13.39	−3.548
0.2102	1087.95	1325.0	523.5	1441.6	163.60	5.121	11.04	−21.45	−5.526
0.3125	1102.00	1346.0	500.9	1483.3	156.52	4.003	11.87	−25.06	−6.287
0.4056	1115.05	1364.1	481.9	1521.1	150.60	2.755	11.66	−26.23	−6.448
0.5142	1130.50	1385.3	461.0	1566.0	144.05	2.199	11.38	−26.00	−6.288
0.6125	1144.73	1404.8	442.6	1608.1	138.32	3.113	11.59	−24.66	−5.939
0.6952	1156.87	1421.5	427.8	1644.5	133.68	4.816	11.99	−22.61	−5.474
0.7859	1170.36	1439.3	412.4	1684.5	128.89	6.845	11.95	−19.01	−4.662
0.8859	1185.41	1456.7	397.6	1726.8	124.24	7.156	9.598	−12.50	−3.130
1.0000	1202.69	1469.6	385.0	1767.4	120.32	0.000	0.000	0.000	0.000
308.15 K									
0.0000	1053.94	1253.0	604.3	1320.6	190.37	0.000	0.000	0.000	0.000
0.1012	1067.73	1281.8	570.0	1368.7	179.55	5.523	8.569	−15.36	−4.153
0.2102	1082.63	1307.3	540.4	1415.4	170.24	6.378	12.220	−23.97	−6.280
0.3125	1096.77	1328.1	516.9	1456.7	162.82	4.683	12.553	−27.39	−6.963
0.4056	1109.86	1346.1	497.2	1494.0	156.63	2.865	11.877	−28.27	−7.018
0.5142	1125.39	1367.4	475.2	1538.9	149.69	2.066	11.423	−27.87	−6.799
0.6125	1139.72	1387.7	455.7	1581.5	143.53	3.391	11.971	−26.63	−6.485
0.6952	1151.99	1405.2	439.6	1618.8	138.48	5.870	12.965	−24.79	−6.092
0.7859	1165.58	1424.1	423.0	1659.9	133.26	8.843	13.684	−21.32	−5.338
0.8859	1180.76	1442.0	407.3	1702.7	128.30	9.558	11.562	−14.44	−3.711
1.0000	1198.02	1453.3	395.2	1741.1	124.49	0.000	0.000	0.000	0.000

Table 4 continued

x_1	ρ (kg·m^{-3})	u (m·s^{-1})	κ_S (TPa^{-1})	Z × 10^{-3} (kg·m^2·s^{-1})	L_f × 10^{-9} (m)	ΔZ (kg·m^{-2}·s^{-1})	Δu (m·s^{-1})	κ_S^E (TPa^{-1})	L_f^E × 10^{-9} (m)
313.15 K									
0.0000	1050.34	1237.2	622.0	1299.5	197.48	0.000	0.000	0.000	0.000
0.1012	1064.10	1266.7	585.7	1347.9	185.95	6.683	9.498	−16.80	−4.628
0.2102	1078.89	1292.5	554.9	1394.4	176.17	8.181	13.64	−26.05	−6.962
0.3125	1092.91	1313.2	530.6	1435.2	168.46	6.757	14.13	−29.61	−7.689
0.4056	1105.86	1331.0	510.4	1471.9	162.06	5.050	13.53	−30.48	−7.734
0.5142	1121.25	1352.2	487.8	1516.1	154.87	4.421	13.16	−30.06	−7.503
0.6125	1135.45	1372.3	467.6	1558.2	148.47	5.955	13.87	−28.82	−7.190
0.6952	1147.58	1389.9	451.1	1595.0	143.22	8.582	15.03	−26.97	−6.795
0.7859	1161.09	1408.6	434.1	1635.5	137.82	11.61	15.75	−23.36	−5.997
0.8859	1176.06	1425.8	418.2	1676.9	132.79	11.74	13.23	−15.95	−4.202
1.0000	1193.02	1435.2	406.9	1712.2	129.20	0.000	0.000	0.000	0.000
318.15 K									
0.0000	1045.68	1224.6	637.7	1280.5	204.70	0.000	0.000	0.000	0.000
0.1012	1059.54	1255.3	598.9	1330.0	192.26	8.438	11.04	−18.80	−5.301
0.2102	1074.35	1281.3	566.9	1376.6	181.99	10.75	15.88	−28.80	−7.886
0.3125	1088.39	1301.8	542.2	1416.8	174.04	9.493	16.46	−32.39	−8.617
0.4056	1101.32	1319.2	521.8	1452.8	167.49	7.711	15.77	−33.08	−8.600
0.5142	1116.71	1339.9	498.8	1496.2	160.12	7.042	15.36	−32.49	−8.309
0.6125	1130.95	1359.8	478.2	1537.8	153.51	8.743	16.17	−31.23	−7.985
0.6952	1143.12	1377.2	461.2	1574.3	148.06	11.61	17.49	−29.39	−7.598
0.7859	1156.64	1395.6	443.9	1614.2	142.48	14.77	18.33	−25.70	−6.775
0.8859	1171.61	1412.1	428.0	1654.4	137.40	14.39	15.37	−17.76	−4.804
1.0000	1188.49	1418.9	417.9	1686.3	134.15	0.000	0.000	0.000	0.000

