Binary Mixtures of Butanol + Pentane, + Hexane, + Heptane, + Octane, + 2,2,4-Trimethylpentane, and + Carbon Tetrachloride. 1. Excess Molar Volumes at 288.15 K and 298.15 K and Refractive Indexes at 298.15 K

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Excess molar volumes, V_m^E, have been measured for binary mixtures of butanol (n-C_4H_9OH) + pentane (n-C_5H_{12}), + hexane (n-C_6H_{14}), + heptane (n-C_7H_{16}), + octane (n-C_8H_{18}), + 2,2,4-trimethylpentane (2,2,4-TMP), and + carbon tetrachloride (CCl_4) at 288.15 K and 298.15 K, and refractive indexes, n_D, have been measured for these mixtures at 298.15 K. At both temperatures V_m^E has been found to be positive throughout the entire range of composition for xn-C_4H_9OH + $(1 - x)n$-C_7H_{16} and + $(1 - x)n$-C_8H_{18}. At both temperatures 288.15 K and 298.15 K, V_m^E is positive at low mole fractions of n-C_4H_9OH and negative at its higher mole fractions in the case of mixtures of n-C_4H_9OH with n-C_5H_{12}, n-C_6H_{14}, 2,2,4-TMP, and CCl_4. Values of V_m^E and n_D for the various mixtures of n-C_4H_9OH have been fitted in smoothing equations.

Introduction

Mixtures of alkanes with alkanols are of particular interest from a theoretical viewpoint of the models of the hydrogen-bonded systems. Systematic studies of the thermodynamic excess functions provide important information concerning the deeper understanding of the molecular liquid structure and intermolecular interactions predominated by the self-association of alkanol molecules due to hydrogen bonding. These studies are also important from the viewpoint of prediction of thermodynamic properties of alkane + alkanol mixtures of components having varying numbers of $-CH_2-$ units in the alkyl chain or varying numbers of $-CH_3$ substituents attached to the alkyl chain in alkanols or alkanes. With this viewpoint, Wagner and Heintz (1986) and Heintz et al. (1986) measured excess molar volumes, V_m^E, of binary mixtures of nonane and hexane with five different 1-alkanols at various temperatures. Fuente et al. (1992) have determined excess molar volumes of (nonan-1-ol + decane or tetradecane) at the temperatures 298.15 K, 308.15 K, and 318.15 K. Zielkiewicz (1994a,b) measured total vapor pressures and V_m^E of (heptane + propan-2-ol or 2-methylpropan-1-ol, 2-methylpropan-2-ol, butan-1-ol, or pentan-1-ol) at the temperature 313.15 K. Franjo et al. (1994) have measured excess molar volumes of $xCH_3(CH_2)_5OH + (1 - x)CH_3(CH_2)_4CH_3$ at 298.15 K. In this paper we wish to know the variation of V_m^E and n_D of mixtures of an alkanol with alkanes of varying molecular complexities. Hence, in view of the above considerations, the measurements of excess molar volumes, V_m^E, and refractive indexes, n_D, have been made in this work, for binary liquid mixtures of n-C_4H_9OH with pentane (n-C_5H_{12}), hexane (n-C_6H_{14}), heptane (n-C_7H_{16}), octane (n-C_8H_{18}), and 2,2,4-trimethylpentane (2,2,4-TMP). The system of n-C_4H_9OH with carbon tetrachloride (CCl_4) has also been studied, since it is interesting from the viewpoint of the existence of the donor−acceptor interaction between the components in the liquid state, and the results of V_m^E and n_D for the mixtures of n-C_4H_9OH + n-C_5H_{12}, + n-C_6H_{14}, + n-C_7H_{16}, + n-C_8H_{18}, + 2,2,4-TMP, and + CCl_4 are reported and discussed in this paper.

