EXCESS MOLAR VOLUMES AND DEVIATIONS IN VISCOSITY OF THE BINARY MIXTURES OF 1-PENTANOL + AROMATIC HYDROCARBONS AT T = 298.15 K

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Abstract

The densities, ρ and viscosities, η of pure 1-Pentanol, toluene, ethylbenzene, propylbenzene, and those of their binary mixtures covering the whole composition range have been measured at 298.15 K under atmospheric pressure. Excess molar volumes, V_m^E, partial molar volumes, V_i, deviations in viscosity, $\Delta\eta$ and excess free energies of activation, $\Delta G^{\#E}$, have also been calculated from the ρ and η data. To obtain correlation coefficients and standard deviations V_m^E, $\Delta\eta$ and $\Delta G^{\#E}$ were fitted to Redlich–Kister type function in terms of mole fraction. In the whole range of composition, V_m^E were found to be positive for 1-pentanol + ethylbenzene and + propylbenzene, but 1-pentanol + toluene system showed negative V_m^E in the alcohol-rich region for. The $\Delta\eta$ and $\Delta G^{\#E}$ values were the negative for all the binary mixtures.

Keywords: Density, Viscosity, 1-pentanol, Aromatic hydrocarbon

1. Introduction:

In general, thermodynamic and transport properties are adequately employed in understanding the nature of molecular systems and physico-chemical behavior of liquid-liquid mixtures. In recent years several studies for alcohol + hydrocarbon (P S Nikam et al., 2000; M A Chowdhury et al., 2001; G Ouyang et al., 2003; S L Oswal et al., 2005) have been made on these properties of binary liquid mixtures in addition to their volumetric properties. Due to the presence of both hydrophilic and hydrophobic groups, alcohols are interesting polar solvents, self-associated through hydrogen bonding creating multimers of different degrees. And so, the dipolar association of alcohols decreases when they are mixed with aromatic hydrocarbons, due to some sort of specific intermolecular interactions between alcohols and aromatic hydrocarbons (S C Bhatia, R Rani and R Bhatia, 2011). Literature survey further revealed that although many attempts have already been made to study the binary mixtures of alcohols and aromatic hydrocarbons, systematic studies focusing their dependence on composition and number position as well as length of substituent on aromatic hydrocarbons are still scarce. In this paper we have reported on densities, excess molar volumes, viscosities, deviations in viscosity, interaction parameter and excess free energy of activation of the binary mixtures of 1-pentanol + toluene, 1-pentanol + ethylbenzene and 1-pentanol + propylbenzene systems.

2. Materials and Methods:

1-Pentanol (PL): Aldrich, mole fraction purity > 0.99%, toluene (TN): Merck, mole fraction purity > 0.998, ethylbenzene (EB): Merck, mole fraction purity > 0.996 and propylbenzene (PB): Aldrich, mole fraction purity > 0.997, were kept over molecular sieves for 2-3 weeks and binary mixtures of various compositions were prepared by mass mixing pure components at different proportions with the help of an electronic balance (College B 204-S, METTLER TOLEDO) accurate up to ±0.0001 g. Densities, ρ were measured by using a 10 cm^3 bi-capillary pycnometer and viscosity, η by an Ostwald viscometer (Technico, PSL BS/U, A-type) both being calibrated previously with twice distilled water. The flow time of liquids was recorded by an electronic stopwatch reading up to ±0.01 s. For all measurements a thermostatic water bath (Thermo Haake) controlled up to ±0.01 K was used. The mole fraction was estimated accurate up to 10^{-4}, while the uncertainty in measured ρ and η were ±1.3×10^{-4} g.cm^{-3} and ±1.8×10^{-4} mPa.s respectively. The purities were further checked by comparing measured ρ and η of pure liquids with the literature data as tabulated in Table 1.

Table 1. Comparison of experimental densities (ρ /g.cm^{-3}) and viscosities (η /mPa.s) of pure liquidswith literature values at 298.15 K.