The sign and magnitude of the observed values of Δu, ΔZ, and L_f^E on mixing were found to depend upon several contributions, which are physical or/and chemical in nature. The physical contributions comprise the dispersion forces and non-specific physical weak interactions that lead to negative values in Δu, ΔZ or positive L_f^E, hence in order (intermolecular interaction) in the system. Chemical contributions involve breaking up of H-bonded structures, if any, resulting in positive L_f^E or negative values in Δu, and ΔZ and specific interactions such as formation of new H-bonds, charge transfer complex and strong dipole–dipole interactions between component molecules result in positive Δu, and ΔZ and negative L_f^E values, making the system more ordered due to increased intermolecular interactions (Table 3).

An examination of data in the Table 4 shows that ΔZ values are positive for all binary systems over the entire composition range at 303.15, 308.15, 313.15 and 318.15 K. The positive values of deviation in acoustic impedance for the binary systems investigated suggest that the specific interaction between unlike molecules in mixtures are dominant over the breaking of dipolar interactions between like molecules in the mixture.

The variation of V^E, κ_S^E and $\Delta\eta$ with mole fraction were fitted to the Redlich–Kister polynomial equation [28] of the type:

$$Y^E = x_1 x_2 \left\{ a_0 + a_1(x_1 - x_2) - a_2(x_1 - x_2)^2 \right\}. \tag{11}$$

The values of a_0, a_1 and a_2 are the coefficients of the polynomial equation and the corresponding standard deviations, with σ obtained by the method of least-squares with equal weights assigned to each point. The standard deviation (σ) is defined as:

$$\sigma\left(Y^E\right) = \sqrt{\sum \frac{\left(Y_{obs}^E - Y_{cal}^E\right)^2}{n - m}} \tag{12}$$

where n is the total number of experimental points and m is the number of coefficients. The values of a_0, a_1 and a_2 are determined by a multiple-regression analysis using the least-squares method and summarized along with the standard deviations between the experimental and fitted values of V^E, κ_S^E, and $\Delta\eta$ in Table 5.

Knowledge of the viscosities of the pure liquids and respective mixtures and study of the viscosity calculation methods are important for practical and theoretical purposes. Over the last years, numerous equations for liquid mixture viscosity have been proposed. Methods concerning viscosity modeling can be found in the literature [29, 30]. In the present work, three typical semi-empirical relations are used to correlate the experimental viscosity data of the investigated binary systems.

Grunberg–Nissan provided the following empirical equation containing one adjustable parameter [31]:

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 d_{12} \tag{13}$$

where d_{12} may be regarded as a parameter proportional to the interchange energy, also an approximate measure of the strength of the interaction between the components.

