Excess molar volumes of the systems *m*-xylene + 1-propanol, +2-propanol, +1-butanol, +2-methyl-2-propanol

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Densities, ρ, of the systems *m*-xylene + 1-propanol, + 2-propanol, + 1-butanol and + 2-methyl-2-propanol have been determined from 303.15 to 323.15 K with an interval of 5 K. Excess molar volumes, V_m^E, have been calculated from density data. ρ and V_m^E have been fitted to appropriate polynomial equations and plotted against mole fraction of alkanols. For the systems, *m*-xylene + 1-propanol and *m*-xylene + 1-butanol, sigmoid type of curves have been observed, but for the systems, *m*-xylene + 2-propanol and *m*-xylene + 2-methyl-2-propanol, positive V_m^E have been observed for the whole range of composition. The observed volumetric properties have been explained by the concepts: (a) dissociation of alkanols in *m*-xylene-rich region, (b) partial accommodation of *m*-xylene into interstitial positions of 1-alkanols in alkanol-rich region, (c) donor–acceptor interaction between alkanols and *m*-xylene and (d) steric hindrance of branched alkanols.

Keywords: Excess molar volume; *m*-Xylene; 1-Propanol; 2-Propanol; 1-Butanol and 2-Methyl-2-propanol

1. Introduction

In recent years, a number of studies have been made on the volumetric properties of the mixtures of alkanols + aromatic hydrocarbons. Singh *et al.* [1] reported on excess molar volumes of 1-propanol and 2-propanol with aromatic hydrocarbons at 298.15 K. They observed V_m^E curves to be sigmoid for 1-propanol + toluene, +*o*-xylene, +*m*-xylene and +*p*-xylene, but for mixtures with 2-propanol, V_m^E have been found to be positive for most of the concentration range, except for slightly negative values at highly rich alkanol region. The excess molar volumes for the systems 2-propanol and 2-methyl-2-propanol with xylenes were reported at 298.15 K by Ouyang *et al.* [2]. Nikam *et al.* [3] studied the excess volumes for mixtures of toluene

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with a number of alkanols, such as methanol, ethanol, 1-propanol, 1-butanol, 1-pentanol and 2-methyl-2-propanol at 303.15, 308.15 and 313.15 K. They observed negative V_m^E for toluene + methanol for the whole range of composition, both positive and negative V_m^E for the systems, toluene + ethanol, + 1-propanol, + 1-butanol, and positive V_m^E for the systems toluene + 1-pentanol, + 2-methyl-2-propanol, at 303.15 K. In our earlier study [4] we had observed a very slight negative V_m^E for toluene + 1-pentanol system in alkanol-rich region at the same temperature, contrasting the observation of Nikam et al. For the systems, toluene + 2-pentanol, + 3-pentanol, however, we observed positive V_m^E for the whole range of composition. In another study [5], we found positive V_m^E for the systems, 1-hexanol + benzene, + cyclohexane, but negative V_m^E for the system, 1-hexanol + n-hexane, for the whole range of composition.

On the analysis of the results so far reported by different workers, the following points emerge:

(i)   For 1-alkanols + xylenes, V_m^E show generally sigmoid type of curves.
(ii) Mixtures of branched alkanols with xylenes generally show positive V_m^E for the whole range of composition and that the magnitude of the V_m^E depends largely on the extent of branching.
(iii) The relative position of the two methyl groups in xylenes does not have much influence on the magnitude and pattern of V_m^E curve for a particular alkanol.

In our present system we have chosen two isomers of propanol, 1-propanol and 2-propanol; and two isomers of butanol, 1-butanol and the most extensively branched isomer, 2-methyl-2-propanol, with a view to examine the branching effect of the alkanols on V_m^E within the respective isomers.

2. Experimental

The chemicals used for this investigation have been procured from Aldrich with quoted purities: 1-propanol (99.5%), 2-propanol (99.5%), 1-butanol (99.8%), 2-methyl-2-propanol (99.5%) and *m*-xylene (99+%). These have been used without any further treatment, except that the alkanols were kept over molecular sieves (4 A) for at least three weeks, prior to their use. As measures of purity check, the densities of pure liquids were compared with the available literature values, which show a satisfactory agreement.

