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Acoustic Properties of Binary Liquid Mixtures Containing Dimethyl Formamide with Anilines At Different Temperatures

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Abstract

Experimental work was carried to find the values of densities (ρ), viscosity (η) and speed of sound (U) for binary mixture systems of Dimethyl formamide (DMF) with Aniline(AN), N-methyl Aniline(NMA) and NN Dimethylaniline (NNDMA) for all the mole fractions at different temperatures T=303.15 K, 308.15 K and at 313.15 K. These values were used for the purpose of interpreting the intermolecular interactions present between them. Excess values of molar volume (V^E), isentropic compressibility (K_s^E), free length (L_f^E), Gibb's free energy (ΔG^{*E}) and enthalpy (H^E) were determined. The Redlich–Kister type polynomial equation was used for the purpose of fitting of the values. The presence of interactions in the component mixtures were studied with the help of deviations observed in the excess parameters.

***Key words*:** Speed of sound, Viscosity, Redlich-Kister polynomial, molar volume.

INTRODUCTION

The experimental measurements of speed of sound along with density, viscosity for pure and mixtures of some liquids are important apart from being simple and easy to measure. They find applications in different fields and industries[1,2] Previously many people[3-9] worked in a similar fashion by measuring the density, viscosity, and speed of sound for so many liquid mixtures and their combinations. The chemicals studied here have great role to play in the fields of chemistry, biology etc. The insight into thermodynamic properties for liquid mixtures containing two or more components along with data for analysis in terms of various models are of great significance in different fields at industry level[10]. The excess functions [11] are varied by different factors like differences in intermolecular forces and the size of the molecules. The interactions can be analyzed on the basis of the signs and magnitudes of excess values. Taking into consideration the experimentally determined values, several other parameters like excess values of isentropic compressibility (K_s^E), free length (L_f^E), Gibbs free energy (G^{*E}), molar volume (V^E) and enthalpy (H^E) were calculated. With the help of these excess values the intermolecular interactions were predicted.

EXPERIMENTAL WORK

Experimental work was carried on to find out the values of speed of sound, density, and viscosity for all the mixtures of DMF with Anilines at different temperatures 303.15K, 308.15K, and 313.15K for all mole fractions. Here we have calculated certain excess parameters to check the applicability of the ideal mixing rules to the present systems. The excess values of isentropic compressibility K_s^E, excess values of molar volumes, V^E, free length (L_f^E), Gibbs free energy (ΔG^{*E}) and enthalpy (H^E), were calculated by using the expressions given in literature [15] as follows,,

$$K_s^E = K_s - K_s^{id} \quad (1)$$

$$K_s = \frac{1}{\rho U^2} \quad (2)$$

$$K_s^{id} = \sum \phi_i \left\{ K_{s,i}^o + \frac{T V_i^o (\alpha_i^o)^2}{C_{p,i}^o} \right\} - T \left(\sum x_i V_i^o \right) \left(\frac{\sum \phi_i \alpha_i^{o2}}{\sum x_i C_{p,i}^o} \right) \quad (3)$$

here K_s is calculated value of isentropic compressibility for the mixture, K_s^E is its excess value, K_s^{id} is the ideal isentropic compressibility value. A relation given by Benson and Kiyohara[12, 13] and Douheret et al[14] was used to calculate K_s^{id} for an ideal mixture. $K_{s,i}^o$, V_o^i, α_i^o, $C_{p,i}^o$ are the isentropic compressibility, molar volume, isobaric thermal expansion coefficient and molar isobaric heat capacity of pure component i, T represents temperature, ϕ_i is the volume fraction and x_i represents the mole fraction of i in the mixture.

$$V^E = \frac{x_1 M_1 + x_2 M_2}{\rho} - \left(\frac{x_1 M_1}{\rho_1} + \frac{x_2 M_2}{\rho_2} \right) \quad (4)$$

where ρ is the density of the mixture and x_1, M_1, and ρ_1 and x_2, M_2, and ρ_2 are the mole fraction, molar mass, and density of pure components 1 and 2, respectively.

$$L_f^E = L_f - K_T (K_s^{id})^{1/2} \quad (5)$$

Where K_T is a temperature dependent constant $K_T=(91.368+0.3565T)x10^{-8}$.

$$\Delta G^{*E} = RT \left[\ln \left(\frac{\eta V}{\eta_2 V_2} \right) - x_1 \ln \left(\frac{\eta_1 V_1}{\eta_2 V_2} \right) \right] \quad (6)$$

$$H^E = H - (x_1 H_1 + x_2 H_2) \quad (7)$$

Where R represents gas constant, T is absolute temperature, ηis the viscosity of the mixture and η_1,η_2 are the viscosities of the pure compounds, V is the molar volume of mixture and V_1, V_2 are the molar volumes of the pure compounds, H represents the calculated value of enthalpy for the mixture and H_1, H_2 represent enthalpy of pure components 1 and 2, respectively.