Sample	ρ /g.cm^{-3}		η /mPa.s	
	This work	Literature	This work	Literature
1-Pentanol	0.8115	0.81103[a]	3.2698	3.37[b]
Toluene	0.8621	0.8622[c]	0.5446	0.552[d]
Ethylbenzene	0.8625	0.862433[e]	0.6206	0.635[f]
Propylbenzene	0.8577	0.85776[g]	0.7827	0.7995[h]

[a]Ref-[6], [b]Ref-[7], [c]Ref-[8], [d]Ref-[9], [e]Ref-[10], [f]Ref-[5], [g]Ref-[11], [h]Extrapolated value of Ref-[12]

3. Results and Discussion:
3.1 Excess molar volumes
Excess molar volumes, V_m^E, for a solution is expressed as (M Page, J Huot and C Jolicoeur, 1993):

$$V_m^E = V_m - x_1 V_1^* - x_2 V_2^* \qquad (1)$$

where, V_m is the molar volume of the mixture, V_1^* and V_2^* are the molar volumes and x_1 and x_2 are the mole fractions of the pure components 1 and 2, respectively. V_m^E of the binary mixtures were calculated from experimental densities of pure liquids and their mixtures by using the equation (M A Tejraj, 1999):

$$V_m^E = \left[\frac{x_1 M_1 + x_2 M_2}{\rho} \right] - \left[\frac{x_1 M_1}{\rho_1} + \frac{x_2 M_2}{\rho_2} \right] \qquad (2)$$

Here, ρ is the density of the mixture, M_1 and M_2 represent the molar masses and ρ_1 and ρ_2 are the densities of components 1 and 2, respectively. V_m^E valuesfor the systems of PL + TN, PL + EB and PL + PB are listed in Table 2. Fig. 1represents the comparative diagram for V_m^E of the three systems against mole fraction of aromatic hydrocarbons, x_2 at 298.15 K. Each set of results were fitted by the Redlich-Kister equation (O J. Redlich and A T. Kister, 1948):

$$Y^E = \left[(1 - x_2) x_2 \sum_{i=0}^{n} A_i (1 - 2x_2)^i \right] \qquad (3)$$

The standard deviation, σ, followed the equation:

$$\sigma = \left[\frac{\sum \left(Y_{\exp} - Y_{cal} \right)^2}{n - p - 2} \right]^{\frac{1}{2}} \qquad (4)$$

where, $Y^E \equiv V_m^E$ and A_i is the i-th polynomial fitting coefficient, $Y_{\exp}$ and Y_{cal} are the experimental and calculated of properties, n the total number of

compositions for a particular system and p is the number of coefficients. In the whole range of composition V_m^E are found to be positive for PL + EB and PL + PB, whereas, only small negative values are observed in the alcohol-rich region for PL+TN system.

Table 2. Densities, ρ (g.cm^{-3}) and excess molar volumes, V_m^E (cm^3.mol^{-1}) of the systems of PL (x_1)+ TN(x_2), PL (x_1)+ EB (x_2) and PL (x_1)+ PB (x_2) for different molar ratios at 298.15 K.

x_2	ρ	V_m^E	x_2	ρ	V_m^E	x_2	ρ	V_m^E
PL (x_1)+ TN(x_2)			PL (x_1)+ EB(x_2)			PL (x_1)+ PB(x_2)		
0.0000	0.8115	0.0000	0.0000	0.8115	0.0000	0.0000	0.8115	0.0000
0.1002	0.8169	-0.0536	0.1004	0.8171	0.0170	0.1004	0.8172	0.0157
0.1995	0.8216	-0.0179	0.2008	0.8225	0.0410	0.1977	0.8224	0.0341
0.3007	0.8264	0.0188	0.3002	0.8277	0.0659	0.3007	0.8276	0.0549
0.4002	0.8311	0.0589	0.4005	0.8328	0.0926	0.3999	0.8323	0.0812
0.4997	0.8358	0.1005	0.4991	0.8377	0.1171	0.5001	0.8368	0.1087
0.6004	0.8407	0.1258	0.6001	0.8426	0.1413	0.6000	0.8411	0.1304
0.6983	0.8457	0.1220	0.6998	0.8474	0.1535	0.6999	0.8453	0.1354
0.7994	0.8511	0.0904	0.8000	0.8522	0.1527	0.8001	0.8493	0.1426
0.9018	0.8567	0.0447	0.9008	0.8572	0.1111	0.8999	0.8534	0.1016
1.0000	0.8621	0.0000	1.0000	0.8625	0.0000	1.0000	0.8577	0.0000