The density was measured by a 25-mL specific gravity bottle, previously calibrated with redistilled water. An analytical balance weighing up to ±0.0001 g was used in the density measurement. The same balance was used to make solutions, the calculated mole fractions were accurate up to the fourth decimal place. Temperature was controlled by a thermostatic water bath with a fluctuation of ±0.05 K. The average uncertainty in the measured density was $1.4 \times 10^{-1}\,\mathrm{kg\,m^{-3}}$.

3. Results and discussion

Densities of the pure compounds, *m*-xylene, 1-propanol, 2-propanol, 1-butanol and 2-methyl-2-propanol at temperatures ranging from 298.15 to 323.15 K have been shown in table 1. The literature values have been cited which compare well with our

Table 1.   Densities, $\rho \times 10^{-3}$ (kg m^{-3}), of pure compounds at different temperatures.

Compound	Temperatures/K					
	298.15	303.15	308.15	313.15	318.15	323.15
m-Xylene	0.8597 (0.85979)[a] (0.85986)[b]	0.8554 (0.8553)[c]	0.8511 (0.8521)[c]	0.8468	0.8425	0.8381
1-Propanol	0.7994 (0.7996)[d,e]	0.7953 (0.7956)[e,f] (0.7954)[g] (0.79584)[h] (0.7955)[i]	0.7912 (0.7912)[e] (0.7913)[g]	0.7871 (0.7874)[f] (0.7871)[g] (0.78747)[h]	0.7830	0.7789 (0.7789)[j]
2-Propanol	0.7814 (0.78126)[b,k] (0.7816)[d]	0.7768 (0.7805)[d] (0.7769)[l]	0.7724 (0.7725)[e] (0.7726)[l]	0.7679 (0.7683)[l]	0.7634 (0.7636)[l]	0.7588 (0.7589)[l]
1-Butanol	0.8054 (0.8057)[e] (0.80575)[k]	0.8019 (0.8018)[e,i] (0.80194)[h]	0.7980 (0.7981)[e,m]	0.7941 (0.79405)[h] (0.7943)[m]	0.7902	0.7863 (0.7862)[j] (0.7865)[n]
2-Methyl-2-propanol	0.7807 (0.7809)[d]	0.7754 (0.77541)[k]	0.7704 (0.7643)[e]	0.7650	0.7599	0.7546

[a]Ref. [2]; [b]Ref. [9]; [c]Ref. [6]; [d]Ref. [10]; [e]Ref. [11]; [f]Ref. [13]; [g]Ref. [3]; [h]Ref. [14]; [i]Ref. [17]; [j]Ref. [18]; [k]Ref. [8]; [l]Ref. [12]; [m]Ref. [15]; [n]Ref. [16]

experimental values. The densities have been fitted to a polynomial equation of the form,

$$\rho/(\text{kg m}^{-3}) = \sum_{i=0}^{n} A_i x_2^i \tag{1}$$

The coefficients of this equation and the squares of the regression coefficients are shown in table 3. Figure 1 shows the variation of densities for all the systems at 303.15 K, as a function of mole fraction of alkanols.

Excess molar volumes, V_m^E, have been calculated by the following equation,

$$V_m^E = \left[(x_1 M_1 + x_2 M_2)/\rho - \{ (x_1 M_1)/\rho_1 + (x_2 M_2)/\rho_2 \} \right] \tag{2}$$

where x_1, M_1, ρ_1 represent the mole fraction, molar mass and the density of *m*-xylene, respectively, and x_2, M_2 and ρ_2 are the corresponding quantities of alkanols. The densities and the excess molar volumes of the systems at 303.15, 308.15, 313.15, 318.15 and 323.15 K are listed in table 2. The V_m^E of the systems are represented by the Redlich–Kister polynomial equation of the form,

$$V_m^E = x_1 x_2 \sum_{i=0}^{n} a_i (x_2 - x_1)^i \tag{3}$$

The coefficients, a_i, and the standard deviations, σ, of the polynomial equation are shown in table 4. V_m^E for the systems, *m*-xylene + 1-propanol, *m*-xylene + 2-propanol, *m*-xylene + 1-butanol and *m*-xylene + 2-methyl-2-propanol at different temperatures