Katti and Chaudhri [32] proposed the following equation:

$$\ln V\eta = x_1 \ln V_1\eta_1 + x_2 \ln V_2\eta_2 + x_1 x_2 \frac{W_{vis}}{RT} \tag{14}$$

where W_{vis}/RT is an interaction term.

Table 5 Coefficients of Redlich–Kister equation and standard deviation (σ) values

Binary mixtures	Functions	a_o	a_1	a_2	σ
303.15 K					
2-Chloroaniline + *o*-chlorotoluene	V^E (cm^3·mol^{-1})	−0.586	−0.047	0.033	0.001
	$\Delta\eta$ (mPa·s)	0.595	0.446	0.136	0.001
	κ_S^E (TPa^{-1})	−90.37	13.67	−1.081	0.047
2-Chloroaniline + *m*-chlorotoluene	V^E (cm^3·mol^{-1})	−0.609	−0.031	−0.077	0.001
	$\Delta\eta$ (mPa· s)	0.688	0.582	0.204	0.001
	κ_S^E (TPa^{-1})	−97.5	13.74	−27.73	0.001
2-Chloroaniline + *p*-chlorotoluene	V^E (cm^3·mol^{-1})	−0.636	−0.035	−0.173	0.001
	$\Delta\eta$ (mPa· s)	0.754	0.667	0.394	0.001
	κ_S^E (TPa^{-1})	−104.4	13.71	−50.12	0.001
308.15 K					
2-Chloroaniline + *o*-chlorotoluene	V^E (cm^3·mol^{-1})	−0.621	−0.047	−0.195	0.001
	$\Delta\eta$ (mPa· s)	0.626	0.463	0.14	0.001
	κ_S^E (TPa^{-1})	−96.27	12.3	−29.37	0.001
2-Chloroaniline + *m*-chlorotoluene	V^E (cm^3·mol^{-1})	−0.648	−0.052	−0.266	0.002
	$\Delta\eta$ (mPa· s)	0.758	0.613	0.188	0.001
	κ_S^E (TPa^{-1})	−103.9	13.58	−50.34	0.001
2-Chloroaniline + *p*-chlorotoluene	V^E (cm^3·mol^{-1})	−0.677 1.024	−0.029	−0.351	0.001
	$\Delta\eta$ (mPa· s)	0.869	0.681	0.197	0.001
	κ_S^E (TPa^{-1})	−111.9 57.82	14.75	−71.08	0.001
313.15 K					
2-Chloroaniline + *o*-chlorotoluene	V^E (cm^3·mol^{-1})	−0.658	−0.057	−0.293	0.001
	$\Delta\eta$ (mPa· s)	0.675	0.492	0.142	0.001
	κ_S^E (TPa^{-1})	−102.2	16.15	−49.06	0.001
2-Chloroaniline + *m*-chlorotoluene	V^E (cm^3·mol^{-1})	−0.692	−0.042	−0.381	0.001
	$\Delta\eta$ (mPa· s)	0.812	0.632	0.176	0.001
	κ_S^E (TPa^{-1})	−111.9	15.81	−69.18	0.001
2-Chloroaniline + *p*-chlorotoluene	V^E (cm^3·mol^{-1})	−0.725	−0.038	−0.453	0.001
	$\Delta\eta$ (mPa· s)	0.92	0.697	0.188	0.001
	κ_S^E (TPa^{-1})	−120.7	15.03	−81.87	0.001
318.15 K					
2-Chloroaniline + *o*-chlorotoluene	V^E (cm^3·mol^{-1})	−0.687	−0.045	−0.448	0.002
	$\Delta\eta$ (mPa· s)	0.744	0.549	0.152	0.001
	κ_S^E (TPa^{-1})	−109.2	15.48	−63.81	0.001
2-Chloroaniline + *m*-chlorotoluene	V^E (cm^3·mol^{-1})	−0.725	−0.033	−0.534	0.002
	$\Delta\eta$ (mPa·s)	0.904	0.675	0.172	0.001
	κ_S^E (TPa^{-1})	−118.1	15.18	−88.9	0.001
2-Chloroaniline + *p*-chlorotoluene	V^E (cm^3·mol^{-1})	−0.771	−0.032	−0.564	0.001
	$\Delta\eta$ (mPa·s)	1.045	0.772	0.184	0.001
	κ_S^E (TPa^{-1})	−130.4	17.22	−98.32	0.001

