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Application of Flory-Treszczanowicz-Benson model and Prigogine-Flory-Patterson theory to Excess Molar Volumes of Isomers of Propanol with Cyclohexane or n-Hexane

Suman Gahlyan, Sweety Verma, Manju Rani* and Sanjeev Maken†

Department of Chemistry, Deenbandhu Chhotu Ram University of Science and Technology, Murthal, Sonepat 131039, India
**Department of Chemical Engineering, Deenbandhu Chhotu Ram University of Science and Technology, Murthal, Sonepat 131039, India*
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Abstract – Excess molar volumes (V_m^E) of binary mixtures of 1-propanol or 2-propanol (1) + cyclohexane or n-hexane (2) were measured with V-shaped dilatometer at 303.15 K. The V_m^E data for these mixtures varied as: 2-propanol > 1-propanol and were higher for cyclohexane than n-hexane for both propanol systems. The experimental data were correlated with Redlich-Kister polynomial. The V_m^E data were interpreted qualitatively as well as quantitatively in terms of Flory-Treszczanowicz-Benson model and Prigogine-Flory-Patterson theory. Both models correctly described the sign and shape of V_m^E *vs* x_1 curves. The values calculated by both the models agree well with the experimental data.

Key words: Excess volume, Propanol, Cyclohexane, n-Hexane, Flory-Treszczanowicz-Benson model, Prigogine-Flory-Patterson theory

1. Introduction

Oxygenates are oxygen-containing compounds that can be blended with fuel to increase fuel efficiency, decrease fossil fuel dependence as well as CO_2 and toxic emission. Oxygenates produced from renewable sources, specially propanol and higher alkanol, may have acceptable properties as gasoline blend components and are considered as potential second generation bio-fuel additives [5]. Though propanol is rarely discussed as a biofuel, it can be produced by microbial fermentation of cellulose [6]. Excess thermodynamic properties of such model oxygenated fuels are quite important for the motor fuel formulation as well as understanding molecular interactions [1-4]. We are working on the thermodynamics of mixtures containing oxygenate, alkanes and aromatic hydrocarbons [5-18]. Excess function of binary mixtures with alkanol were considered to be of two parts: physical and chemical contribution. In this paper, excess molar volume (V_m^E) of isomers of propanol (1) + cyclohexane and n-hexane (2) at 303.15 K are reported. The V_m^E data were analyzed in the light of Flory-Treszczanowicz-Benson (FTB) model and Prigogine-Flory-Patterson (PFP) theory. FTB model assumes the V_m^E as sum of physical contribution calculated by Flory theory and chemical contribution due to Mecke-Kempter (MK) type of association in alcohol [19], whereas PFP theory considers the excess volume as a sum of three contributions due to interaction between unlike molecules, free volume change and change in internal pressure and reduced volume [20].

† To whom correspondence should be addressed.
E-mail: sanjeevmakin@gmail.com

2. Experimental Section

Cyclohexane , n-hexane and isomer of propanol (Merck) were purified by standard procedure [9,21]. Refractive index (±0.001) and density ($\pm 5\times10^{-5}$ g cm^{-3}) of pure components were measured by Abbe refractometer (OSAW, India) and dilatometric technique, respectively, as described elsewhere [22,23]. The temperature of water thermostat was controlled to ±0.01 K by a mercury-in-toluene regulator. The V_m^E data were determined with V-shaped dilatometer in the manner described previously [24]. The V_m^E data were accurate up to ± 0.5%.

Experimentally measured densities and refractive indices of the purified compounds compared well with literature and are reported in Table 1 [25-30].

The mixtures were made by weight using a balance (Shimadzu UniBloc 321-62900-64, AUX 120) having an accuracy of ±0.00001 g and mole fraction was reported to ±0.0001.