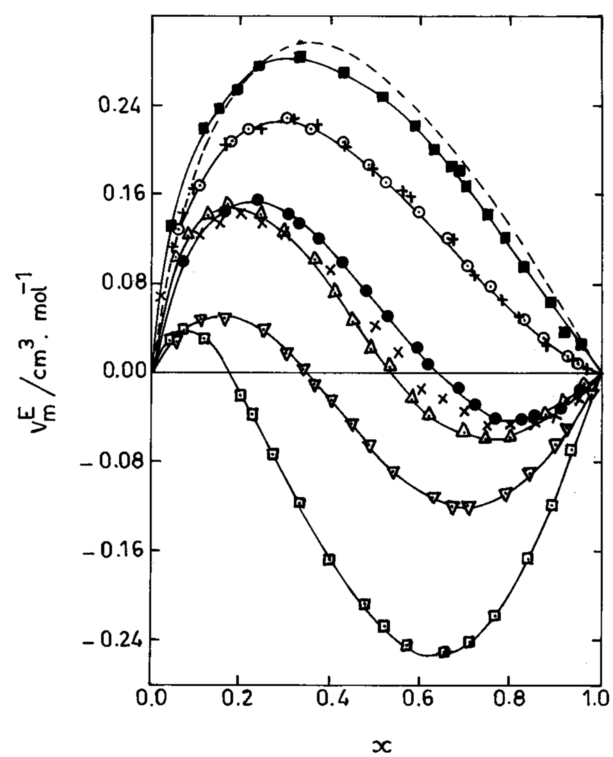

Figure 1. V_m^E plotted versus x for the following systems at 298.15 K: (⊡) xn-C_4H_9OH + $(1 - x)n$-C_5H_{12}; (◬) (this work), and (x) (Heintz et al., 1986) xn-C_4H_9OH + $(1 - x)n$-C_6H_{14}; (⊙) (this work) and (+) (Berro and Péneloux, 1984) xn-C_4H_9OH + $(1 - x)$-n-C_7H_{16}; (■) (this work) and (- - - -) (curve based on results of Liu et al., 1988) xn-C_4H_9OH + $(1 - x)n$-C_8H_{18}; (●) xn-C_4H_9OH + $(1 - x)$2,2,4-TMP; (▽) xn-C_4H_9OH + $(1 - x)CCl_4$.

Experimental Section

Materials. Pentane, hexane, heptane, and carbon tetrachloride all of HPLC quality and stated purity of the order of 99.98% and 1-butanol of HPLC quality with stated purity of 99.95% were obtained from Sisco Research Laboratories, Pvt. Ltd., Bombay. 2,2,4-Trimethylpentane of HPLC quality and stated purity of the order of 99.8%

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Table 1. Experimental Values of the Excess Molar Volumes, V_m^E, for n-C_4H_9OH + n-C_5H_{12}, + n-C_6H_{14}, + n-C_7H_{16}, + n-C_8H_{18}, + 2,2,4-TMP, and + CCl_4 at 288.15 and 298.15 K