The above results have been explained as follows. When small amount of PL is added to aromatic hydrocarbons (TN, EB and PB) separately, it is expected that it would disperse within aromatic hydrocarbons causing depolymerization of the network structures and disrupt their H-bonding and resulting into volume expansion. So that, V_m^E become positive for all the systems. Similar type of behavior in V_m^E values has also been observed for 1-decanol + ethylbenzene(S C Bhatia, R Rani and R Bhatia, 2011; R K Dewan and S K Mehta, 1987), 1-nonanol + ethylbenzene (R Rani and S C Bhatia, 2013), 1-pentanol + benzene (N G Tsierkezos, M MPalaiologou and I E Molinou, 2000). But small negative values in the alcohol region of PL + TN system leads to suggest that "partial interstitial accommodation" of the component molecules might have occurred favorably which suppressed the dispersive forces.

Djordjevic*et. al*., 2009 [18] has established that, with the increased steric hindrance of an alkyl group in the aromatic hydrocarbons the positive V_m^E becomes more positive. But strength of hydrogen bond interaction becomes greater for PB compared to EB due to the presence of larges alkyl group and so, the V_m^E values of PL + PB system become less than that of PL + EB system.

x_2
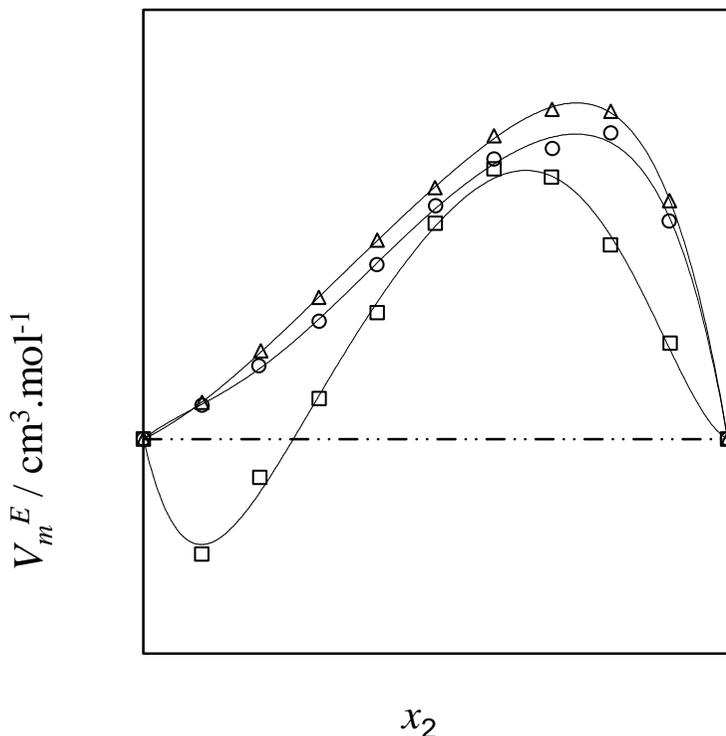
Figure 1. Comparison of excess molar volumes, V_m^E as a function of x_2 for the systems of PL (x_1)+ TN(x_2) (□), PL (x_1)+ EB (x_2) (Δ) andPL (x_1)+ PB (x_2) (o) at 298.15 K.

3.2. Partial molar volumes

The partial molar volume of the ith component for a solution, V_i, is defined as:

$$V_i = \left(\frac{\partial V_m^E}{\partial n_i} \right)_{T,P,n_j} \tag{5}$$

Considering equation (1) in (5) equations obtained for the partial molar volumes V_1 and V_2 of components 1 and 2 respectively are:

$$V_1 = V_1^* + V_m^E - x_2 \left(\frac{\partial V_m^E}{\partial x_2} \right)_{T,P} \tag{6}$$

$$V_2 = V_2^* + V_m^E + \left(1 - x_2\right) \left(\frac{\partial V_m^E}{\partial x_2} \right)_{T,P} \tag{7}$$

Following the procedure (Maham*et al*., 2002), V_1 and V_2 were obtained as:

$$V_1 = V_1^* + x_2^2 \sum_{i=0}^{n} A_i \left(1 - 2x_2\right)^i + 2x_2^2 \left(1 - x_2\right) \sum_{i=0}^{n} iA_i \left(1 - 2x_2\right)^{i-1} \tag{8}$$

$$V_2 = V_2^* + (1 - x_2)^2 \sum_{i=0}^{n} A_i (1 - 2x_2)^i - 2x_2 (1 - x_2)^2 \sum_{i=0}^{n} i A_i (1 - 2x_2)^{i-1} \quad (9)$$

