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Thermodynamic Properties of the Binary Mixtures of 1,2-Dichloroethane with Chlorobenzene and Bromobenzene from (298.15 to 313.15) K[†]

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Densities, viscosities, and refractive indices for the binary mixtures of 1,2-dichloroethane with chlorobenzene and bromobenzene have been determined from (298.15 to 313.15) K over the entire range of concentration at atmospheric pressure. Excess molar volumes have been calculated from the experimental data and fitted to a Redlich−Kister polynomial function. The ability of the Prigogine−Flory−Patterson (PFP) theory to predict V_m^E at 303.15 K has been tested.

Introduction

The thermodynamic and transport properties of liquids and liquid mixtures[1] are used to study the molecular interactions between the various components of the mixture and also to understand engineering applications concerning heat transfer, mass transfer, and fluid flow. Properties such as density, viscosity, and refractive index of binary liquid mixtures over the whole concentration range are useful for a theoretical and practical understanding of their thermodynamic and transport properties.

Following our systematic investigation of thermodynamic properties of binary mixtures containing chloroalkanes[2,3] as one of the components, we report in this work density, viscosity, and refractive index for binary mixtures of 1, 2-dichloroethane with chlorobenzene and bromobenzene over the whole concentration range at T = (298.15, 303.15, 308.15, and 313.15) K and atmospheric pressure. From these experimental results, excess molar volume was calculated.

Excess molar volumes of 1,2-dichloroethane with chlorobenzene and bromobenzene have been studied by Reddy et al.[4] by a dilatometric method at (303.15 and 313.15) K. However, the authors did not report any density or other thermodynamic properties.

Experimental Section

Materials. All compounds used in this work were supplied by Shanghai Chemicals Factory and were used as received, without any further purification. The stated density, viscosity, and refractive index of the pure components at (298.15, 303.15, 308.15, and 313.15) K are reported in Table 1 and compared with available literature data. Gas chromatographic studies showed no evidence of appreciable impurities in reagents.

Apparatus and Procedure. Binary mixtures were prepared by mass in airtight glass bottles. The mass measurements were performed on a Dhona 100 DS (India) single pan analytical balance, with a precision of $\pm 1 \cdot 10^{-4}$ g. The required properties were measured on the same day of the mixture. The possible error in mole fraction was estimated to be less than $\pm 1 \cdot 10^{-4}$.

Densities, ρ, of the pure compounds and their mixtures were determined with an Anton Paar DMA4500 vibrating tube densimeter automatically thermostatted within ± 0.01 K. The densimeter was calibrated with doubly distilled, degassed water and with dry air at atmospheric pressure. After each series of measurements, the densimeter should be washed by two kinds of lotions and dried. The estimated uncertainty is $2 \cdot 10^{-5}$ g·cm^{-3}.

An Ubbelohde viscometer with a capacity of about 15 mL and the capillary tube with a length of about 90 mm and 0.5 mm internal diameter, calibrated by using conductivity water (conductivity less than $1 \cdot 10^{-6}$) using (0.8902, 0.7972, 0.7190, and 0.6526) mPa·s as its viscosities at (298.15, 303.15, 308.15, and 313.15) K (Riddick et al., 1986[5]), respectively, is used to measure the flow times of pure liquids and liquid mixtures.[6] The viscometer was thoroughly cleaned and perfectly dried, and care was taken to reduce evaporation during the measurements. The viscometer is kept vertically in a transparent walled bath with a thermal stability of ± 0.01 K for about 30 min to attain the thermal equilibrium. The efflux time between the marks on either side of the bulb is measured with a stop watch of ± 0.01 s precision. The average of four or five sets of flow times for each fluid was taken for the purpose of the calculation of viscosity. Since all flow times were greater than 300 s and capillary radius (0.5 mm) was far less than its length (90 mm), the kinetic energy and end corrections, respectively, were found to be negligible. The uncertainty of viscosity values is $\pm 3 \cdot 10^{-3}$ mPa·s. The performance of the viscometer was checked by measuring and comparing the viscosities of the pure liquids with the value reported in Table 1.

