Densities, Viscosities, and Ultrasonic Velocity Studies of Binary Mixtures of Chloroform with Propan-1-ol and Butan-1-ol at (303.15 and 313.15) K

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Densities, viscosities, and ultrasonic velocities of binary mixtures of chloroform with propan-1-ol and butan-1-ol have been measured over the entire range of composition, at (303.15 and 313.15) K and at atmospheric pressure. From the experimental data, excess molar volumes (V^E), deviations in viscosity ($\Delta\eta$), and excess isentropic compressibility (κ_S^E) have been calculated. The excess molar volumes and isentropic compressibility for propan-1-ol and butan-1-ol are positive while deviations in viscosity are negative. The results have been interpreted in terms of intermolecular interactions. These are further fitted to the Redlich−Kister polynomial equation.

Introduction

The effect of molecular size, shape, chain length, and degree of molecular association of normal alkanols and branched alkanols on the volumetric, viscometric, and acoustic properties of binary mixtures containing acetonitrile, dimethyl sulfoxide, ethyl acetate, and benzonitrile have been reported earlier.[1−5] Alkanols and chloroform are widely used solvents in chemical industry. Knowledge of the excess molar volumes and thermodynamic properties is of importance in the design of storage and handling facilities of such mixtures. Literature provides information on these properties at 298.15 K, but there is a lack of combined study of the excess molar volumes, deviation in viscosity, and isentropic compressibility at other temperatures, particularly for the systems propan-1-ol + chloroform and butan-1-ol + chloroform. In the present paper we report density, viscosity, and ultrasonic velocity data for the binary mixtures of chloroform with propan-1-ol and butan-1-ol at (303.15 and 313.15) K.

Experimental Section

Propan-1-ol (E. Merck, purity 99.5 %) and butan-1-ol (s.d., fine chemical, purity 99 %) were used after single distillation. Chloroform (s.d., fine chemical, purity 99 %) was shaken several times with distilled water to remove ethanol. It was dried over anhydrous calcium chloride and then fractionally distilled. The purity of the solvents, after purification, was ascertained by comparing their densities, viscosities, and ultrasonic velocities with the corresponding literature values at 303.15 K (Table 1). Binary mixtures were prepared by mass in airtight stoppered glass bottles. The masses were recorded on an Adairdutt balance to an accuracy of $\pm 1 \times 10^{-4}$ g. Care was taken to avoid evaporation and contamination during mixing. The estimated uncertainty in mole fraction was $< 1 \times 10^{-4}$.

Densities were determined by using a 15 cm^3 bicapillary pycnometer as described earlier.[6,7] The pycnometer was calibrated using conductivity water with 0.99705 g·cm^{-3} as its density[8] at 298.15 K. The pycnometer filled with air-bubble-free experimental liquids was kept in a transparent walled water bath (maintained constant to ± 0.01 K) for (10 to 15) min to attain thermal equilibrium. The positions of the liquid levels in the two arms were recorded with the help of a traveling microscope, which could read to 0.01 mm. The estimated uncertainty of density measurements of solvent and binary mixtures was 0.00005 g·cm^{-3}. At least three to four measurements were made, which had an average deviation of ± 0.00005 g·cm^{-3}.

Table 1. Comparison of Experimental Density, Viscosity, and Ultrasonic Velocity of Pure Liquids with Literature Values at 303.15 K

	$\rho \times 10^{-3}$/(kg·cm^{-3})		η/(mPa·s)		u/(m·s^{-1})	
pure liquid	exptl	lit	exptl	lit	exptl	lit
chloroform	1.46921	1.47060[a]	0.534	0514[a]	969	968[f]
		1.4692[b]		0.534[b]		
propan-1-ol	0.79666	0.7963[a]	1.719	1.725[a]	1193	1191[g]
		0.7963[b]		1.719[c]		
butan-1-ol	0.80231	0.80195[a]	2.274	2.271[a]	1227	1224[g]
		0.8023[d]		2.273[e]		

[a] Ref 12. [b] Ref 15. [c] Ref 16. [d] Ref 17. [e] Ref 18. [f] Ref 19. [g] Ref 20.

