NON-IDEAL BEHAVIOUR OF BINARY LIQUID MIXTURES AND DIFFERENT APPROACHES TO CALCULATE IDEAL EXPANSIBILITY

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Densities for binary mixtures of cyclopentane with 1-propanol, 1-pentanol, and 1-heptanol over the whole composition range have been measured at temperature 318.15 K. From these densities, excess molar volumes were calculated. All the experimental excess molar volumes were fitted to Redlich–Kister equation and were analyzed with the help of Prigogine–Flory–Patterson theory. Also isobaric expansibilities and their excess counterparts were calculated. Further different approaches were analysed for calculating ideal expansibilities.
Keywords: Binary liquid mixtures; Ideal expansibility; Cyclopentane; 1-Alkanols; Prigogine–Flory–Patterson theory; Thermodynamics.

Molecular interactions that are present in solutions and liquid mixtures are characterised by the help of excess quantities. In earlier studies[1–4] results on various thermodynamic properties of binary liquid mixtures have been reported. In a continuing effort to understand the mixing behaviour and to extend the database for binary liquid mixtures, new densities for the mixtures cyclopentane (1) + 1-propanol (2), + 1-pentanol (2), + 1-heptanol (2) at 318.15 K over whole composition range are reported in this paper. From these density measurements, we calculated the excess molar volumes V_m^E, and these were fitted to the Redlich–Kister polynomial equation. Also, the results of excess molar volumes were analysed with the help of Prigogine–Flory–Patterson (PFP) theory.

In order to study and understand the behaviour of cycloalkane + 1-alkanol systems under different conditions of temperature and compositions their thermophysical properties are needed. Earlier Dominguez et al.[5] have reported densities, speed of sound and viscometric measurement for cycloalkane with 1-alkanol at different temperatures.

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Excess thermal coefficients α^E have been neglected in interrelating thermodynamic properties, because they have usually been thought too small to be significant for thermodynamic consideration. The values are certainly small for non-polar mixtures, but the values are considerably larger for polar mixtures[6]. So, isobaric expansibility α_P and excess isobaric expansibility $\alpha_P{}^E$ were also calculated from excess molar volumes. The excess properties are useful in the characterization of molecular interactions present in liquid mixtures, which are calculated with the evaluation of ideal quantities. From the literature[7–12], it is revealed that many approximations are used for expressing ideal mixing laws, which gave different set of excess quantities. For better understanding, different approaches for calculating ideal expansibilities are analysed with the help of our experimental values.

EXPERIMENTAL

Cyclopentane, 1-propanol, 1-pentanol, and 1-heptanol were obtained from SD Fine Chemicals, India. All the liquids were stored in dark bottles over 0.4 nm molecular sieves to reduce water content, and were partially degassed under vacuum. The estimated purities as per gas chromatographic analysis were better than 99.5 mole % for all liquid samples. Further the purities of liquids were checked by comparing densities of pure solvents with their corresponding literature values[13–17] reported in Table I.

TABLE I
Experimental densities, ρ, at 318.15 K and isobaric expansibilities, α_P, at 308.15 K of the pure component liquids

Solvent	ρ, kg m^{-3}		$\alpha_P \times 10^3$, K^{-1}[a]
	exp	lit.	
Cyclopentane	719.307		0.979
1-Propanol	783.409	783.31[13] 783.25[14]	1.031
1-Pentanol	796.128	796.2[15] 796.19[16]	0.921
1-Heptanol	804.536	804.8[15] 804.43[17]	0.876

[a] Calculated from densities from this work, and densities measured at 298.15 and 308.15 K and published in ref.[1]

The densities of the pure liquids and their mixtures were measured using an Anton Paar DSA 5000 densimeter provided with two integrated Pt 100 Platinum thermometers. The temperature was controlled to ±0.001 K by built-in solid state thermostat. Before each series of measurements, the apparatus was calibrated with double distilled and degassed water, n-hexane, n-heptane, n-octane, cyclohexane, and benzene. The reproducibility of density measurements was $\pm 1 \times 10^{-6}$ g cm^{-3} and uncertainties of these assumed to be less than 5×10^{-6} g cm^{-3}.

