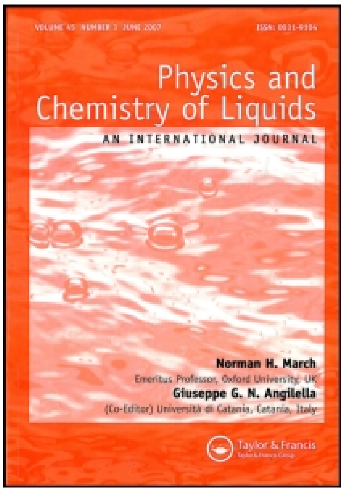

Physics and Chemistry of Liquids
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713646857

Density and excess molar volumes of binary mixtures of sulfolane with methanol, <i>n</i>-propanol, <i>n</i>-butanol, and <i>n</i>-pentanol at 298.15-323.15 K and atmospheric pressure

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To cite this Article Motin, M. A. , Ali, M. Azhar and Sultana, Shila(2007) 'Density and excess molar volumes of binary mixtures of sulfolane with methanol, <i>n</i>-propanol, <i>n</i>-butanol, and <i>n</i>-pentanol at 298.15-323.15 K and atmospheric pressure', Physics and Chemistry of Liquids, 45: 2, 221 — 229

To link to this Article: DOI: 10.1080/00319100601088713

URL: http://dx.doi.org/10.1080/00319100601088713

Physics and Chemistry of Liquids
Vol. 45, No. 2, April 2007, 221–229

Density and excess molar volumes of binary mixtures of sulfolane with methanol, *n*-propanol, *n*-butanol, and *n*-pentanol at 298.15–323.15 K and atmospheric pressure

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(*Received 14 September 2006; in final form 27 October 2006*)

Densities, ρ and excess molar volumes, V^E of the binary mixtures of sulfolane, +methanol, +*n*-propanol, +*n*-butanol, and +*n*-pentanol were measured at temperatures 298.15, 303.15, 308.15, 313.15, and 318.15 K, respectively, covering the whole composition range except methanol at 303.15–323.15 K. The V^E for the systems were found to be negative and large in magnitude. The values of V^E of the sulfolane, +*n*-butanol and sulfolane, +*n*-pentanol mixtures are being positive at lower and higher mole fractions of the alkanols (x_2). The magnitudes of the V^E values of the mixtures are in the order sulfolane + methanol > sulfolane + *n*-propanol > sulfolane + *n*-butanol > sulfolane + *n*-pentanol. The observed values of V^E for the mixtures have been explained in terms of (i) effects due to the differences in chain length of the alcohols, (ii) dipole–dipole interactions between the polar molecules, and (iii) geometric effect due to the differences in molar volume of the component molecules. These are more noticeable in the case of lower alcohols. All these properties have been expressed satisfactorily by appropriate polynomials.

Keywords: Excess molar volumes; Sulfolane; Methanol; *n*-Propanol; *n*-Butanol; *n*-Pentanol

1. Introduction

Sulfolane is an important industrial solvent having several advantageous physicochemical properties and the ability to extract monocyclic aromatic hydrocarbons from petroleum products. Interactions between sulfolane and alcohols may be complex. Alcohols are self-associated liquids through H-bonding. Alcohols possess hydrophilic OH group as well as hydrophobic group. Sulfolane is a globular molecule in which

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Physics and Chemistry of Liquids
ISSN 0031-9104 print/ISSN 1029-0451 online © 2007 Taylor & Francis
DOI: 10.1080/00319100601088713

the negative end of its large dipole moment is exposed and cannot act as proton acceptor/donor [13]. Therefore, the mode of interactions of alcohols and sulfolane is of vital importance in the field of solution chemistry as it can provide us with important information regarding hydrophilic and hydrophobic interactions.

It is necessary to have complete thermodynamic data for these mixtures. Research on their thermophysical properties has been reported by several authors [1–8]. Jannelli *et al.* [9,10] have studied the viscosity, dielectric constant, and solid liquid phase diagram of sulfolane–benzene binary mixtures. Several other researchers [11–17] have studied the vapor liquid and liquid–liquid equilibrium for binary and ternary mixtures containing sulfolane and aromatic hydrocarbons. Neverthless, to our knowledge no density and excess molar volume data on these mixtures were previously reported.