3. Results

The V_m^E is defined by

$$V_m^E = V_m - x_1 V_1^o - x_2 V_2^o \quad (1)$$

where V_m, V_1^o and V_2^o are the molar volume of a (1+2) mixture,

Table 1. Measured densities (ρ) and refractive indices (n_D) of the pure components at 303.15 K

Compound	ρ/g cm^{-3}		n_D	
	Exptl.	Literature	Exptl.	Literature
1-propanol	0.79565	0.79548 [25]	1.382	1.3814 [26]
2-propanol	0.78085	0.77695 [25]	1.371	1.37278 [27]
cyclohexane	0.76875	0.76845 [28]	1.421	1.42073 [29]
n-hexane	0.65110	0.65018 [28]	1.368	1.3698 [30]

Table 2. Experimental excess molar volume (cm^3mol^{-1}) of binary liquid mixtures of propanol (1) + hydrocarbon (2) at 303.15 K

x_1	V^E	x_1	V^E	x_1	V^E	x_1	V^E
1-propanol (1) + cyclohexane (2)		1-propanol (1) + n-hexane (2)		2-propanol (1) + cyclohexane(2)		2-propanol (1) + n-hexane (2)	
0.0456	0.057	0.0338	0.081	0.0496	0.115	0.0331	0.120
0.0923	0.109	0.0746	0.156	0.1236	0.278	0.1021	0.319
0.1422	0.176	0.1319	0.236	0.1847	0.388	0.1752	0.445
0.1847	0.246	0.1864	0.273	0.2535	0.498	0.2338	0.498
0.2345	0.298	0.2594	0.293	0.3180	0.568	0.2971	0.541
0.2712	0.342	0.3176	0.297	0.3548	0.594	0.3569	0.546
0.3548	0.395	0.3819	0.284	0.4125	0.619	0.4342	0.526
0.4278	0.431	0.4641	0.257	0.4478	0.640	0.4944	0.508
0.4900	0.429	0.5139	0.249	0.4900	0.637	0.5651	0.461
0.5446	0.416	0.5714	0.225	0.5446	0.633	0.6039	0.442
0.6011	0.395	0.6372	0.191	0.6011	0.596	0.6670	0.388
0.6549	0.349	0.7014	0.142	0.6549	0.565	0.7138	0.354
0.7168	0.315	0.7722	0.107	0.7168	0.502	0.7704	0.296
0.7782	0.265	0.8304	0.065	0.7782	0.432	0.8531	0.212
0.8561	0.198	0.8924	0.032	0.8561	0.306	0.8898	0.162
0.9427	0.086	0.9794	0.009	0.9470	0.131	0.9509	0.083

Table 3. Adjustable parameters of Redlich-Kister equation and standard deviation (σ) in (cm^3mol^{-1}) at 303.15 K

System	A_0	A_1	A_2	A_3	σ_m
1-propanol (1) + cyclohexane (2)	1.706	-0.371	-0.366	0.825	0.005
1-propanol (1) + n-hexane (2)	1.005	-0.754	0.382	-0.648	0.004
2-propanol (1) + cyclohexane (2)	2.549	-0.216	-0.018	0.341	0.004
2-propanol (1) + n-hexane (2)	2.006	-1.019	0.874	-0.133	0.004

propanol and hydrocarbon, respectively.

The V_m^E values are tabulated in Table 2 and correlated by Redlich and Kister equation:

$$V_m^E = x_1x_2\left[\sum_{j=0}^{n} A_j(x_1-x_2)^j\right] \quad (2)$$

where A_j are the adjustable parameters, and x_1 is the mole fraction of propanol in propanol (1) + hydrocarbon (2) mixture. These parameters were evaluated by fitting V_m^E data to Eq. (2) by the least squares method and reported in Table 3 with the standard deviations $\sigma(V_m^E)$ of V_m^E.

$$\sigma(V_m^E)(cm^3mol^{-1}) = \{[\sum(V_{expt.}^E - V_{cal.(Eq.2)}^E)^2]/(m-n)\}^{1/2} \quad (3)$$

where m is the number of experimental values, and n is the number of adjustable parameters in Eq. (2). The choice of n to have 1-4 values was dictated by the consideration that the maximum deviation $\sigma_{max}(V^E)$ of V_m^E (as calculated from Eq. (2) from the corresponding experimental V_m^E values satisfied the relation $\sigma_{max}(V_m^E) \le 2\sigma(V_m^E)$. Comparison of V_m^E experimental data for the studied system together with smoothing curves using Eq. (2) is shown in Fig. 1.

