Studies of Thermodynamics and Transport Properties of Binary solutions of o-Xylene with 2-Alkanols (C_3-C_5) at Different Temperatures

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ABSTRACT

Densities and viscosities for the binary mixtures of o-xylene with propan-2-ol, butan-2-ol and pentan-2-ol have been measured at a number of mole fractions at different temperatures (298.15, 303.15, 308.15 and 313.15) K and at atmospheric pressure. From the experimental data, excess molar volumes (V^E) and deviations in viscosities ($\Delta\eta$) have been calculated. The excess molar volumes for o-xylene with propan-2-ol and butan-2-ol system are negative at lower mole fractions of o-xylene and positive at higher mole fractions. Binary mixtures of o-xylene with pentan-2-ol give positive V^E values at all temperatures while deviations in viscosity are negative. The results have been interpreted in terms of molecular interactions. These are further fitted to the Redlich-Kister type of equation.

KEYWORDS- Density, Viscosity, o-Xylene, Propan-2-ol, Butan-2-ol, Pentan-2-ol.

I. INTRODUCTION

Thermodynamic and Transport properties of binary liquid mixtures are frequently needed in chemical processes. Specific and non–specific interactions taking place between the components of mixtures. Alcohols are strongly self-associated molecules through Hydrogen–bonding and for binary solutions rich in alcohols. Xylenes are non-associated and potential electron donors. Molecular interactions between toluene, xylenes having -CH_3 as electron donating group, and alkanols have been reported [1]-[5]. Measured densities and viscosities of xylene (o-, m- and p-), normal and branched alkanols(C_1-C_{10}) and their binary mixtures with xylene in the liquid state were reported [6]-[11] over the whole range of composition at different temperatures and atmospheric pressure. In the present investigation, we report density and viscosity studies of binary mixtures of o-xylene with propan-2-ol, butan-2-ol and pentan-2-ol over entire range of composition at (298.15, 303.15, 308.15 and 313.15) K at atmospheric pressure. More work has been reported about excess, transport and thermodynamic properties of binary liquid mixtures [12]-[17].

II. EXPERIMENTAL SECTION

o-xylene, propan-2-ol, butan-2-ol and pentan-2-ol (s. d. fine chemicals, Lancaster, Purity>99) were purified by standard procedures [18]. The purity of the solvents, after purification, was ascertained by comparing their densities and viscosities with the corresponding literature values at (298.15, 303.15, 308.15 and 308.15) K. Binary mixtures were prepared by mass in air-tight stoppered glass bottles. The masses were recorded on an Adair dutt balance to an accuracy of ± 1 x 10^{-4} g. Densities of pure liquids and their mixtures were determined by using a 15 cm^3 bicapillary pycnometer as described earlier [19]-[20]. The pycnometer was calibrated using conductivity water with 0.99705 g cm^{-3} as its density [21] at 298.15K. The dynamic viscosities were measured using an Ubbelohde suspended level viscometer [22], 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 sec and capillary radius (0.5mm) was far less than its length (50 to 60) mm, the kinetic energy and end corrections respectively, were found to be negligible. The viscosity, (η) of the liquids was calculated by ,

$$\frac{\eta}{\eta_w} = \frac{\rho t}{\rho_w t_w} \qquad ---(1)$$

Where, ρ, ρ_w and t, t_w refer to the density and flow time of the experimental liquids and water, respectively. The uncertainties in dynamic viscosities are of the order of ± 0.001 m Pa.s.

III. RESULT AND DISCUSSION

Experimental values of densities (ρ), and viscosities (η) of mixtures at (298.15, 303.15, 308.15 and 313.15) K are listed as a function of mole fraction in Table I, II and III. The density values have been used to calculate excess molar volumes (V^E) using the following equation.

$$V^E = (x_1M_1 + x_2M_2)/\rho_{12} - (x_1M_1/\rho_1) - (x_2M_2/\rho_2) \qquad --(2)$$

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 (Δη) were calculated using

$$\Delta\eta = \eta_{12} - x_1\eta_1 - x_2\eta_2 \qquad -----(3)$$

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 molar volumes and deviations in viscosity were fitted to a Redlich-Kister [23] equation of the type

$$Y = X_1X_2\sum_{i}^{n} a_i(X_2 - X_1)^i \qquad -----(4)$$

Where, Y is either V^E or Δη and n is the degree of polynomial. The calculated values of the coefficients (ai) along with the standard deviations (σ) are given in Table IV.