RESULTS AND DISCUSSION

From the experimentally determined values of density, the speed of sound and viscosity[1], the values of excess isentropic compressibility (K_s^E), excess molar volume (V^E), excess free length (L_f^E), excess Gibbs free energy (G^{*E}) and excess enthalpy (H^E) were calculated. These excess values were plotted against mole fractions separately at three different temperatures as shown in Figures 1-5. The observed deviations in the excess values infer us about the strength of interactions present between these compounds[16]. The variations in these excess parameter values reflect the interactions between the mixing species, depending upon the composition, molecular sizes, and shapes of the components and temperature.

The excess isentropic compressibility (K_s^E) for the liquid mixtures under study are as shown in figure 1. It can be observed that the K_s^E values are positive over the entire mole fraction range indicating the presence of weak interactions in these mixtures and also that the molecules are loosely packed in the mixtures due to their shape and size. Fort and Moore[17] suggested that there will be a reduction in volume if the liquids having different molecular sizes and shape mix well this causes the values of K_s^E to be negative and if the liquids do not mix well because they are almost same size then there is a lot of free space in between. So, it can be said that the molecular interactions are weak in these liquid mixtures and that the medium is loosely packed.

TABLE 1 Acoustic impedance (Z), isentropic compressibility (K_s), molar volume (V), free length (L_f), free volume (V_f), internal pressure (π_i) and enthalpy (H) for the liquid mixtures at T=303.15 K.

X_1	**Z** x10^6 kg $m^{-2}s^{-1}$	**K_s** x10^{-10} $kg^{-1}ms^{-2}$	**V** x10^{-5} m^3mol^{-1}	**L_f** x10^{-11} m	**V_f** x10^{-8} m^3	**π_i** x10^8 Pa	**H** x10^4 J.mol^{-1}	**τ** x10^{-10} sec
			(Dimethyl Formamide + Aniline)					
0.0000	1.3777	4.9415	9.9296	4.4335	4.2267	3.4003	3.3764	6.5887
0.1240	1.4055	4.7951	9.5687	4.3673	3.1452	4.0393	3.8651	6.3935
0.2415	1.4337	4.6545	9.2284	4.3028	2.4889	4.6506	4.2917	6.2060
0.3532	1.4621	4.5193	8.9071	4.2398	2.0491	5.2467	4.6733	6.0257
0.4593	1.4909	4.3888	8.6025	4.1782	1.7344	5.8354	5.0199	5.8518
0.5602	1.5199	4.2638	8.3149	4.1182	1.4984	6.4207	5.3388	5.6850
0.6565	1.5492	4.1434	8.0424	4.0597	1.3151	7.0060	5.6345	5.5245
0.7483	1.5789	4.0271	7.7830	4.0023	1.1688	7.5940	5.9105	5.3695
0.8359	1.6087	3.9156	7.5375	3.9465	1.0495	8.1853	6.1697	5.2208
0.9197	1.6388	3.8081	7.3039	3.8920	0.9505	8.7817	6.4141	5.0774
1.0000	1.6693	3.7041	7.0810	3.8385	0.8671	9.3845	6.6451	4.9389
			(Dimethyl Formamide +N-Methylaniline)					
0.0000	1.3605	5.0770	7.7771	1.1249	3.2702	4.5422	3.5326	6.7693
0.1400	1.3638	5.1364	8.1187	1.1315	2.9532	4.6461	3.7720	6.8486
0.2681	1.3664	5.2000	8.4677	1.1385	2.7144	4.7134	3.9912	6.9334
0.3858	1.3685	5.2680	8.7401	1.1459	2.5041	4.8058	4.2003	7.0240
0.4942	1.3699	5.3404	8.9725	1.1537	2.3235	4.9035	4.3996	7.1205
0.5945	1.3706	5.4175	9.1706	1.1620	2.1659	5.0061	4.5909	7.2233
0.6874	1.3708	5.4994	9.3383	1.1708	2.0264	5.1140	4.7756	7.3326
0.7738	1.3703	5.5865	9.4800	1.1800	1.9017	5.2267	4.9549	7.4486
0.8543	1.3691	5.6789	9.5986	1.1897	1.7894	5.3442	5.1297	7.5719
0.9295	1.3674	5.7769	9.6971	1.2000	1.6875	5.4663	5.3008	7.7026
1.0000	1.3650	5.8809	9.7782	1.2107	1.5944	5.5928	5.4688	7.8412
			(Dimethyl Formamide +NN-Dimethylaniline)					
0.0000	1.3777	4.9422	1.2919	4.4338	5.5549	2.4932	3.2209	6.5896
0.1555	1.3794	4.9344	1.2110	4.4303	4.9335	2.7609	3.3434	6.5793
0.2930	1.3813	4.9262	1.1394	4.4266	4.4105	3.0399	3.4635	6.5682
0.4153	1.3832	4.9179	1.0756	4.4229	3.9672	3.3294	3.5811	6.5573
0.5250	1.3851	4.9097	1.0185	4.4192	3.5873	3.6296	3.6967	6.5463
0.6237	1.3870	4.9015	9.6705	4.4155	3.2596	3.9402	3.8104	6.5354
0.7132	1.3889	4.8934	9.2045	4.4118	2.9753	4.2608	3.9219	6.5245
0.7946	1.3908	4.8852	8.7806	4.4081	2.7264	4.5917	4.0318	6.5136
0.8689	1.3927	4.8771	8.3932	4.4045	2.5078	4.9324	4.1398	6.5028
0.9371	1.3945	4.8690	8.0379	4.4008	2.3144	5.2831	4.2465	6.4920
1.0000	1.3964	4.8609	7.7107	4.3972	2.1426	5.6435	4.3515	6.4812