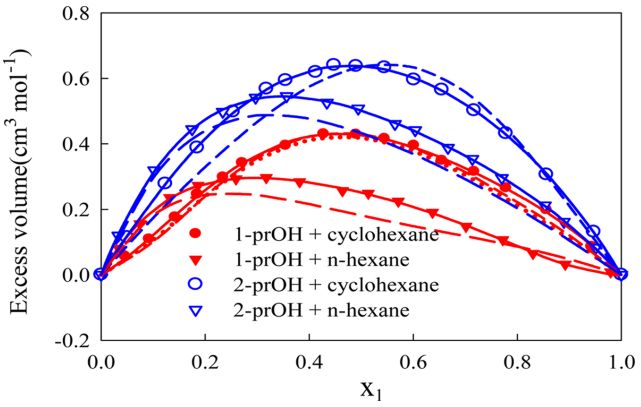

Fig. 1. Excess molar volume (V_m^E) of isomer of propanol (1) + cyclohexane or n-hexane (2) mixture as a function of mole fraction of propanol (x_1) at 303.15 K; Experimental symbols represent experimental values, lines represent values calculated from Eq. (2), and long dash (1- + n-hexane [31] and 2-propanol + n-hexane at 298.15 K [32]), small dash (2-propanol + cyclohexane) [33], dotted line (1-propanol + cyclohexane [28]).

4. Discussion

The V_m^E versus x_A plots for all these systems are positive and symmetrical about $x_1 = 0.5$ for cyclohexane mixtures whereas these are skewed toward the y-axis for the binary systems with n-hexane. The present mixtures were also compared with literature for propanol with cyclohexane mixtures at 303.15 K, but for propanol with n-hexane no data was found at 303.15 K, so we compared this system at 298.15 K [29,31-33]. It was observed from Fig. 1 that the V_m^E increases with increase in temperature due to weakening of bond between propanol molecules. The V_m^E data for propanol + cyclohexane or n-hexane mixtures vary as 2-propanol > 1-propanol and are

higher for cyclohexane than n-hexane. This may be due to the rupturing of H-bonded network in propanol in the pure state owing to mixing with cyclohexane and n-hexane that results in increase in volume [34]. As straight chain n-hexane gets fitted into linearly associated propanol molecules leading to negative contribution for excess volume, so excess volume for cyclohexane is larger than n-hexane. Furthermore, the 2-propanol mixture has higher V_m^E values than 1-propanol; this may be due to branching of alkyl group attached to hydroxyl group that would offer more steric hindrance [35].

4-1. The Prigogine-Flory-Patterson (PFP) theory

According to this theory, V_m^E is the combination of three factors [20]:

$$V_m^E = V_{Inter}^E + V_{free\ vol}^E + V_{P*}^E \tag{4}$$

$$\frac{V_m^E}{x_1V_1^* + x_2V_2^*} = \frac{(\tilde{V}^{-1/3}-1)\tilde{V}^{2/3}\psi_1\theta_2(\chi_{12}^*/P_1^*)}{((4/3)\tilde{V}^{-1/3}-1)} - \frac{(\tilde{V}_1-\tilde{V}_2)^2((14/9)\tilde{V}^{-1/3}-1)\psi_1\psi_2}{((4/3)\tilde{V}^{-1/3}-1)\tilde{V}} + \frac{(\tilde{V}_1-\tilde{V}_2)(P_1^*-P_2^*)\psi_1\psi_2}{P_2^*\psi_1+P_1^*\psi_2} \tag{5}$$

where all the terms have their usual meaning [36,37]. Flory theory parameters of pure compounds employed in these calculations are recorded in Table 4. The V_m^E data were calculated from Eq. (4) in the manner described elsewhere [39].

The three contributions to V_m^E at $x_1 = 0.5$ and Flory interaction parameters χ_{12}^* are reported in Table 5. The V_m^E values calculated by PFP theory not only predict the sign and shape of V_m^E versus x_1 curves, but also the agreement between experimental and calculated excess volume is very impressive (Fig. 2). This comparison is also shown quantitatively in terms of standard deviation (σ_{PFP}) in Table 6.