Refractive indices of the mixtures at the sodium D-line were determined with an Abbe refractometer equipped with a circulating water bath, permitting us to maintain the sample at constant temperature to within ± 0.01 K. The instrument was calibrated by measuring the refractive index of deionized water. The sample support was rinsed with acetone and dried with a paper towel. The estimated uncertainty is $2 \cdot 10^{-4}$.

Results and Discussion

The experimental values of the density, viscosity, and refractive index for binary mixtures at different temperatures and at atmospheric pressure are listed in Table 2. Excess molar volumes were calculated from our measurements according to the following equation

$$V_\mathrm{m}^\mathrm{E} = x_1M_1(1/\rho - 1/\rho_1) + x_2M_2(1/\rho - 1/\rho_2) \qquad (1)$$

where x_1 and x_2 are the mole fractions of components 1 and 2; ρ_1 and ρ_2 are the densities of pure components 1 and 2; ρ is the density of

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Table 1. Comparison of Measured Values of Density, ρ, Viscosity, η, and Refractive Index, n_D, of the Pure Substances with Those in the Literature at 298.15 K

		ρ/g·cm^{-3}		η/mPa·s		n_D	
component	T/K	exptl	lit.	exptl	lit.	exptl	lit.
dichloroethane	298.15	1.24670	1.24637[5]	0.7902	0.7890[15]	1.4416	1.44135[15]
	303.15	1.24080		0.7291		1.4383	1.43866[15]
	308.15	1.23049		0.6739		1.4336	1.43599[15]
	313.15	1.22560		0.6441		1.4311	1.43341[15]
chlorobenzene	298.15	1.10040	1.1010[14]	0.7568	0.758[16]	1.5222	1.5223[14]
	303.15	1.09490	1.095[14]	0.7258	0.725[14]	1.5192	1.5195[14]
	308.15	1.08938	1.090[14]	0.6770	0.683[14]	1.5163	1.5166[14]
	313.15	1.08390	1.09394[16]	0.6240	0.6469[16]	1.5115	
bromobenzene	298.15	1.48940	1.4890[14]	1.0820	1.084[17]	1.5578	1.5577[14]
	303.15	1.48110	1.481[14]	1.0146	1.013[14]	1.5538	1.5542[14]
	308.15	1.47580	1.473[14]	0.9540	0.953[14]	1.5514	1.5514[14]
	313.15	1.46734		0.8780		1.5468	

the mixture; and M_1 and M_2 are the molecular weights of components 1 and 2.

The values of V_m^E of the studied binary mixtures, at all measured temperatures, are given in Table 2.

The excess molar volumes were fitted by means of a Redlich−Kister type equation[7] of the form

$$Y^E = x_1(1 - x_1)\sum_{j=0}^{j=n} a_j(1 - 2x_1)^j \tag{2}$$

where Y^E represents either V_m^E, and x_1 is the mole fraction of 1,2-dichloroethane. Numerical values of the coefficients a_j, obtained from a least-squares analysis of the data, with standard deviation, σ, are given in Table 3. The number of coefficients used for each property was determined as the minimum number needed to adequately represent the data. The standard deviation, σ, between the experimental and calculated values was defined as

$$\sigma = [\sum(Y_{\text{exptl}}^E - Y_{\text{calcd}}^E)^2/(N - p)]^{1/2} \tag{3}$$

where N and p are the numbers of experimental points and parameters, respectively.

Figures 1 and 2 show the excess molar volumes, plotted against the mole fraction of 1,2-dichloroethane for the binary systems at several temperatures, respectively. These plots show that the excess molar volumes are always positive over the whole studied concentration and temperature ranges.

As can be seen in Figures 1 and 2, the V_m^E values for binary mixtures of chlorobenzene and bromobenzene with 1,2-dichloroethane are positive over the whole composition range and become more positive with an increase of temperature. The values of V_m^E are also in agreement with the conclusion reported by Reddy et al.[4] by a dilatometric method at (303.15 and 313.15) K. The positive V_m^E values indicate that there is a volume expansion on mixing. The positive V_m^E values can be attributed to the predominance of the dispersion forces and dipolar dissociation over the specific interactions in the binary mixtures.