TABLE 2 Acoustic impedance (Z), isentropic compressibility (K_s), molar volume (V), free length (L_f), free volume (V_f), internal pressure (π_i) and enthalpy (H) for the liquid mixtures at T=308.15 K.

X_1	Z $x10^6$ kg $m^{-2}s^{-1}$	K_s $x10^{-10}$ $kg^{-1}ms^{-2}$	V $x10^{-5}$ m^3mol^{-1}	L_f $x10^{-11}$ m	V_f $x10^{-5}$ m^3	π_i $x10^8$ Pa	H $x10^4$ J.mol^{-1}	τ $x10^{-10}$ sec
(Dimethyl Formamide + Aniline)								
0.0000	1.3398	5.2093	9.9594	4.5927	4.3879	3.3856	3.3719	6.9458
0.1240	1.3660	5.0514	9.6175	4.5226	3.3578	3.9605	3.8090	6.7352
0.2415	1.3924	4.8998	9.2945	4.4542	2.7053	4.5130	4.1946	6.5330
0.3532	1.4190	4.7542	8.9890	4.3875	2.2555	5.0527	4.5419	6.3390
0.4593	1.4460	4.6139	8.6997	4.3223	1.9273	5.5854	4.8591	6.1519
0.5602	1.4732	4.4796	8.4244	4.2589	1.6775	6.1153	5.1518	5.9728
0.6565	1.5006	4.3505	8.1639	4.1971	1.4814	6.6439	5.4240	5.8006
0.7483	1.5282	4.2263	7.9165	4.1367	1.3234	7.1736	5.6789	5.6350
0.8359	1.5561	4.1068	7.6807	4.0778	1.1936	7.7056	5.9184	5.4757
0.9197	1.5842	3.9918	7.4563	4.0203	1.0852	8.2409	6.1447	5.3224
1.0000	1.6127	3.8807	7.2417	3.9640	0.9934	8.7811	6.3590	5.1742
(Dimethyl Formamide + N-Methyl Aniline)								
0.0000	1.3366	5.2372	4.8135	1.1540	2.1173	7.8757	3.7910	6.9830
0.1400	1.3392	5.3014	5.7059	1.1610	2.1460	6.9921	3.9897	7.0686
0.2681	1.3412	5.3700	6.5249	1.1685	2.1434	6.4036	4.1783	7.1600
0.3858	1.3427	5.4430	7.2290	1.1764	2.1077	6.0354	4.3630	7.2574
0.4942	1.3435	5.5208	7.8490	1.1848	2.0566	5.7869	4.5422	7.3611
0.5945	1.3436	5.6034	8.3966	1.1936	1.9970	5.6169	4.7163	7.4712
0.6874	1.3432	5.6912	8.8796	1.2029	1.9326	5.5028	4.8863	7.5883
0.7738	1.3422	5.7843	9.3067	1.2128	1.8661	5.4291	5.0527	7.7124
0.8543	1.3405	5.8831	9.6840	1.2231	1.7991	5.3862	5.2160	7.8441
0.9295	1.3383	5.9878	10.017	1.2339	1.7325	5.3674	5.3767	7.9837
1.0000	1.3354	6.0987	10.312	1.2453	1.6673	5.3676	5.5354	8.1316
(Dimethyl formamide+ N N Dimethyl Aniline)								
0.0000	1.3399	5.2079	1.2960	4.5921	5.6684	2.5035	3.2446	6.9439
0.1555	1.3426	5.1911	1.2150	4.5847	5.0138	2.7778	3.3750	6.9215
0.2930	1.3455	5.1738	1.1432	4.5770	4.4664	3.0637	3.5026	6.8984
