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Volumetric Properties of Binary Mixtures of *n*-Butylacetate with 1-Butanol, 2-Butanol, 1,2-Butanediol and 1,3-Butanediol at Different Temperatures

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Densities for binary mixtures formed by *n*-butylacetate with 1-butanol, 2-butanol, 1,2-butanediol and 1,3-butanediol have been determined over the full range of compositions in the temperature range 298.15 to 308.15 K at the ambient pressure 81.5 kPa, using an Anton Paar DMA 4500 oscillating density meter. The excess molar volumes (V_m^E), excess partial molar volumes (V_i^E), were calculated from experimental densities. The excess molar volumes for all binary mixtures positive and increase with increasing temperatures from 298.15 to 308.15 K. This means that the interaction of like moleculars is stronger than the unlike molecular intractions. The binary data of V_m^E were correlated as a function of the mole fraction by using the Redlich-Kister type function to determine the fitting parameters and the standard deviations. The experimental data of the constituent binaries are analyzed to discuss the nature and strength of intermolecular intractions in these mixtures.

Key Words: Excess molar volume, Excess partial molar volume, *n*-Butylacetate, 1-Butanol, 2-Butanol, 1,2-Butanediol, 1,3-Butanediol.

INTRODUCTION

Resarch activities of our laboratory comprise, the systematic measurements of volumetric properties of different groups of organic compounds[1-6]. Studies of the thermodynamic properties of binary mixtures are of considerable importance in the fundamental understanding of the nature of the interactions between unlike molecules. In recent years there has been considerable interest in theoretical and experimental investigations of the excess thermodynamic properties of binary mixtures[7,8]. In principle, interactions between the molecules can be established from the study of the deviations from ideal behaviour of physical properties such as, excess molar volume and isentropic compressibility. The negative or positive deviations from the ideal value depend on the type and the extent of the interactions between the unlike molecules, as well as on the composition and the temperature. In the case of

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excess molar volume, when the interactions between the molecules of the mixed components are weaker than in the pure component, the excess molar volume will be positive, whereas when the association between the mixed components (the packing effect) predominates, the excess molar volume would be negative[9]. Geometrical solution models are considered to predict excess molar volumes for the ternary mixture from binary contribution because of dependence of interactions in ternary systems on the interaction in binary systems[10-13]. This paper reports the densities (ρ), excess molar volumes (V_m^E) and excess partial molar volume (V_i^E) binary mixture of n-butylacetate with for 1-butanol, 2-butanol, 1,2-butanediol, 1,3-butanediol atmospheric pressure and at different temperatures 298.15, 303.15 and 308.15 K.

EXPERIMENTAL

n-Butylacetate, 1-butanol, 2-butanol, 1,2-butanediol and 1,3-butanediol were high purity grade reagents from Merck. The purity of reagents is summarized in Table-1. It was checked by comparing the measured densities and refractive indices of the compounds with those reported in the literature[14-32], showing a good agreement (Table-1). No further purification was attempted owing to their high purity grade.

The density of the pure compounds and mixtures was measured by means of an Anton Parr DMA 4500 oscillating U-tube density meter, provided with automatic viscosity correction. The densities of the samples were observed with a reproducibility of $\pm 1 \times 10^{-5}$ g cm^{-3}. Total uncertainty in the density measurement, as reported by the equipment manufacturers, was $\pm 1 \times 10^{-5}$ g cm^{-3} at a confidence level of 95 % and the accuracy for the density measurement was $\pm 5 \times 10^{-5}$ g cm^{-3}.

The temperature in the cell was regulated to ± 0.01 K with a solid state thermostat. The apparatus was calibrated once a day with dry air and double distilled freshwater. Air tight stopper bottles were used for the preparation of the mixtures. The mass of the dry bottle was first determined. The less volatile component of the mixture was introduced in the bottle and the mass of the bottle along with the two components was determined. All the weighing was performed on an electronic balance (AB 204-N Mettler) precise to ± 0.1 mg. Refractive indices were measured at 298.15 to 308.15 K with an Abbe´ refractometer. The accuracy of the refractive index measured is in the order of ± 0.0002 and for the temperature of measurement it was ± 0.01 K. Each mixture was used immediately after it was well mixed by shaking.