TABLE I
DENSITIES (ρ) , VISCOSITIES (η) , EXCESS MOLAR VOLUMES (V^E) , AND DEVIATION IN VISCOSITY (Δη) FOR THE O-XYLENE (1) + PROPAN-2-OL (2) SYSTEM.

Temp(K)	X_1	ρ ($g.cm^{-3}$)	η (Poise)	V^E (cm^3mol^{-1})	Δη (cP)
298.15	0.0000	0.78070	0.0210	0.0000	0.000
	0.1010	0.79563	0.0162	-0.0600	-0.350
	0.2007	0.80806	0.0134	-0.0321	-0.496
	0.3002	0.81889	0.0109	0.0333	-0.609
	0.4007	0.82865	0.0097	0.1164	-0.593
	0.5002	0.83749	0.0091	0.1924	-0.520
	0.6008	0.84589	0.0082	0.2401	-0.471
	0.7007	0.85376	0.0078	0.2598	-0.377
	0.8004	0.86155	0.0076	0.2075	-0.266
	0.9015	0.86990	0.0074	0.1350	-0.143
	1.0000	0.87620	0.0075	0.0000	0.000
303.15	0.0000	0.77640	0.0182	0.0000	0.000
	0.1010	0.79110	0.0142	-0.0400	-0.282
	0.2007	0.80341	0.0118	-0.0009	-0.414
	0.3002	0.81414	0.0098	0.0751	-0.503
	0.4007	0.82400	0.0087	0.1470	-0.497
	0.5002	0.83290	0.0083	0.2156	-0.432
	0.6008	0.84133	0.0075	0.2590	-0.397
	0.7007	0.84921	0.0072	0.2766	-0.312
	0.8004	0.85683	0.0070	0.2451	-0.219
	0.9015	0.86471	0.0069	0.1126	-0.122
	1.0000	0.87173	0.0070	0.0000	0.000
308.15	0.0000	0.77200	0.0159	0.0000	0.000
	0.1010	0.78652	0.0125	-0.0254	-0.246
	0.2007	0.79868	0.0106	0.0277	-0.346
	0.3002	0.80933	0.0084	0.1117	-0.470
	0.4007	0.81921	0.0080	0.1806	-0.417
	0.5002	0.82820	0.0076	0.2377	-0.369
	0.6008	0.83659	0.0069	0.2850	-0.342
	0.7007	0.84450	0.0067	0.2976	-0.272
	0.8004	0.85207	0.0066	0.2711	-0.196
	0.9015	0.85953	0.0065	0.1937	-0.111
	1.0000	0.86712	0.0067	0.0000	0.000
313.15	0.0000	0.76730	0.0138	0.0000	0.000
	0.1010	0.78178	0.0115	-0.0123	-0.147
	0.2007	0.79384	0.0094	0.0620	-0.285
	0.3002	0.80460	0.0080	0.1447	-0.354
	0.4007	0.81469	0.0074	0.1995	-0.340
	0.5002	0.82371	0.0070	0.2628	-0.306
	0.6008	0.83220	0.0064	0.3073	-0.282
	0.7007	0.84019	0.0063	0.3184	-0.224
	0.8004	0.84776	0.0062	0.3003	-0.161
	0.9015	0.85521	0.0061	0.2325	-0.091
	1.0000	0.86312	0.0063	0.0000	0.000

TABLE II
DENSITIES (ρ) , VISCOSITIES (η) , EXCESS MOLAR VOLUMES (V^E) , AND DEVIATION IN VISCOSITY (Δη) FOR THE O-XYLENE (1) + BUTAN-2-OL (2) SYSTEM.

