

Excess molar volumes & viscosity deviations of liquid mixtures of N, N, Dimethylaniline & N, N Diethylaniline in methanol and ethanol.

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Aim:

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Abstract:

The values of density & viscosity for the binary liquid mixtures of N, N dimethylaniline & N, N diethylaniline alkanols (methanol & ethanol) have been measured over entire range of composition at $T = 298\text{K}$ to 313K .

Using these excess molar volumes (V^E) & deviations in viscosity have been correlated with Redlich - Kister type polynomial equation to derive the binary coefficients & standard deviation.

Introduction:

Experimental viscosities provide information on the structures of liquids & are required in the design processes which involve mass transfer, fluidflow etc. The thermodynamic properties of the solvents such as density (ρ) and viscosity (η), which are two important physical

properties of solvent systems and are often used to explain the medium effects of solvent on transport phenomenon. These properties are functionally dependent on temp. and at least for binary mixtures, on the component of solvent systems. The change in these properties with mole fraction often provides more detailed evidence about the structural character of the system themselves.

Alcohols are strongly self-associated molecules through hydrogen bonding and for a binary solutions rich in alcohols, three dimensional network of hydrogen bond is believed to be present, substituted anilines also exist as associated structure in liquid state. Alcohol is proton donating & aniline is proton accepting molecules. The molecules N,N Dimethylaniline (DMA) & N, N Diethylaniline (DEA) has many Industrial applications, of which, DMA is a key precursor to commercially important triarylmethane dyes have specific associations with alcohols & they have interesting solvent properties, thus upon mixing these anilines with alcohols of varying chain lengths & branching interesting properties due to specific interaction arising from the charge transfer. Dipole-dipole, donor-acceptor & hydrogen bond formation forces may be observed. In view of this we report here in the density and viscosity data for the Fourth binary systems formed by methanol & ethanol with DMA & DEA at atmospheric pressure over the entire composition range at temp.

$T = 298\text{K}$ to 313K . These results have been used to calculate excess volumes & deviations in viscosity.

Experimental:

N, N Dimethyl aniline (99.5% pure) and N, N Diethyl aniline (99.5% pure) procured from S.D. Fine chemical Ltd. India were used after single distillation of Methanol (99.5% pure) and Ethanol (99% pure) were purchased from S.D.Fine chemical Ltd. India. These samples were used after single distillation. The purities of solvents were further ascertained by GLC and comparing their densities and viscosities at $T = 298\text{k}$, which agreed reasonably with corresponding to reported values (Table 1).

Table - 1:

Comparison of Experimental densities (ρ /g cm⁻³) and viscosities (η /mpa.S) of pure liquids at different temperature with literature data.

Component	ρ (g cm ⁻³)		η (mpa.S)	
	Expt.	Lit.	Expt.	Lit.
		298k		
N,N DMA	0.9523	0.9522	1.288	1.282
N,N DEA	0.9302	0.9307	1.935	1.933
Methanol	0.7866	0.7866	0.5510	0.5510
Ethanol	0.7849	0.7849	1.082	1.082

Binary mixtures were prepared by mass in air-tight glass bottles. The mass measurements were performed on a Dhona 160 D (India) singlepan analytical balance, with a precision of 0.01 mg. The required properties were measured within a day of preparing the mixture. The possible error in mple fraction is estimated to be less than $\pm 1 \times 10^{-4}$.

Densities of pure liquids & their mixtures were determined by using 15cm³ bicapillary pycnometer as reported earlier. The pycnometer was calibrated using conductivity water with 0.99795cm⁻³ as its density at 298.15k. The uncertainty in density & excess molar volume values is found to be $\pm 0.00005\text{gcm}^{-3}$ & $\pm 0.001\text{ cm}^3\text{ mol}^{-1}$ respectively.

Ubbelonde viscometer having a capacity of about 15ml was used to measure the flow times of pure liquids & liquid mixtures. The dynamic viscosities were measured using this viscometer with an electronic stopwatch with readability of $\pm 0.015\text{s}$ for flow time measurement was repeated number of times (usually 5-6 times).