The mixtures were prepared by mass and were kept in special air tight stoppered glass bottles to avoid evaporation. The weighings were done with an electronic balance with precision of ±0.01 mg. The mole fraction of each mixture was obtained with an uncertainty of 1×10^{-4} from the measured masses of the components. All molar quantities were based on the IUPAC relative atomic mass table[18].

THEORETICAL

Density Measurements

Experimental densities, ρ, for binary mixtures cyclopentane (1) + 1-propanol (2), + 1-pentanol (2), and + 1-heptanol (2) at 318.15 K are reported in Table II. Excess molar volumes for binary mixtures were calculated from our measurements according to following equation

$$V_m^E = (x_1M_1 + x_2M_2)/\rho - x_1M_1/\rho_1 - x_2M_2/\rho_2 \qquad (1)$$

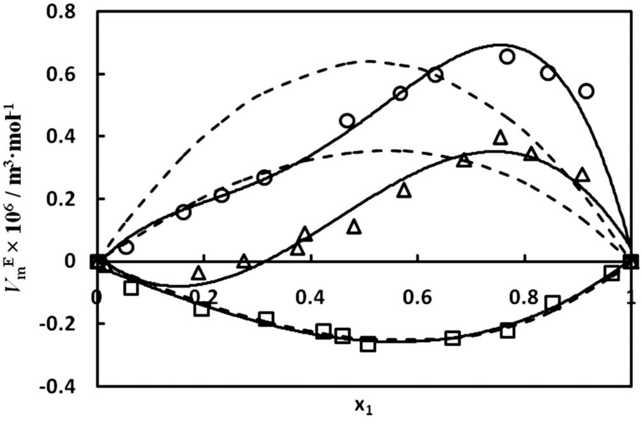

FIG. 1
Excess molar volumes, V_m^E, at 318.15 K for cyclopentane (1) + 1-alkanol (2): ○, 1-propanol; △, 1-pentanol; □, 1-heptanol. Solid lines have been obtained from Eq. (*2*). Dotted lines have been obtained from PFP theory

TABLE II
Densities, ρ, and excess molar volume, V_m^E, for cyclopentane (1) + 1-alkanol (2) mixtures at 318.15 K

x_1	$\rho \times 10^{-3}$, kg m^{-3}	$V_m^E \times 10^6$, m^3 mol^{-1}
	Cyclopentane (1) + 1-Propanol (2)	
0.0553	0.778506	0.0464
0.1627	0.769213	0.1562
0.2341	0.763485	0.2125
0.3137	0.757424	0.2669
0.4691	0.745603	0.4519
0.5666	0.738891	0.5388
0.6338	0.734471	0.5959
0.7688	0.726442	0.6545
0.8446	0.722803	0.6012
0.9167	0.719498	0.5438
	Cyclopentane (1) + 1-Pentanol (2)	
0.0155	0.795169	–0.0127
0.1886	0.783336	–0.0359
0.2742	0.776936	0.0021
0.3746	0.769286	0.0432
0.3884	0.767938	0.0877
0.4798	0.760881	0.1121
0.5738	0.752792	0.2280
0.6868	0.743137	0.3255
0.7543	0.737140	0.3977
0.8131	0.732668	0.3467
0.9079	0.725187	0.2798
	Cyclopentane (1) + 1-Heptanol (2)	
0.0632	0.801306	–0.0852
0.1965	0.793351	–0.1521
0.3173	0.785294	–0.1837
0.4244	0.777623	–0.2260
0.4606	0.774892	–0.2412
0.5084	0.771202	–0.2668
0.6664	0.757237	–0.2451
0.7675	0.747250	–0.2241
0.8534	0.737525	–0.1316
0.9644	0.724016	–0.0398

where x_1 and x_2 are mole fractions, M_1 and M_2 are molar masses and ρ_1 and ρ_2 are the densities of pure components 1 and 2, respectively, ρ is the density of the binary mixtures. The values of excess molar volumes are reported in Table II and are graphically presented in Fig. 1.

