



Thermo-Physical Properties of Binary Mixtures of Dec + Xylenes at Different Temperatures

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ABSTRACT

Internal pressure (π_i), free volume (V_f), enthalpy (H) and entropy (T_s) have been derived from the measured values of densities (ρ) and speeds of sound (u) for binary liquid mixtures of diethylcarbonate with *o*-xylene, *m*-xylene and *p*-xylene at $T = (298.15 \text{ to } 323.15) \text{ K}$ over the entire composition range and at atmospheric pressure. By using these values, excess internal pressure (π_i^E), excess free volume (V_f^E), excess enthalpy (H^E), excess free energy (G^E) and excess entropy (T_s^E) have been calculated. The calculated excess values have been fitted to the Redlich-Kister polynomial equations and the results are analysed in terms of molecular interactions due to physical, chemical and structural effects between the unlike molecules.

Keywords: Thermophysical properties; Internal pressure; Excess Gibbs energy of activation; Molecular interactions; Diethylcarbonate; Xylene

INTRODUCTION

Over the years the thermodynamic functions such as internal pressure, free volume, enthalpy and entropy have gained significant interest from chemists, physicists and chemical engineers, as they provide a measure of explaining various phenomenon like molecular interaction, clustering, internal structure, ionic and dipolar interactions¹ and hence have been employed subsequently to investigate the intermolecular interactions in binary mixtures. Excess enthalpies data of liquid mixtures are required for many engineering applications such as heat exchanger design, process design and simulation^{2,3}. The sign, magnitude and symmetry of excess molar enthalpies are a cumulative effect of bond breaking and rearranging of the constituent molecules during the mixing process. Rate process in liquids is determined by the free volume, V_f . So, V_f plays an important role in ultrasonic wave propagation. Any effect arising due to molecular interaction is directly reflected in excess values.

We have previously reported the excess molar volumes, excess isentropic compressibilities, excess partial molar volume, and excess partial molar compressibilities in binary mixtures of Diethylcarbonate (DEC) with xylenes⁴. In this work we calculate the excess internal pressure (π_i^E), excess free volume (V_f^E), excess enthalpy (H^E), excess free energy (G^E) and excess entropy (T_s^E) at temperatures (298.15, 303.15, 308.15, 313.15, 318.15 and 323.15) K. This work forms part of an investigation into the thermophysical properties of DEC and its mixtures. As far as we know, no measurements on above parameters have been reported for the mixtures investigated in this work. The study of propagation of ultrasonic waves in liquids and liquid mixtures is of great importance for examining the nature of intermolecular and intramolecular interactions in binary systems⁵.

Diethyl carbonate (DEC) is an important solvent, which has been used in a variety of liquid extraction processes, especially in the synthesis of pharmaceuticals and agricultural chemicals^{6,7}. This compound is also a good gasoline additive⁸, in place of methyl tert-butyl ether (MTBE), to prevent the crack of abnormal burning and promote the blending octane values. Moreover, mixtures of DEC with aromatic possess certain properties, for example, miscible with gasoline, stable in storage and allowable by the emission restriction of the clean air legislation to benefit specific applications.

From the experimental values of density and speed of sound the internal pressure (π_i), free volume (V_f), enthalpy (H), entropy (Ts), excess internal pressure (π_i^E), excess free volume (V_f^E), excess enthalpy (H^E), excess free energy (G^E) and excess entropy (T_s^E) at temperatures (303.15 to 323.15) K over the entire composition range have been evaluated. The results are fitted to the Redlich-Kister polynomial equation.