The different readings did not deviate from the mean by more than 0.2s and the results were arranged since all flow times were greater than 200s and capillary radius (0.5mm) was far less than its length (50 to 60mm), the kinetic energy & end corrections, respectively, were found to be negligible. Viscosity value (η) of pure liquids and mixtures were calculated using the relation;

$$\eta = \frac{a}{t} \rho \quad \text{-----1}$$

Where ‘a’ and ‘b’ represent the characteristics constants of the viscometer, ρ is the density and t the flow time. The uncertainty in viscosity and viscosity deviation values was found to be ± 0.001mpa.S and ±0.005 mpa.S respectively.

Table-2:

The values of excess molar volumes ($V^E / \text{cm}^3 \text{mol}^{-1}$) & deviations in viscosity ($\Delta\eta / \text{g cm}^{-1} \text{S}^{-1}$) for the binary liquid mixtures of N, N Dimethylaniline (1) with alkanols (2) at different temperatures.

N, N Dimethylaniline (1) + methanol (2)

X ¹	$V^E / \text{cm}^3 \text{mol}^{-1}$				$\Delta\eta / \text{g cm}^{-1} \text{S}^{-1}$			
	298k	303k	308k	313k	298k	303k	308k	313k
0.0000	0.0000	0.0000	0.0000	0.0000	0.55100	0.52300	0.49900	0.44800
0.1017	-0.1522	-0.1276	-0.0898	-0.0586	0.63250	0.59410	0.56360	0.50960
0.2026	-0.2418	-0.1970	-0.1597	-0.1242	0.71210	0.66510	0.62840	0.57110
0.3014	-0.2852	-0.2494	-0.2152	-0.1764	0.78910	0.73310	0.69170	0.63160
0.4047	-0.3055	-0.2726	-0.2435	-0.2106	0.86690	0.80230	0.75590	0.69336
0.5047	-0.3138	-0.2853	-0.2553	-0.2221	0.94110	0.86830	0.81710	0.75260
0.6073	-0.3034	-0.2704	-0.2405	-0.2079	1.01600	0.93440	0.87760	0.81050
0.7041	-0.2768	-0.2392	-0.2115	-0.1815	1.08500	0.99510	0.93350	0.86307
0.8053	-0.2345	-0.1919	-0.1673	-0.1292	1.15600	1.05700	0.99020	0.91880
0.9016	-0.1493	-0.1267	-0.0944	-0.0609	1.22100	1.11400	1.04300	0.97130
1.0000	0.0000	0.0000	0.0000	0.0000	1.28800	1.17400	1.09900	1.02600

N, N Dimethylaniline (1) + Ethanol (2)

0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0511	-0.1098	-0.0764	-0.0600	-0.0358	0.0135	0.0091	0.0050	0.0026
0.1125	-0.2048	-0.1677	-0.1330	-0.0988	0.0078	0.0023	-0.0018	-0.0055
0.2329	-0.3135	-0.2738	-0.2234	-0.1747	0.0020	-0.0048	-0.0089	-0.0129
0.3536	-0.3796	-0.3441	-0.3039	-0.2576	-0.0038	-0.0099	-0.0131	-0.0164
0.4772	-0.3913	-0.3589	-0.3200	-0.2760	-0.0053	-0.0115	-0.0143	-0.0169
0.5973	-0.3676	-0.3265	-0.2787	-0.2378	-0.0041	-0.0095	-0.0131	-0.0151
0.7179	-0.2989	-0.2586	-0.2130	-0.1641	0.0021	-0.0056	-0.0089	-0.0125
0.8385	-0.2041	-0.1642	-0.3143	-0.0899	0.0083	0.0013	-0.0037	-0.0075
0.9590	-0.1096	-0.0698	-0.0454	-0.0337	0.0134	0.0092	0.0044	0.0022
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

Table: 3 — The Value of excess molar volumes ($V^E / \text{cm}^3 \text{mol}^{-1}$) & deviations in viscosity ($(\Delta\eta / \text{g cm}^{-1} \text{S}^{-1})$) for the binary liquid mixtures of N, N Dimethyl aniline (1) with alkanols (2) at different temperatures.