The calculated values of V_m^E of the binary mixtures were fitted to a Redlich-Kister[19] type polynomial equation

$$Y^E = x_1 x_2 \sum_{i=0}^{n} A_i (x_1 - x_2)^i \qquad (2)$$

where Y^E stands for V_m^E. The coefficients A_i for the correlation of V_m^E – composition data, evaluated using least-squares method are given in Table III along with the resulting standard deviations in excess molar volume $\sigma(Y^E)$. The standard deviation was calculated by equation

$$\sigma(Y^E) = [\sum_{1}^{n} (Y^E_{exp} - Y^E_{calc})^2/(n - m)]^{1/2} \qquad (3)$$

where n and m are the numbers of experimental points and parameters, respectively.

TABLE III
Standard deviations, σ, and parameters A_i in Eq. (2) for excess molar volumes, V_m^E, and excess isobaric compressibilities, α_p^E

Y^E	T, K	A_0	A_1	A_2	A_3	A_4	σ
			Cyclopentane (1) + 1-Propanol (2)				
$V_m^E \times 10^6$, m^3 mol^{-1}	318.15	1.9376	1.7387	0.6696	2.1848	3.0501	0.0352
$\alpha_p^E \times 10^3$, K^{-1}	308.15	0.0226	0.0260	0.0306	0.0156		0.0006
			Cyclopentane (1) + 1-Pentanol (2)				
$V_m^E \times 10^6$, m^3 mol^{-1}	318.15	0.6200	1.8631	1.2775	0.6273		0.0244
$\alpha_p^E \times 10^3$, K^{-1}	308.15	–0.0073	0.0114	0.0313	0.0382	–0.0456	0.0003
			Cyclopentane (1) + 1-Heptanol (2)				
$V_m^E \times 10^6$, m^3 mol^{-1}	318.15	–1.0061	–0.5465	–0.1768	1.0242		0.0141
$\alpha_p^E \times 10^3$, K^{-1}	308.15	–0.0243	–0.0069	–0.0069			0.0004

Prigogine–Flory–Patterson Theory[20–23]

The PFP theory in the following form has been used to analyze the experimental excess molar volumes of the studied binary liquid mixtures

$$\frac{V_m^E}{x_1 V_1{}^* + x_2 V_2{}^*} = \frac{(\tilde{v}^{1/3} - 1)\tilde{v}^{2/3}}{4/3\,\tilde{v}^{1/3} - 1}\Psi_1\theta_2\frac{\chi_{12}}{P_1{}^*}$$

$$-\frac{(\tilde{v}_1 - \tilde{v}_2)^2\,(14/9\,\tilde{v}^{-1/3} - 1)}{4/3\,\tilde{v}^{1/3} - 1}\Psi_1\Psi_2$$

$$+\frac{(\tilde{v}_1 - \tilde{v}_2)^2\,(P_1{}^* - P_2{}^*)}{P_1{}^*\,\Psi_2 + P_2{}^*\,\Psi_1}\Psi_1\Psi_2 \qquad (4)$$

where Ψ represents the contact energy fraction, given by

$$\Psi_1 = 1 - \Psi_1 = \frac{\phi_1 P_1{}^*}{\phi_1 P_1{}^* + \phi_2 P_2{}^*} \qquad (5)$$

and x_i is the mole fraction, P_i^* (= $\gamma T\tilde{v}^2$ = $(\alpha_{P,i}/\kappa_{T,i})T\tilde{v}_i^2$) is the characteristic pressure, $\tilde{v}_i$ is the reduced volume (i = 1 and 2), $\kappa_{T,i}$ is the isothermal compressibility, θ_2 (= $\phi_2/[\phi_2 + \phi_1(s_1/s_2)]$) are the segment fractions and ϕ_2 (= $1 - \phi_1 = x_2/[x_2 + x_1(r_1/r_2)]$) are the site fractions. Also, r_1 and r_2 are in the ratio of the respective molar core volumes V_1^* and V_2^*, i.e. $r_1/r_2 = V_1^*/V_2^*$. Similarly s_1 and s_2 are the ratios of the molecular surface areas of contact per segment ($s_1/s_2 = (V_2^*/V_1^*)^{1/3}$). The subscripts 1 and 2 refer to the two pure components, whereas quantities without a subscript refer to the mixture. The first term of Eq. (*4*) represents the interactional contribution to V_m^E, i.e. χ_{12} parameter is often called cross interaction parameter. The second term is the difference in "free volume", while the third term is the internal pressure contribution, i.e. P^* effect of V_m^E. The values of the parameters for pure components and mixtures, which are required to compute V_m^E from Eq. (*4*), have been calculated using Flory theory[23] and are reported in Table IV for pure components. The cross interaction parameter χ_{12} required for the calculation of V_m^E using PFP theory was derived from excess molar volumes for each mixture. The calculated values of three contributions, viz. interactional, free volume and P^* effect along with cross interaction parameter are given in Table V.