Table 2. Densities, $\rho \times 10^{-3}$ (kg m^{-3}), excess molar volumes, $V_m^E \times 10^6$ (m^{3}mol^{-1}), of the systems, m-xylene +1-propanol, +2-propanol, +1-butanol and + 2-methyl-2-propanol, at different temperatures.

T/K	303.15		308.15		313.15		318.15		323.15	
X_2	ρ	V_m^E	ρ	V_m^E	ρ	V_m^E	ρ	V_m^E	ρ	V_m^E
				m-Xylene (x_1) + 1-propanol (x_2)						
0.0000	0.8554	0.0000	0.8511	0.0000	0.8468	0.0000	0.8425	0.0000	0.8381	0.0000
0.1010	0.8508	0.1054	0.8465	0.1082	0.8421	0.1121	0.8378	0.1284	0.8334	0.1326
0.1999	0.8466	0.1169	0.8421	0.1489	0.8376	0.1697	0.8331	0.2149	0.8286	0.2369
0.2988	0.8421	0.1197	0.8376	0.1525	0.8331	0.1755	0.8286	0.2202	0.8241	0.2444
0.3969	0.8374	0.1011	0.8330	0.1219	0.8285	0.1469	0.8240	0.1905	0.8195	0.2168
0.4996	0.8322	0.0574	0.8277	0.0912	0.8232	0.1183	0.8187	0.1610	0.8142	0.1894
0.5988	0.8266	0.0215	0.8221	0.0558	0.8176	0.0849	0.8132	0.1148	0.8087	0.1451
0.6993	0.8202	−0.0026	0.8158	0.0213	0.8114	0.0413	0.8070	0.0706	0.8026	0.0915
0.7931	0.8136	−0.0287	0.8093	−0.0144	0.8050	−0.0027	0.8007	0.0155	0.7963	0.0387
0.8961	0.8057	−0.0786	0.8013	−0.0524	0.7970	−0.0375	0.7926	−0.0088	0.7882	0.0174
1.000	0.7953	0.0000	0.7912	0.0000	0.7871	0.0000	0.7830	0.0000	0.7789	0.0000
				m-Xylene (x_1) + 2-propanol (x_2)						
0.0000	0.8554	0.0000	0.8511	0.0000	0.8468	0.0000	0.8425	0.0000	0.8381	0.0000
0.1078	0.8483	0.2239	0.8439	0.2389	0.8394	0.2675	0.8350	0.2822	0.8305	0.2972
0.1980	0.8429	0.2749	0.8385	0.2887	0.8340	0.3150	0.8294	0.3559	0.8248	0.3834
0.2991	0.8365	0.3117	0.8320	0.3401	0.8274	0.3771	0.8229	0.4012	0.8182	0.4397
0.3999	0.8299	0.3090	0.8254	0.3329	0.8207	0.3791	0.8161	0.4133	0.8115	0.4484
0.4994	0.8228	0.3016	0.8182	0.3358	0.8135	0.3783	0.8088	0.4216	0.8039	0.4600
0.6002	0.8152	0.2597	0.8106	0.2909	0.8059	0.3290	0.8012	0.3677	0.7963	0.4199
0.6992	0.8072	0.1921	0.8025	0.2219	0.7978	0.2655	0.7932	0.2880	0.7883	0.3349
0.8024	0.7978	0.1366	0.7934	0.1402	0.7887	0.1688	0.7840	0.1982	0.7791	0.2397
0.9010	0.7879	0.0706	0.7835	0.0723	0.7789	0.0862	0.7742	0.1111	0.7695	0.1258
1.0000	0.7768	0.0000	0.7724	0.0000	0.7679	0.0000	0.7634	0.0000	0.7588	0.0000