X ¹	$V^E / \text{cm}^3 \text{mol}^{-1}$				$\Delta\eta / \text{g cm}^{-1} \text{S}^{-1}$			
	298k	303k	308k	313k	298k	303k	308k	313k
N, N Diethylaniline (1) + methanol (2)								
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1004	-0.6016	-0.4852	-0.3622	-0.2880	0.0101	-0.0029	-0.0071	-0.0200
0.2006	-0.7645	-0.6472	-0.5028	-0.3933	0.0499	0.0369	-0.0249	0.0061
0.3007	-0.8280	-0.6823	-0.5428	-0.4090	0.0753	0.0600	0.0500	0.0320
0.4008	-0.7682	-0.5920	-0.4250	-0.2747	0.0873	0.0789	0.0701	0.0550
0.5002	-0.4811	-0.2937	-0.0736	-0.1515	0.0912	0.0857	0.0777	0.0683
0.6004	0.0946	0.3299	0.4985	0.7352	0.0870	0.0787	0.0691	0.0546
0.7007	0.6374	0.8116	0.9641	0.1485	0.0737	0.0596	0.0494	0.0299
0.8005	0.8437	0.9646	1.0895	0.2637	0.0492	0.0360	0.0253	0.0046
0.9004	0.6086	0.7571	0.9093	0.9980	0.0094	-0.0017	-0.0070	-0.0189
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

N, N Diethylaniline (1) + Ethanol (2)								
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1025	-0.1616	-0.1042	-0.0708	-0.0440	0.0032	-0.0081	-0.0150	-0.0199
0.2043	-0.2676	-0.2210	-0.1570	-0.0948	0.0302	0.0213	-0.0116	0.0020
0.3058	-0.2766	-0.2304	-0.1723	-0.1079	0.0547	0.0458	0.0317	0.0221
0.4073	-0.1732	-0.1260	-0.0761	-0.0314	0.0664	0.0615	0.0519	0.0397
0.5088	-0.0563	-0.0076	0.0555	0.1153	0.0720	0.0661	0.0601	0.0534
0.6097	0.0947	0.1725	0.2357	0.3273	0.0661	0.0601	0.0497	0.0403
0.7099	0.3011	0.3690	0.4601	0.5435	0.0538	0.0436	0.0308	0.0197
0.8098	0.5233	0.5951	0.6690	0.7421	0.0269	0.0204	0.0090	0.0003
0.9095	0.3922	0.4478	0.5137	0.6046	-0.0020	-0.0047	-0.0135	-0.0191
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

In all of the property measurements, the temp. was controlled with in $\pm 0.01\text{k}$ using a constant temp. bath (INSREF model IRI-0166, India).

Result & Discussion:

The values of density and viscosity of binary mixtures of DMA & DEA with Ethanol, and methanol at T = 298k to 313k along with the molefraction are given in the Table 2 & 3. The excess molar volumes (V^E) have been evaluated from density using,

$$V^E = V_m - (X_1V_2 + X_2V_2) \text{-----} 2$$

$$V_m = (X_1M_1 + X_2M_2) / \rho_m \text{-----} 3$$

Where V_m and ρ_m are the molar volume and density of the mixture, X_1 , V_1 , M_1 , and X_2 , V_2 , M_2 are the mole fraction, molar volume and molecular weight of pure components 1 and 2 respectively. The deviation in viscosity is calculated using the relation;

$$\Delta\eta = \eta_m - (X_1 \eta_1 + X_2 \eta_2) \text{-----} 4$$

Where η_m , η_1 and η_2 are the pure viscosities of the liquid mixtures & of the pure components 1 and 2 respectively; X_1 & X_2 are the mole fractions of the pure components, 1 & 2 in the liquid state.

The excess properties y^E are fitted by the method of non-linear least squares to a Redlich - Kister type polynomial;

$$y^E = X_1 X_2 \sum A_i (X_1 - X_2)^i \text{-----} 5$$

In each cases, the optimum number of coefficients A_i was determined from examination of the variation of standard deviation (δ) as calculated by;

$$\delta(y) = \left[\frac{\sum (y^{E_{obs}} - y^{E_{cal}})^2}{n-m} \right]^{1/2} \text{-----} 6$$

Where n represents the number of experimental points and m is the number of coefficients used in fitting the data. The estimated values of A_i & δ for V^E & $\Delta\eta$ are given in the Tables 4 and 5. In all cases, the best fit was found by using fine adjustable fitting coefficients in equation (5).

Excess Molar volume (V^E):

The data included in Table 2 shows that excess molar volumes V^E are negative in the mixture of DMA with ethanol over a whole range of temp. The V^E values for the binary mixtures of DMA + methanol shows negative peaks over a whole range of temp.