0.4153	1.3483	5.1572	1.0795	4.5697	4.0045	3.3605	3.6276	6.8762
0.5250	1.3512	5.1400	1.0224	4.5621	3.6112	3.6682	3.7500	6.8534
0.6237	1.3539	5.1235	9.7086	4.5547	3.2735	3.9863	3.8702	6.8313
0.7132	1.3568	5.1065	9.2417	4.5472	2.9817	4.3150	3.9878	6.8087
0.7946	1.3596	5.0901	8.8179	4.5399	2.7272	4.6538	4.1036	6.7869
0.8689	1.3625	5.0733	8.4297	4.5324	2.5042	5.0030	4.2174	6.7644
0.9371	1.3653	5.0571	8.0745	4.5251	2.3077	5.3618	4.3294	6.7428
1.0000	1.3682	5.0404	7.7467	4.5176	2.1337	5.7309	4.4395	6.7205

The nature of the intermolecular interactions in liquid mixtures can be explained by the variation of excess molar volume (V^E), with respect to mole fraction, x_1, as shown in Figure 2. The expansion in molar volume can be attributed to the presence of weak forces of attraction [18] between the molecules. Garcia et al [19] reported similar results were. The negative trend in the values of V^E indicates about the compact packing of molecules which indicate strong intermolecular interactions, the other way it implies a loose packing of molecules. Sastry et al., [20] observed some similar results.

It can be observed from Figure 3 that the L_f^E values are negative which suggest that presence of specific interactions between unlike molecules of our systems[21].From figure-4 it can be observed, where the values are positive over the entire range of mole fraction.

The strength of bond formation in DMF+ NN Dimethylaniline system is less compared to that of another

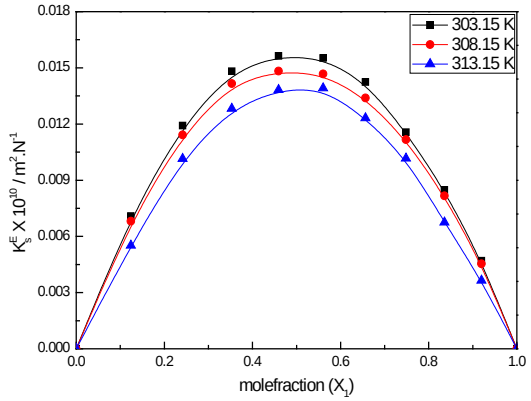
system, this is indicated by the maximum deviation observed in G^{*E}. Similar results were observed by earlier workers[22].

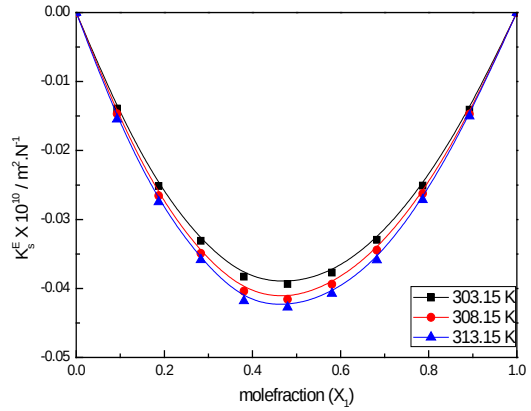
The excess values of Enthalpy (H^E) move towards positive from negative as shown in figure 5 with respect to the mole fraction, x_1. The positive values of H^E also increase, this is a clear indication that there are dispersion forces between molecules in this liquid mixtures [23]. It also suggests the existence of weak dipole-dipole interactions in these systems.