Temp(K)	X_1	ρ ($g.cm^{-3}$)	η (Poise)	V^E (cm^3mol^{-1})	Δη (cP)
298.15	0.0000	0.79795	0.0344	0.0000	0.000
	0.1006	0.80834	0.0255	-0.0501	-0.620
	0.2003	0.81723	0.0191	-0.0007	-0.990
	0.3014	0.82531	0.0153	0.1005	-1.093
	0.4019	0.83298	0.0125	0.1902	-1.107
	0.5071	0.84075	0.0108	0.2598	-0.995
	0.6009	0.84746	0.0095	0.3033	-0.871
	0.7013	0.85455	0.0086	0.3152	-0.693
	0.8025	0.86180	0.0079	0.2653	-0.488
	0.9047	0.86930	0.0076	0.1433	-0.253
	1.0000	0.87620	0.0075	0.0000	0.000
303.15	0.0000	0.79366	0.0293	0.0000	0.000
	0.1006	0.80382	0.0220	-0.0262	-0.505
	0.2003	0.81244	0.0167	0.0544	-0.811
	0.3014	0.82056	0.0136	0.1501	-0.897
	0.4019	0.82826	0.0112	0.2354	-0.909
	0.5071	0.83606	0.0098	0.3002	-0.821
	0.6009	0.84283	0.0087	0.3349	-0.719
	0.7013	0.84989	0.0079	0.3493	-0.572
	0.8025	0.85716	0.0074	0.2946	-0.400
	0.9047	0.86459	0.0071	0.1795	-0.210
	1.0000	0.87173	0.0070	0.0000	0.000
308.15	0.0000	0.78976	0.0254	0.0000	0.000
	0.1006	0.79954	0.0192	0.0085	-0.436
	0.2003	0.80801	0.0147	0.1000	-0.700
	0.3014	0.81607	0.0122	0.1950	-0.761
	0.4019	0.82378	0.0102	0.2700	-0.773
	0.5071	0.83152	0.0089	0.3350	-0.699
	0.6009	0.83815	0.0080	0.3800	-0.616
	0.7013	0.84541	0.0073	0.3600	-0.493
	0.8025	0.85236	0.0069	0.3395	-0.346
	0.9047	0.85971	0.0066	0.2266	-0.188
	1.0000	0.86712	0.0065	0.0000	0.000
313.15	0.0000	0.78557	0.0222	0.0000	0.000
	0.1006	0.79479	0.0167	0.0795	-0.390
	0.2003	0.80330	0.0130	0.1697	-0.602
	0.3014	0.81140	0.0109	0.2646	-0.652
	0.4019	0.81923	0.0092	0.3298	-0.657
	0.5071	0.82711	0.0081	0.3792	-0.595
	0.6009	0.83374	0.0073	0.4294	-0.525
	0.7013	0.84076	0.0068	0.4446	-0.421
	0.8025	0.84797	0.0065	0.3927	-0.292
	0.9047	0.85527	0.0062	0.2883	-0.156
	1.0000	0.86312	0.0061	0.0000	0.000

TABLE III
DENSITIES (ρ) , VISCOSITIES (η) , EXCESS MOLAR VOLUMES (V^E) , AND DEVIATION IN VISCOSITY (Δη) FOR THE O-XYLENE (1) + PENTAN-2-OL (2) SYSTEM.