RESULTS AND DISCUSSION

The density of pure components and binary mixtures was measured at temperatures 298.15 to 308.15 K at atmospheric pressure. Tables 2 and 3 show the results for pure components, *n*-butylacetate + 1-butanol, *n*-butylacetate + 2-butanol, *n*-butylacetate + 1,2-butanediol, *n*-butylacetate + 1,3-butanediol, respectively.

The values of excess molar volumes (V_m^E) were calculated from the mixture densities (ρ) and the densities (ρ_i) and molar masses (M_i) of the pure components i (i = 1, 2) by using the relation:

TABLE-1
SOURCE, PURITY GRADES, REFRACTIVE INDEX n_D
AND DENSITY ρ, OF PURE COMPONENT

Component	Purity (mass fraction) (%)	Temp. (K)	ρ (g cm^{-1})		n_D^{25}		$10^4\alpha$ (K^{-1})
			Exp.	Lit.	Exp.	Lit.	
n-Butylacetate	99.5	298.15	0.87636	0.876360[**17**] 0.876180[**19**] 0.876120[**20**] 0.876030[**21**] 0.876160[**22**] 0.876190[**23**] 0.876450[**25**] 0.876330[**26**] 0.876350[**27**]	1.3192	1.39180[**17**] 1.39191[**27**]	11.19
		303.15	0.87131	0.871290[**17**] 0.871310[**19**] 0.871340[**24**]			11.84
		308.15	0.86604	0.866240[**19**] 0.865430[**23**] 0.866250[**25**] 0.866220[**26**]			12.52
1-Butanol	99.5	298.15	0.80573	0.805540[**15**] 0.805750[**17**] 0.805750[**28**] 0.805842[**29**] 0.806200[**30**]	1.3976	1.3974[**14**]	8.71
		303.15	0.80218	0.801900[**15**]			8.95
		308.15	0.79855	0.798270[**15**]			9.19
2-Butanol	99.0	298.15	0.80218	0.802280[**15**]	1.3953	1.39550[**14**] 1.39488[**31**] 1.39500[**32**]	9.39
		303.15	0.79826	0.797990[**15**]			10.26
		308.15	0.79399	0.793720[**15**]			11.15
1,2-Butanediol	99.5	298.15	0.99881	0.998870[**18**]	1.4381	1.4382[**16**]*	6.79
		303.15	0.99526	0.995310[**18**]			7.45
		308.15	0.99139	0.991430[**18**]			8.13
1,3-Butanediol	99.5	298.15	1.00005	1.000080[**15**]	1.4389	1.43900[**17**]	5.85
		303.15	0.99712	0.997160[**15**]			5.91
		308.15	0.99326	0.994230[**15**]			5.98

*T = 293.15 K

TABLE-2
DENSITIES, EXCESS MOLAR VOLUMES AND EXCESS PARTIAL MOLAR VOLUMES FOR THE BINARY MIXTURES OF *x n*-BUTYLACETATE + (1-*x*) 1-BUTANOL AND *x n*-BUTYLACETATE + (1-*x*) 2-BUTANOL AT 298.15 K to 308.15 K AND ATMOSPHERIC PRESSURE