THEORY

The internal pressure of a fluid is related to the thermal pressure coefficient $(\partial P/\partial T)_V$ by the following well-known thermodynamic equation of state,

$$\pi_i = \left(\frac{\partial E}{\partial V}\right)_T = T\left(\frac{\partial P}{\partial T}\right)_V - P = T\left(\frac{\alpha_p}{k_T}\right) - P \quad \text{--- (1)}$$

Where α_p is the isobaric expansivity and k_T is the isothermal compressibility of the mixture. For most of the liquids, the thermal pressure coefficient multiplied by absolute temperature, i.e., $T(\alpha_p/k_T)$ is very high so that the external pressure (P) becomes negligible in comparison¹⁰, therefore it may be neglected in Eq.(1) in the present calculations. Thus, the internal pressure can be shown to be equal to the following relationship^{9,10}.

$$\pi_i = \frac{\alpha_p T}{k_T} \quad \text{---- (2)}$$

The free volume, V_f of the mixtures are calculated from the relation^{9,10}

$$V_f = \frac{RT}{(P + \pi_i)} \quad \text{---- (3)}$$

Since P is very small as compared to π_i , it has been neglected in the Eq.(3) in the present calculations. k_T is calculated using the well-known thermodynamic relationship^{11,12},

$$k_T = k_s + \frac{TV\alpha_p^2}{C_p} \quad \text{--- (4)}$$

Where k_s [$=1/(\rho u^2)$] is isentropic compressibility, V is the molar volume and C_p is the heat capacity of the mixture. The α_p values for the mixtures were evaluated from temperature dependence of density data⁴. The C_p values of pure liquids have been taken from the literature^{13,14} and the C_p values for the mixtures have been calculated by using the following relationship.

$$C_p = x_1 C_{p,1} + x_2 C_{p,2} \quad \text{--- (5)}$$

The values of various parameters, C_p , α_p , k_s and k_T of pure liquids used in the calculations are taken from our earlier paper⁴. The π_i^E and V_f^E of binary mixtures have been calculated using the relationship,

$$Y^E = Y - (x_1 Y_1 + x_2 Y_2) \quad \text{--- (6)}$$

Where Y is π_i or V_f and subscripts 1 and 2 refers to pure DEC and xylenes respectively. The excess enthalpies (H^E) and excess entropies (T_s^E) are calculated from π_i and V_f by using the following relations based on regular solution theory^{15,16,17}.

$$-H^E = \pi_i V - [x_1 \pi_{i,1} V_1 + x_2 \pi_{i,2} V_2] \quad \text{--- (7)}$$

$$T_s^E = R [x_1 \ln V_{f,1} + x_2 \ln V_{f,2} - \ln V_f] \quad \text{--- (8)}$$

The excess free energy (G^E) of mixtures is given by the relationship,

$$G^E = H^E - T_s^E \quad \text{--- (9)}$$

The values of π_i^E , V_f^E , H^E , T_s^E and G^E were fitted to a Redlich-Kister¹⁸ type polynomial equation

$$Y^E = x_1 x_2 \sum_{i=0}^j A_i (1 - 2x_1)^i \quad \text{--- (10)}$$

Where Y^E is π_i^E or V_f^E or H^E or T_s^E or G^E . The values of A_i are the coefficients of the polynomial equation and the corresponding standard deviations, $\sigma(Y^E)$ obtained by the method of least – squares with equal weights assigned to each point are calculated. The standard deviation $\sigma(Y^E)$ is defined as follows:

$$\sigma(Y^E) = \left[\frac{\sum (Y_{\text{obs}}^E - Y_{\text{cal}}^E)^2}{(n-m)} \right]^{1/2} \quad \text{--- (11)}$$

Where n is the total number of experimental points and m is the number of coefficients.

RESULTS AND DISCUSSION

The values of coefficients, A_i of Eq.(10) for the excess functions and the corresponding standard deviations, σ are listed in Table 1. The variation of π_i^E , V_f^E , H^E , T_s^E and G^E with composition for the three systems at temperature 298.15K are presented graphically in Figures 1 - 5, respectively.