Also, the excess values correlate with one another and also supports the inferences drawn for these liquid mixtures.

TABLE 3 Acoustic impedance (Z), isentropic compressibility (K_s), molar volume (V), free length (L_f), free volume (V_f), internal pressure (π_i) and enthalpy (H) for the liquid mixtures at T=313.15 K.

X_1	**Z** x10^6 kg $m^{-2}s^{-1}$	**K_s** x10^{-10} $kg^{-1}ms^{-2}$	**V** x10^{-5} m^3mol^{-1}	**L_f** x10^{-11} m	**V_f** x10^{-5} m^3	**π_i** x10^8 Pa	**H** x10^4 $J.mol^{-1}$	**τ** x10^{-10} sec
(Dimethyl Formamide+ Aniline)								
0.0000	1.3199	5.3435	10.004	4.6927	4.5774	3.3586	3.3600	7.1246
0.1240	1.3453	5.1836	9.6616	4.6219	3.6111	3.8694	3.7384	6.9115
0.2415	1.3712	5.0296	9.3368	4.5528	2.9671	4.3661	4.0765	6.7061
0.3532	1.3972	4.8816	9.0296	4.4853	2.5085	4.8545	4.3834	6.5088
0.4593	1.4234	4.7399	8.7395	4.4197	2.1654	5.3387	4.6658	6.3199
0.5602	1.4499	4.6032	8.4636	4.3555	1.8998	5.8218	4.9273	6.1376
0.6565	1.4767	4.4717	8.2016	4.2929	1.6881	6.3056	5.1716	5.9623
0.7483	1.5035	4.3457	7.9534	4.2319	1.5157	6.7907	5.4009	5.7942
0.8359	1.5308	4.2239	7.7164	4.1722	1.3728	7.2794	5.6171	5.6319
0.9197	1.5583	4.1067	7.4907	4.1139	1.2525	7.7717	5.8215	5.4756
1.0000	1.5860	3.9938	7.2755	4.0570	1.1499	8.2685	6.0157	5.3251
(Dimethyl formamide + N-Methyl aniline)								
0.0000	1.3117	5.4058	4.8422	1.1840	2.2051	7.8121	3.7826	7.2078
0.1400	1.3123	5.4838	5.7453	1.1925	2.2129	6.9662	4.0018	7.3117
0.2681	1.3124	5.5666	6.5752	1.2015	2.1937	6.4004	4.2080	7.4222
0.3858	1.3118	5.6546	7.2955	1.2110	2.1443	6.0471	4.4083	7.5394
0.4942	1.3106	5.7478	7.9214	1.2209	2.0822	5.8093	4.6017	7.6638
0.5945	1.3099	5.8378	8.4848	1.2304	2.0151	5.6452	4.7874	7.7837
0.6874	1.3065	5.9515	8.9752	1.2423	1.9418	5.5398	4.9719	7.9353
0.7738	1.3036	6.0625	9.4133	1.2539	1.8691	5.4715	5.1505	8.0833
0.8543	1.3002	6.1790	9.8025	1.2659	1.7970	5.4332	5.3255	8.2387
0.9295	1.2959	6.3045	10.153	1.2787	1.7260	5.4190	5.4981	8.4060
1.0000	1.2912	6.4364	10.452	1.2920	1.6570	5.4232	5.6683	8.5819
(Dimethyl formmide +NN dimethyl aniline)								
0.0000	1.3200	5.3407	1.3023	4.6915	5.8607	2.4941	3.2480	7.1210
0.1555	1.3224	5.3248	1.2210	4.6845	5.2368	2.7531	3.3616	7.0997
0.2930	1.3250	5.3083	1.1490	4.6772	4.7084	3.0223	3.4727	7.0778
0.4153	1.3275	5.2925	1.0851	4.6703	4.2569	3.3010	3.5818	7.0567
0.5250	1.3301	5.2762	1.0277	4.6630	3.8679	3.5895	3.6888	7.0349
0.6237	1.3326	5.2605	9.7605	4.6561	3.5305	3.8871	3.7940	7.0140
0.7132	1.3352	5.2443	9.2921	4.6489	3.2357	4.1942	3.8973	6.9924
0.7946	1.3377	5.2288	8.8669	4.6420	2.9768	4.5101	3.9991	6.9717
0.8689	1.3404	5.2127	8.4774	4.6349	2.7480	4.8353	4.0991	6.9503
0.9371	1.3429	5.1973	8.1210	4.6280	2.5449	5.1691	4.1978	6.9297
1.0000	1.3455	5.1814	7.7921	4.6209	2.3635	5.5121	4.2951	6.9085