x_1	ρ (g cm^{-3})	V_m^E (cm^3 mol^{-1})	V_1^E (cm^3 mol^{-1})	V_2^E (cm^3 mol^{-1})
x n-Butylacetate + (1-*x*) 1-butanol, T = 298.15 K				
0.0496	0.81035	0.037	0.709	0.003
0.0994	0.81481	0.072	0.588	0.013
0.1500	0.81925	0.095	0.479	0.028
0.2003	0.82350	0.116	0.389	0.048
0.3002	0.83158	0.141	0.265	0.088
0.3501	0.83542	0.149	0.228	0.106
0.3978	0.83898	0.153	0.203	0.121
0.4980	0.84609	0.159	0.171	0.147
0.5501	0.84960	0.159	0.157	0.162
0.5993	0.85282	0.158	0.142	0.181
0.6997	0.85909	0.149	0.104	0.254
0.7984	0.86495	0.126	0.058	0.396
0.8997	0.87076	0.078	0.016	0.633
x n-Butylacetate + (1-*x*) 1-butanol, T = 303.15 K				
0.0496	0.80673	0.034	0.757	-0.002
0.0994	0.81107	0.072	0.708	0.000
0.1500	0.81535	0.103	0.630	0.011
0.2003	0.81945	0.132	0.533	0.032
0.3002	0.82725	0.172	0.360	0.089
0.3501	0.83100	0.181	0.299	0.118
0.3978	0.83446	0.189	0.258	0.143
0.4980	0.84142	0.195	0.204	0.187
0.5501	0.84487	0.194	0.182	0.210
0.5993	0.84803	0.192	0.160	0.240
0.6997	0.85421	0.178	0.104	0.347
0.7984	0.86005	0.140	0.043	0.529
0.8997	0.86582	0.079	0.004	0.743
x n-Butylacetate + (1-*x*) 1-butanol, T = 308.15 K				
0.0496	0.80290	0.045	0.941	0.001
0.0994	0.80704	0.095	0.844	0.009
0.1500	0.81120	0.129	0.709	0.028
0.2003	0.81518	0.161	0.572	0.057
0.3002	0.82284	0.196	0.362	0.126
0.3501	0.82650	0.206	0.297	0.158
0.3978	0.82990	0.212	0.256	0.181
0.4980	0.83672	0.215	0.215	0.215
0.5501	0.84009	0.214	0.200	0.230
0.5993	0.84318	0.211	0.184	0.253
0.6997	0.84921	0.197	0.132	0.352
0.7984	0.85488	0.165	0.065	0.555
0.8997	0.86057	0.096	0.013	0.849

x n-Butylacetate + (1-*x*) 2-butanol T = 298.15 K				
0.0493	0.80645	0.102	1.955	0.006
0.1005	0.81080	0.194	1.720	0.025
0.1494	0.81485	0.273	1.503	0.056
0.1991	0.81892	0.338	1.302	0.098
0.2997	0.82695	0.435	0.965	0.209
0.3500	0.83084	0.470	0.831	0.274
0.4004	0.83470	0.491	0.716	0.343
0.5004	0.84209	0.512	0.526	0.498
0.6000	0.84919	0.499	0.366	0.694
0.6991	0.85609	0.443	0.224	0.959
0.7501	0.85955	0.403	0.159	1.130
0.7997	0.86292	0.348	0.104	1.323
0.9000	0.86968	0.199	0.025	1.770
x n-Butylacetate + (1-*x*) 2-butanol T = 303.15 K				
0.0493	0.80244	0.104	2.076	0.000
0.1005	0.80663	0.207	1.933	0.013
0.1494	0.81057	0.293	1.719	0.043
0.1991	0.81451	0.367	1.482	0.093
0.2997	0.82230	0.480	1.061	0.232
0.3500	0.82610	0.520	0.902	0.309
0.4004	0.82991	0.540	0.774	0.385
0.5004	0.83719	0.563	0.581	0.542
0.6000	0.84423	0.545	0.415	0.747
0.6991	0.85102	0.492	0.245	1.066
0.7501	0.85450	0.442	0.162	1.286
0.7997	0.85785	0.383	0.091	1.529
0.9000	0.86472	0.203	0.007	1.989
x n-Butylacetate + (1-*x*) 2-butanol T = 308.15 K				
0.0493	0.79803	0.115	2.161	0.002
0.1005	0.80217	0.217	2.017	0.014
0.1494	0.80605	0.304	1.841	0.039
0.1991	0.80985	0.388	1.645	0.081
0.2997	0.81735	0.526	1.249	0.213
0.3500	0.82105	0.572	1.068	0.300
0.4004	0.82475	0.601	0.902	0.400
0.5004	0.83192	0.627	0.623	0.628
0.6000	0.83898	0.594	0.405	0.894
0.6991	0.84575	0.530	0.237	1.207
0.7501	0.84925	0.471	0.166	1.392
0.7997	0.85255	0.413	0.109	1.590
0.9000	0.85938	0.225	0.028	2.050