Table 1: Coefficients (A_i) of Eq. (10) for π_i^E , V_f^E , H^E , T_s^E and G^E along with standard deviations (σ) for DEC + o-/m-/p-xylene binary mixtures at different temperatures

Property	T(K)	A_1	A_2	A_3	A_4	A_5	σ
DEC + o-xylene							
$\pi_i^E / 10^8 \text{ Nm}^{-2}$	298.15	0.0404	0.0001	0.0010	0.0003	0.0013	3.55E-05
	303.15	0.0372	-0.0020	0.0015	-0.0004	0.0000	2.11E-05
	308.15	0.0332	-0.0029	0.0017	-0.0004	0.0001	2.04E-05
	313.15	0.0291	-0.0044	0.0024	0.0014	-0.0009	2.98E-05
	318.15	0.0244	-0.0041	0.0035	0.0002	-0.0030	2.39E-05
$V_f^E / 10^{-6} \text{ m}^3 \text{ mol}^{-1}$	298.15	-7.8537	-0.0951	-0.3141	-0.0234	0.0109	4.12E-05
	303.15	-7.5099	0.3582	-0.3607	-0.0211	0.0076	2.63E-05
	308.15	-6.9842	0.7202	-0.4057	-0.0214	0.0082	5.56E-05
	313.15	-6.3949	0.9205	-0.4412	-0.0206	0.0076	2.74E-05
	318.15	-5.6369	0.9519	-0.4669	-0.0190	0.0072	2.89E-05
$H^E / \text{kJ mol}^{-1}$	298.15	-0.4496	-0.0861	-0.0001	-0.0007	-0.0010	3.55E-05
	303.15	-0.4199	-0.0631	-0.0044	-0.0017	0.0033	1.30E-05
	308.15	-0.3809	-0.0478	-0.0037	0.0003	-0.0008	2.25E-05
	313.15	-0.3401	-0.0406	-0.0057	-0.0002	0.0022	2.57E-05
	318.15	-0.2916	-0.0425	-0.0052	-0.0020	-0.0004	9.66E-06
$T_s^E / 10^{-2} \text{ J mol}^{-1}$	298.15	-0.0281	-0.0009	-0.0011	0.0014	0.0000	9.28E-06
	303.15	-0.0264	0.0013	-0.0013	-0.0008	0.0001	5.97E-06
	308.15	-0.0244	0.0024	-0.0020	-0.0004	0.0020	1.20E-05
	313.15	-0.0220	0.0033	-0.0007	-0.0004	-0.0016	3.75E-05
	318.15	-0.0191	0.0028	-0.0015	0.0004	-0.0001	3.20E-05
$G^E / \text{kJ mol}^{-1}$	298.15	-0.0157	0.0027	-0.0018	0.0002	0.0005	2.42E-05
	303.15	-0.4215	-0.0863	0.0002	0.0007	0.0015	2.38E-05
	308.15	-0.3934	-0.0653	-0.0026	0.0007	0.0019	1.78E-05
	313.15	-0.3564	-0.0501	-0.0037	-0.0007	0.0012	1.63E-05
	318.15	-0.3180	-0.0443	-0.0026	0.0008	-0.0021	1.08E-05
DEC + m-xylene	298.15	-0.2725	-0.0461	-0.0036	-0.0008	-0.0005	3.57E-05
	303.15	-0.2218	-0.0578	-0.0045	-0.0003	0.0012	6.44E-06
	298.15	0.0747	-0.0048	-0.0022	0.0063	-0.0038	3.15E-05
	303.15	0.0802	-0.0053	-0.0016	0.0058	-0.0045	6.37E-05
	308.15	0.0831	-0.0055	0.0018	0.0062	-0.0090	3.46E-05
$V_f^E / 10^{-6} \text{ m}^3 \text{ mol}^{-1}$	313.15	0.0846	-0.0052	0.0004	0.0055	-0.0074	5.45E-05
	318.15	0.0831	-0.0046	0.0000	0.0050	-0.0061	2.80E-05
	323.15	0.0851	-0.0055	0.0006	0.0069	-0.0081	3.72E-05
	298.15	-20.6862	-0.3326	-0.1274	-1.0937	1.5164	0.0069
	303.15	-22.4110	-0.2392	-0.1498	-1.0459	1.4516	0.0066
$H^E / \text{kJ mol}^{-1}$	308.15	-23.7299	-0.1809	-0.1685	-1.1109	1.5478	0.0070
	313.15	-24.7678	-0.1448	-0.1806	-1.1499	1.6005	0.0073
	318.15	-25.1696	-0.1198	-0.1854	-1.2130	1.6885	0.0077
	323.15	-25.3299	-0.1827	-0.1908	-1.1993	1.6720	0.0076
	298.15	-1.4826	0.0428	-0.0004	0.0014	0.0036	3.06E-05
$T_s^E / 10^{-2} \text{ J mol}^{-1}$	303.15	-1.5576	0.0475	0.0007	0.0003	0.0010	2.84E-05
	308.15	-1.6042	0.0494	-0.0005	-0.0002	0.0019	2.90E-05
	313.15	-1.6279	0.0490	0.0006	0.0005	0.0004	5.20E-05
	318.15	-1.6155	0.0458	-0.0004	0.0004	0.0028	3.82E-05
	323.15	-1.6837	0.0489	0.0010	-0.0020	0.0028	4.26E-05
$G^E / \text{kJ mol}^{-1}$	298.15	-0.0680	0.0006	0.0000	-0.0036	0.0061	4.00E-05
	303.15	-0.0736	0.0010	-0.0002	-0.0029	0.0054	1.88E-05
	308.15	-0.0776	0.0010	0.0001	-0.0033	0.0050	2.78E-05
	313.15	-0.0805	0.0016	0.0000	-0.0043	0.0053	5.57E-05
	318.15	-0.0809	0.0012	0.0002	-0.0032	0.0049	1.93E-05
DEC + p-xylene	323.15	-0.0833	0.0016	0.0006	-0.0044	0.0034	3.46E-05
	298.15	-1.4145	0.0421	-0.0004	0.0050	-0.0025	2.70E-05
	303.15	-1.4840	0.0462	0.0001	0.0045	-0.0020	1.20E-05
	308.15	-1.5267	0.0482	0.0007	0.0023	-0.0035	2.73E-05
	313.15	-1.5476	0.0478	0.0012	0.0035	-0.0043	3.18E-05
$\pi_i^E / 10^8 \text{ Nm}^{-2}$	318.15	-1.5346	0.0445	0.0015	0.0046	-0.0041	1.72E-05
	323.15	-1.6003	0.0478	0.0006	0.0017	-0.0007	3.34E-05
	298.15	0.0837	-0.0071	0.0055	0.0083	-0.0031	0.0004
	303.15	0.0886	-0.0082	0.0042	0.0097	-0.0005	0.0004
$\pi_i^E / 10^8 \text{ Nm}^{-2}$	308.15	0.0911	-0.0083	0.0033	0.0089	0.0004	0.0004
	313.15	0.0915	-0.0092	0.0053	0.0100	-0.0025	0.0004
	318.15	0.0891	-0.0087	0.0045	0.0090	-0.0011	0.0004