The & data included in Table 3 Shows that excess molar volumes V^E are negative in lower mole fraction values for both DEA + methanol & DEA + Ethanol binary systems, which further decreased at higher temp. that is positive V^E versus molar fraction, (X_1) of the binary liquid mixtures of DEA with methanol & ethanol is depicted in the fig. 2 .

The observed values of excess molar explained in terms of several effects, which may arbitrarily be divided into physical, chemical and geometrical contributions. Treszcanowicz et al¹⁵ & Aminabhavi et al¹⁶ suggested that the V^E is the resultant contribution from several opposing effects. The observed negative V^E values is of DMA + Methanol and

ethanol due to specific interactions between constituent molecules occur through hydrogen bonding of the type.

Table – 4 : The binary coefficient (Ai) and Standard deviation (δ) of N,N Dimethyl aniline with alkanols at different temp.

Property	Temp(k)	A ₀	A ₁	A ₂	A ₃	A ₄	A ₅	(δ)
N,N , Dimethyl aniline (1) + Methanol (2)								
V ^E (cm ³ mol ⁻¹)	298	-1.2457	0.0367	-0.6305	-0.0836	-0.1066	0.0763	0.0028
	303	-1.1380	0.0474	0.0007	-0.1047	-0.6678	0.0116	0.0033
	308	-1.0125	0.0650	-0.0503	-0.5536	0.0486	0.5805	0.0026
	313	-0.8824	-0.0247	0.0822	-0.1989	0.4050	0.2947	0.0026
oη (g cm ⁻¹)	298	0.0725	-0.0069	0.0160	0.0408	-0.0377	-0.0616	0.0003
	303	0.0662	0.0016	0.0092	-0.0024	-0.0550	-0.0120	0.0002
	308	0.0604	0.0007	-0.0173	0.0171	-0.0308	-0.0359	0.0002
	313	0.0509	-0.0023	-0.0518	0.0004	0.0246	-0.0034	0.0002

N,N Dimethyl aniline (1) + Ehanol(2)								
V ^E (cm ³ . mol ⁻¹)	298	- 1.6025	0.1164	0.6845	1.3478	-2.1171	-2.1479	0.0107
	303	- 1.4306	0.1553	0.3498	1.1143	0.7484	1.7053	0.0070
	308	- 1.2534	0.2998	0.4753	0.0215	-0.4947	-0.3755	0.0080
	313	- 1.0823	0.2144	0.8305	0.6032	-0.5792	-1.1204	0.0103
oη (g cm ⁻³)	298	0.0944	0.3041	-2.2072	-3.1376	5.0086	5.7344	0.00327
	303	- 0.0325	-0.0154	-0.1803	0.1525	0.5692	-0.1416	0.0037
	308	- 0.0477	-0.0093	-0.1497	0.1239	0.3993	-0.1405	0.0028
	313	- 0.0575	-0.0042	-0.1178	0.1135	0.3770	-0.1385	0.0027

Table-5: The binary coefficients (Ai) and Standard deviation (δ) of N,N Diethyl aniline with alkanols at different temp.

Property	Temp(k)	A ₀	A ₁	A ₂	A ₃	A ₄	A ₅	(δ)
N,N Diethyl aniline (1) + methanol (2)								
V ^E (cm ³ .mol ⁻¹)	298	-1.8614	9.3233	10.9354	-5.6624	- 13.9764	8.5582	0.0141

	303	-0.9469	9.7748	8.1194	-6.5685	-6.7180	7.4828	0.0450
	308	-0.1514	10.0193	6.8736	-8.5439	-2.9770	10.4638	0.0337
	313	0.6731	10.6430	6.7695	-9.6080	-2.5869	10.8720	0.0214
η (g cm ⁻¹)	298	0.3585	-0.0092	0.1557	0.0299	-0.8515	-0.0305	0.0017
	303	0.3293	0.0035	-0.0295	-0.0482	-0.8116	0.0901	0.0042
	308	0.2996	-0.0113	-0.2176	-0.0714	-0.5761	-0.0802	0.0031
	313	0.2546	-0.0046	-0.5369	-0.0381	-0.3035	0.0906	0.0040