Here we report density and excess molar volumes of four binary mixtures, viz, sulfolane, +methanol, +*n*-propanol, +*n*-butanol, and +*n*-pentanol at 298.15–318.15 K except methanol at 303.15–323.15 K. The knowledge of interactions of simple smaller hydrophobic molecules systems may be useful sometimes to interpret many complex systems. The data are also useful for the design of mixing, storage, and process equipment.

2. Experimental

Materials: The chemicals used were purchased from Aldrich chemical Co. with the quoted purities: methanol (99.5%), *n*-propanol (99.5%), *n*-butanol (99.5%), *n*-pentanol (99.0%), and sulfolane (99%). These chemicals were used without any further purification.

2.1. *Density measurements*

The densities of the pure liquid and liquid–liquid mixtures were measured using a high precision vibrating tube digital density meter (model DMA 5000, Anton Paar, Austira). The temperature of the sample tube is controlled by two integrated inbuilt P + 100 platinum thermometers to a level of highest accuracy and traceable to national standards. The temperature of the sample tube is controlled to ±0.001 K.

The design of the cell ensures identical volumes to be used for the measurement on different samples. Using a polyethylene syringe the sample was continuously and slowly injected from the upper port of the U-tube until the excess fluid flowed out of the lower part. This ensured that the inner surface of the cell was completely wet and there were no micro bubbles inside the U-tube. The syringe was kept plugged in. After the measurement, the sample was removed and air was passed, by built-in pump, through the tube to remove excess liquid. The tube was then rinsed several times with the solution of higher concentration, and finally the solution was injected for the measurement. All the measurements were made, starting from the lowest to the highest solute concentrations. The density meter was checked by measuring the density of standard water sample supplied with the density meter.

2.2. *Determination of excess molar volume*

The excess molar volumes, $V_{\mathrm{m}}^{\mathrm{E}}$ were calculated using the following equation:

$$\frac{V^{\mathrm{E}}}{\mathrm{m}^3}\ \mathrm{mol}^{-1} = \frac{(X_1M_1 + X_2M_2)}{\rho_{\mathrm{mix}}} - \left(\frac{X_1M_1}{\rho_1} + \frac{X_2M_2}{\rho_2}\right) \tag{1}$$

where X_1, M_1, and ρ_1 are the mole fraction, molar mass and density of component 1 (solvent), X_2, M_2, and ρ_2 are the corresponding values of component 2 (organic solutes), and ρ_{mix} is the density of the mixture, respectively. The excess molar volumes were fitted to a Redlich–Kister polynomial equation of the form,

$$\frac{V^{\mathrm{E}}}{\mathrm{m}^3}\ \mathrm{mol}^{-1} = X_1X_2\sum_{i=0}^{n} a_i(1-2X_1)^i \tag{2}$$

where, a_i is the ith fitting coefficient. Using $n = 3$, four a_i coefficients and the standard deviation σ were obtained through the least squares method.

3. Results and discussion

Densities of sulfolane, +methanol, +*n*-propanol, +*n*-butanol, and +*n*-pentanol were measured at temperatures at 298.15, 303.15, 308.15, 313.15, and 318.15 K, respectively, covering the whole composition range except methanol at 303.15–323.15 K. The densities of the pure components are shown in table 1 together with the literature values for all temperature range, wherever possible for comparison. The agreement between the measured values and literature values has been found to be satisfactory. The densities of the binary systems have been shown in tables 2–5 at different temperatures. It shows continuous decrease in density on addition of methanol, *n*-propanol, *n*-butanol, and *n*-pentanol in sulfolane. In pure state the density of alcohol has been found to be in the order of,