	323.15	0.0846	-0.0086	0.0059	0.0088	-0.0030	0.0004
$V_f^E / 10^{-6} \text{ m}^3 \text{ mol}^{-1}$	298.15	-21.1531	0.3602	-0.9498	-1.9141	0.0219	0.0775
	303.15	-22.8588	0.5616	-1.0140	-2.0613	0.0680	0.0824
	308.15	-24.1435	0.6739	-1.1674	-2.0568	0.1434	0.0865
	313.15	-25.1272	0.7952	-1.2484	-2.1328	0.1550	0.0918
	318.15	-25.4769	0.9240	-1.3352	-2.2629	0.2095	0.0969
	323.15	-25.2191	1.0343	-1.3866	-2.3958	0.1745	0.1031
$H^E / \text{kJ mol}^{-1}$	298.15	-1.1522	0.1255	-0.1006	-0.1430	0.0431	0.0045
	303.15	-1.2261	0.1306	-0.1021	-0.1430	0.0438	0.0046
	308.15	-1.2706	0.1340	-0.1047	-0.1455	0.0467	0.0046
	313.15	-1.2917	0.1346	-0.1058	-0.1455	0.0470	0.0046
	318.15	-1.2768	0.1338	-0.1061	-0.1484	0.0462	0.0047
	323.15	-1.2309	0.1299	-0.1052	-0.1498	0.0453	0.0047
$T_s^E / 10^{-2} \text{ J mol}^{-1}$	298.15	-0.0710	0.0028	-0.0016	-0.0056	-0.0015	0.0003
	303.15	-0.0761	0.0038	-0.0037	-0.0074	0.0010	0.0003
	308.15	-0.0798	0.0038	-0.0052	-0.0062	0.0027	0.0003
	313.15	-0.0823	0.0041	-0.0037	-0.0060	-0.0004	0.0003
	318.15	-0.0826	0.0050	-0.0039	-0.0074	0.0004	0.0003
	323.15	-0.0810	0.0052	-0.0040	-0.0086	0.0010	0.0003
$G^E / \text{kJ mol}^{-1}$	298.15	-1.0817	0.1223	-0.0972	-0.1367	0.0429	0.0042
	303.15	-1.1499	0.1267	-0.1005	-0.1374	0.0461	0.0043
	308.15	-1.1909	0.1298	-0.0989	-0.1385	0.0433	0.0043
	313.15	-1.2094	0.1301	-0.1010	-0.1388	0.0454	0.0043
	318.15	-1.1944	0.1285	-0.1015	-0.1398	0.0461	0.0044
	323.15	-1.1500	0.1247	-0.1012	-0.1412	0.0443	0.0044