Temp(K)	X_1	ρ ($g.cm^{-3}$)	η (Poise)	V^E (cm^3mol^{-1})	Δη (cP)
298.15	0.0000	0.80544	0.0345	0.0000	0.000
	0.1050	0.81318	0.0239	0.0542	-0.777
	0.2025	0.81995	0.0180	0.1388	-1.100
	0.3068	0.82703	0.0141	0.2292	-1.212
	0.4036	0.83360	0.0119	0.2934	-1.166
	0.5067	0.84047	0.0102	0.3600	-1.057
	0.6035	0.84694	0.0093	0.4000	-0.888
	0.7078	0.85394	0.0084	0.4200	-0.699
	0.8020	0.86083	0.0079	0.3400	-0.496
	0.9036	0.86858	0.0076	0.1938	-0.256
	1.0000	0.87620	0.0075	0.0000	0.000
303.15	0.0000	0.80110	0.0285	0.0000	0.000
	0.1050	0.80854	0.0202	0.0936	-0.604
	0.2025	0.81526	0.0155	0.1836	-0.861
	0.3068	0.82234	0.0125	0.2738	-0.941
	0.4036	0.82888	0.0106	0.3418	-0.920
	0.5067	0.83583	0.0093	0.3950	-0.824
	0.6035	0.84233	0.0085	0.4300	-0.701
	0.7078	0.84928	0.0078	0.4550	-0.551
	0.8020	0.85616	0.0074	0.3750	-0.385
	0.9036	0.86388	0.0071	0.2290	-0.203
	1.0000	0.87173	0.0070	0.0000	0.000
308.15	0.0000	0.79690	0.0239	0.0000	0.000
	0.1050	0.80404	0.0174	0.1294	-0.475
	0.2025	0.81065	0.0136	0.2300	-0.683
	0.3068	0.81777	0.0112	0.3100	-0.745
	0.4036	0.82426	0.0097	0.3800	-0.727
	0.5067	0.83120	0.0085	0.4300	-0.665
	0.6035	0.83767	0.0079	0.4635	-0.559
	0.7078	0.84534	0.0072	0.3804	-0.453
	0.8020	0.85133	0.0068	0.4210	-0.327
	0.9036	0.85899	0.0069	0.2759	-0.147
	1.0000	0.86712	0.0067	0.0000	0.000
313.15	0.0000	0.79250	0.0201	0.0000	0.000
	0.1050	0.79945	0.0151	0.1640	-0.358
	0.2025	0.80603	0.0121	0.2742	-0.520
	0.3068	0.81316	0.0100	0.3596	-0.586
	0.4036	0.81980	0.0087	0.4142	-0.578
	0.5067	0.82678	0.0078	0.4650	-0.525
	0.6035	0.83332	0.0072	0.4950	-0.451
	0.7078	0.84034	0.0067	0.5095	-0.360
	0.8020	0.84704	0.0064	0.4550	-0.261
	0.9036	0.85466	0.0062	0.3198	-0.410
	1.0000	0.86312	0.0061	0.0000	0.000

TABLE IV

COEFFICIENTS OF THE REDLICH-KISTER EQUATION AND STANDARD DEVIATION FOR EXCESS MOLAR VOLUMES AND VISCOSITY DEVIATIONS OF MIXTURES.

Temp (K)	Property	a_0	a_1	a_2	a_3	σ
o-xylene + propan-2-ol						
298.15	$V^E/cm^3\ mol^{-1}$	0.889523	1.758064	-1.56994	-1.87386	0.035916
	Δη/mPa.s	-2.15371	1.33569	-0.86484	-------	0.016182
303.15	$V^E/cm^3\ mol^{-1}$	0.915012	1.33233	-0.68886	-0.36947	0.016263
	Δη/mPa.s	-1.80028	1.067743	-0.65709	-------	0.012544
308.15	$V^E/cm^3\ mol^{-1}$	0.965779	0.974772	-0.04711	0.86185	0.004228
	Δη/mPa.s	-1.55735	0.904958	-0.62801	--------	0.023635
313.15	$V^E/cm^3\ mol^{-1}$	1.046841	0.810514	0.277512	1.37894	0.009298
	Δη/mPa.s	-1.31235	0.513082	-0.08492	-------	0.019871
o-xylene + butan-2-ol						
298.15	$V^E/cm^3\ mol^{-1}$	1.073066	1.291134	-0.77171	0.16936	0.009861
	Δη/mPa.s	-4.06234	2.479029	-1.36595	--------	0.012627
303.15	$V^E/cm^3\ mol^{-1}$	1.227251	1.05708	-0.47966	0.63780	0.009396
	Δη/mPa.s	-3.34771	2.0064	-1.08357	---------	0.010229
308.15	$V^E/cm^3\ mol^{-1}$	1.058412	-0.49688	0.352223	3.35229	0.13417
	Δη/mPa.s	-2.84467	1.689749	-1.06014	---------	0.011416
313.15	$V^E/cm^3\ mol^{-1}$	1.552511	0.883609	0.846454	0.96600	0.01002
	Δη/mPa.s	-2.41397	1.540782	-1.01895	---------	0.010981
o-xylene + pentan-2-ol						
298.15	$V^E/cm^3\ mol^{-1}$	1.484076	1.068549	-0.08545	--	0.013999
	Δη/mPa.s	-4.22504	2.793339	-2.19217	0.902651	0.006359
303.15	$V^E/cm^3\ mol^{-1}$	1.615567	1.029648	0.326103	--	0.011131
	Δη/mPa.s	-3.3132	2.182458	-1.69068	0.655191	0.004848
308.15	$V^E/cm^3\ mol^{-1}$	1.643002	0.979806	0.952235	--	0.034917
	Δη/mPa.s	-2.67691	1.522534	-1.18226	0.930967	0.009382
313.15	$V^E/cm^3\ mol^{-1}$	1.834226	1.071105	1.329266	--	0.012696
	Δη/mPa.s	-2.11207	1.257926	-0.94644	0.213744	0.001998