4-2. Flory-Treszczanowicz-Benson (FTB) association model

Treszczanowicz and Benson developed an association model [19] for alkanol (1) + alkane (2) systems in which alkanol is self-associated through Mecke-Kempter (MK) association and alkane

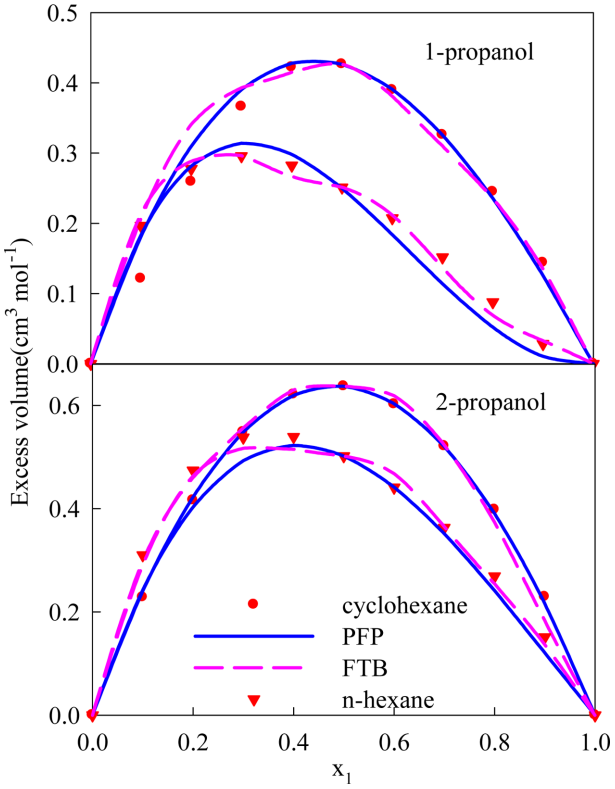

Fig. 2. Excess molar volume (V_m^E) of isomer of propanol (1) + cyclohexane or n-hexane (2) as a function of mole fraction of propanol (x_1) at 303.15 K.

behaves as inert entity. According to this model V_m^E is the result of two contributions: (i) a chemical contribution arises due to MK type of association, and (ii) a physical contribution given by Flory theory [36,37].

$$V_m^E = V_{MK}^E + V_{Phys}^E \tag{6}$$

The chemical contribution V_{MK}^E is expressed as [36,37]

$$V_{MK}^E = \Delta v_H^o x_1 h(K^\phi, \phi_1) \tag{7}$$

The values of association parameters Δv_H^0, Δh_H^0 and Δs_H^0 used

Table 4. Molar volume (V) isobaric expansivity (α) and isothermal compressibility (κ_T) characteristic pressure (P*) characteristic molar volume (V*) and characteristic temperature (T*) obtained from Flory theory for the pure liquids at 303.15 K

Compound	V ($cm^3\ mol^{-1}$)	$10^3\ \alpha\ (K^{-1})$	$10^6\ \kappa_T\ (cm^3\ J^{-1})$	P* (J cm^{-3})	V* ($cm^3\ mol^{-1}$)	T* (K)
1-propanol	75.503	1.0344	679.79	730.272	60.01	5174.85
2-propanol	77.359	1.1437	828.66	730.272	60.01	5174.85
cyclohexane	109.401	1.232	1237	508.147	84.329	4732.17
n-hexane	131.610	1.404	780.9	422.277	99.011	4452.122

Table 5. Values of the various contributions to excess volume ($cm^3\ mol^{-1}$), PFP interaction parameter χ_{12}^* at 303.15 K as well as Treszczanowicz-Benson (TB) association model parameters K^ϕ and χ_{12}

System	V_{Inter}^E	$V_{Free\ vol}^E$	V_{P*}^E	χ_{12}^*	K^ϕ	χ_{12}
1-propanol (1) + cyclohexane (2)	0.783	0.041	-0.254	64.359	234.368	51.500
1-propanol (1) + n-hexane (2)	1.154	0.150	-0.726	77.557	234.368	67.853
2-propanol (1) + cyclohexane (2)	0.724	0.008	-0.094	52.363	232.904	44.879
2-propanol (1) + n-hexane (2)	1.051	0.071	-0.442	65.443	232.904	53.928