x	V_m^E/cm³·mol⁻¹	x	V_m^E/cm³·mol⁻¹	x	V_m^E/cm³·mol⁻¹	x	V_m^E/cm³·mol⁻¹
			xn-C_4H_9OH + $(1 - x)n$-C_5H_{12}				
			T = 288.15 K				
0.0442	0.025	0.3024	−0.096	0.5398	−0.276	0.7976	−0.192
0.0709	0.037	0.3232	−0.117	0.5823	−0.284	0.8466	−0.144
0.1186	0.036	0.3727	−0.160	0.6240	−0.282	0.9392	−0.048
0.1733	0.017	0.4118	−0.198	0.7055	−0.259		
0.2811	−0.071	0.5141	−0.261	0.7536	−0.228		
			T = 298.15 K				
0.0440	0.031	0.2769	−0.073	0.5696	−0.245	0.8914	−0.119
0.0700	0.039	0.3341	−0.116	0.6539	−0.251	0.9388	−0.069
0.1193	0.030	0.3960	−0.167	0.7108	−0.243		
0.2046	−0.020	0.4780	−0.209	0.7652	−0.219		
0.2302	−0.037	0.5180	−0.229	0.8404	−0.168		
			xn-C_4H_9OH + $(1 - x)n$-C_6H_{14}				
			T = 288.15 K				
0.0332	0.041	0.2990	0.106	0.5073	0.021	0.7316	−0.043
0.0604	0.067	0.3158	0.100	0.5457	0.005	0.7681	−0.043
0.1159	0.104	0.3651	0.083	0.5796	−0.010	0.8294	−0.036
0.1707	0.121	0.3830	0.076	0.6119	−0.022	0.8821	−0.025
0.2162	0.121	0.4299	0.053	0.6518	−0.034	0.9328	−0.014
0.2678	0.116	0.4598	0.040	0.6887	−0.042	0.9517	−0.007
			T = 298.15 K				
0.0538	0.096	0.2953	0.127	0.5299	0.006	0.7989	−0.057
0.0825	0.124	0.3626	0.100	0.5828	−0.021	0.8719	−0.039
0.1227	0.140	0.4100	0.073	0.6204	−0.039	0.9179	−0.028
0.1677	0.150	0.4487	0.048	0.6975	−0.052	0.9605	−0.012
0.2442	0.142	0.4899	0.023	0.7417	−0.058		
			xn-C_4H_9OH + $(1 - x)n$-C_7H_{16}				
			T = 288.15 K				
0.0361	0.064	0.3028	0.200	0.5673	0.142	0.8419	0.033
0.0516	0.081	0.3391	0.201	0.6355	0.115	0.8675	0.025
0.0988	0.134	0.3825	0.197	0.6794	0.099	0.9006	0.011
0.1421	0.157	0.4212	0.187	0.7178	0.081	0.9379	0.005
0.1864	0.177	0.4687	0.176	0.7667	0.062	0.9789	0.001
0.2461	0.192	0.5134	0.161	0.7952	0.049		
			T = 298.15 K				
0.0581	0.127	0.3560	0.219	0.6600	0.119	0.9273	0.014
0.1077	0.167	0.4226	0.205	0.7066	0.095	0.9481	0.008
0.1753	0.205	0.4834	0.185	0.7547	0.076		
0.2163	0.217	0.5255	0.170	0.8300	0.047		
0.3029	0.226	0.5969	0.143	0.8723	0.030		
			xn-C_4H_9OH + $(1 - x)n$-C_8H_{18}				
			T = 288.15 K				
0.0483	0.099	0.2464	0.237	0.5937	0.199	0.8310	0.085
0.0763	0.139	0.3297	0.251	0.6598	0.172	0.8761	0.061
0.1085	0.168	0.3855	0.252	0.6982	0.151	0.9510	0.019
0.1418	0.189	0.4432	0.243	0.7362	0.135		
0.1941	0.217	0.5273	0.226	0.7914	0.107		
			T = 298.15 K				
0.0423	0.130	0.3345	0.282	0.6677	0.186	0.8262	0.094
0.1168	0.216	0.4286	0.268	0.6745	0.182	0.8847	0.062
0.1497	0.234	0.5124	0.246	0.7015	0.166	0.9259	0.038
0.1907	0.252	0.5857	0.221	0.7432	0.142	0.9534	0.026
0.2450	0.273	0.6278	0.199	0.7845	0.120		
			xn-C_4H_9OH + $(1 - x)$2,2,4-TMP				
			T = 288.15 K				
0.0468	0.032	0.1712	0.086	0.5036	0.003	0.8158	−0.055
0.0638	0.046	0.2757	0.078	0.5564	−0.020	0.8544	−0.045
0.0852	0.060	0.3362	0.064	0.5913	−0.033	0.8914	−0.035
0.1234	0.076	0.3677	0.056	0.6385	−0.050	0.9415	−0.020
0.1369	0.079	0.4077	0.042	0.7859	−0.061	0.9787	−0.008
			T = 298.15 K				
0.0640	0.096	0.3741	0.120	0.6169	0.008	0.8559	−0.043
0.1624	0.144	0.4259	0.097	0.6847	−0.015	0.9115	−0.034
0.2333	0.152	0.4762	0.073	0.7187	−0.029	0.9541	−0.015
0.3085	0.140	0.5283	0.050	0.7686	−0.041		
0.3286	0.132	0.5941	0.021	0.8283	−0.045		
			xn-C_4H_9OH + $(1 - x)CCl_4$				
			T = 288.15 K				
0.0465	0.024	0.2774	−0.007	0.5205	−0.087	0.7969	−0.072
0.0850	0.037	0.3239	−0.023	0.5573	−0.100	0.8387	−0.056
0.1243	0.037	0.3562	−0.036	0.5915	−0.106	0.8973	−0.033
0.1568	0.029	0.3846	−0.046	0.6409	−0.109	0.9551	−0.015
0.2358	0.010	0.4309	−0.061	0.6866	−0.104		
0.2589	0.002	0.4699	−0.074	0.7389	−0.092		
			T = 298.15 K				
0.0501	0.031	0.3442	0.004	0.5427	−0.089	0.8470	−0.090
0.1149	0.047	0.3687	−0.010	0.6337	−0.112	0.9005	−0.064
0.1633	0.049	0.4082	−0.025	0.6715	−0.119	0.9246	−0.049
0.2528	0.038	0.4489	−0.045	0.7041	−0.120	0.9727	−0.018
0.3078	0.018	0.4923	−0.064	0.7896	−0.108		