Prigogine−Flory−Patterson (PFP) Theory

In recent years, PFP[8−12] theory has been used extensively to predict excess thermodynamic functions of nonelectrolyte systems for different kinds of mixtures. In this paper, we have applied PFP theory for prediction of the V_m^E values of binary liquid mixtures (1,2-dichloroethane + chlorobenzene and bromobenzene) at $T = 303.15$ K.

Quantitative estimation of different contributions to the V_m^E values can be obtained by using the well-known Prigogine−Flory−Patterson theory.[8−12] According to the PFP theory, V_m^E calculations include three contributions: (i) interactional, which is proportional to the χ_{12} parameters; (ii) the free volume contribution, which arises from the dependence of the reduced volume upon the reduced temperature as a result of the difference between the degree of expansion of the two components; and (iii) the P^* contribution, which depends both on the differences of internal pressures and on the differences of reduced volumes of the components.

V_m^E was correlated by means of the PFP theory using the following equation:

$$\begin{aligned}\frac{V_m^E}{x_1V_1^* + x_2V_2^*} &= \frac{(\tilde{V}^{1/3} - 1)\tilde{V}^{2/3}\psi_1\theta_2(\chi_{12}/P_1^*)}{((4/3)\tilde{V}^{-1/3} - 1)}\ (\chi_{12}\text{ contribution})\\ &- \frac{(\tilde{V}_1 - \tilde{V}_2)^2((14/9)\tilde{V}^{-1/3} - 1)\psi_1\psi_2}{((4/3)\tilde{V}^{-1/3} - 1)\tilde{V}}\ (\tilde{V}\text{ contribution})\\ &+ \frac{(\tilde{V}_1 - \tilde{V}_2)(P_1^* - P_2^*)\psi_1\psi_2}{P_1^*\psi_2 + P_2^*\psi_1}\ (P^*\text{ contribution})\end{aligned} \tag{4}$$

The $\tilde{V}$ of the solution is approximated in eq 7 by

$$\tilde{V} = \psi_1\tilde{V}_1 + \psi_2\tilde{V}_2 \tag{5}$$

Here, the molecular contact energy fraction is given by

$$\psi_1 = 1 - \psi_2 = \phi_1P_1^*/(\phi_1P_1^* + \phi_2P_2^*) \tag{6}$$

with the hard-core volume fractions defined by

$$\phi_1 = 1 - \phi_2 = x_1V_1^*/(x_1V_1^* + x_2V_2^*) \tag{7}$$

The thermal expansion coefficient α_i is used to compute the reduced volume by the equation

$$\tilde{V}_i = \left(\frac{1 + (4/3)\alpha_iT}{1 + \alpha_iT}\right)^3 \tag{8}$$

The characteristic volume is $V_i^* = V_i/\tilde{V}_i$, and the characteristic pressure is given by

Table 2. Densities, Viscosities, Refractive Indices, and Excess Molar Volumes for Binary Mixtures at the Temperature (298.15, 303.15, 308.15, and 313.15) K and Atmospheric Pressure