Table 6. Comparison of excess molar volume (V^E) (in cm^3 mol^{-1}) values calculated from Flory-Treszczanowicz-Benson (FTB) and Prigogine-Flory-Patterson theory (PFP) model with their corresponding experimental values for the various binary mixtures as functions of mole fraction of propanol x_1, at 303.15 K. Standard deviations (σ) (in cm^3 mol^{-1}) of both the models are also included

x_1	V^E			V^E		
	Exptl	FTB	PFP	Exptl	FTB	PFP
	1-propanol (1) + cyclohexane (2)			2-propanol (1) + cyclohexane(2)		
0.1	0.172	0.255	0.212	0.228	0.298	0.128
0.2	0.318	0.380	0.361	0.416	0.460	0.256
0.3	0.423	0.457	0.452	0.548	0.556	0.384
0.4	0.479	0.489	0.493	0.621	0.630	0.511
0.5	0.489	0.489	0.489	0.637	0.637	0.637
0.6	0.459	0.436	0.446	0.602	0.618	0.763
0.7	0.397	0.364	0.370	0.521	0.524	0.888
0.8	0.304	0.278	0.267	0.398	0.374	1.013
0.9	0.177	0.161	0.142	0.229	0.183	1.136
		σ = 0.061	σ = 0.085		σ = 0.049	σ = 0.021
	1-propanol (1) + n-hexane (2)			2-propanol (1) + n-hexane (2)		
0.1	0.196	0.213	0.173	0.310	0.290	0.235
0.2	0.278	0.288	0.268	0.474	0.463	0.393
0.3	0.294	0.296	0.303	0.538	0.517	0.485
0.4	0.278	0.267	0.292	0.539	0.515	0.518
0.5	0.250	0.249	0.250	0.502	0.502	0.502
0.6	0.220	0.212	0.189	0.441	0.468	0.446
0.7	0.189	0.137	0.122	0.363	0.365	0.358
0.8	0.151	0.069	0.061	0.269	0.270	0.248
0.9	0.094	0.033	0.017	0.151	0.145	0.126
		σ = 0.017	σ = 0.065		σ = 0.025	σ = 0.117

to calculate V^E_{MK} from Eq. (7) are taken as -10 cm^3 mol^{-1}, -24400 J mol^{-1} and -33 J $K^{-1}mol^{-1}$, respectively [19].

The physical contribution is given as [36-40]

$$V^E_{Phys} = V^E_{Flory} = V^E_{Interaction} + V^E_{Free\ volume} \quad (8)$$

According to Flory theory

$$V^E_{Flory} = V^*[\tilde{V} - (\tilde{V}_1\phi_1 + \tilde{V}_2\phi_2)] \quad (9)$$

$$\tilde{T}\tilde{V}^{4/3} - \tilde{V}^{1/3} + 1 = 0 \quad (10)$$

$$\tilde{T} = \sum_{i=1}^{2}(\phi_i P^*_i \tilde{T}_i)/\left(\sum_{i=1}^{2}(\phi_i P^*_i) - \phi_1\theta_2\chi_{12}\right) \quad (11)$$

Calculation of V^E_{Flory} from Eq. (9) requires the reduced volume of mixtures $\tilde{V}$ from Eq. (10), which in turn involves Flory's interaction parameter χ_{12}. The parameter χ_{12} was computed from Eqs. (9)-(11) and using equimolar values of experimental V_m^E (Table 5). In these equations all the terms have their usual meaning [19], [36-40]. The calculated V_m^E values agree well with experimental data as shown in Fig. 2 and Table 6 and also in terms of standard deviation (σ_{FTB}) in Table 6.

5. Conclusion

Excess molar volumes (V_m^E) for isomers of propanol (1) + cyclohexane or n-hexane (2) systems were determined with v-shaped dilatometer at 303.15 K. The V_m^E data for propanol + cyclohexane or n-hexane mixtures varied as: 2-propanol > 1-propanol and were higher for cyclohexane than n-hexane. The V_m^E data were analyzed in the light of FTB model and PFP theory. Both the models, FTB and PFP, correctly predicted the sign and shape of V_m^E versus x_1 curves. The V_m^E values calculated by FTB and PFP model compared well with the respective measured data for all the present systems and this was also confirmed quantitatively in terms of standard deviations.

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