The results are as listed in Table 3. The difference between partial molar volume, V_i and molar volume V_i^* of each of component was also estimated at 298.15 K and thus the values obtained for $(V_i - V_i^*)$ are graphically represented by Figs. 2 & 3. For both the components, these differences are almost positive for all systems, indicating that there is volume expansion in the systems. This is exactly in support of positive V_m^E observed experimentally.
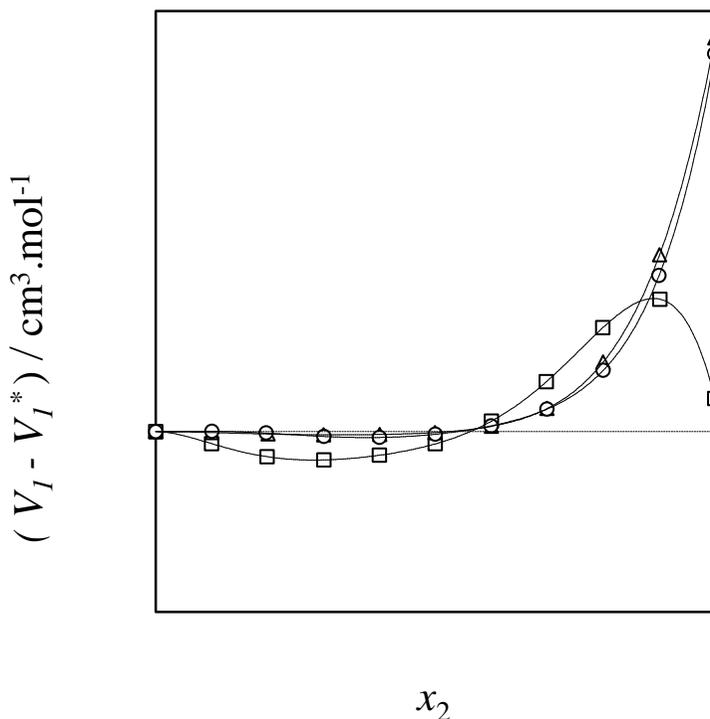
Figure 2. Plot of $(V_1 - V_1^*)$ of component 1 as a function of x_2 in the systems of PL (x_1)+ TN(x_2) (□), PL (x_1)+ EB (x_2) (Δ) andPL (x_1)+ PB (x_2) (o) at 298.15 K.

Table 3. Partial molar volumes of component 1, V_1(cm^3.mol^{-1}) and of component 2, V_2 (cm^3.mol^{-1}) in the systems of PL (x_1)+ TN(x_2), PL (x_1)+ EB (x_2) andPL (x_1)+ PB (x_2) for different molar ratios at 298.15 K.

x_2	V_1	V_2	x_2	V_1	V_2	x_2	V_1	V_2
PL (x_1)+ TN (x_2)			PL (x_1)+ EB (x_2)			PL (x_1)+ PB (x_2)		
0.0000	108.63	105.70	0.0000	108.63	123.23	0.0000	108.63	140.36
0.1002	108.58	106.84	0.1004	108.62	123.30	0.1004	108.63	140.29
0.1995	108.52	107.17	0.2008	108.62	123.34	0.1977	108.62	140.33