The dynamic viscosities were measured using an Ubbelohde suspended level viscometer,[7] calibrated with conductivity water. An electronic digital stop watch with readability of ± 0.01 s was used for the flow time measurements. At least three repetitions of each data reproducible to ± 0.05 s were obtained, and the results were averaged. Since all flow times were greater than 200 s and capillary radius (0.5 mm) was far less than its length (50 to 60) mm, the kinetic energy and end corrections, respectively, were found to be negligible. The uncertainties in dynamic viscosities are of the order of ± 0.003 mPa·s.

The ultrasonic velocities (u) were measured at a frequency of 2 MHz in these solutions through interferometric method (using Mittal's F-81 model) at (303.15 and 313.15) K (± 0.05 K). The error in velocity measurements is ± 0.1 %.The other experimental details are the same as reported earlier.[2−4]

Results and Discussion

Experimental values of densities (ρ), viscosities (η), and ultrasonic velocities (u) of mixtures at (303.15 and 313.15) K are listed as a function of mole fraction in Table 2. The density

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Table 2. Density (ρ), Viscosity (η), Ultrasonic Velocity (u), Isentropic Compressibility (κ_s), Excess Molar Volume (V^E), Deviation in Viscosity ($\Delta\eta$), and Excess Isentropic Compressibility (κ_s^E) for Chloroform (1) + Alkanols (2) at (303.15 and 313.15) K