The internal pressure is an interesting and valuable quantity that describes the macroscopic result of molecular interactions. The significance of the internal pressure has been discussed in a number of review articles^{19,20}. Figure 1 shows the excess internal pressure, π_i^E for the three binary mixtures over the entire composition range and at temperature 298.15K. The π_i^E values are observed to be positive for all the three mixtures. It has been established that the sign and magnitude of excess functions give good estimate of the strength of the unlike interactions in a binary mixture. A closer packing of molecules, resulting in a contraction in volume of the mixture, leading to an increase in the internal pressure of the mixture, hence, positive π_i^E values. The magnitude of values at equal-molar concentration are in the order: p-xylene (0.0213) > m-xylene (0.0187) > o-xylene (0.0100), which indicates the order of interactions in these systems.

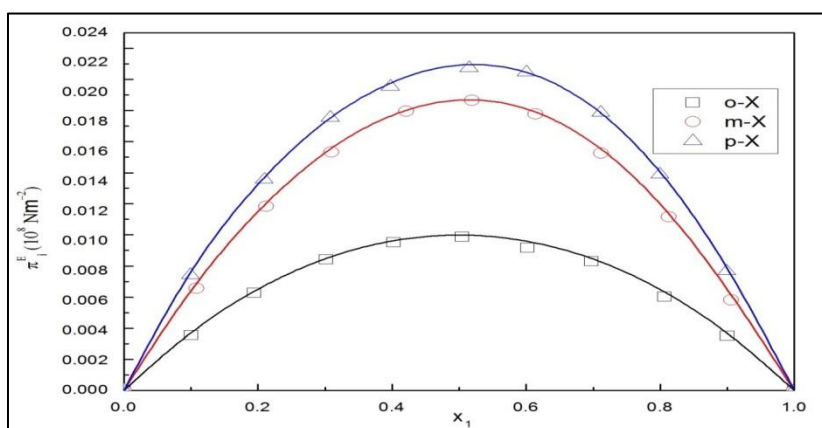


Figure 1: Variation of excess internal pressure, π_i^E with mole fraction of DEC for the three binary mixture DEC + o-xylene / + m-xylene / + p-xylene at 298.15K. Solid lines have been drawn from equation (10) using the coefficients given in Table 1