V^E (cm ³ . mol ⁻¹)	298	0.3467	2.3024	2.8855	7.7503	-0.0342	-7.9495	0.0271
	303	-0.1178	2.5265	3.0135	7.4918	0.7713	-8.0676	0.0266
	308	0.1101	2.8164	3.7270	6.2038	0.4528	-6.1590	0.0216
	313	0.3702	3.3145	4.1247	3.4047	0.7469	-1.8952	0.0164
η (g cm ⁻¹)	298	0.2819	0.0207	-0.0207	-0.1019	-0.7065	0.1752	0.0012
	303	0.2611	0.0009	-0.1739	0.0217	-0.5216	0.0580	0.0011
	308	0.2265	0.0123	-0.3114	-0.0382	-0.4374	0.0777	0.0036
	313	0.1752	0.0982	-0.0788	-0.9244	-0.8286	1.2274	0.0095

N – H.....O & O – H.....N while at intermediate concentration of DMA the dispersion forces prevail due to different sizes of DMA & methanol and ethanol molecules.

The variation of V^E with X_1 , of DEA + methanol and ethanol systems at all temp. yield a sigmoidal nature of graph (fig 2). The V^E for the mixture of DEA with alkanols have negative values at concentration of DEA <0.6 mole fraction. In this region the added DEA molecules rupture the hydrogen bonds of aggregates of alkanols and form new hydrogen bond proton of the alkanols and unpaired electron pair of Nitrogen of DEA. However in the binary mixture with DEA concentration >0.6 mole fraction, the excess DEA molecules ruptures the hydrogen bonds of very few alkanols molecules & exert Steric hindrance in positive V^E values.

Viscosity Deviation (η):

The data included in The Table 2 & 3 show that η values for mixture DMA + ethanol are positive in lower and higher concentration ranges & negative in the intermediate concentration region ($X_1 = 0.1125$ to 0.8385). The plots of η versus X_1 shows the variation with temp. In fig. 3 & 4.

The η values for the mixture DEA + ethanol & DEA + methanol are mostly positive. The positive η values are attributed to strong intermolecular interactions. However, there is general decrease in viscosity deviation with increasing temp.

The correlation between the sign of η & V^E has been observed for a number of binary solvent systems, η being negative whereas V^E is positive or vice versa. The observed data of V^E and η do not go by the general rule. For such systems, Rastogi et al 18, suggested

that the observed excess property is a combination of an interaction & non interaction part. Thus we can safely say,

$$Y^E (\text{observed}) = Y^E (\text{interaction}) + Y^E (\text{size effect})$$

Where Y^E refers to the excess of deviation in the property. The non- interaction part in the form of the size effect can be comparable to the interaction part. Based on this theory, the observed incongruity may be accredited to the size effect. The molecular interactions between the binary solvent systems appear to be dominating by physical forces.