				m-Xylene (x_1) + 1-butanol (x_2)						
0.0000	0.8554	0.0000	0.8511	0.0000	0.8468	0.0000	0.8425	0.0000	0.8381	0.0000
0.0994	0.8506	0.1047	0.8462	0.1115	0.8418	0.1310	0.8375	0.1368	0.8332	0.1428
0.2000	0.8461	0.1257	0.8416	0.1501	0.8373	0.1631	0.8329	0.1883	0.8285	0.2140
0.3000	0.8415	0.1302	0.8371	0.1486	0.8327	0.1638	0.8284	0.1934	0.8240	0.2236
0.4003	0.8368	0.1113	0.8324	0.1354	0.8281	0.1551	0.8237	0.1889	0.8194	0.2096
0.5001	0.8320	0.0728	0.8271	0.1020	0.8232	0.1394	0.8189	0.1640	0.8146	0.1891
0.6003	0.8267	0.0578	0.8223	0.0799	0.8182	0.0954	0.8139	0.1244	0.8096	0.1539
0.7002	0.8213	0.0162	0.8171	0.0315	0.8129	0.0516	0.8086	0.0849	0.8044	0.1060
0.8002	0.8155	−0.0201	0.8112	0.0136	0.8072	0.0139	0.8030	0.0391	0.7988	0.0649
0.8998	0.8089	−0.0048	0.8049	−0.0005	0.8009	0.0054	0.7969	0.0115	0.7928	0.0298
1.0000	0.8019	0.0000	0.7980	0. 0000	0.7941	0.0000	0.7902	0.0000	0.7863	0.0000
				m-Xylene (x_1) + 2-methyl-2-propanol (x_2)						
0.0000	0.8554	0.0000	0.8511	0.0000	0.8468	0.0000	0.8425	0.0000	0.8381	0.0000
0.0997	0.8471	0.2751	0.8428	0.2834	0.8383	0.2999	0.8337	0.3232	0.8293	0.3417
0.1997	0.8393	0.4405	0.8349	0.4411	0.8303	0.4723	0.8257	0.4877	0.8212	0.5077
0.3000	0.8316	0.5384	0.8271	0.5449	0.8225	0.5609	0.8178	0.5821	0.8132	0.6023
0.3998	0.8238	0.6035	0.8192	0.6154	0.8145	0.6301	0.8098	0.6423	0.8051	0.6624
0.4999	0.8159	0.6258	0.8113	0.6284	0.8065	0.6409	0.8016	0.6715	0.7969	0.6765
0.5997	0.8078	0.6227	0.8031	0.6294	0.7982	0.6391	0.7934	0.6460	0.7885	0.6634
0.7002	0.7995	0.5798	0.7947	0.5898	0.7899	0.5691	0.7848	0.6063	0.7800	0.5937
0.8000	0.7911	0.4971	0.7863	0.4968	0.7813	0.4851	0.7762	0.5111	0.7713	0.4959
0.9004	0.7829	0.3188	0.7780	0.3197	0.7729	0.3023	0.7677	0.3291	0.7626	0.3228
1.0000	0.7754	0.0000	0.7704	0.0000	0.7650	0.0000	0.7599	0.0000	0.7546	0.0000

Table 3. Coefficients, A_i, of equation expressing density, $\rho \times 10^{-3}$ (kg m^{-3}) and the square of the regression coefficient, r^2, for the systems, *m*-xylene +1-propanol, +2-propanol, +1-butanol and + 2-methyl-2-propanol.