x_1	ρ / $\mathrm{g \cdot cm^{-3}}$	η / $\mathrm{mPa \cdot s}$	n_D	V_m^E / $\mathrm{cm^3 \cdot mol^{-1}}$
$x_1$1,2-dichloroethane + (1 − x_1)chlorobenzene				
T = 298.15 K				
0.1022	1.11156	0.7456	1.5149	0.0643
0.2015	1.12305	0.7387	1.5080	0.1147
0.3013	1.13529	0.7351	1.5012	0.1514
0.4039	1.14864	0.7364	1.4935	0.1775
0.4999	1.16191	0.7372	1.4860	0.1883
0.5977	1.17636	0.7410	1.4781	0.1811
0.6986	1.19224	0.7445	1.4696	0.1597
0.8000	1.20930	0.7536	1.4607	0.1230
0.8990	1.22712	0.7659	1.4516	0.0722
T = 303.15 K				
0.1022	1.10604	0.7146	1.5120	0.0633
0.2015	1.11748	0.7049	1.5052	0.1150
0.3013	1.12968	0.6991	1.4983	0.1528
0.4039	1.14296	0.6969	1.4908	0.1810
0.4999	1.15620	0.6963	1.4832	0.1913
0.5977	1.17062	0.6973	1.4752	0.1831
0.6986	1.18646	0.6975	1.4666	0.1614
0.8000	1.20348	0.7009	1.4576	0.1238
0.8990	1.22128	0.7100	1.4484	0.0712
T = 308.15 K				
0.1022	1.10015	0.6659	1.5090	0.0659
0.2015	1.11122	0.6595	1.5022	0.1177
0.3013	1.12302	0.6542	1.4951	0.1565
0.4039	1.13591	0.6505	1.4873	0.1827
0.4999	1.14868	0.6506	1.4797	0.1955
0.5977	1.16262	0.6500	1.4715	0.1885
0.6986	1.17792	0.6493	1.4626	0.1670
0.8000	1.19440	0.6530	1.4534	0.1276
0.8990	1.21162	0.6574	1.4440	0.0729
T = 313.15 K				
0.1022	1.09468	0.6190	1.5049	0.0685
0.2015	1.10579	0.6151	1.4983	0.1214
0.3013	1.11762	0.6143	1.4915	0.1619
0.4039	1.13048	0.6119	1.4838	0.1941
0.4999	1.14337	0.6125	1.4763	0.2016
0.5977	1.15736	0.6158	1.4682	0.1948
0.6986	1.17273	0.6191	1.4595	0.1727
0.8000	1.18928	0.6241	1.4503	0.1328
0.8990	1.20659	0.6314	1.4410	0.0767
$x_1$1,2-dichloroethane + (1 − x_1)bromobenzene				
T = 298.15 K				
0.1014	1.46957	1.0410	1.5479	0.0560
0.2002	1.44941	1.0036	1.5398	0.1023
0.3004	1.42809	0.9677	1.5313	0.1368
0.3999	1.40601	0.9348	1.5218	0.1549
0.5008	1.38250	0.9036	1.5107	0.1642
0.6001	1.35818	0.8752	1.4984	0.1630
0.6999	1.33244	0.8496	1.4847	0.1512
0.7995	1.30543	0.8268	1.4704	0.1249
0.8998	1.27672	0.8058	1.4559	0.0808
T = 303.15 K				
0.1014	1.46155	0.9749	1.5439	0.0530
0.2002	1.44165	0.9388	1.5355	0.0957
0.3004	1.42051	0.9040	1.5266	0.1329
0.3999	1.39859	0.8714	1.5170	0.1546
0.5008	1.37526	0.8409	1.5060	0.1675
0.6001	1.35121	0.8141	1.4940	0.1641
0.6999	1.32577	0.7886	1.4808	0.1485
0.7995	1.29912	0.7646	1.4671	0.1154
0.8998	1.27073	0.7445	1.4528	0.0677
T = 308.15 K				
0.1014	1.45577	0.9162	1.5412	0.0534
0.2002	1.43538	0.8809	1.5321	0.0980
0.3004	1.41375	0.8474	1.5227	0.1352
0.3999	1.39137	0.8150	1.5128	0.1557
0.5008	1.36754	0.7839	1.5015	0.1684
0.6001	1.34297	0.7583	1.4893	0.1656
0.6999	1.31702	0.7321	1.4761	0.1500
0.7995	1.28984	0.7110	1.4622	0.1171
0.8998	1.26096	0.6904	1.4479	0.0678
T = 313.15 K				
0.1014	1.44763	0.8463	1.5372	0.0537
0.2002	1.42753	0.8155	1.5279	0.0994
0.3004	1.40626	0.7870	1.5182	0.1357
0.3999	1.38419	0.7607	1.5079	0.1579
0.5008	1.36071	0.7355	1.4967	0.1703
0.6001	1.33650	0.7120	1.4849	0.1680
0.6999	1.31092	0.6917	1.4722	0.1524
0.7995	1.28414	0.6728	1.4590	0.1183
0.8998	1.25565	0.6569	1.4452	0.0685