n-pentanol > *n*-butanol > *n*-propanol > methanol

The corresponding V^{E} data are also presented in tables 2–5. For V^{E} the fitting coefficients (a_i) are shown in table 6 along with standard deviations. The V^{E} for the systems of methanol, *n*-propanol, *n*-butanol, and *n*-pentanol in sulfolane systems have been plotted in figures 1–4, respectively. The lines are generated by the polynomial equation (2). The V^{E} values for the sulfolane + methanol and *n*-propanol mixtures are negative over the whole range of mole fractions, and for the sulfolane + *n*-butanol and sulfolane + *n*-pentanol mixtures are being positive at lower and higher mole fractions (x_2). The magnitudes of V^{E} values are in the order, sulfolane + methanol > sulfolane + *n*-propanol > sulfolane + *n*-butanol > sulfolane + *n*-pentanol. The V^{E} *versus* x_2 plots are symmetrical minima around 0.5 mole fractions for *n*-butanol and *n*-pentanol sulfolane systems. The observed V^{E} values of the mixtures under investigation can be explained in terms of the following contributions:

(a) Effect due to differences in the chain length of the alkanols,
(b) Dipole–dipole interaction between the unlike polar molecules,
(c) Geometric effect due to differences in molar volumes of the component molecules.

Table 1. Comparison of experimental and literature values of density, $\rho \times 10^3$ (kg m^{-3}) of pure components at different temperatures.

Component	Temperature (K)	Density (kg m^{-3})	
		ρ_{lit}	ρ_{exp}
Sulfolane	298.15	1.2640 [24] 1.26705 [25] 1.266654 [26]	1.265815
	303.15	1.26140 [22] 1.2614 [23]	1.261413
	308.15		1.257009
	313.15	1.25190 [27] 1.2532 [28]	1.252609
	318.15		1.24821
Methanol	298.15	0.7869 [35]	
	303.15	0.781266 [29]	0.782364
	308.15	0.778325 [29]	0.777618
	313.15	0.773944 [29] 0.7733 [36]	0.772847
	318.15	0.769285 [29]	0.768043
	323.15	0.763559 [29]	0.763201
n-Propanol	298.15	0.79941 [30] 0.7995 [31] 0.7996 [32]	0.801379
	303.15	0.801745 [29]	0.79732
	308.15	0.797499 [29]	0.793227
	313.15	0.793515 [29] 0.787598 [20] 0.7875 [21]	0.789093
	318.15	0.784714 [29]	0.784907
n-Butanol	298.15	0.80606 [30] 0.8059 [32] 0.80573 [33]	0.807366
	303.15	0.80203 [30] 0.8018 [31] 0.8119 [32] 0.80559 [33]	0.803495
	308.15		0.799589
	313.15	0.79329 [30] 0.7941 [32] 0.745 [31] 0.79432 [33]	0.795646
	318.15	0.8018 [31]	0.791662
n-Pentanol	298.15	0.81073 [30] 0.8101 [31]	0.811394
	303.15	0.80677 [30] 0.8070 [31] 0.80711 [34]	0.807701
	308.15		0.803977
	313.15	0.78805 [30] 0.7995 [31] 0.79978 [34] 0.79266 [33]	0.800219
	318.15		0.796422

The excess molar volumes of the binary mixtures under investigations may be considered to be the resultant of the above-mentioned competing interactions of the component molecules. All the components are polar compounds; the value of dipole moment (μ) being 4.8, 1.7, 1.69, 1.66, 1.68, and 1.64 D for sulfolane, methanol,

Table 2. Densities, $\rho \times 10^3$ (kg m^{-3}), excess molar volumes, $V^E \times 10^6$ (m^3 mol^{-1}), of methanol–sulfolane systems (303.15, 308.15, 313.15, 318.15, and 323.15) K.