| System	Temperature/K	a_0	a_1	a_2	a_3	a_4	a_5	r^2
m-Xylene + 1-propanol	303.15	1.2668	−2.6428	−6.6558	28.2120	−33.916	13.734	0.9855
	308.15	1.3044	−1.7693	−9.8581	32.0640	−35.370	13.628	0.9992
	313.15	1.3567	−1.5626	−9.9843	30.5340	−32.826	12.483	0.9993
	318.15	1.5483	−1.2944	−11.453	30.5330	−29.959	10.627	0.9961
	323.15	1.6013	−1.1041	−11.077	26.9100	−24.392	8.0638	0.9931
m-Xylene + 2-propanol	303.15	3.4702	−17.88	52.047	−83.669	66.453	−20.421	0.9988
	308.15	3.7276	−19.604	58.09	−93.293	73.05	−21.97	0.9981
	313.15	4.1162	−21.601	63.822	−101.71	78.904	−23.529	0.9976
	318.15	4.5208	−24.397	74.546	−122.42	97.823	−30.073	0.9995
	323.15	4.5061	−21.874	61.317	−94.19	70.931	−20.691	0.9993
m-Xylene + 1-butanol	303.15	1.785	−9.753	27.86	−45.176	36.848	−11.562	0.9885
	308.15	1.7508	−7.8543	18.732	−27.232	20.94	−6.3372	0.9967
	313.15	2.3407	−13.785	42.709	−71.588	58.743	−18.419	0.995
	318.15	2.1149	−9.3167	22.627	−32.601	24.061	−6.8853	0.9995
	323.15	2.0638	−7.1212	12.643	−14.14	8.7397	−2.1848	0.999
m-Xylene + 2-methyl-2-propanol	303.15	3.6749	−11.283	26.181	−40.21	32.944	−11.306	0.9999
	308.15	3.6878	−10.734	22.63	−32.014	24.946	−8.5163	0.9997
	313.15	4.0787	−13.203	30.192	−44.315	34.551	−11.304	0.9995
	318.15	4.5198	−16.414	40.6	−61.356	48.554	−15.905	0.9998
	323.15	4.9279	−19.476	51.508	−81.05	65.383	−21.293	0.9999
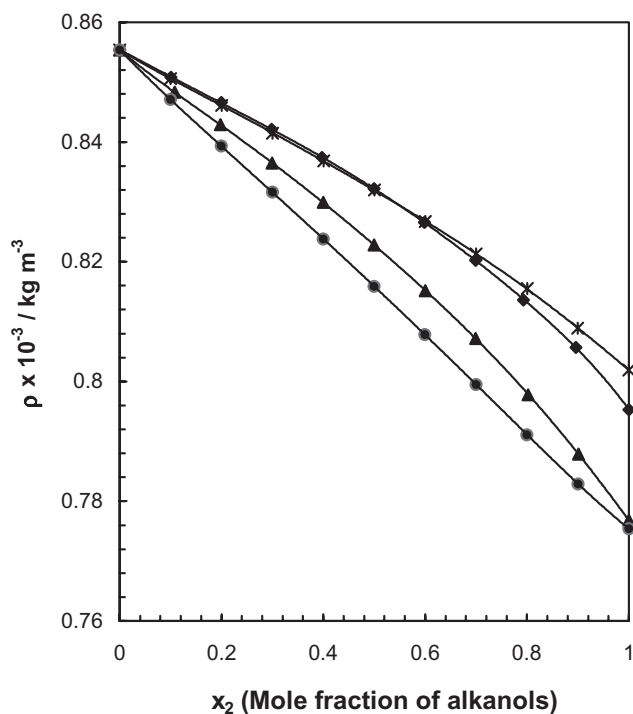
Figure 1. Variation of densities of *m*-xylene + 1-propanol (◆), + 2-propanol (▲), + 1-butanol (∗), and + 2-methyl-2-propanol (●) at 303.15 K as a function of mole fraction of alkanols.

Table 4. Coefficients, a_i, of Redlich–Kister equation expressing $V_m^E \times 10^6$ (m^3 mol^{-1}), and standard deviation, σ, for the systems.