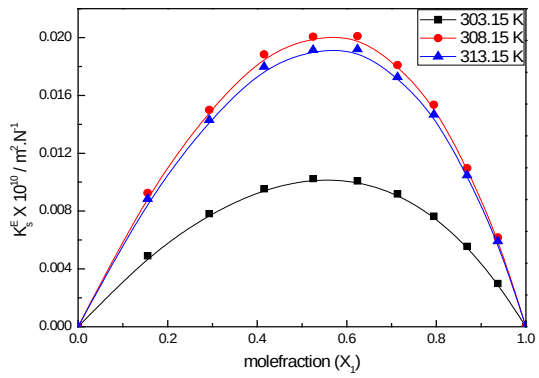
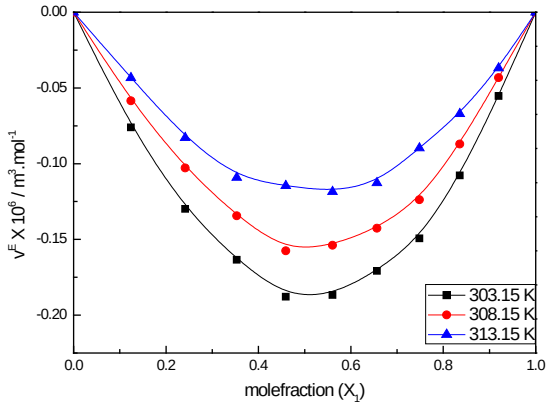
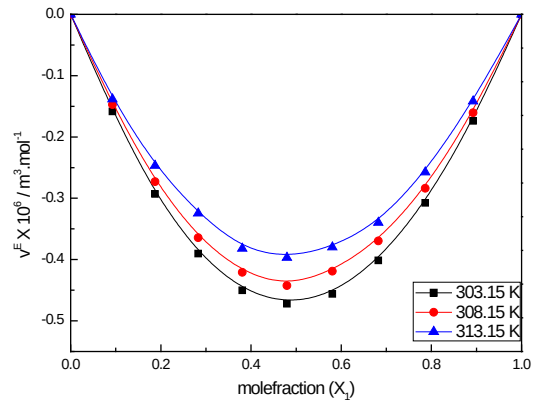


Fig.1 Variation of Excess Isentropic compressibility with respect to mole fraction at various temperatures for (a) Dimethyl formamide + Aniline ; (b) Dimethyl formamide+N-Methyl aniline (c) Dimethyl formamide + NN Dimethylaniline.





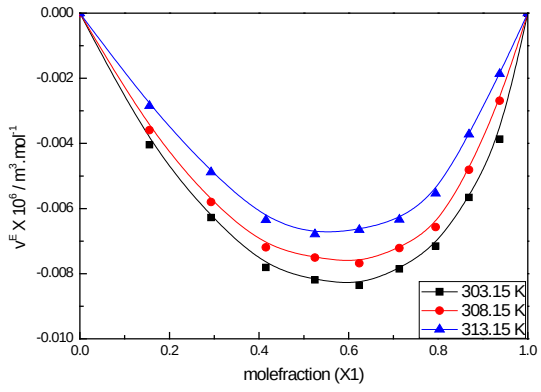


Fig.2 Variation of Excess Volumes with respect to mole fraction at various temperatures for (a) Dimethyl formamide + Aniline ; (b) Dimethyl formamide+N-Methyl aniline (c) Dimethyl formamide + NN Dimethylaniline.