Excess and Ideal Expansibility

By definition, an excess quantity Z^E (in present case it is expansibility) is the excess of the given property Z of a real mixture over Z^{id}, the value for an ideal mixture under same set of conditions

$$Z^E = Z - Z^{id} \ . \qquad (6)$$

The accurate experimental value of Z and correct approximation used for Z^{id} gives accurate value of Z^E. There are many approximations which have been proposed for ideal contribution to excess speeds of sound, excess viscosities, excess isentropic compressibilities and excess expansibilities. In present paper, different approaches used for evaluating ideal expansibilities have been analysed.

TABLE IV
Values of isothermal compressibility, κ_T, reduced volume, $\tilde{v}$, characteristic volume, V^*, characteristic temperature, T^*, and characteristic pressure, P^*, of pure components at 318.15 K

Component	κ_T, T Pa^{-1}	$\tilde{v}$	$V^* \times 10^6$, m^3 mol^{-1}	T^*, K	$P^* \times 10^6$, J m^{-3}
Cyclopentane	1367	1.2617	77.2772	5383	371.2
1-Propanol	1157	1.2723	60.2926	5247	468.3
1-Pentanol	974	1.2488	88.6658	5566	384.3
1-Heptanol	906	1.2373	116.7290	5745	386.7

TABLE V
Values of interaction parameter, χ_{12}, and contributions to the excess molar volume from the PFP theory at 318.15 K calculated from Eq. (*4*)

Component	$\chi_{12} \times 10^6$, J m^{-3}	Caculated contribution		
		interaction	free volume	P^* effect
1-Propanol	0.3258	0.5597	0.0027	0.0389
1-Pentanol	0.2252	0.3912	0.0049	–0.0628
1-Heptanol	–0.0469	–0.0858	0.0202	–0.1391

The values of expansibilities are mainly discussed in terms of isobaric expansibility α_P and isentropic expansibility α_S. The values of isobaric expansibilities α_P for the binary mixtures at a particular composition were calculated by adding the contribution of the expansion coefficients of each component in the mixtures

$$\alpha_P = 1/V_m[(x_1M_1\alpha_{P,1}/\rho_1 + x_2M_2\alpha_{P,2}/\rho_2) + (\partial V^E/\partial T)_{P,x}] \qquad (7)$$

where $\alpha_{P,1}$ and $\alpha_{P,2}$ are isobaric expansion coefficients of pure components 1 and 2, respectively. These values of isobaric expansibilities α_P are reported in Table VI and are graphically presented in Fig. 2.

The values of isentropic expansibility α_S (= $1/V_m(\partial V_m/\partial T)_S$) can be calculated by equation

$$\alpha_S = -\kappa_S\beta_S \qquad (8)$$

where β_S (= $(\partial P/\partial T)_S$) and κ_S (= $-(1/V)(\partial V/\partial P)_S$) are isentropic thermal pressure coefficient and isentropic compressibility, respectively.