System	Temperature/K	A_0	A_1	A_2	A_3	σ
m-Xylene + 1-propanol	303.15	0.2594	−0.5710	−0.0502	−0.9145	0.01360
	308.15	0.3814	−0.6495	0.0107	−0.6846	0.00771
	313.15	0.4912	−0.6916	−0.0103	−0.5690	0.00714
	318.15	0.6465	−0.8813	0.1191	−0.2270	0.00964
	323.15	0.7560	−0.9360	0.1949	0.0405	0.01193
m-Xylene + 2-propanol	303.15	1.1607	−0.5496	0.4790	−0.6272	0.01199
	308.15	1.2920	−0.5006	0.3648	−0.8655	0.01391
	313.15	1.4654	−0.4642	0.4195	−1.0132	0.01569
	318.15	1.6077	−0.5008	0.8334	−0.9147	0.01613
	323.15	1.7861	−0.3830	0.5908	−1.0830	0.01242
m-Xylene + 1-butanol	303.15	0.3157	−0.6287	0.2166	−0.2649	0.01072
	308.15	0.4156	−0.6290	0.2687	−0.2417	0.00608
	313.15	0.5120	−0.6063	0.2162	−0.4367	0.00999
	318.15	0.6412	−0.6078	0.2269	−0.4261	0.00437
	323.15	0.7448	−0.6477	0.3325	−0.2712	0.00413
m-Xylene + 2-methyl-2-propanol	303.15	2.4945	0.2208	1.2184	0.1557	0.00468
	308.15	2.5238	0.2365	1.2108	0.0734	0.00701
	313.15	2.5637	0.0800	1.1633	−0.0824	0.00990
	318.15	2.6333	0.1383	1.4250	−0.1083	0.00986
	323.15	2.6801	−0.0049	1.3847	−0.1881	0.01237
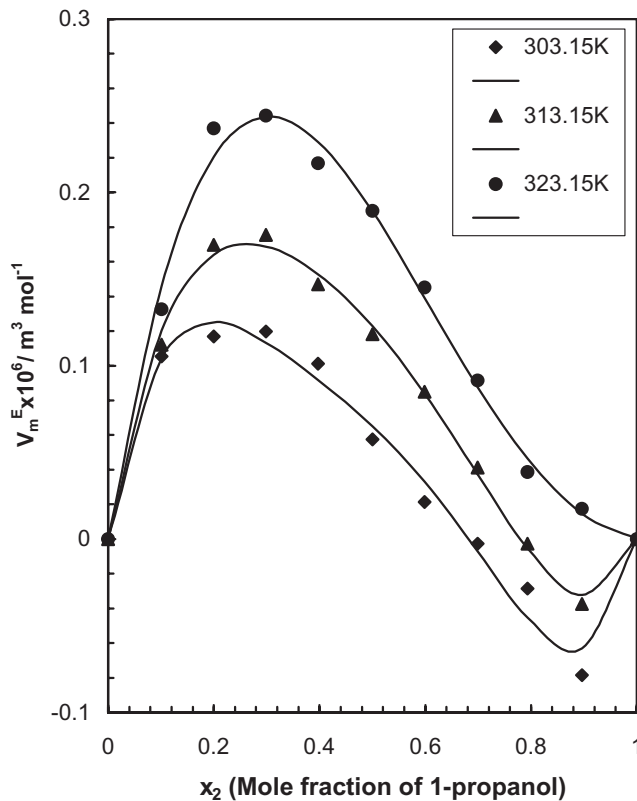
Figure 2. Excess molar volumes for the system, *m*-xylene+1-propanol at different temperatures against mole fraction of 1-propanol.
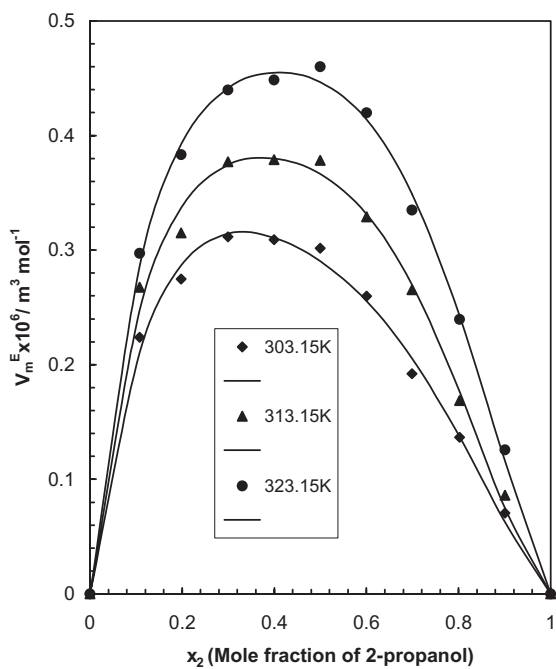
Figure 3.   Excess molar volumes for the system, *m*-xylene+2-propanol at different temperatures against mole fraction of 2-propanol.
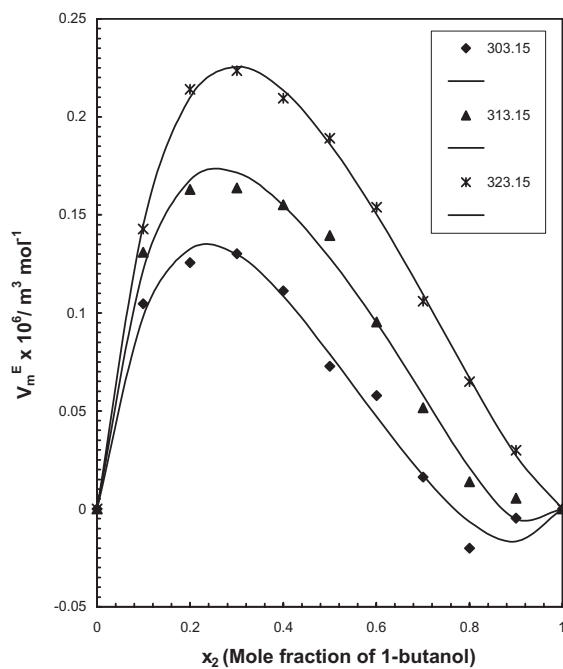