Hind et al. [33] suggested an equation for the viscosity of binary liquid mixtures as:

$$\eta = x_1^2\eta_1 + x_2^2\eta_2 + 2x_1x_2H_{12} \tag{15}$$

where H_{12} is Hind's interaction parameter and is attributed to unlike pair interactions.

Heric and Brewer [34] proposed the following equation:

$$\ln\eta = x_1\ln M_1\eta_1 + x_2\ln M_2\eta_2 - \ln(x_1M_1 + x_2M_2)2x_1x_2\Delta_{12} \tag{16}$$

where Δ_{12} is the interaction term and other symbols have their usual meaning.

The one-parameter equation due to Tamura and Kurata [35] has the form:

$$\eta = x_1\Phi_1\eta_1 + x_2\Phi_2\eta_2 + 2(x_1x_2\Phi_1\Phi_2)^{1/2}T_{12} \tag{17}$$

where Φ_1 and Φ_2 are the volume fractions of components 1 and 2, respectively, T_{12} is Tamura and Kurata constant.

The interaction parameter d_{12} *i*s positive for binary systems. Nigam and Mahl [36] concluded from the study of binary mixtures that (i) if $\Delta\eta > 0$, $d_{12} > 0$ and magnitude of both are large then strong specific interaction, (ii) if $\Delta\eta < 0$, $d_{12} > 0$ then weak specific interaction, and (iii) if $\Delta\eta < 0$, $d_{12} < 0$ magnitude of both are large then the dispersion force will be dominant. Fort and Moore [19] reported that for any binary liquid mixture, a positive value of d_{12} indicates the presence of specific interactions and a negative value of d_{12} indicates the presence of weak interactions between the unlike molecules. On this basis, we can say that there is a strong interaction in the binary systems studied.

The interaction parameter W_{vis}/RT shows almost the same trend as that of d_{12}. In fact, one could say that the parameters d_{12} and W_{vis}/RT exhibit almost similar behavior, which is not unlikely in view of logarithmic nature of both equations. The values of interaction parameters of Tamara and Kurata (T_{12}) and Hind et al. (H_{12}) do not differ appreciably from each other. This is in agreement with the view put forward by Fort and Moore [19] in regard to the nature of parameters T_{12} and H_{12}. The experimental and theoretical values of viscosity of the liquid mixtures calculated using Eqs. 13–16, including standard deviation, are presented in Table 3. The Grunberg–Nissan relation gives better results than the comparison to other theoretical relations. The differences between experimental and theoretical values are greater for other relations studied here.

4 Conclusions

Experimental data of viscosity, speed of sound, and density are reported for binary mixtures of 2-chloroaniline with *o*-chlorotoluene, *m*-chlorotoluene, *p*-chlorotoluene over the entire range of mole fraction at 303.15, 308.15, 313.15 and 318.15 K. Calculated excess molar volume, excess isentropic compressibility, deviation in viscosity, and excess Gibbs energy of activation of viscous flow are fitted with Redlich–Kister type polynomial equation. Positive values of the deviation in viscosity and excess Gibbs energy of activation of viscous flow, and negative values of excess molar volume and excess isentropic compressibility over the whole composition range are observed for all the investigated binary systems. Thermodynamic functions of activation have been estimated for each binary mixture. The viscosity data have been correlated with several semi-empirical equations (Grunberg–Nissan, Katti–Chaudhri, and Hind et al.). The Grunberg–Nissan relation gives better results than the other theoretical relations. The excess/deviation properties and positive values of viscosity interaction parameter can be interpreted by considering the intermolecular charge transfer complexes, molecular size and shapes of the components. The strong intermolecular interactions have significant effect on the thermodynamic and transport properties of the investigated binary mixtures.

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