Table 2. Experimental Values of the Refractive Indexes, n_D, for n-C_4H_9OH + n-C_5H_{12}, + n-C_6H_{14}, + n-C_7H_{16}, + n-C_8H_{18}, + 2,2,4-TMP, and + CCl_4 at 298.15 K

x	n_D	x	n_D	x	n_D	x	n_D
			xn-C_4H_9OH + (1 − x)n-C_5H_{12}				
0.1458	1.3598	0.3639	1.3690	0.6336	1.3812	0.8696	1.3920
0.1527	1.3602	0.4366	1.3722	0.6862	1.3840	0.8805	1.3925
0.2006	1.3622	0.4432	1.3725	0.7447	1.3865	0.9780	1.3962
0.2539	1.3644	0.5564	1.3780	0.7834	1.3885		
0.2782	1.3654	0.5881	1.3795	0.8557	1.3915		
			xn-C_4H_9OH + (1 − x)n-C_6H_{14}				
0.0143	1.3722	0.3252	1.3796	0.6264	1.3874	0.8883	1.3940
0.1083	1.3744	0.4295	1.3824	0.6714	1.3886	0.9194	1.3946
0.1313	1.3750	0.4955	1.3842	0.7260	1.3898	0.9654	1.3958
0.2343	1.3774	0.5119	1.3844	0.7875	1.3915		
0.2932	1.3790	0.5771	1.3862	0.8261	1.3925		
			xn-C_4H_9OH + (1 − x)n-C_7H_{16}				
0.0719	1.3862	0.4037	1.3900	0.6684	1.3932	0.8517	1.3954
0.1392	1.3870	0.4647	1.3908	0.7487	1.3942	0.8670	1.3955
0.2987	1.3888	0.5682	1.3920	0.7889	1.3946	0.9189	1.3962
0.3671	1.3896	0.6111	1.3924	0.8276	1.3952	0.9591	1.3966
			xn-C_4H_9OH + (1 − x)n-C_8H_{18}				
0.1383	1.3950	0.3400	1.3956	0.5294	1.3960	0.7834	1.3965
0.1439	1.3950	0.4314	1.3958	0.5586	1.3962	0.8000	1.3965
0.2274	1.3952	0.4600	1.3958	0.6509	1.3962	0.9677	1.3965
0.3346	1.3955	0.5271	1.3960	0.7028	1.3964	0.9900	1.3968
			xn-C_4H_9OH + (1 − x)2,2,4-TMP				
0.1250	1.3900	0.3893	1.3918	0.5913	1.3938	0.7634	1.3952
0.1395	1.3902	0.3993	1.3920	0.6500	1.3942	0.8087	1.3955
0.1667	1.3905	0.5017	1.3930	0.7045	1.3945	0.8730	1.3960
0.2684	1.3912	0.5437	1.3935	0.7450	1.3950	0.9289	1.3965
			xn-C_4H_9OH + (1 − x)CCl_4				
0.0204	1.4556	0.3132	1.4378	0.5260	1.4250	0.7824	1.4100
0.0760	1.4522	0.3710	1.4340	0.5593	1.4230	0.8401	1.4068
0.0868	1.4515	0.4107	1.4318	0.6129	1.4200	0.8826	1.4040
0.1751	1.4460	0.4676	1.4282	0.6643	1.4170	0.9413	1.4005
0.2764	1.4400	0.5230	1.4255	0.7512	1.4120	0.9976	1.3975

was obtained from S. D. Fine Chemicals Ltd., Bombay. These chemicals were used without any further purification. Octane (Spectrochem product) of AR quality was distilled fractionally before use. The densities of these liquids were measured using a single-capillary pycnometer and have been reported recently (Nath, 1996) as 0.626 22, 0.659 40, 0.683 78, 0.702 55, 0.691 92, 1.594 08, and 0.809 65 g·cm^{-3} for n-C_5H_{12}, n-C_6H_{14}, n-C_7H_{16}, n-C_8H_{18}, 2,2,4-TMP, CCl_4, and n-C_4H_9OH, respectively, at T = 293.15 K, as compared with the corresponding literature (Riddick and Bunger, 1970; Timmermans, 1950) values 0.626 24, 0.659 37, 0.683 76, 0.702 52, 0.691 93, 1.5940, and 0.8097 g·cm^{-3} for the various liquids in the same order.