T (K)	303.15		308.15		313.15		318.15		323.15	
X_2	ρ	V^E	ρ	V^E	ρ	V^E	ρ	V^E	ρ	V^E
0	1.261413	0	1.257009	0	1.252609	0	1.24821	0	1.243804	0
0.1029	1.240925	−0.1462	1.236484	−0.1498	1.232018	−0.1515	1.227575	−0.1551	1.223156	−0.1613
0.2018	1.220248	−0.4004	1.215731	−0.4055	1.211348	−0.4201	1.206703	−0.417	1.202381	−0.4376
0.3039	1.195546	−0.6477	1.190945	−0.6542	1.186214	−0.6525	1.181794	−0.6722	1.17711	−0.6756
0.4052	1.165494	−0.7915	1.160835	−0.8005	1.156209	−0.8122	1.151575	−0.8244	1.146957	−0.8392
0.5018	1.131074	−0.8664	1.126389	−0.8788	1.121723	−0.8933	1.117034	−0.9076	1.112339	−0.9234
0.6045	1.087014	−0.8932	1.082321	−0.9095	1.077615	−0.9261	1.072876	−0.9424	1.068114	−0.9593
0.7026	1.036222	−0.8919	1.031338	−0.9006	1.026609	−0.919	1.021816	−0.9356	1.017007	−0.9534
0.8034	0.971316	−0.8066	0.966782	-0.8347	0.961994	−0.8509	0.957182	−0.8677	0.952338	−0.8851
0.9018	0.890267	−0.5761	0.885506	−0.5885	0.880693	−0.5997	0.875859	−0.6117	0.871	−0.6247
1	0.782364	0	0.777618	0	0.772847	0	0.768043	0	0.763201	0

Table 3. Densities, $\rho \times 10^3$ (kg m^{-3}), excess molar volumes, $V^E \times 10^6$ (m^3 mol^{-1}), of *n*-propanol–sulfolane systems (298.15, 303.15, 308.15, 313.15, and 318.15) K.

T (K)	298.15		303.15		308.15		313.15		318.15	
X_2	ρ	V^E	ρ	V^E	ρ	V^E	ρ	V^E	ρ	V^E
0	1.265815	0	1.261413	0	1.257009	0	1.252609	0	1.24821	0
0.1032	1.228583	−0.1022	1.224138	−0.1016	1.219693	−0.1013	1.215252	−0.1044	1.210809	−0.1074
0.2025	1.192696	−0.3092	1.188245	−0.3131	1.183779	−0.3178	1.179314	−0.3203	1.174842	−0.3264
0.3021	1.152917	−0.5106	1.148434	−0.5217	1.143945	−0.5326	1.139451	−0.5363	1.13495	−0.5461
0.403	1.113787	−0.6529	1.109427	−0.6708	1.104948	−0.6867	1.100436	−0.6942	1.095896	−0.7076
0.5025	1.068878	−0.7176	1.064678	−0.7394	1.060336	−0.7573	1.055893	−0.7694	1.0514	−0.7851
0.6085	1.015931	−0.713	1.011524	−0.734	1.007078	−0.7501	1.002607	−0.7649	0.998111	−0.7806
0.7024	0.97262	−0.6575	0.968234	−0.6732	0.963825	−0.685	0.95938	−0.6981	0.954906	−0.7112
0.7999	0.919444	−0.549	0.91512	−0.5562	0.910766	−0.5621	0.90638	−0.5692	0.901958	−0.5773
0.8958	0.865291	−0.3652	0.861101	−0.365	0.856857	−0.3659	0.852582	−0.366	0.848271	−0.3686
1	0.801379	0	0.797321	0	0.793227	0	0.789093	0	0.784907	0

Table 4. Densities, $\rho \times 10^3$ (kg m^{-3}), excess molar volumes, $V^E \times 10^6$ (m^3 mol^{-1}), of butanol–sulfolane systems (298.15, 303.15, 308.15, 313.15, and 318.15) K.