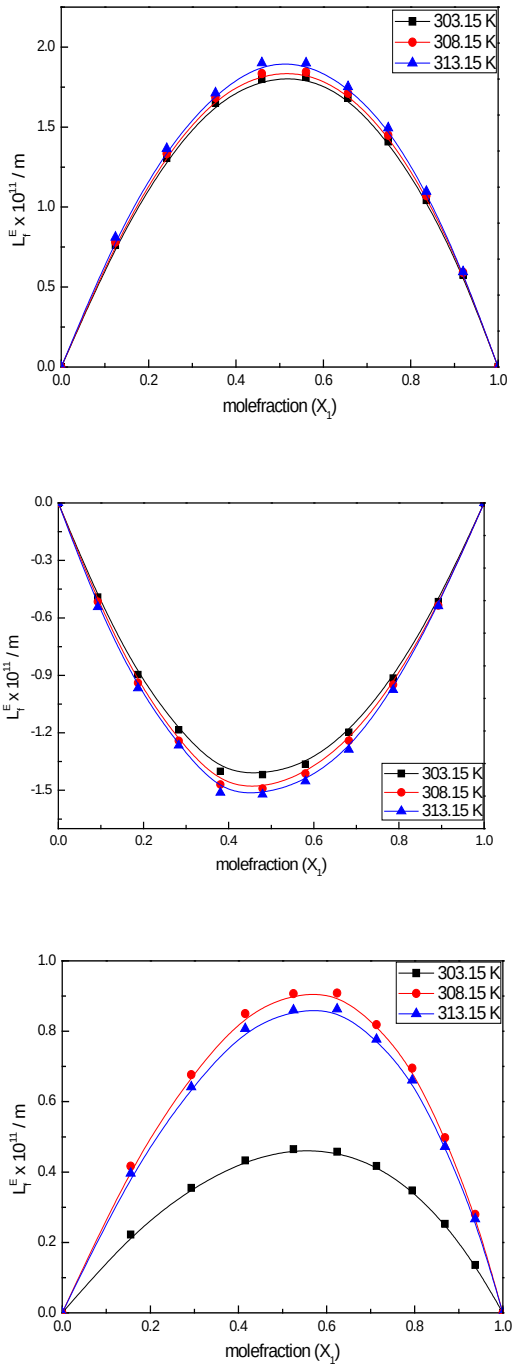


Fig.3 Variation of excess free length with respect to mole fraction at various temperatures for (a) Dimethyl formamide + Aniline ; (b) Dimethyl formamide+N-Methyl aniline (c) Dimethyl formamide + NN Dimethylaniline.