x_1	$\rho \times 10^{-3}$ / kg·m^{-3}	$V^E \times 10^6$ / m^3·mol^{-1}	η / mPa·s	$\Delta\eta$ / mPa·s	u / m·s^{-1}	κ_s / TPa^{-1}	κ_s^E / TPa^{-1}	x_1	$\rho \times 10^{-3}$ / kg·m^{-3}	$V^E \times 10^6$ / m^3·mol^{-1}	η / mPa·s	$\Delta\eta$ / mPa·s	u / m·s^{-1}	κ_s / TPa^{-1}	κ_s^E / TPa^{-1}
Chloroform (1) + Propan-1-ol (2) 303.15 K															
0.0000	0.79666	0.000	1.719	0.000	1193	882	0	0.5541	1.17918	0.157	0.760	−0.302	1017	820	18
0.0521	0.83423	0.002	1.628	−0.029	1171	874	1	0.5994	1.20910	0.169	0.723	−0.286	1007	816	21
0.0996	0.86817	0.002	1.541	−0.060	1153	866	2	0.6511	1.24311	0.165	0.680	−0.268	997	809	23
0.1504	0.90410	0.016	1.435	−0.106	1134	860	3	0.7033	1.27726	0.166	0.654	−0.232	988	802	24
0.2010	0.93951	0.037	1.331	−0.149	1116	855	4	0.7506	1.30810	0.152	0.629	−0.201	980	796	25
0.2498	0.97347	0.052	1.233	−0.190	1100	849	4	0.8000	1.34017	0.136	0.608	−0.163	974	787	25
0.3057	1.01195	0.081	1.113	−0.243	1083	842	5	0.8507	1.37301	0.115	0.596	−0.115	970	774	22
0.3499	1.04215	0.098	1.044	−0.260	1070	838	7	0.9001	1.40492	0.087	0.579	−0.074	967	761	17
0.4031	1.07828	0.114	0.956	−0.285	1056	832	8	0.9484	1.43603	0.048	0.562	−0.034	966	745	11
0.4510	1.11046	0.130	0.884	−0.301	1042	829	12	1.0000	1.46921	0.000	0.534	0.000	969	725	0
0.5056	1.14702	0.143	0.813	−0.307	1028	825	15								
Chloroform (1) + Propan-1-ol (2) at 313.15 K															
0.0000	0.78831	0.000	1.363	0.000	1159	944	0	0.5541	1.16450	0.190	0.662	−0.219	981	892	25
0.0521	0.82528	0.009	1.302	−0.017	1136	939	2	0.5994	1.19391	0.201	0.635	−0.206	972	887	27
0.0996	0.85868	0.014	1.243	−0.033	1117	933	2	0.6511	1.22727	0.201	0.605	−0.191	961	882	31
0.1504	0.89403	0.032	1.163	−0.070	1098	928	4	0.7033	1.26087	0.197	0.592	−0.158	951	877	33
0.2010	0.92892	0.054	1.077	−0.111	1080	923	4	0.7506	1.29108	0.187	0.570	−0.139	944	869	33
0.2498	0.96229	0.073	1.013	−0.133	1064	918	6	0.8000	1.32266	0.165	0.558	−0.108	939	857	30
0.3057	1.00009	0.109	0.919	−0.178	1047	912	9	0.8507	1.35488	0.145	0.547	−0.075	933	848	29
0.3499	1.02985	0.123	0.872	−0.187	1034	908	11	0.9001	1.38624	0.114	0.531	−0.048	931	832	24
0.4031	1.06526	0.149	0.811	−0.201	1018	906	15	0.9484	1.41696	0.061	0.517	−0.019	931	814	13
0.4510	1.09700	0.159	0.752	−0.218	1006	901	17	1.0000	1.44974	0.000	0.491	0.000	934	791	0
0.5056	1.13288	0.176	0.703	−0.220	992	897	21								
Chloroform (1) + Butan-1-ol (2) at 303.15 K															
0.0000	0.80231	0.000	2.274	0.000	1227	828	0	0.5502	1.14588	0.155	0.877	−0.440	1043	802	13
0.0513	0.83244	0.010	2.125	−0.060	1207	825	0	0.6011	1.18026	0.162	0.820	−0.409	1030	799	15
0.0999	0.86144	0.026	1.989	−0.111	1189	821	−2	0.6527	1.21551	0.163	0.754	−0.385	1018	794	16
0.1490	0.89091	0.040	1.829	−0.186	1170	820	0	0.7020	1.24987	0.157	0.709	−0.344	1007	789	17
0.2009	0.92247	0.059	1.683	−0.242	1152	817	0	0.7519	1.28510	0.142	0.676	−0.290	997	783	18
0.2506	0.95306	0.077	1.531	−0.308	1134	816	2	0.8020	1.32098	0.135	0.636	−0.243	989	774	16
0.2999	0.98375	0.096	1.394	−0.359	1118	813	3	0.8517	1.35717	0.116	0.622	−0.171	981	766	15
0.3659	1.02550	0.115	1.224	−0.414	1097	810	5	0.9023	1.39470	0.087	0.587	−0.118	975	754	11
0.4070	1.05186	0.124	1.137	−0.430	1084	809	6	0.9509	1.43143	0.050	0.569	−0.051	971	741	7
0.4515	1.08069	0.136	1.045	−0.444	1071	807	9	1.0000	1.46921	0.000	0.534	0.000	969	725	0
0.4997	1.11225	0.151	0.960	−0.445	1056	806	11								
Chloroform (1) + Butan-1-ol (2) at 313.15 K															
0.0000	0.79437	0.000	1.783	0.000	1193	884	0	0.5502	1.13203	0.197	0.758	−0.314	1006	873	32
0.0512	0.82395	0.024	1.670	−0.046	1172	884	8	0.6012	1.16578	0.204	0.712	−0.295	993	870	34
0.1000	0.85245	0.048	1.566	−0.088	1154	881	7	0.6527	1.20035	0.207	0.671	−0.269	981	866	36
0.1490	0.88147	0.064	1.450	−0.140	1135	881	10	0.7021	1.23400	0.205	0.633	−0.244	970	861	37
0.2009	0.91253	0.084	1.341	−0.182	1116	880	12	0.7519	1.26852	0.192	0.615	−0.197	961	854	36
0.2506	0.94258	0.108	1.230	−0.229	1098	880	15	0.8021	1.30385	0.175	0.579	−0.168	952	846	34
0.2999	0.97285	0.121	1.136	−0.260	1081	880	18	0.8518	1.33940	0.151	0.572	−0.111	945	836	32
0.3659	1.01385	0.146	1.018	−0.292	1060	878	21	0.9024	1.37618	0.122	0.547	−0.070	939	824	27
0.4070	1.03975	0.157	0.943	−0.314	1047	877	23	0.9510	1.41242	0.069	0.524	−0.031	936	808	19
0.4515	1.06795	0.181	0.878	−0.322	1034	876	26	1.0000	1.44974	0.000	0.491	0.000	934	791	0
0.4997	1.09900	0.193	0.820	−0.318	1019	876	30								

values have been used to calculate excess molar volumes (V^E) using the following equation:

$$V^E/\mathrm{m^3{\cdot}mol^{-1}} = (x_1M_1 + x_2M_2)/\rho_{12} - (x_1M_1/\rho_1) - (x_2M_2/\rho_2) \quad (1)$$

where ρ_{12} is the density of the mixture and x_1, M_1, ρ_1 and x_2, M_2, ρ_2 are the mole fraction, the molecular weight, and the density of pure components 1 and 2, respectively.