T (K)	298.15		303.15		308.15		313.15		318.15	
X_2	ρ	V^E	ρ	V^E	ρ	V^E	ρ	V^E	ρ	V^E
0	1.265815	0	1.261413	0	1.257009	0	1.252609	0	1.24821	0
0.1032	1.220818	0.0781	1.21617	0.0978	1.211586	0.1121	1.20716	0.1140	1.202501	0.1340
0.2025	1.176103	−0.1600	1.171313	−0.1290	1.166395	−0.0982	1.161971	−0.0880	1.157247	−0.0640
0.3021	1.134929	−0.3900	1.130305	−0.3720	1.125632	−0.3510	1.120997	−0.3340	1.116265	−0.3100
0.403	1.112896	−0.467	1.108321	−0.4520	1.103634	−0.4288	1.099056	−0.4060	1.09424	−0.3840
0.5025	1.043936	−0.567	1.039298	−0.5500	1.034952	−0.5314	1.030574	−0.5120	1.025881	−0.4960
0.6085	0.997973	−0.4861	0.993549	−0.4620	0.988895	−0.4386	0.984649	−0.4160	0.980086	−0.3960
0.7024	0.949637	−0.2949	0.945124	−0.2629	0.940432	−0.2354	0.936208	−0.2170	0.93169	−0.19230
0.7999	0.90991	−0.1428	0.905455	−0.1070	0.900824	−0.0855	0.896658	−0.0550	0.892214	−0.0290
0.8958	0.854373	0.0124	0.850224	0.0310	0.845852	0.0509	0.841814	0.0702	0.837626	0.0820
1	0.807366	0	0.803495	0	0.799589	0	0.795646	0	0.791662	0

Table 5. Densities, $\rho \times 10^3$ (kg m^{-3}), excess molar volumes, $V^E \times 10^6$ (m^3 mol^{-1}), of pentanol–sulfolane systems (298.15, 303.15, 308.15, 313.15, and 318.15) K.

T (K)	298.15		303.15		308.15		313.15		318.15	
X_2	ρ	V^E	ρ	V^E	ρ	V^E	ρ	V^E	ρ	V^E
0	1.265815	0	1.261413	0	1.257009	0	1.252609	0	1.24821	0
0.0989	1.214124	0.0781	1.209513	0.0978	1.204966	0.1121	1.200571	0.124	1.19595	0.134
0.206	1.163689	−0.16	1.158982	−0.129	1.154153	−0.0882	1.149796	−0.068	1.145151	−0.054
0.3028	1.119353	−0.39	1.114841	−0.372	1.110284	−0.351	1.10576	−0.334	1.101145	−0.31
0.355	1.095332	−0.467	1.090884	−0.452	1.086332	−0.4288	1.081879	−0.416	1.077204	−0.394
0.509	1.024742	−0.547	1.020288	−0.53	1.016098	−0.5214	1.011878	−0.512	1.00737	−0.496
0.6029	0.981735	−0.4661	0.9775	−0.452	0.973061	−0.4296	0.968982	−0.416	0.964618	−0.396
0.7033	0.936434	−0.2949	0.932146	−0.2629	0.9277	−0.2354	0.923662	−0.217	0.919363	−0.1923
0.7851	0.900324	−0.1428	0.896105	−0.107	0.891731	−0.0755	0.887758	−0.055	0.883541	−0.029
0.909	0.847851	0.0124	0.843921	0.031	0.839797	0.0509	0.835959	0.0702	0.831988	0.082
1	0.811394	0	0.807701	0	0.803977	0	0.800219	0	0.796422	0

Table 6. Coefficient, a_i, of Redlich–Kister equation (2), expressing V^E and standard deviation, σ for the sulfolane, +methanol, +n-propanol, +n-butanol, +n-pentanol systems.