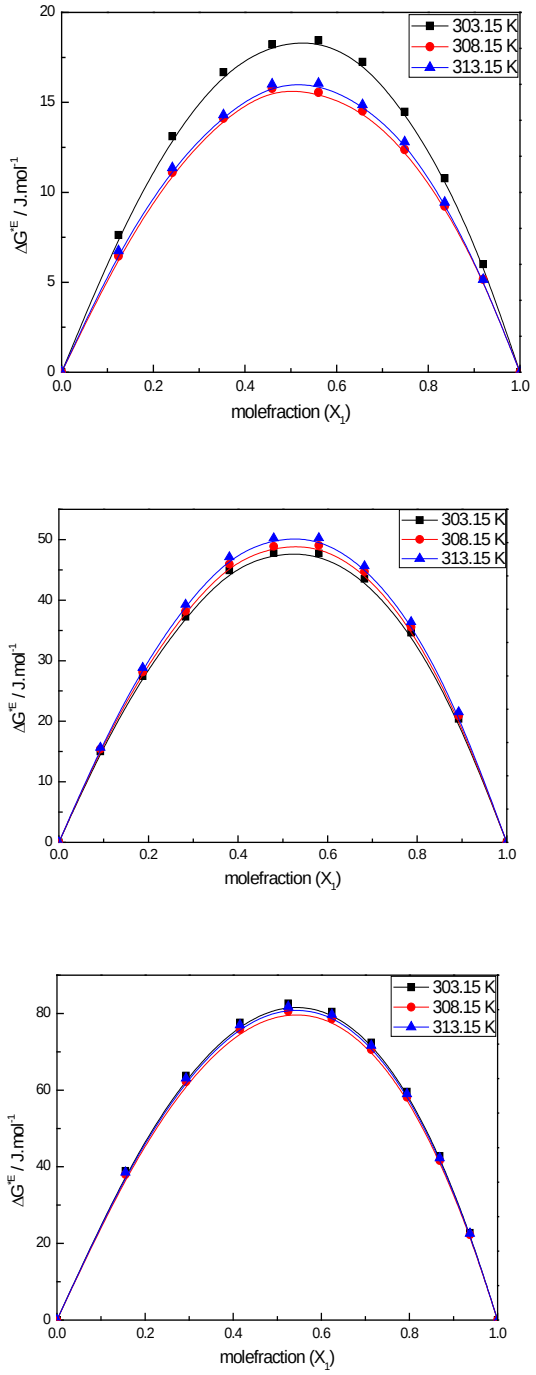


Fig.4 Variation of excess Gibb's free energy with respect to mole fraction at various temperatures for (a) Dimethyl formamide + Aniline ; (b) Dimethyl formamide+N-Methyl aniline (c) Dimethyl formamide + NN Dimethylaniline

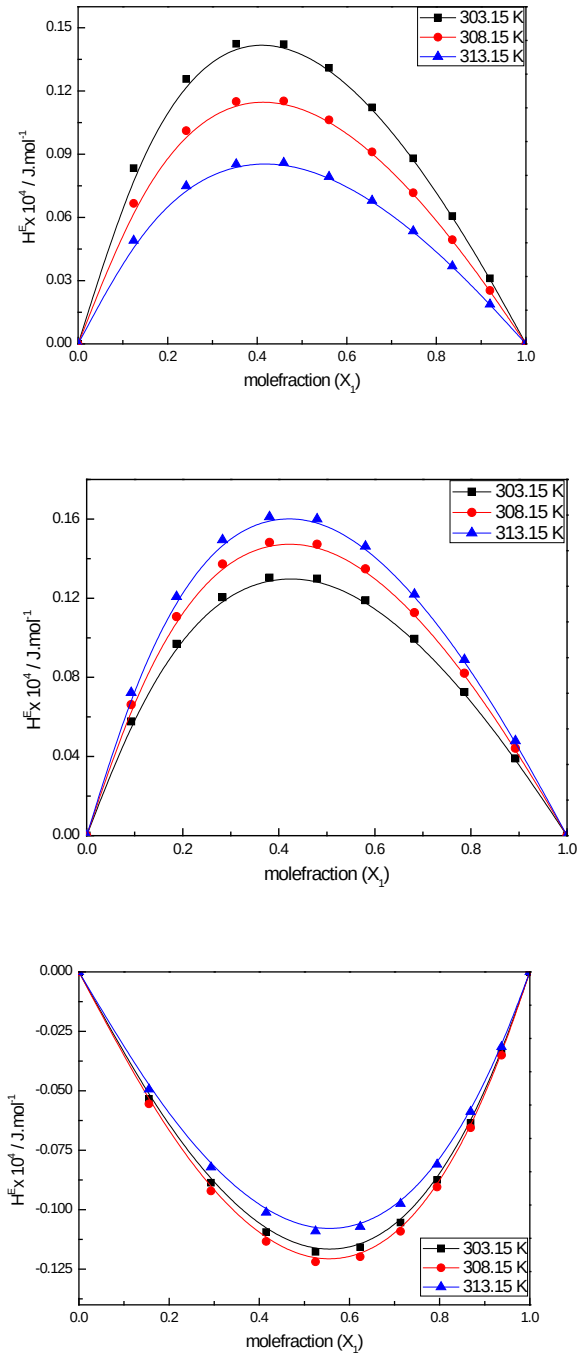


Fig.5 Variation of excess enthalpy with respect to mole fraction at various temperatures for (a) Dimethyl formamide + Aniline ; (b) Dimethyl formamide+N-Methyl aniline (c) Dimethyl formamide + NN Dimethylaniline.

CONCLUSION

From the experimentally determined values of the speed of sound, density and viscosity, the excess values of K_S^E, V^E, L_f^E, G^{*E} and H^E were calculated. Interactions between these mixtures were identified by studying these excess values. Positive values were observed in the case of excess isentropic compressibility, Gibbs free energy of activation while negative values were obtained in excess free length and excess molar volume. The presence of weak bonding between the component molecules can be confirmed with these values. Apart from the above-said values/reasons, the difference in molar masses of the liquid molecules is also responsible for the non-existence of specific interactions.

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