Methods. (i) Excess molar volumes, V_m^E, were measured with an imprecision of the order of ±0.002 cm^3·mol^{-1}, using a two-limbed Pyrex glass dilatometer which was similar to that used in earlier measurements (Nath and Chaudhary, 1992; Nath and Rashmi, 1990). Known amounts of the two liquid components were confined over mercury in the absence of air spaces in the two limbs of the dilatometer. The dilatometer (mounted on a stand) was immersed in a thermostat which was controlled to ±0.01 K. The mixing of the components was achieved by rocking the cell back and forth through a definite angle, and the mercury levels in the capillary of the dilatometer were noted with a cathetometer which could read correct to ±0.001 cm. The working of the dilatometer was checked by measuring V_m^E for C_6H_6 + c-C_6H_{12} at 298.15 K. The values of V_m^E for C_6H_6 + c-C_6H_{12} obtained in this work were found to be in good agreement with the data of Stokes et al. (1970).

(ii) The refractive indexes (sodium D line), n_D, of the pure samples and the various liquid mixtures, were measured with an accuracy of ±0.0002 at (298.15 ± 0.01) K, using a thermostated Abbe refractometer. The values of n_D of n-C_5H_{12}, n-C_6H_{14}, n-C_7H_{16}, n-C_8H_{18}, 2,2,4-TMP, CCl_4, and n-C_4H_9OH at 298.15 K were found to be 1.3545, 1.3720, 1.3852, 1.3948, 1.3890, 1.4572, and 1.3970, respectively, which can be compared with the literature (Riddick and Bunger, 1970) values 1.354 72, 1.372 26, 1.385 11, 1.395 05, 1.388 98, 1.457 39, and 1.3973, respectively, for the above liquids in the same order.

Results and Discussion

The experimental values of V_m^E for the mixtures of n-C_4H_9OH + n-C_5H_{12}, + n-C_6H_{14}, + n-C_7H_{16}, + n-C_8H_{18}, + 2,2,4-TMP, and + CCl_4 at 288.15 K and 298.15 K are given in Table 1, whereas the values of n_D for these mixtures at 298.15 K are given in Table 2. Values of V_m^E for these mixtures at 298.15 K are plotted against the mole fraction of n-C_4H_9OH, x, in Figure 1, where the values of V_m^E for xn-C_4H_9OH + (1 − x)n-C_6H_{14}, xn-C_4H_9OH + (1 − x)n-C_7H_{16}, and xn-C_4H_9OH + (1 − x)n-C_8H_{18} at 298.15 K, obtained by other workers, are also presented. V_m^E has been fitted by the method of least squares to the equation

Table 3. Values of the Coefficients A_j of Eq 1 and the Standard Deviations, $\delta(V_m^E)$, for the Various Mixtures

		cm^3·mol^{-1}				
system	T/K	A_1	A_2	A_3	A_4	$\delta(V_m^E)$
xn-C_4H_9OH + (1 − x)n-C_5H_{12}	288.15	0.053 29	−2.540 40	−1.498 16	3.577 95	0.0023
	298.15	−0.019 33	−2.188 81	−0.276 87	1.389 07	0.0032
xn-C_4H_9OH + (1 − x)n-C_6H_{14}	288.15	0.819 02	−1.665 21	−0.822 91	1.560 36	0.0027
	298.15	1.032 65	−2.454 65	−0.023 70	1.160 18	0.0031
xn-C_4H_9OH + (1 − x)n-C_7H_{16}	288.15	1.187 13	−1.245 64	0.129 13	−0.092 95	0.0029
	298.15	1.388 23	−2.042 02	1.457 03	−0.751 11	0.0019
xn-C_4H_9OH + (1 − x)n-C_8H_{18}	288.15	1.441 23	−1.471 70	0.839 81	−0.450 86	0.0031
	298.15	1.698 18	−2.327 92	2.032 34	−0.939 82	0.0023
xn-C_4H_9OH + (1 − x)2,2,4-TMP	288.15	0.592 86	−1.103 15	−1.304 86	1.478 62	0.0025
	298.15	1.019 30	−1.871 11	−0.114 57	0.479 32	0.0033
xn-C_4H_9OH + (1 − x)CCl_4	288.15	0.181 85	−1.307 17	−0.424 22	1.374 13	0.0029
	298.15	0.344 21	−1.280 11	−0.941 69	1.181 93	0.0031