The viscosity deviations ($\Delta\eta$) were calculated using

$$\Delta\eta/\mathrm{mPa{\cdot}s} = \eta_{12} - x_1\eta_1 - x_2\eta_2 \quad (2)$$

where η_{12} is the viscosity of the mixture and x_1, x_2 and η_1, η_2 are the mole fraction and the viscosity of pure components 1 and 2, respectively.

The excess isentropic compressibility (κ_s^E) was obtained using

$$\kappa_s^E/\mathrm{TPa^{-1}} = \kappa_s - \kappa_s^{id} \quad (3)$$

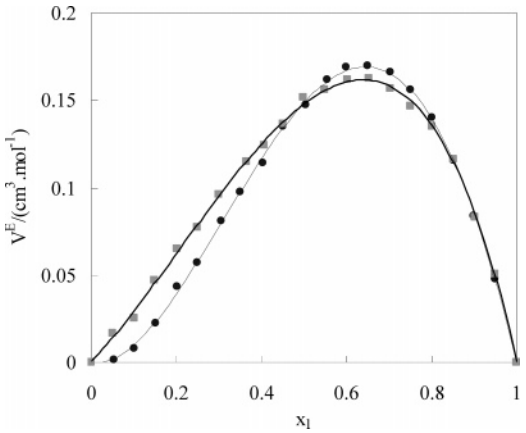

Figure 1. Excess molar volumes (V^E) at 303.15 K for x_1 chloroform + (1 − x_1) alkanols: ●, propan-1-ol; ■, butan-1-ol.

where κ_s is the isentropic compressibility and was calculated using the Laplace relation, that is $\kappa_s = (1/u^2\rho)$ and κ_s^{id} was

Table 3. Parameters and Standard Deviations (σ) of Equations 5 and 6 for Chloroform + Propan-1-ol and Chloroform + Butan-1-ol

	T/K	a_0	a_1	a_2	a_3	a_4	σ
			Chloroform + Propan-1-ol				
V^E/(cm^3·mol^{-1})	303.15	0.583	0.5448	−0.1142			0.0031
	313.15	0.7083	0.5826	−0.0057	0.0685		0.0042
$\Delta\eta$/(mPa·s)	303.15	−1.2287	−0.0373	0.7358	−0.0506		0.0032
	313.15	−0.8838	0.0118	0.5837	−0.0859	0.0935	0.0047
κ_s^E/(TPa^{-1})	303.15	56.258	110.190	79.011			0.6112
	313.15	82.866	138.973	89.372			0.8145
			Chloroform + Butan-1-ol				
V^E/(cm^3·mol^{-1})	303.15	0.5901	0.398	−0.2141	−0.0380	0.4178	0.0038
	313.15	0.7337	0.4653	0.2363			0.0050
$\Delta\eta$/(mPa·s)	303.15	−1.7925	0.0343	0.7766			0.0054
	313.15	−1.2887	0.1291	0.5999			0.0038
κ_s^E/(TPa^{-1})	303.15	42.569	25.796	30.114			0.5691
	313.15	103.246	105.847	172.161	40.367		2.725

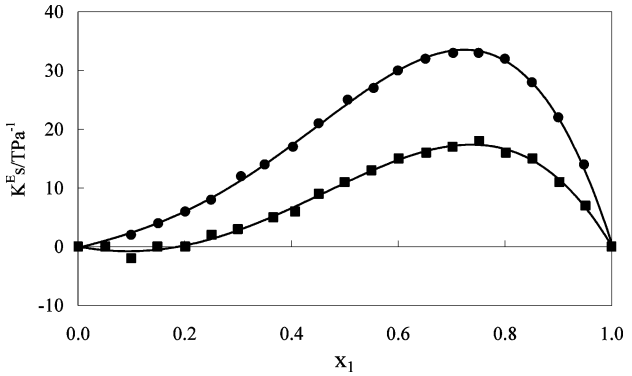

Figure 2. Excess isentropic compressibility (κ_s^E) at 303.15 K for x_1 chloroform + (1 − x_1) alkanols: ●, propan-1-ol; ■, butan-1-ol.