Table 3. Parameters of Equation 2 and Standard Deviations, *σ*, for Binary Mixtures at different Temperature

T/K	a_0	a_1	a_2	a_3	σ
$x_1$1,2-dichloroethane + (1−x_1)chlorobenzene					
298.15	0.7440	−0.0397	0.0008	−0.0296	0.0014
303.15	0.7572	−0.0401	−0.0348	−0.0295	0.0013
308.15	0.7741	−0.0631	−0.0235	0.0166	0.0010
313.15	0.8036	−0.0540	−0.0184	−0.0145	0.0019
$x_1$1,2-dichloroethane + (1−x_1)bromobenzene					
298.15	0.6576	−0.0555	0.1515	−0.1875	0.0010
303.15	0.6664	−0.0934	−0.0041	−0.0203	0.0010
308.15	0.6740	−0.0905	−0.0067	−0.0211	0.0011
313.15	0.6827	−0.0954	−0.0108	−0.0017	0.0009

$$P_i^* = T\tilde{V}_i^2 \alpha_i/\kappa_i \tag{9}$$

where κ is the isothermal compressibility.

The molecular surface fraction is given by

$$\theta_2 = \phi_2 S_2/(\phi_1 S_1 + \phi_2 S_2) \tag{10}$$

where S_i, the molecular surface vs volume ratio for the components, is determined usually from approximation of Abe and Flory[13]

$$S_1/S_2 = (V_1^*/V_2^*)^{-1/3} \tag{11}$$

The parameters involved in eq 4 for the pure components are obtained through Flory's theory and are given in Table 4. The interaction parameter, χ_{12}, can be evaluated if values of the excess molar enthalpies are known. Because these values are not available for these binary systems, it is a common practice to use χ_{12} as the only adjustable parameter to fit the experimental V_m^E values to eq 4. In such a way, possible discrepancies between model and experiment are mostly charged on the χ_{12} parameter. The obtained interaction parameters from the fitting of excess molar volume values to eq 4 qualitatively represent the intermolecular interaction between components of mixtures. Table 5 gives the calculated equimolar values of the three contributions to V_m^E, according to eq 4, together with Flory's parameter χ_{12}.

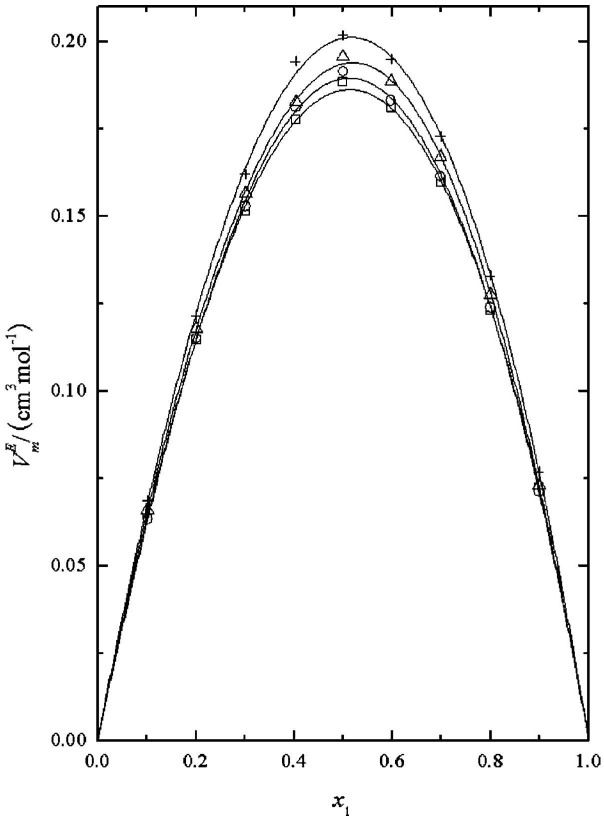

Figure 1. Excess molar volume vs mole fraction for {$x_1$1,2-dichloroethane + (1 − x_1)chlorobenzene} mixtures at □, T = 298.15 K; ○, T = 303.15 K; △, T = 308.15 K; +, T = 313.15 K; the lines are from the Redlich−Kister type equation.