Douheret et al.[24,25] have given ideal mixing rule for isentropic expansibility, α_S^{id}, on the basis of volume fraction weighted average

$$\alpha_S^{id} = \sum_i \varphi_i[1 - (\kappa_{T,i}/\alpha_{S,i})(\beta_S^{id} - \beta_{S,i})]\alpha_{S,i} \qquad (9)$$

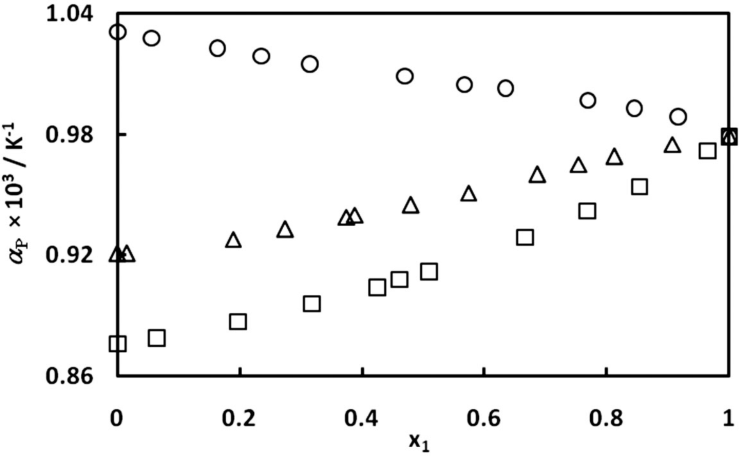

FIG. 2
Isobaric expansibility, α_P, at 308.15 K for cyclopentane (1) + 1-alkanol (2): ○, 1-propanol; △, 1-pentanol; □, 1-heptanol

TABLE VI
Values of isobaric expansibility, α_P, and excess isobaric expansibility, α_P^E, for cyclopentane (1) + 1-alkanol (2) mixtures at 308.15 K

x_1	$\alpha_P \times 10^3$, K^{-1}	$\alpha_P^E \times 10^3$, K^{-1}
	Cyclopentane (1) + 1-Propanol (2)	
0.0553	1.028	0.0006
0.1627	1.023	0.0023
0.2341	1.019	0.0026
0.3137	1.015	0.0031
0.4691	1.009	0.0055
0.5666	1.005	0.0065
0.6338	1.003	0.0078
0.7688	0.997	0.0081
0.8446	0.993	0.0074
0.9167	0.989	0.0065
	Cyclopentane (1) + 1-Pentanol (2)	
0.0155	0.921	–0.0008
0.1886	0.928	–0.0028
0.2742	0.933	–0.0025
0.3746	0.939	–0.0020
0.3884	0.940	–0.0018
0.4798	0.945	–0.0020
0.5738	0.951	–0.0015
0.6868	0.960	0.0008
0.7543	0.965	0.0017
0.8131	0.969	0.0020
0.9079	0.975	0.0020
	Cyclopentane (1) + 1-Heptanol (2)	
0.0632	0.879	–0.0015
0.1965	0.887	–0.0036
0.3173	0.896	–0.0046
0.4244	0.904	–0.0062
0.4606	0.908	–0.0056
0.5084	0.912	–0.0063
0.6664	0.929	–0.0061
0.7675	0.942	–0.0051
0.8534	0.954	–0.0041
0.9644	0.972	–0.0017

where φ_i is the volume fraction and $\kappa_{T,i}$ is the isothermal compressibility.

Also, $\beta_{S,i}$ and β_S^{id} isentropic thermal pressure coefficient for pure component and ideal isentropic thermal pressure coefficient, respectively, are calculated as

$$\beta_{S,i} = C_{P,i}/T\, V_i\, \alpha_{P,i} \tag{10}$$

$$\beta_S^{id} = C_{P,}{}^{id}{}_{m}/T\, V_m^{id}\, \alpha_P^{id}\ . \tag{11}$$

Baumgartner and Atkinson[26] gave following ideal mixing rules for different thermodynamic quantities

$$C_{P,}{}^{id}{}_{m} = \sum_i x_i C_{P,i} \tag{12}$$

$$V_m^{id} = \sum_i x_i V_i \tag{13}$$

$$\alpha_P^{id} = \sum_i \varphi_i \alpha_{P,i}\ . \tag{14}$$

Equations (*6*), (*7*), and (*14*) have been used to calculate the excess isobaric expansibilities α_P^E. The values of α_P^E were fitted to Eq. (*2*), where Y^E stands for α_P^E and standard deviations were calculated with the help of Eq. (*3*). The coefficients A_i of Eq. (*2*) for the correlation of α_P^E – composition data, evaluated using least-squares method are given in Table III along with the resulting standard deviations in excess isobaric expansibilities $\sigma(\alpha_P^E)$.