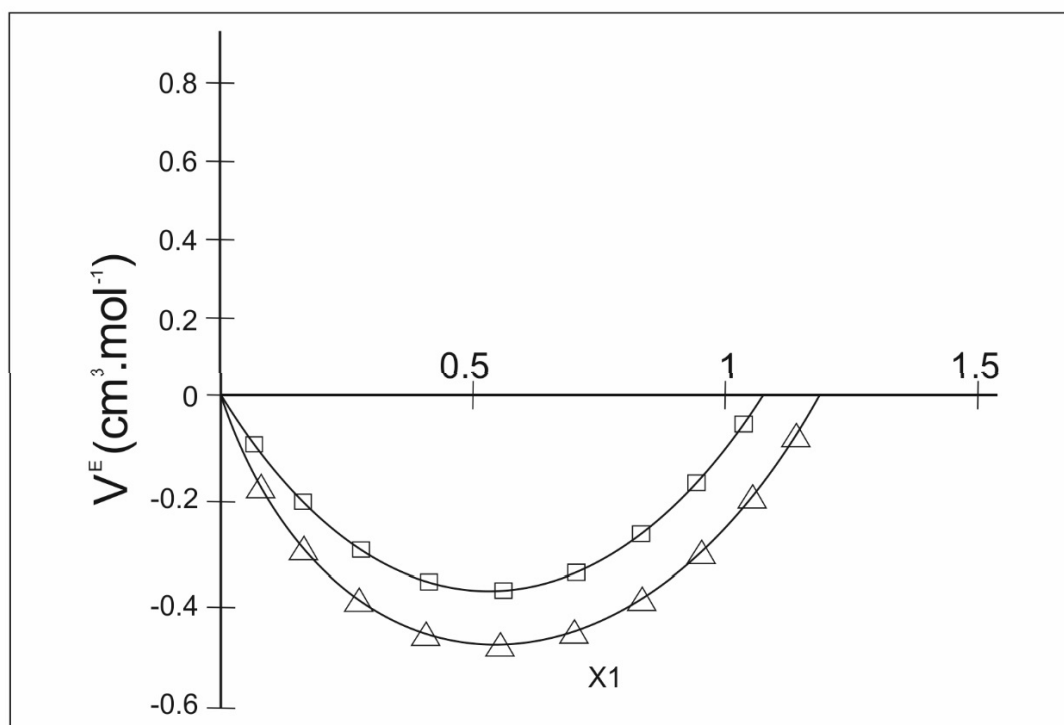


Fig 1: Variation of excess molar volume V^E versus mole fraction (X_1) of the binary mixtures of (Δ) DMA + methanol and (\square) DMA + ethanol attemp 303k.

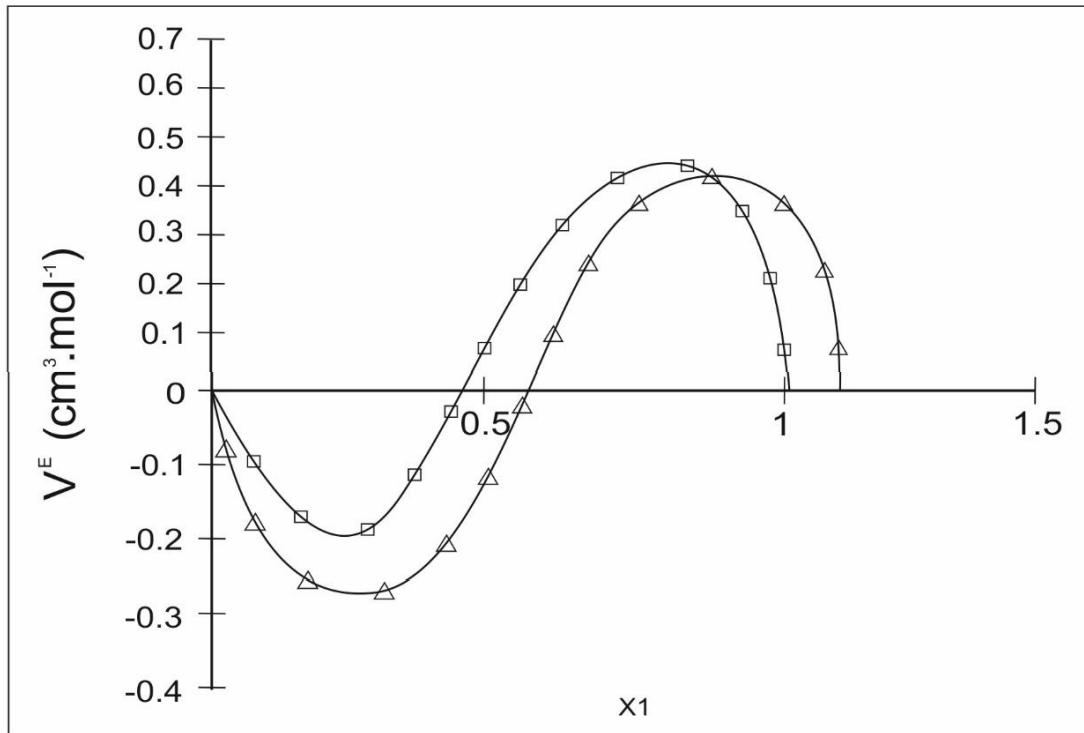


Fig 2: Variation of excess molar volume V^E versus mole fraction (X_1) of the binary mixtures of (Δ) DEA + methanol and (\square) DEA + ethanol at temp 303k.