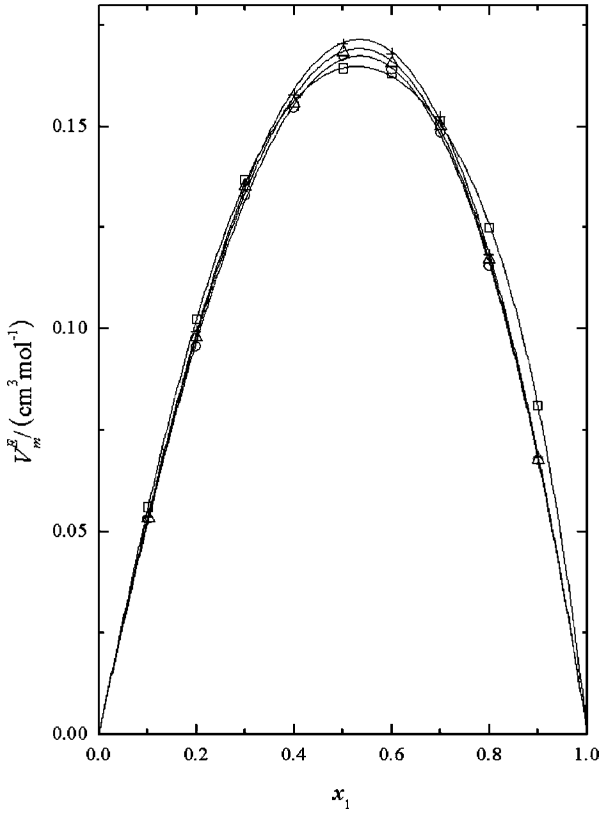

Figure 2. Excess molar volume vs mole fraction for {$x_1$1,2-dichloroethane + (1 − x_1)bromobenzene} mixtures at □, T = 298.15 K; ○, T = 303.15 K; △, T = 308.15 K; +, T = 313.15 K; the lines are from the Redlich−Kister type equation.

Table 4. Parameters of the Pure Components Used in PFP Theory Calculations at T = 298.15 K

	$10^4\alpha$	$10^4\kappa_T$		P^*	V^*
	$\mathrm{K^{-1}}$	$\mathrm{MPa^{-1}}$	$\tilde{V}$	$\mathrm{J{\cdot}cm^{-3}}$	$\mathrm{cm^3{\cdot}mol^{-1}}$
1,2-dichloroethane	11.37	8.17[18]	1.2752	674.2	62.64
chlorobenzene	9.85	7.846[17]	1.248	590.56	82.33
bromobenzene	9.09	6.891[17]	1.231	605.9	86.82

Table 5. Calculated Values of Three Contributions of the PFP Theory to V_m^E for (1,2-Dichloroethane + Chlorobenzene or Bromobenzene) Mixtures at x = 0.5 and T = 298.15 K

	χ_{12}			
	$\mathrm{J{\cdot}cm^{-3}}$	interactional	free volume	P^* effect
$x_1$1,2-dichloroethane + (1 − x_1)chlorobenzene	13.38	0.1460	−0.0208	0.0659
$x_1$1,2-dichloroethane + (1−x_1)bromobenzene	13.04	0.1352	−0.0553	0.0877

An analysis of each of the three theoretical contributions to V_m^E shows that the interactional contribution is always positive for all the investigated systems, and it seems to be the most important to explain the V_m^E behavior for the (1,2-dichloroethane + chlorobenzene or bromobenzene) system. The contribution shows the same sign as Flory's parameter χ_{12}. The free volume effect, which is a measure of geometrical accommodation, is always negative in all mixtures and has the least influence for the studied systems in the three contributions. The third contribution, due to differences in internal pressure and in reduced volume of the components, is always positive in all mixtures.