TABLE-3
DENSITIES, EXCESS MOLAR VOLUMES AND EXCESS PARTIAL MOLAR VOLUMES FOR THE BINARY MIXTURES OF *x n*-BUTYLACETATE + (1-*x*)1,2-BUTANEDIOL AND *x n*-BUTYLACETATE + (1-*x*) 1,3-BUTANEDIOL AT 298.15 K TO 308.15 K AND ATMOSPHERIC PRESSURE

x_1	ρ (g cm^{-3})	V_m^E (cm^3 mol^{-1})	V_1^E (cm^3 mol^{-1})	V_2^E (cm^3 mol^{-1})
x n--Butylacetate +(1-*x*) 1,2-butanediol T = 298.15 K				
0.0996	0.97935	0.225	1.866	0.041
0.1513	0.97032	0.306	1.533	0.089
0.1995	0.96240	0.368	1.274	0.144
0.3000	0.94732	0.454	0.872	0.276
0.4000	0.93389	0.497	0.611	0.415
0.4917	0.92283	0.494	0.446	0.547
0.5995	0.91095	0.468	0.297	0.726
0.7000	0.90092	0.412	0.179	0.945
0.7997	0.89195	0.312	0.083	1.237
0.8978	0.88392	0.181	0.020	1.587
x n-Butylacetate +(1-*x*) 1,2-butanediol T = 303.15 K				
0.0496	0.98518	0.117	2.475	0.006
0.0996	0.97530	0.252	2.194	0.029
0.1513	0.96600	0.350	1.875	0.075
0.1995	0.95790	0.422	1.584	0.136
0.3000	0.94242	0.536	1.070	0.306
0.4000	0.92885	0.581	0.711	0.498
0.4917	0.91765	0.583	0.487	0.677
0.5995	0.90575	0.544	0.300	0.901
0.7000	0.89582	0.462	0.163	1.156
0.7997	0.88694	0.336	0.060	1.465
0.8978	0.87892	0.189	0.007	1.755
x n-Butylacetate +(1-*x*) 1,2-butanediol T = 308.15 K				
0.0496	0.98075	0.160	2.967	0.016
0.0996	0.97079	0.295	2.432	0.058
0.1513	0.96128	0.405	1.974	0.124
0.1995	0.95307	0.482	1.626	0.198
0.3000	0.93750	0.591	1.094	0.372
0.4000	0.92385	0.630	0.749	0.556
0.4917	0.91255	0.631	0.524	0.737
0.5995	0.90056	0.589	0.317	0.986
0.7000	0.89062	0.494	0.162	1.273
0.7997	0.88170	0.360	0.053	1.597
0.8978	0.87370	0.196	0.004	1.864
x n-Butylacetate +(1-*x*) 1,3-butanediol T = 298.15 K				
0.0495	0.98950	0.163	3.166	0.005
0.0978	0.97985	0.306	2.857	0.030
0.1500	0.97015	0.439	2.458	0.087
0.1974	0.96180	0.551	2.088	0.165
0.3000	0.94589	0.689	1.389	0.395
0.3997	0.93228	0.755	0.904	0.654
0.4992	0.92032	0.750	0.581	0.916
0.6000	0.90952	0.688	0.350	1.197