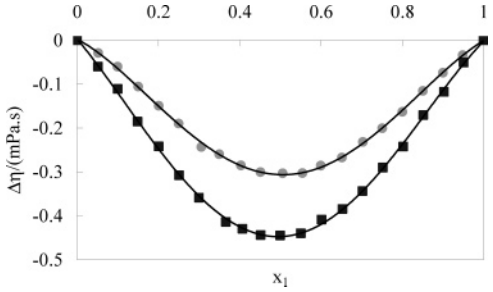

Figure 3. Deviations in viscosity ($\Delta\eta$) at 303.15 K for x_1 chloroform + (1 − x_1) alkanols: ●, propan-1-ol; ■, butan-1-ol.

calculated from[9,10]

$$\kappa_s^{id} = \sum \phi_i[\kappa_{s,i} + TV^\circ_i(\alpha^\circ_i{}^2)/C_{p,i}] - [T(\sum x_i V^\circ_i)(\sum \phi_i \alpha^\circ_i)^2/\sum x_i C_{p,i}] \quad (4)$$

where ϕ_i is the volume fraction of the component i in the mixture stated, T is the temperature, and $\kappa_{s,i}$, V°_i, α°_i, and $C_{p,i}$ are the isentropic compressibility, molar volume, coefficient of thermal expansion, and molar heat capacity, respectively, for pure component i. The values required were taken from the literature.[11,12]

The excess molar volumes, excess isentropic compressibility, and deviations in viscosity were fitted to a Redlich−Kister[13] equation of the type

$$Y = x_1 x_2 \sum_i^n a_i (x_1 - x_2)^i \quad (5)$$

where Y is either V^E, $\Delta\eta$, or κ_s^E, and n is the degree of polynomial. Coefficients a_i were obtained by fitting eq 5 to experimental results using a least-squares regression method. In each case, the optimum number of coefficients is ascertained from an examination of the variation in standard deviation (σ).

σ was calculated using

$$\sigma(Y) = \left[\frac{\sum (Y_{expt} - Y_{calc})^2}{N - n}\right]^{1/2} \quad (6)$$

where N is the number of data points and n is the number of coefficients. The calculated values of the coefficients (a_i) along with the standard deviations (σ) are given in Table 3.

The volume changes arising due to addition of a second component to an alkanol results from several effects. The main volume effects accompanying the addition of chloroform to an alkanols result from changes of free volume, interstitial accommodation of chloroform within the hydrogen bonded structure of alkanol, disruption of the alkanols structure, and the so-called condensation effect due to restriction of the rotational motion of the alkanols molecule.[14] The break-up of the alkanol structure tends to increase V^E. In the present investigation, positive V^E values, at 303.15 K, for propan-1-ol and butan-1-ol (Figure 1) may be attributed to predominance of the declustering of alkanols in the presence of chloroform. The V^E values at equimolar mixtures are almost the same, but the curve for butan-1-ol is slightly lower than for propan-1-ol in the low mole fraction region of chloroform, suggesting that initially some of the chloroform molecules may be occupying interstitial spaces in the slight bulkier butan-1-ol. A similar nature of the curve is obtained for κ_s^E at 303.15 K (Figure 2).

The $\Delta\eta$ values for both the alkanols are negative at 303.15 K (Figure 3) with butan-1-ol more negative than propan-1-ol, suggesting more specific interaction for butan-1-ol than for propan-1-ol. This observation supports the inference drawn from V^E and κ_s^E values. The self-association of alkanols decreases as the chain length increases, so that the breaking of intramolecular interactions in butan-1-ol is much easier than in propan-1-ol. Thus, the establishment of intermolecular interactions between chloroform and butan-1-ol is stronger than for chloroform and propan-1-ol mixtures. This fact is also supported, in general, by the observation that the excess properties curves for the butan-1-ol mixture are less asymmetric than for the propan-1-ol mixture (Figures 1 and 2).

Conclusion

There are specific interactions present in the mixtures studied, and the strength of interaction is more for butan-1-ol than for propan-1-ol.

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