The quantities molar isobaric and molar isentropic expansibilities $A_{P,m}$ and $A_{S,m}$ is given by equations

$$A_{P,m} = (\partial V_m/\partial T)_P = V_m \alpha_P \tag{15}$$

$$A_{S,m} = (\partial V_m/\partial T)_S = V_m \alpha_S\ . \tag{16}$$

The corresponding ideal molar isobaric expansibility and ideal molar isentropic expansibility[27] are calculated using equations

$$A_{P,}{}^{id}{}_{m} = V_m^{id}\, \alpha_P^{id} \tag{17}$$

$$A_{S,}{}^{id}{}_{m} = \sum x_i[1 - (K_{T,i}/A_{S,i})(\beta_S{}^{id} - \beta_{S,i})]A_{S,i} \tag{18}$$

where $K_{T,i}$ is the molar isothermal compressibility for pure components.

Excess molar isobaric expansibility has been calculated using the equation

$$A_{P,}{}^{E}{}_{m} = A_{P,m} - A_{P,}{}^{id}{}_{m} \; . \tag{19}$$

The excess quantities $\alpha_P{}^E$ and $A_{P,}{}^{E}{}_{m}$ are presented in Fig. 3. Due to non-availability of some parameters, the excess quantities $\alpha_S{}^E$ and $A_{S,}{}^{E}{}_{m}$ have not been calculated in the present study. Further, values of $\alpha_P{}^{id}$ and $\alpha_S{}^{id}$, and corresponding molar quantities $A_{P,}{}^{id}{}_{m}$ and $A_{S,}{}^{id}{}_{m}$ are graphically presented in Fig. 4 for one binary mixture cyclopentane (1) + 1-propanol (2) as other mixtures show similar behaviour.

RESULTS AND DISCUSSION

It is observed from Table II and Fig. 1 that the excess molar volumes are negative for the system cyclopentane (1) + 1-heptanol (2) at 318.15 K over the whole composition range. Excess molar volumes values change sign from negative to positive for the system cyclopentane (1) + 1-pentanol (2)

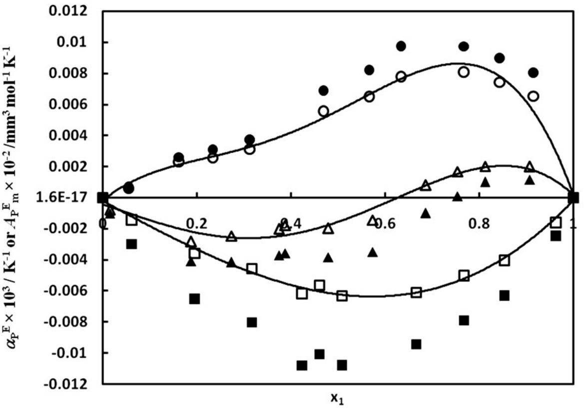

FIG. 3

Excess isobaric expansibilities, $\alpha_P{}^E$, for cyclopentane (1) + 1-alkanol (2): ○, 1-propanol; △, 1-pentanol; □, 1-heptanol. Solid lines have been obtained from Eq. (*2*). Excess molar isobaric expansibility, $A_{P,}{}^{E}{}_{m}$, for cyclopentane (1) + 1-alkanol (2): ●, 1-propanol; ▲, 1-pentanol; ■, 1-heptanol and are positive for the system cyclopentane (1) + 1-propanol (2) at 318.15 K. The negative excess molar volumes can be attributed to strong unlike interactions between molecules. The excess molar volume increases with the decrease in the chain length of 1-alkanol. The behaviour is same as observed for cyclopentane (1) + 1-alkanol (2) systems[1] at 298.15 and 308.15 K. Weaker dipole–dipole interactions in lower 1-alkanol results due to the decrease in their polarizability with decreasing chain length[28]. Excess molar volumes increase with increase in temperature for the binary mixtures.