0.6973	0.90018	0.581	0.177	1.516
0.8000	0.89131	0.420	0.048	1.900
0.8981	0.88372	0.217	-0.004	2.176
x n-Butylacetate +(1-*x*) 1,3-butanediol T = 303.15 K				
0.0495	0.98608	0.193	3.396	0.018
0.0978	0.97635	0.331	2.879	0.058
0.1500	0.96650	0.464	2.471	0.115
0.1974	0.95810	0.570	2.188	0.175
0.3000	0.94151	0.755	1.714	0.331
0.3997	0.92746	0.850	1.304	0.552
0.4992	0.91500	0.885	0.894	0.889
0.6000	0.90383	0.853	0.506	1.364
0.6973	0.89450	0.729	0.210	1.909
0.8000	0.88596	0.504	0.029	2.438
0.8981	0.87852	0.260	-0.016	2.659
x n-Butylacetate +(1-*x*) 1,3-butanediol T = 308.15 K				
0.0495	0.98298	0.190	3.569	0.008
0.0978	0.97292	0.344	3.224	0.036
0.1500	0.96268	0.500	2.833	0.091
0.1974	0.95398	0.623	2.492	0.163
0.3000	0.93705	0.818	1.855	0.373
0.3997	0.92262	0.932	1.367	0.634
0.4992	0.91006	0.957	0.964	0.963
0.6000	0.89872	0.924	0.603	1.406
0.6973	0.88914	0.812	0.302	1.964
0.8000	0.88048	0.581	0.077	2.629
0.8981	0.87308	0.309	-0.008	3.076

$$V_m^E = [xM_1 + (1-x)M_2]/\rho - [xM_1/\rho_1 + xM_2/\rho_2] \quad (1)$$

where subscript 1 and 2 refers to *n*-butylacetate and 1-butanol,2-butanol, 1,2-butanediol and 1,3-butanediol respectively and 'x' stands for the mole fraction of the *n*-butylacetate. The accuracy in the calculation of excess molar volume was estimated as ± 0.0008 (cm^3 mol^{-1}). Calculated excess molar volumes at temperatures between 298.15 and 308.15 K and atmospheric pressure appear in Tables 2 and 3 and are graphically represented in Figs. 1-4.

Each set of results were fitted using Redlich-Kister[33] expression for the binary mixtures as follows:

$$Y = x_i x_j \sum_{p=0}^{4} \left(\sum_{q=0}^{2} A_{pq} T^q \right) (x_i - x_j)^p \quad (2)$$

where x_i and x_j are the mole fraction; A_{pq} are the temperature independent parameters for the binary mixtures and T is the absolute temperature. These parameters were obtained by the unweighted least-squares method. The parameters A_{pq} for all the binary mixtures are listed in Table-4.

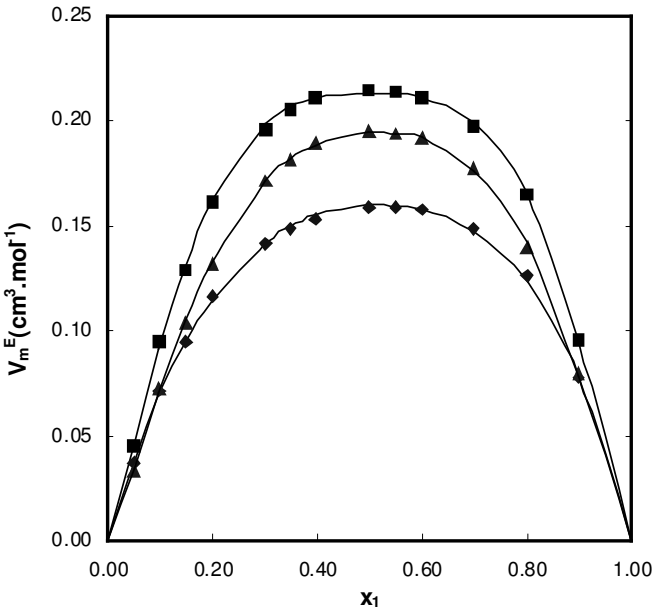

Fig. 1. Excess molar volumes of *x n*-butylacetate + (1-*x*) 1-butanol at atmospheric pressure and different temperatures against mole fraction. Solid curves were calculated from eqn. 2 in Table-2; (◆), T =298.15 K; (▲), T =303.15 K; (■), T =308.15 K