Figures 3 and 4 indicate the composition dependence of V_m^E values calculated from PFP theory and Redlich−Kister polynomial eq 2 for (1,2-dichloroethane + chlorobenzene or

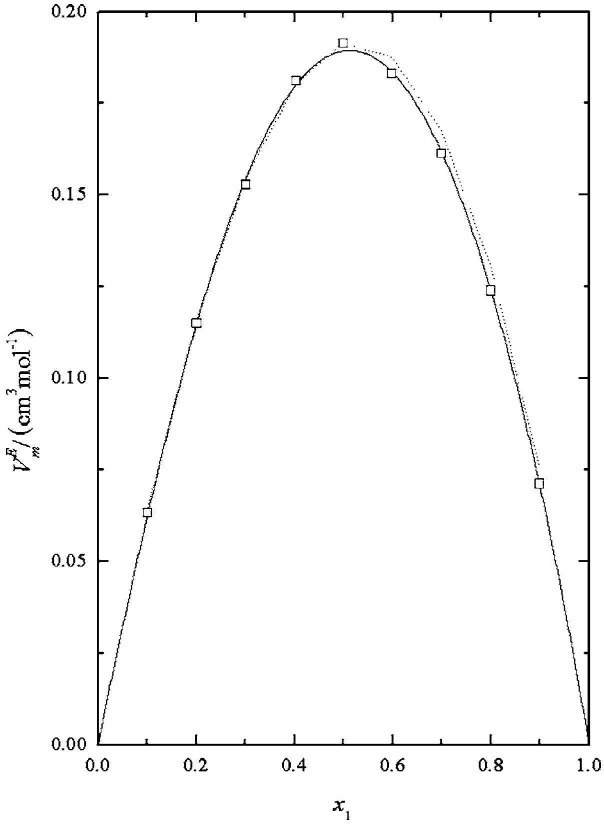

Figure 3. PFP theory predicting excess molar volume for the {$x_1$1,2-dichloroethane + (1 − x_1)chlorobenzene} mixtures at T = 303.15 K. □, experimental; ..., PFP.

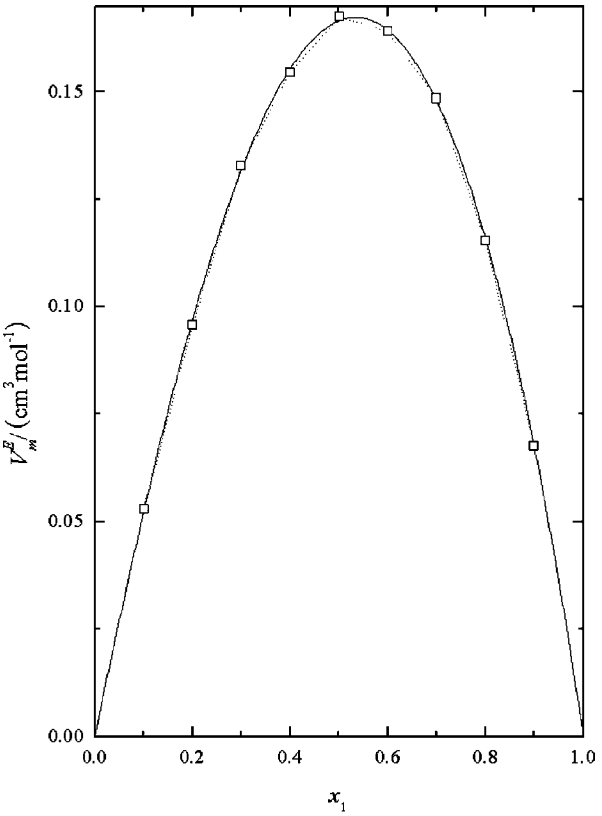

Figure 4. PFP theory predicting excess molar volume for the {$x_1$1,2-dichloroethane + (1 − x_1)bromobenzene} mixtures at T = 303.15 K. □, experimental; ..., PFP.

bromobenzene) mixtures compared with experiments at 303.15 K. From the figures, we conclude that there is a good agreement in general between the experimental V_m^E values and theoretical values. Because χ_{12} is an adjustable parameter, the plots in Figures 3 and 4 might not reflect correctly the agreement between model and theory. In this respect, it might be also useful to report the uncertainty associated to the fitted value of χ_{12}.

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