Systems	T (K)	a_o	a_1	a_2	a_3	σ
Sulfolane–methanol systems	303.15	−3.4744	−0.9820	−0.8663	−3.2772	0.0443
	308.15	−3.5175	−1.0284	−0.9641	−3.3527	0.0048
	313.153	−3.5690	−1.1126	−1.0220	−3.2760	0.0042
	318.15	−3.6322	−1.1224	−1.0326	−3.4057	0.0048
	323.15	−3.6887	−1.1704	−1.1364	−3.3573	0.0043
Sulfolane–propanol systems	298.15	−2.8675	−0.6138	0.5648	−1.8411	0.0000
	303.15	−2.9545	−0.6531	0.7105	−1.7819	0.0455
	308.15	−3.0259	−0.6587	0.8190	−1.7859	0.0442
	313.153	−3.0741	−0.7421	0.8689	−1.6205	0.0413
	318.15	−3.1368	−0.7710	0.9208	−1.5705	0.0372
Sulfolane–butanol systems	298.15	−2.2970	0.7045	4.2162	−1.7463	0.0107
	303.15	−2.2264	0.8063	4.5226	−1.9226	0.0090
	308.15	−2.1434	0.8025	4.7245	−1.9017	0.0088
	313.153	−2.0591	0.7917	4.8038	−1.6545	0.0094
	318.15	−1.9833	0.8115	5.0074	−1.7402	0.0105
Sulfolane–pentanol systems	298.15	−2.2444	0.7703	4.0624	−1.8901	0.0107
	303.15	−2.1856	0.8411	4.3993	−1.9988	0.0097
	308.15	−2.1199	0.8376	4.7285	−1.9784	0.0086
	313.153	−2.0768	0.8305	4.9570	−1.9000	0.0082
	318.15	−1.9942	0.8374	5.0667	−1.8402	0.0081

n-propanol, n-butanol, and n-pentanol, respectively. Due to dipole–dipole interactions between the components negative V^E values are generally expected. Sulfolane + n-butanol and sulfolane + n-pentanol mixtures are being positive at lower and higher mole fractions. Unfavorable packing may, however, result due to disruption of the closely associated sulfolane multimers on addition of alkanols and formation of new association between the unlike sulfolane and alkanol molecules. Reorganization of the pure components in the mixtures due to formation of different type H-bonding may also result unfavorable packing and lead to expansion in volume.

As far the magnitude of V^E, it has been observed that the value of V^E increases with increasing chain lengths [18] and size [19] of the components. This is not in agreement

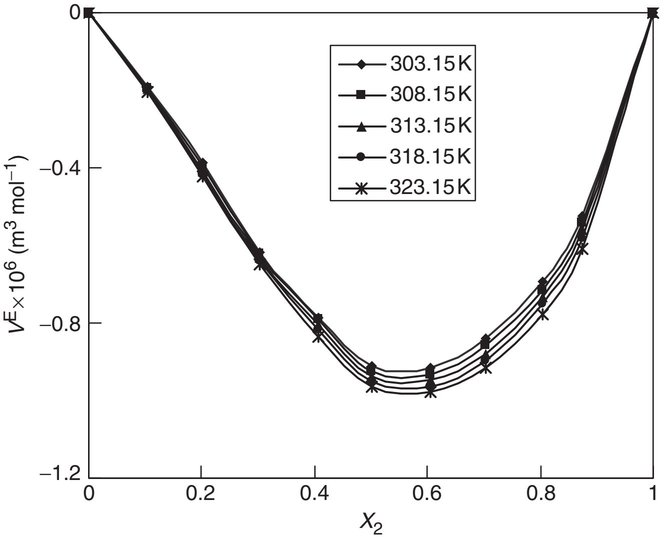

Figure 1. Plots of excess molar volume (V^E) *vs.* X_2 for methanol at ◆, 298.15 K; ■, 303.15 K; ▲, 308.15 K; ●, 313.15 K; ⁎, 318.15 K.

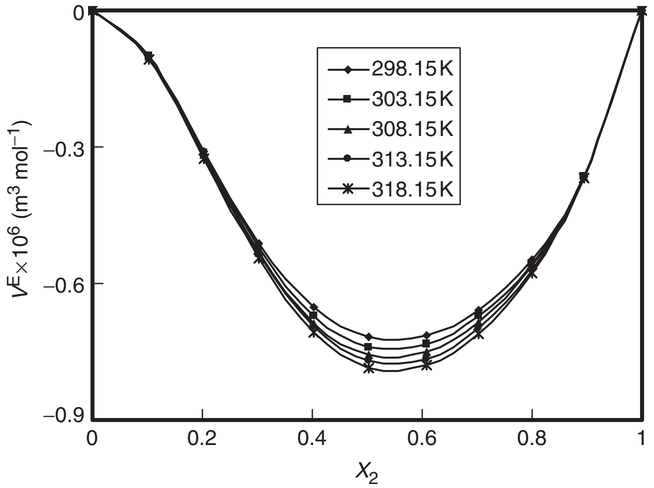

Figure 2. Plots of excess molar volume (V^E) *vs.* X_2 for *n*-propanol at ◆, 298.15 K; ■, 303.15 K; ▲, 308.15 K; ●, 313.15 K; ⁎, 318.15 K.