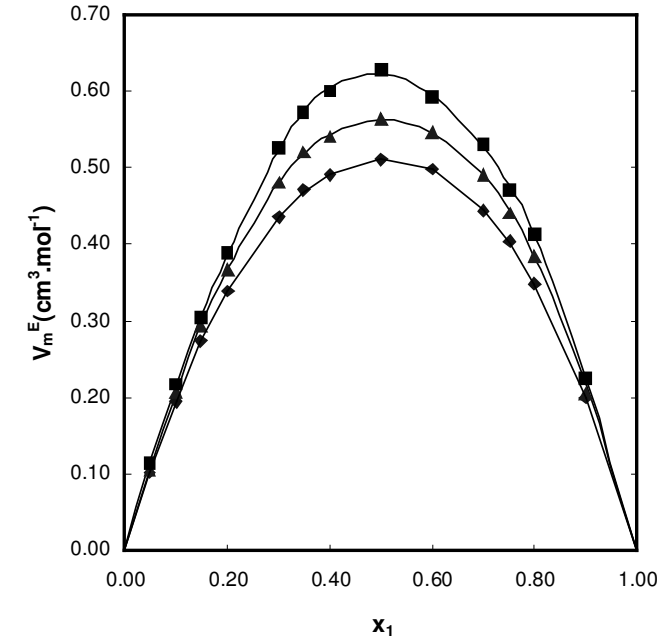

Fig. 2. Excess molar volumes of *x n*-butylacetate + (1-*x*) 2-butanol at atmospheric pressure and different temperatures against mole fraction. Solid curves were calculated from eqn. 2 in Table-2; (◆), T =298.15 K; (▲), T =303.15 K; (■), T =308.15 K

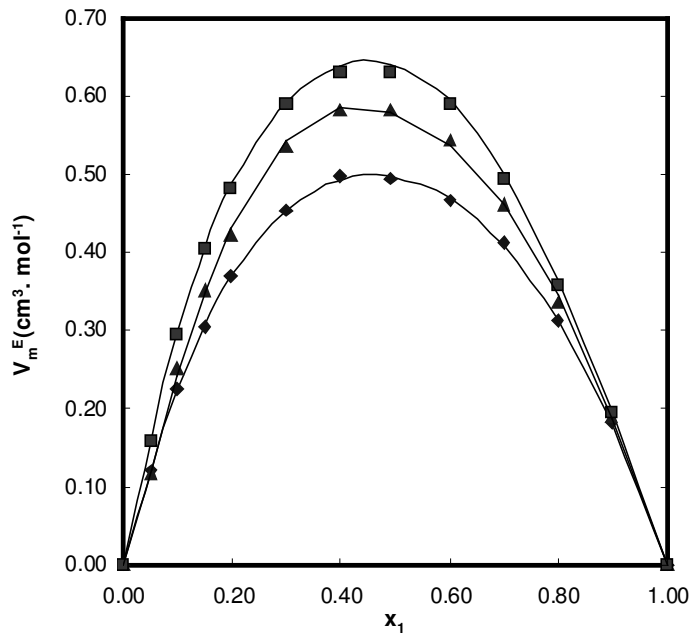

Fig. 3. Excess molar volumes of *x n*-butylacetate + (1-*x*) 1,2-butandiol at atmospheric pressure and different temperatures against mole fraction. Solid curves were calculated from eqn. 2 in Table-2; (◆), T =298.15 K; (▲), T =303.15 K; (■), T =308.15 K