x_2	V_1	V_2	x_2	V_1	V_2	x_2	V_1	V_2
0.3007	108.51	107.22	0.3002	108.61	123.35	0.3007	108.61	140.37
0.4002	108.53	107.18	0.4005	108.61	123.34	0.3999	108.60	140.38
0.4997	108.58	107.13	0.4991	108.62	123.33	0.5001	108.62	140.37
0.6004	108.67	107.05	0.6001	108.65	123.31	0.6000	108.65	140.34
0.6983	108.83	106.96	0.6998	108.72	123.28	0.6999	108.72	140.30
0.7994	109.06	106.89	0.8000	108.92	123.21	0.8001	108.88	140.25
0.9018	109.18	106.86	0.9008	109.36	123.14	0.8999	109.28	140.18
1.0000	108.76	106.88	1.0000	110.26	123.10	1.0000	110.20	140.14
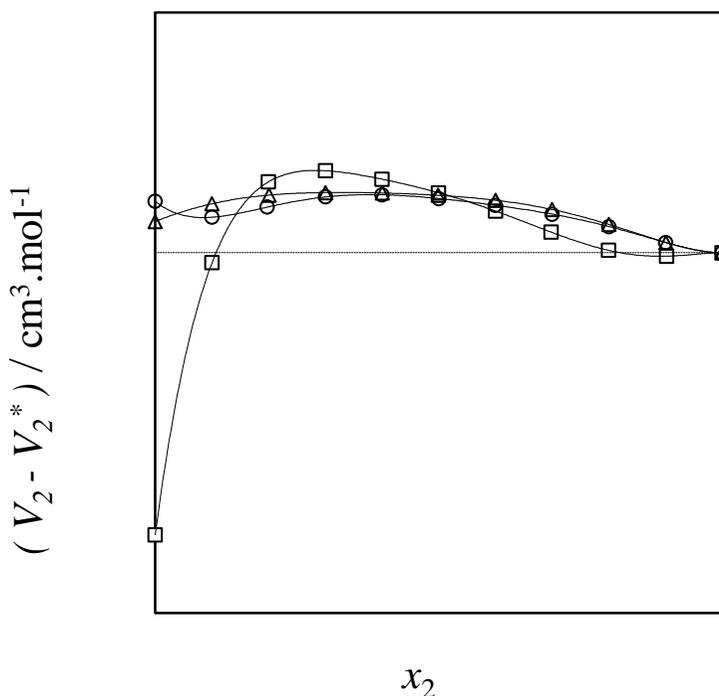
Figure 3. Plot of ($V_2 - V_2^*$) of component 2 as a function of x_2 in the systems of PL (x_1)+ TN(x_2) (□), PL (x_1)+ EB (x_2) (Δ) andPL (x_1)+ PB (x_2) (o) at 298.15 K.

3.3.Viscosities and Deviations in Viscosity

Measured viscosities, η, of PL + TN, PL + EB and PL + PB in the range 0≤ x_2 ≤ 1 composition at 298.15 K are as summarized in Table 4. η of pure liquids varies in the order: PL > PB > EB > TN. From experimental η data, deviations in viscosity, $\Delta\eta$ for the binary mixtures were calculated as follows (C Yang, P Ma and Q Zhou, 2004; G Li et al., 2013):

$$\Delta\eta = \eta - \eta_1 x_1 - \eta_2 x_2 \qquad (10)$$

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where, η is the viscosity of solution, η_1 and η_2 are viscosities of pure components 1 and 2, respectively. The estimated $\Delta\eta$ were correlated well to equation (3) and the adjustable parameters and standard deviations are as given in Table 5. Fig. 4 represents the comparative diagram of $\Delta\eta$ against x_2 at 298.15 K. For all these systems, $\Delta\eta$ are found to be negative in the whole range of composition forming a single lobe with minima at x_2 ~0.4 and their order varied as: PL + PB > + EB > + TN. Negative values of $\Delta\eta$ indicated that all the species get favorable conditions to flow. This is because addition of PL to TN/EB/PB causes mainly dispersion due to break up of weak forces. Also, aromatic hydrocarbons with longer chain lengths encounter less inconvenience to flow even through intermolecular H-bonded species may also be present in the solutions.

The Grunberg - Nissan interaction parameter (ε) (L Grunberg and A H Nissan, 1949) estimated by following the equation

$$\ln\eta = x_1 \ln\eta_1 + x_2 \ln\eta_2 + x_1 x_2 \varepsilon \qquad (11)$$

ε are as listed in Table 4. For all the systems, ε are observed to be negative. According to Fort and Moore (R J Fort and W R Moore, 1966), ε may be termed as an approximate index for estimating the strength of interaction between dissimilar components of a mixture in flow: If $\Delta\eta < 0$ and $\varepsilon < 0$ and magnitudes of both are large, specific interaction would be absent and only dispersion force may be dominant there. The negative values of ε indicates that any interaction between the alcohol and aromatic hydrocarbons is of non-specific type, which also supports their positive V_m^E behavior.