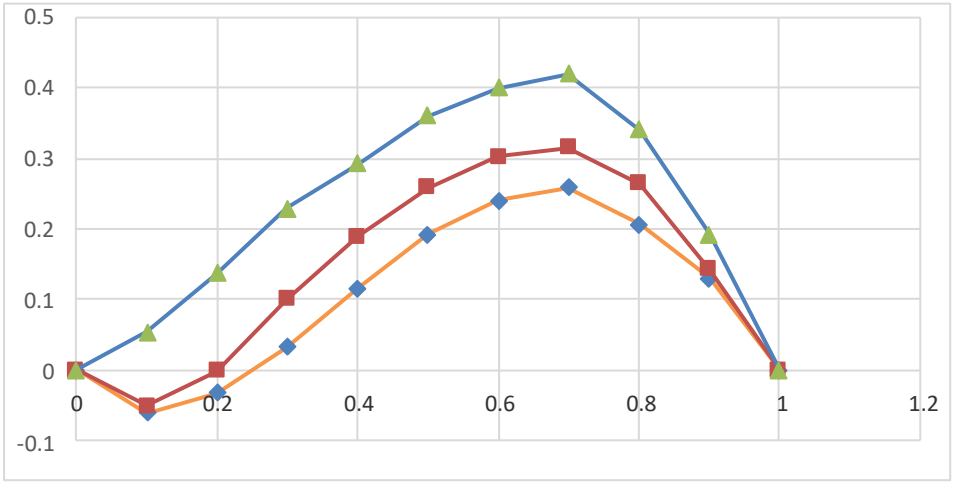

Fig. 1 Excess Molar Volume V^E at 298.15K for (X_1) o-xylene + (1- X_1) Propan- 2-ol(♦), Butan-2-ol(■), Pentan-2-ol(▲).

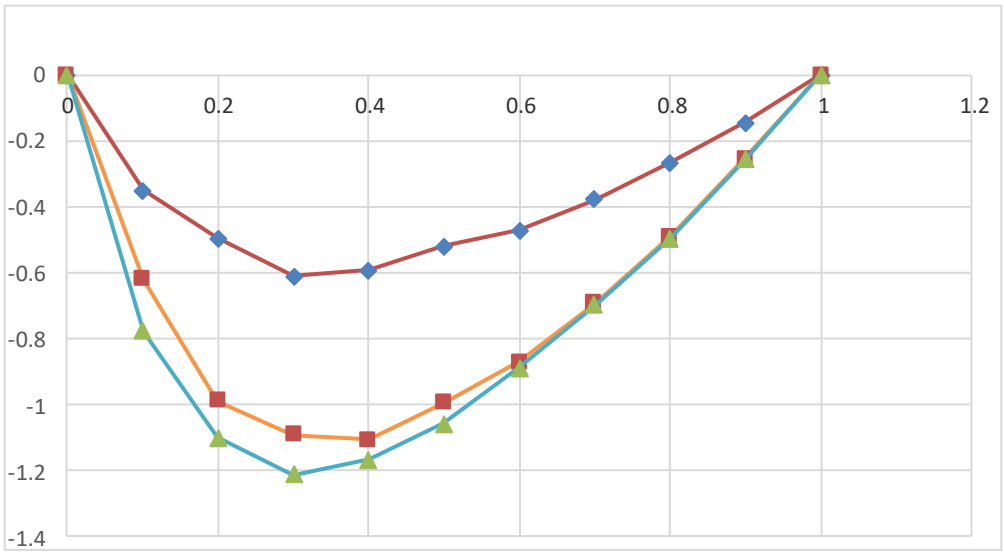

Fig. 2 Deviation in viscosity Δη at 298.15K for (X_1) o-xylene +(1- X_1) Propan-2-ol (♦), Butan-2-ol (■), Pentan-2-ol(▲).