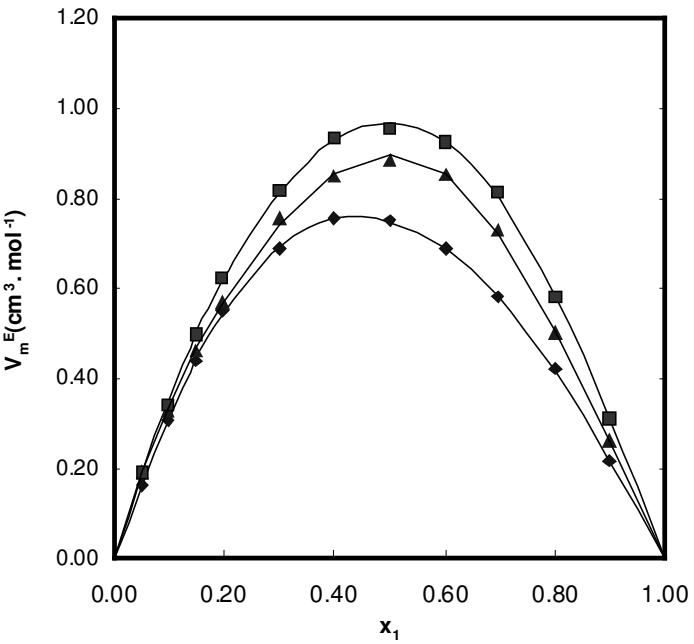

Fig. 4. Excess molar volumes of *x n*-butylacetate + (1-*x*) 1,3-butandiol at atmospheric pressure and different temperatures against mole fraction. Solid curves were calculated from eqn. 2 in Table-2; (◆), T =298.15 K; (▲), T =303.15 K; (■), T =308.15 K

TABLE-4
PARAMETERS OF REDLICH-KISTER (2) AND THE STANDARD DEVIATIONS FOR BINARY MIXTURES {(*n*-BUTYLACETATE + 1-BUTANOL, +2-BUTANOL, +1,2-BUTANEDIOL AND 1,3-BUTANEDIOL) AT T = (298.15, 303.15 AND 308.15) K

	p					σ (V_m^E/cm^3 mol^{-1})
q	0	1	2	3	4	
{*n*-Butylacetate (1) + 1-butanol (2)}						
0	-130.056	0.84082	-0.0013	29.6942	-0.2009	
1	0.00034	426.867	-2.8349	0.0047	21.4267	0.001
2	-0.13791	0.0002	598.642	-3.9195	0.0064	
{*n*-Butylacetate (1) + 2-butanol (2)}						
0	89.1451	-0.618656	0.00109519	197.947	-1.31272	
1	0.0022	-1317.78	8.7279	-0.0144	-674.975	0.003
2	4.4753	-0.0074	2267.45	-14.9704	0.0247	
{*n*-Butylacetate (1) + 1,2-butanediol (2)}						
0	-288.696	1.8665	-0.003	-265.684	1.7313	
1	-0.0028	333.913	-2.195	0.00361101	902.753	0.004
2	-5.9821	0.00991	972.418	-6.4119	0.0106	
{*n*-Butylacetate (1) + 1,3-butanediol (2)}						
0	-543.445	3.52261	-0.0057	1278.63	-8.3688	
1	0.0137	2600.66	-17.1358	0.0282	-2457.95	0.005
2	16.1674	-0.0266	-2839.32	18.7304	-0.0309	

The temperature dependence of density of the pure components was fitted to the equation:

$$\rho(T)/g\ cm^{-3} = \sum_{i=0}^{4} a_i T^i \quad (3)$$

The thermal expansion coefficients of pure components at different temperature are presented in Table-1. The average uncertainty in the thermal expansion coefficient is estimated to be 5×10^{-6} K.

$$\alpha = -\rho^{-1}(\partial\rho/\partial T)_P \quad (4)$$

In each case, the optimum number of coefficients was ascertained from an examination of the variation of standard deviation σ(Y) with:

$$\sigma(Y) = \left[\sum \left(Y^E_{mexp.,i} - Y^E_{mcal.,i}\right)^2 \Big/ (n-p)\right]^{1/2} \quad (5)$$

where n and p are the number of experimental points and number of parameters retained in the respective equation, standard deviation for eqn. 2 in Table-4.

The partial molar quantities are important in the study of the dependence of an extensive property on phase composition at constant pressure and temperature, showing its trend with composition. The excess partial molar volumes (V_i^E) for these mixtures can be determined from excess molar volume data using the expression[14].

$$V_i^E = V_m^E + (1 - x_i)\left(\frac{\partial V_m^E}{\partial x_i}\right)_{x_j,p,T} \quad (6)$$

$\left(\frac{\partial V_m^E}{\partial x_i}\right)_{x_j,p,T}$ were calculated from eqn. 2 using the parameters in Table-4. The excess partial molar volumes (V_i^E) are given in Tables 2 and 3 and are graphically represented in Figs. 5 and 6.