x_2
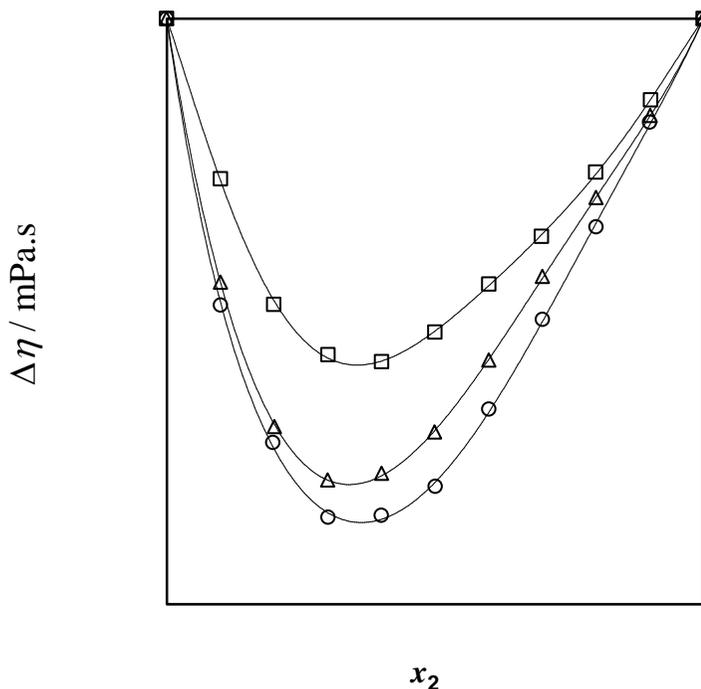
Figure 4. Comparison of deviation in viscosity, $\Delta\eta$ as a function of x_2 for the systems of PL (x_1) + TN (x_2) (□), PL (x_1)+ EB (x_2) (Δ) andPL (x_1)+ PB (x_2) (o) at 298.15 K.

Table 4. Viscosities, η (mPa.s) and interaction parameter (ε) of the systems of PL (x_1)+ TN(x_2), PL (x_1)+ EB (x_2) andPL (x_1)+ PB (x_2) for different molar ratios at 298.15 K.

x_2	η	ε	x_2	η	ε	x_2	η	ε
PL (x_1) + TN (x_2)			PL (x_1) + EB (x_2)			PL (x_1) + PB (x_2)		
0.0000	3.2698	…..	0.0000	3.2698	…..	0.0000	3.2698	…..
0.1002	2.5954	-0.4615	0.1004	2.5423	-0.7600	0.1004	2.5880	-0.8091
0.1995	2.0427	-0.4529	0.2008	1.9938	-0.6403	0.1977	2.1028	-0.6444
0.3007	1.6205	-0.3791	0.3002	1.5917	-0.5153	0.3007	1.7015	-0.5193
0.4002	1.3029	-0.3040	0.4005	1.2923	-0.3933	0.3999	1.4218	-0.3917
0.4997	1.0676	-0.2239	0.4991	1.0736	-0.2853	0.5001	1.2001	-0.2872
0.6004	0.8881	-0.1512	0.6001	0.9146	-0.1844	0.6000	1.0532	-0.1834
0.6983	0.7495	-0.0957	0.6998	0.8020	-0.1040	0.6999	0.9453	-0.1030
0.7994	0.6490	-0.0462	0.8000	0.7124	-0.0486	0.8001	0.8638	-0.0468
0.9018	0.5800	-0.0123	0.9008	0.6491	-0.0132	0.8999	0.8147	-0.0115
1.0000	0.5446	…..	1.0000	0.6206	…..	1.0000	0.7827	…..

Table 5. Fitting coefficients of polynomial equation (3) and standard deviations, σ in equation (4) for the systems of PL (x_1)+ TN(x_2), PL (x_1)+ EB (x_2) andPL (x_1)+ PB (x_2) for different molar ratios at 298.15 K.