The variations of V^E and Δη with mole fraction of o-xylene for the binary systems of o-xylene with propan-2-ol, butan-2-ol and pentan-2-ol at 298.15K are represented in Figure 1 and 2. Similar plots are obtained at other temperatures. The V^E curves for binary mixtures of o-xylene with propan-2-ol and butan-2-ol are negative at lower mole fraction and positive at higher mole fraction of o-xylene. The close perusal of tables 1, 2 and 3 indicates that V^E values are greatly influenced by temperature and chain length and branching of 2-alkanols for the same binary mixtures. Therefore the nature of the graph for the same binary mixture changes with temperature.

The sigmoidal nature of the graph reduces at higher temperature. It is generally observed that V^E values increase with increase of temperature and chain length of 2-alkanols.Treszczanowicz [24] and Aminabhavi [25] observed that V^E may be discussed in terms of several effects which may be arbitrarily divided into physical, chemical and geometrical contributions.

The observed V^E data of binary mixtures of o-xylene with 2-alkanols (C_3-C_5) may be explained qualitatively by postulating that V^E is resultant of opposing effects. Negative V^E values in the lower region of o-xylene may be attributed to two factors (i) due to change of free volumes in the real mixture because of interstitial accommodation of xylene molecules into clusters of alkanol molecules. (ii) The xylene molecules behave as electron donors due to presence of two $–CH_3$ groups which make π electron cloud denser on benzene ring. These electrons from xylene molecules are accepted by protons from alkanols forming donor-acceptor π---HO complexes, resulting in a negative contribution to V^E. The positive values of V^E arise due to the breaking or stretching of H-bond in the self-associated alcohols and physical dipole-dipole interactions between alcohol monomers and multimers and due to the disruption in the favorable orientation order of aromatic hydrocarbons. In other words the positive V^E values may also be observed due to steric hindrance between bulky xylene and alkanol molecules. Figure 1 show that for o-xylene V^E_{max} and V^E_{min} values vary with the length of alkanol in a way similar to that observed by Ortega et al[14].The order of V^E_{max} and V^E_{min} is

$$V^E_m(C_3H_7OH) < V^E_m(C_4H_9OH) < V^E_m(C_5H_{11}OH)$$

Fort and Moore [26] have pointed out that negative Δη values are common in mixtures of molecules of unequal size in which dispersion forces are dominant. The molar volumes of o-xylene are 121.17 and those of propan-2-ol, butan-2-ol and pentan-2-ol are 76.98, 92.88 and 109.44 respectively at 298.15K. Similar molar volume could be obtained for these molecules at other temperatures (303.15, 308.15 and 313.15). These molar volumes of o-xylene and 2-alkanols at 298.15 to 313.15K differ considerably and hence non associated xylene molecules are interstitially accommodated into clusters of alkanols yielding negative Δη values for all binary systems at all temperatures. Figure 2 exhibit that Δη increases with decrease of chain length of alkanols. This implies that dispersion forces are dominant in o-xylene + pentan-2-ol followed by those in o-xylene + butan-2-ol and o-xylene + propan-2-ol. These results suggest that depolymerisation effect, which depends upon self-association of alkanols decreases from propan-2-ol to pentan-2-ol. In other words interstitial accommodation of non-associated xylene molecules into cluster of alkanols follows the order

propan-2-ol > butan-2-ol > pentan-2-ol

IV. CONCLUSION

In the present investigation the excess molar volume (V^E) and deviations in viscosities (Δη) of binary mixtures of o-xylene with 2- alkanols (C_3-C_5) were measured at different temperatures. The excess molar volumes for o-xylene + propan-2-ol, + butan-2-ol are negative at lower mole fraction and positive at higher mole fraction of o-xylene. Binary mixtures of o-xylene with pentan-2-ol give positive V^E values at all temperatures while deviations in viscosity are negative.

The negative V^E values may arise due to π---HO donar-acceptor complex formation between xylene and alkanol molecules. Positive V^E are obtained as a result of dissociation of aggregates of alkanol in presence of xylene molecules. The Δη values for all binary mixtures are negative at all temperatures. These negative Δη values are attributed to difference in molar volumes and free volumes of 2-alkanols and xylene molecules.

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