Figure 4.   Excess molar volumes for the system, *m*-xylene+1-butanol at different temperatures against mole fraction of 1-butanol.
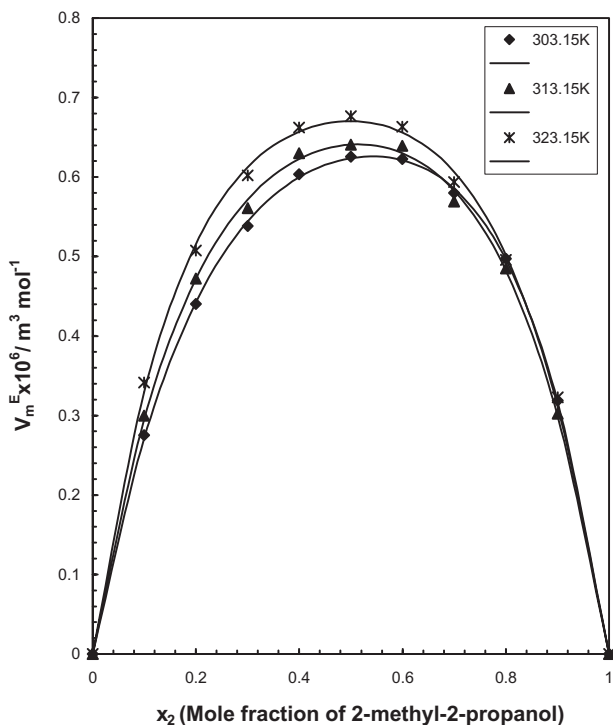
Figure 5. Excess molar volumes for the system, *m*-xylene + 2-methyl-2-propanol at different temperatures against mole fraction of 2-methyl-2-propanol.

have been represented respectively by figures 2–5. An examination of the figures reveals the following characteristics:

(a) V_m^E are positive in *m*-xylene-rich region, but negative in the alkanol-rich region for the systems, *m*-xylene + 1-propanol and *m*-xylene + 1-butanol, although the negative values are very small (figures 2 and 4).