Property	A_0	A_1	A_2	A_3	A_4	σ
PL (x_1)+ TN(x_2)						
V_m^E / cm^3 mol^{-1}	0.3973	-0.6000	-0.2592	-0.0559	-0.6570	0.0060
$\Delta \eta$ (mPa.s)	-1.0661	-0.6843	-0.4611	0.3268	0.5009	0.0028
$\Delta G^{\#E}$ / kJ mol^{-1}	-2.2009	0.6992	-0.4134	0.5926	0.4331	0.0051
PL (x_1)+ EB(x_2)						
V_m^E / cm^3 mol^{-1}	0.4710	-0.4848	0.3320	-0.2694	0.0812	0.0011
$\Delta \eta$ (mPa.s)	-1.4045	-1.0404	-0.3476	0.0848	-0.2133	0.0018
$\Delta G^{\#E}$ / kJ mol^{-1}	-2.7791	0.2466	0.5455	0.5383	-0.9125	0.0047
PL (x_1)+ PB(x_2)						
V_m^E / cm^3 mol^{-1}	0.4299	-0.4724	0.1954	-0.2068	0.2680	0.0040
$\Delta \eta$ (mPa.s)	-1.5825	-0.9462	-0.1712	-0.1669	-0.3774	0.0045
$\Delta G^{\#E}$ / kJ mol^{-1}	-2.7245	0.3287	0.5735	-0.1108	-0.5355	0.0083

3.4.Excess Free Energy

Excess free energies of activation $\Delta G^{\#E}$ for viscous flow have been calculated using the equation (S L Oswal et al., 2005; N V Sastry, S R Patel and S SSoni, 2013):

$$\Delta G^{\#E} = RT\left[\ln(\eta V_m) - \sum x_i \ln\left(\eta_i V_i^*\right)\right] \quad (12)$$

where all the terms have their usual significance. Comparative diagram of $\Delta G^{\#E}$ against x_2 at 298.15 K (Fig. 5) shows that $\Delta G^{\#E}$ are all negative in the whole range of composition. This leads to suggest that irrespective of volume expansion and contraction, all the flowing species surpass energy barriers smaller than expected from the additive rule.
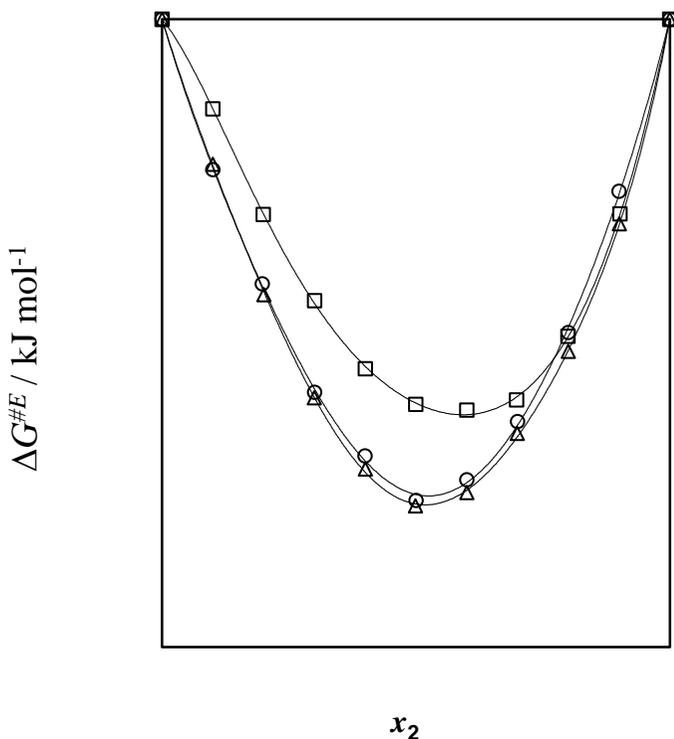
Figure 5. Comparison of excess free energies of activation for viscous flow, $\Delta G^{\#E}$ as a function of x_2 for the systems of PL (x_1)+ TN (x_2) (□), PL (x_1) + EB (x_2) (Δ) and PL (x_1) + PB (x_2) (o) at 298.15 K.

4. Conclusion

Densities and viscosities for the binary mixtures of 1-pentanol + toluene, 1-pentanol + ethylbenzene and 1-pentanol + propylbenzene systems at 298.15 K have measured. From measured ρ , V_m^E and V_i and from η , $\Delta\eta$, ε and $\Delta G^{\#E}$ were calculated. The V_m^E were found to be positive for 1-pentanol + ethylbenzene and 1-pentanol + propylbenzenein the whole range of composition, whereas, for the 1-pentanol + toluene system small negative values are observed in the alcohol-rich region which may be suggested as due to 'partial' interstitial accommodation'.

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