It is observed from Table V that cross interaction parameter χ_{12} is positive for all the mixtures except for cyclopentane (1) + 1-heptanol (2) system for which it gives negative value. Positive values of χ_{12} suggest relatively weak intermolecular interactions between the components of the mixtures which support experimental V_m^E results. The values for free volume are positive for all the mixtures. The third contribution, i.e. P^* effect values are negative for all the mixtures except for cyclopentane (1) + 1-propanol (2) mixture. The V_m^E values calculated from PFP theory are plotted along with experimental V_m^E in Fig. 1 and it is observed that PFP theory explains cyclopentane (1) + 1-heptanol (2) mixture very well and shows some deviations for other two mixtures.

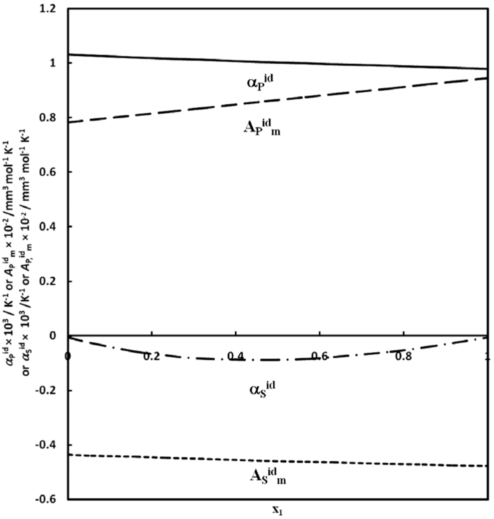

FIG. 4

Ideal expansibilities, α_P^{id}, $A_{P,}{}^{id}{}_m$, α_S^{id}, and $A_{S,}{}^{id}{}_m$, for cyclopentane (1) + 1-propanol (2) system

From Fig. 2 and Table VI it is found that isobaric expansibility increases for all mixtures as concentration of cyclopentane increases except for cyclopentane (1) + 1-propanol (2) for which it decreases. Excess isobaric expansibilities listed in Table VI and shown in Fig. 3 are negative and positive for higher and lower 1-alkanol, respectively. Change in sign in values of excess isobaric expansibility is observed for cyclopentane (1) + 1-pentanol (2). Isobaric expansibility represents the fluctuation of enthalpy and interactive volume, the negative excess isobaric expansibility probably mean a strong interaction between unlike species, and positive excess isobaric expansibility may indicate a breakable and loose interaction between unlike species or strong interaction between like species[29]. The excess isobaric expansibility increases with decreasing chain length of 1-alkanol, the same as observed in the case of excess molar volumes.

Further, from Fig. 4 it is observed that the values obtained of $\alpha_P{}^{id}$ and $A_{P,}{}^{id}{}_m$ are close to each other and there is a close similarity between their excess counterparts $\alpha_P{}^E$ and $A_{P,}{}^E{}_m$ as it is evident from Fig. 3. The $\alpha_S{}^{id}$ and $A_{S,}{}^{id}{}_m$ values are negative and show large deviations from positive values of $\alpha_P{}^{id}$ and $A_{P,}{}^{id}{}_{m.}$ The difference between the values of $\alpha_P{}^{id}$ and $A_{P,}{}^{id}{}_m$ is large as compared to difference between the values of $\alpha_S{}^{id}$ and $A_{S,}{}^{id}{}_m$.

So, it is observed that different ideal mixing rules for expansibility give different set of values which may lead to improper estimates of excess expansibilities which are quite important from thermodynamic point of view.

CONCLUSIONS

The excess molar volumes reported in this study for cyclopentane (1) + 1-alkanol (2) binary liquid mixtures are in accordance with earlier study[1]. The PFP theory used to analyse experimental excess molar volumes gives good results for cyclopentane (1) + 1-heptanol (2) only, and shows deviations for rest of the mixtures. On this basis, it may be said that the PFP theory does not predict well or is not useful for binary liquid mixtures chosen for this study. The analysis of different approaches used to calculate ideal expansibility shows that different sets of values are obtained, that give rise to different excess expansibilities which may not be correct as it may lead to improper estimates.

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