(b) V_m^E are positive for the whole range of composition for the systems, *m*-xylene + 2-propanol and *m*-xylene + 2-methyl-2-propanol (figures 3 and 5).

(c) For all systems, $\delta V_m^E / \delta T$ is positive, irrespective of sign of V_m^E.

Comparison of the V_m^E curves in figure 6 shows that the values of V_m^E of the system, *m*-xylene + 1-butanol are only slightly greater than those for the system *m*-xylene + 1-propanol, throughout the whole range of composition. But for the system, *m*-xylene + 2-methyl-2-propanol, V_m^E are substantially larger than those for the system, *m*-xylene + 2-propanol.

The sign of V_m^E depends upon the resultant of the factors contributing to the expansion of volume and the factors contributing to the contraction of volume. In the present systems, we envisage that the main factors contributing to expansion of volume are:

(a) a dissociation of associated alkanols in solution systems, particularly in *m*-xylene-rich region.

(b) a steric hindrance due to branching of the alkanols in 2-propanol and 2-methyl-2-propanol.
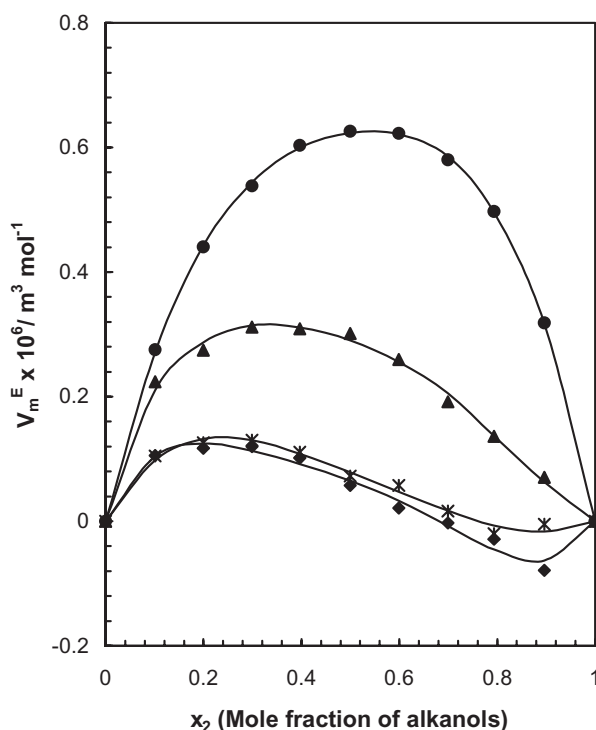
Figure 6. Comparison of excess molar volumes for the systems, *m*-xylene + 1-propanol (◆), + 2-propanol (▲), + 1-butanol (∗), and + 2-methyl-2-propanol (●) at 303.15 K as a function of mole fraction of alkanols.

On the other hand, the following factors are considered responsible for volume contraction:

(a) a donor–acceptor interaction between the π-electrons of the aromatic ring and the alkanols.
(b) a possible inclusion of *m*-xylene, at least partially, into the structural network of alkanols.

In the *m*-xylene-rich region, disintegration of multimers of alkanols into smaller units through disruption of H-bonding in alkanols takes place. This causes the volume to expand. Another important factor that contributes significantly towards volume expansion is the steric hindrance of the branched chain alkanols. The steric effect is clearly evident by much larger positive V_m^E for branched chain alkanols than for straight chain alkanols. In highly rich region of alkanols, where smaller units of alkanols are believed to be reformed into larger units, negative V_m^E are observed for straight chain alkanols at lower temperatures. At higher temperatures, however, these values turn to very small positive. In this region, the combined effects for volume contraction, i.e., donor–acceptor interaction and inclusion of *m*-xylene into structural network of alkanols, as outlined above, exceeds the effects responsible for volume expansion. Singh *et al.* [1] and Akhtar *et al.* [4] suggested the same explanation for the volumetric properties for their systems, 1-propanol + xylenes and 1-pentanol + toluene, respectively.

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