Table 4. Values of the Coefficients B_j of Eq 2 and the Standard Deviations, $\delta(n_D)$, for the Variuos Mixtures at 298.15 K

system	B_1	B_2	B_3	B_4	$\delta(n_D)$
xn-C_4H_9OH + (1 − x)n-C_5H_{12}	1.354 61	0.032 48	0.025 65	−0.015 65	0.000 15
xn-C_4H_9OH + (1 − x)n-C_6H_{14}	1.372 37	0.019 86	0.010 03	−0.005 54	0.000 23
xn-C_4H_9OH + (1 − x)n-C_7H_{16}	1.385 29	0.011 52	0.000 88	−0.000 62	0.000 07
xn-C_4H_9OH + (1 − x)n-C_8H_{18}	1.394 73	0.002 08	0.001 26	−0.001 29	0.000 11
xn-C_4H_9OH + (1 − x)2,2,4-TMP	1.389 09	0.007 21	0.001 46	−0.000 73	0.000 12
xn-C_4H_9OH + (1 − x)CCl_4	1.457 05	−0.064 89	0.010 67	−0.005 72	0.000 16

$$V_m^E/x(1-x) = \sum_{j=1}^{m} A_j Y_j \qquad (1)$$

where $Y_j = [x - 1/(1 + Dx)]^{j-1}$ and where x denotes the mole fraction of n-C_4H_9OH in the various mixtures, and A_j are coefficients characteristic of a system at a given temperature. As pointed out by Berro and Péneloux (1984), the calculations were performed for all the systems with $D = 25$. The values of the coefficients A_j of eq 1, along with the standard deviations, $\delta(V_m^E)$, for the various mixtures are given in Table 3.

The refractive indexes, n_D, of the present mixtures at 298.15 K have been fitted by the method of least-squares to the equation

$$n_D = \sum_{j=1}^{m} B_j x^{j-1} \qquad (2)$$

The values of the coefficients B_j of eq 2, along with the standard deviations, $\delta(n_D)$, are given in Table 4. The values of the quantity Δn_D, which refers to the deviations of the observed refractive indexes n_D of the mixtures from a mole fraction average, are found to be slightly negative throughout the whole composition range for xn-$C_4H_9OH + (1 - x)n$-C_6H_{14}, xn-$C_4H_9OH + (1 - x)n$-C_8H_{18}, and xn-$C_4H_9OH + (1 - x)CCl_4$. Δn_D is slightly negative at low mole fractions of n-C_4H_9OH and slightly positive at its higher mole fractions, for xn-$C_4H_9OH + (1 - x)n$-C_5H_{12} and xn-$C_4H_9OH + (1 - x)$2,2,4-TMP. Δn_D is found to be very slightly positive for xn-$C_4H_9OH + (1 - x)n$-C_7H_{16}.

V_m^E is positive at lower mole fractions of n-C_4H_9OH for xn-$C_4H_9OH + (1 - x)n$-C_5H_{12}, $+ (1 - x)n$-C_6H_{14}, $+ (1 - x)$2,2,4-TMP, and $+ (1 - x)CCl_4$, and an inversion of sign of V_m^E from positive to negative occurs for these mixtures at higher mole fractions of n-C_4H_9OH, at both the temperatures 288.15 K and 298.15 K. V_m^E is found to be positive throughout the entire range of composition for xn-$C_4H_9OH + (1 - x)n$-C_7H_{16} and $+ (1 - x)n$-C_8H_{18} at 288.15 K and 298.15 K. The inversion of sign of V_m^E with increasing values of x may be attributed to the contributions to V_m^E due to the self-association of the butanol molecules through hydrogen bonding. At $x = 0.5$, and at 298.15 K, V_m^E for the various mixtures of n-C_4H_9OH with the alkanes has the sequence

$$n\text{-}C_8H_{18} > n\text{-}C_7H_{16} > 2,2,4\text{-TMP} > n\text{-}C_6H_{14} > n\text{-}C_5H_{12}$$

The data show that V_m^E increases with temperature at lower mole fractions of n-C_4H_9OH for xn-$C_4H_9OH + (1 - x)n$-C_6H_{14}, xn-$C_4H_9OH + (1 - x)n$-C_7H_{16}, xn-$C_4H_9OH + (1 - x)n$-C_8H_{18}, and xn-$C_4H_9OH + (1 - x)$2,2,4-TMP and is almost independent of temperature at high x values for these systems. For xn-$C_4H_9OH + (1 - x)CCl_4$, V_m^E increases with temperature at low mole fractions of n-C_4H_9OH and decreases with an increase of temperature at high mole fractions of n-C_4H_9OH. For xn-$C_4H_9OH + (1 - x)n$-C_5H_{12}, V_m^E increases with temperature at $x \sim 0.5$ and is practically independent of temperature in the other concentration range.

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