with the magnitude of our V^E values which are in the order, sulfolane + methanol > sulfolane + *n*-propanol > sulfolane + *n*-butanol > sulfolane + *n*-pentanol mixtures. The molar volumes of sulfolane, methanol, *n*-propanol, *n*-butanol, and *n*-pentanol at 298.15 K are, respectively, 95.04, 40.70, 74.96, 91.86, and 108.65 cm³ mole⁻¹. The methanol and propanol molecules being smaller, there is possibility of partial accommodation of methanol and propanol molecules in the

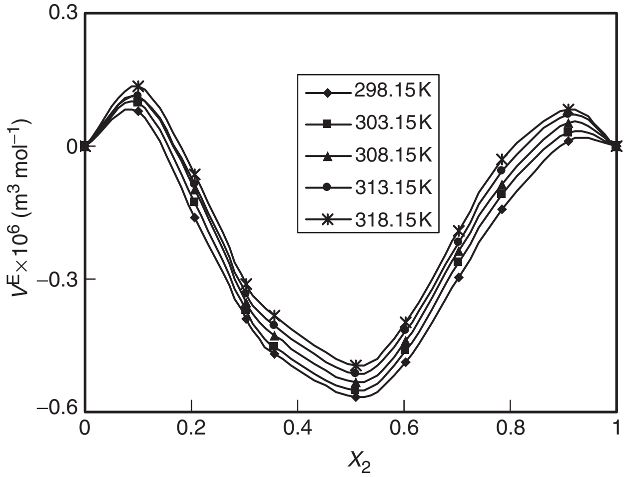

Figure 3. Plots of excess molar volume (V_m^E) *vs.* X_2 for *n*-butanol at ◆, 298.15 K; ■, 303.15 K; ▲, 308.15 K; ●, 313.15 K; ⁎, 318.15 K.

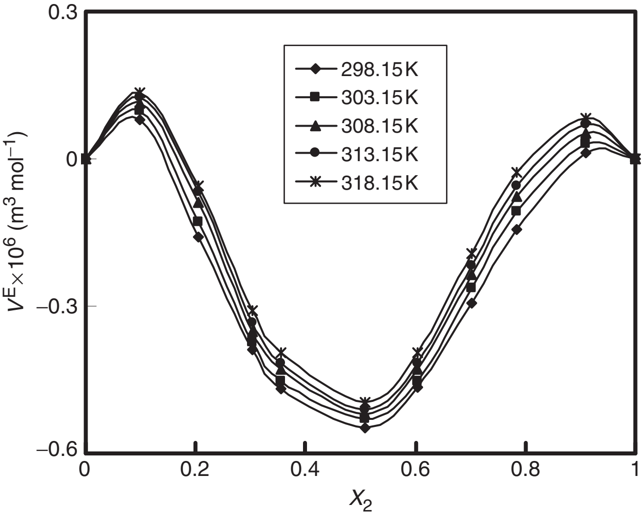

Figure 4. Plots of excess molar volume (V_m^E) *vs.* X_2 for *n*-pentanol at ◆, 298.15 K; ■, 303.15 K; ▲, 308.15 K; ●, 313.15 K; ⁎, 318.15 K.

interstices of the sulfolane molecules. Large and negative V^E value of sulfolane + methanol and *n*-propanol mixtures may be attributed to the occupation of void spaces of one component by an other.

Examination of the results shows that the temperature effects on V^E are not very significant. However, for the systems (i.e. *n*-butanol and *n*-pentanol) V^E values are being positive at lower and higher mole fractions, V^E increases with temperature and for those with negative values (i.e. methanol and *n*-propanol), V^E decreases with temperature.

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