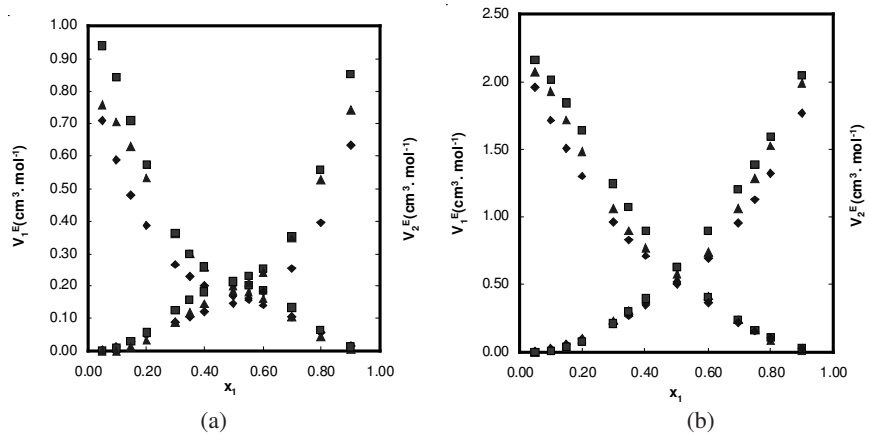

Fig. 5. Plot of: (a) Excess partial molar volumes against mol fraction for {*n*-butylacetate (1) + 1-butanol (2)}, (b) Excess partial molar volumes against mol fraction for {*n*-butylacetate (1) + 2-butanol (2)} at the temperatures 298.15 (◆), 303.15 (▲), 308.15 (■). Solid curves represent the values calculated from eqn. 5 in Table-2

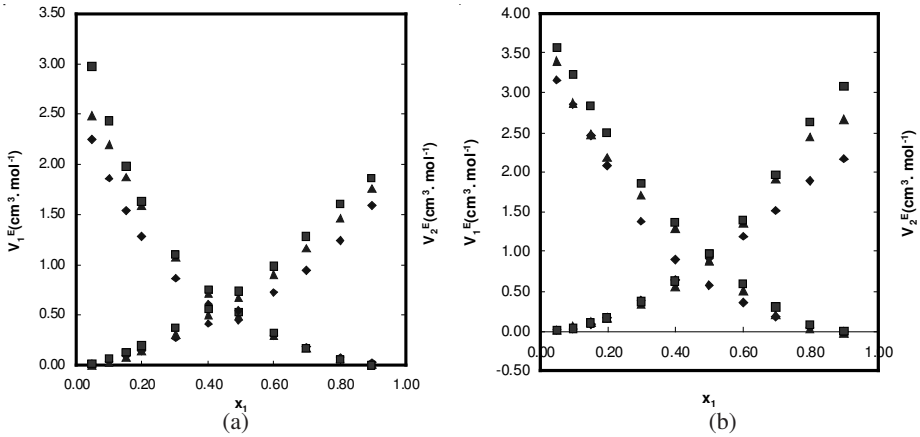

Fig.6. Plot of: (a) Excess partial molar volumes against mol fraction for {*n*-butylacetate (1) + 1,2-butanediol (2)}, (b) Excess partial molar volumes against mol fraction for {*n*-butylacetate (1) + 1,3-butanediol (2)} at the temperatures 298.15 (◆), 303.15 (▲), 308.15 (■). Solid curves represent the values calculated from eqn. 5 in Table-3

The excess molar volumes for all binary mixtures are positive and increase with increasing temperatures from 298.15 to 308.15 K. The positive V_m^E values indicate that there is a volume contraction on mixing. This means the intraction between *n*-butylacetate and 1-butanol, 2-butanol and 2-butanol, 1,2-butanediol and 1,2-butandiol, 1,3-butanediol and 1,3-butandiol is stronger than that of *n*-butylacetate with 1-butanol, 2-butanol, 1,2-butanediol, 1,3-butanediol.

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