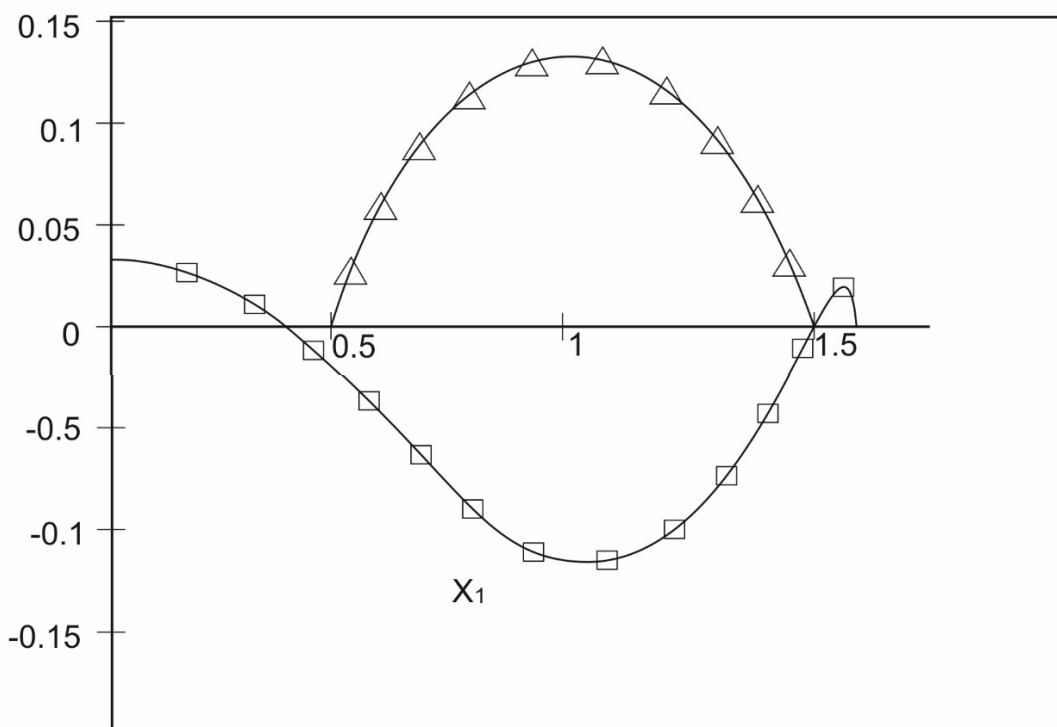


Fig 3: Variation of excess molar volume of Δ (DMA) + methanol & \square (DMA) + ethanol at temp 303k.

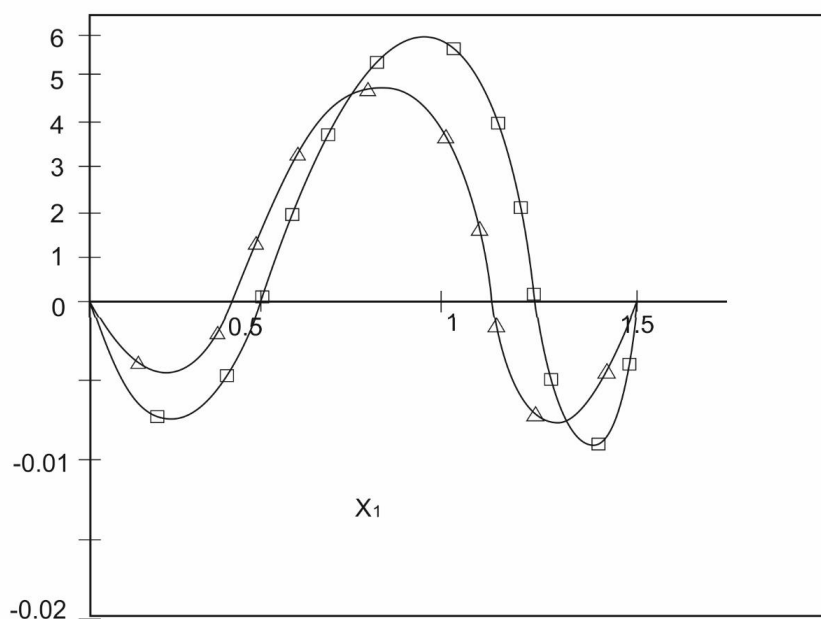


Fig 4: Variation of excess molar volume V^E versus mole fraction (x_1) of the binary mixtures of Δ (DEA) + methanol and \square (DEA) + ethanol attemp 303k.

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