Free volume is a central concept in considering both equilibrium thermodynamic properties and transport properties in liquids. The V_f^E values are observed [Figure 2] to be negative for the three binary mixtures at temperature 298.15 K. The same trend is observed at all the studied temperatures. The negative values of V_f^E are in the order: o-xylene < m-xylene < p-xylene. The minimum value is observed at equal molar region in all the three systems. The observed negative trend in V_f^E values indicates specific interactions between the molecules of the mixtures. The sign of the V_f^E depends on the relative strength between the contractive forces and expansive forces. The factors responsible for volume contraction are (a) specific interactions between the component molecules and (b) weak physical forces, such as dipole-dipole or dipole-induced dipole interactions or Vanderwaal's forces. The factors that cause expansion in volume are dispersive forces, steric hindrance of component molecules, unfavorable geometric fitting and electrostatic repulsion. The negative values of excess

free volume in binary system assert that the combined effects of the factors are responsible for volume contraction and vice-versa²¹. According to these investigations, the negative values of excess free volume may be attributed to the packing effect and ion-dipole interaction of solvent molecules with the DEC in these mixtures^{22,23}.

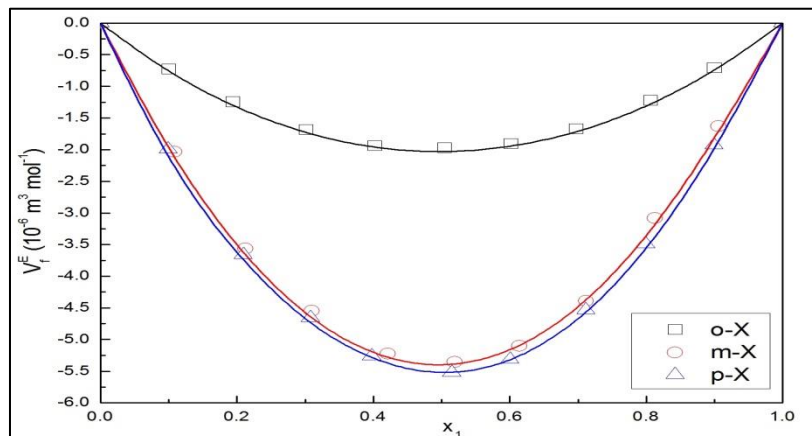


Figure 2: Variation of excess free volume, V_r^E with mole fraction of DEC for the three binary mixture DEC + o-xylene / + m-xylene / + p-xylene at 298.15K. Solid lines have been drawn from equation (10) using the coefficients given in Table 1

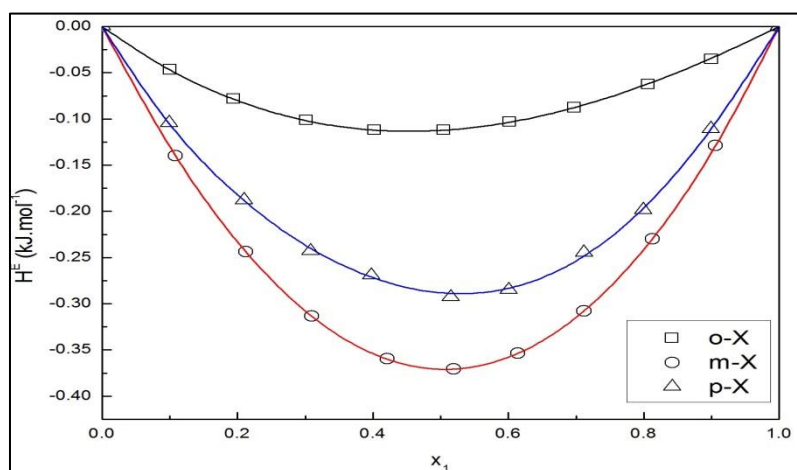


Figure 3: Variation of excess enthalpy, H^E with mole fraction of DEC for the three binary mixture DEC + o-xylene / + m-xylene / + p-xylene at 298.15K. Solid lines have been drawn from equation (10) using the coefficients given in Table 1.

Enthalpy is a thermodynamic quantity equivalent to the total heat content of a system. It is equal to the internal energy of the system plus the product of pressure and volume. The variation of H^E with mole fraction of DEC for all the three systems at temperature 298.15K is illustrated in Figure 3, which shows that the excess molar enthalpies of three binary mixtures are negative over the entire composition range. In general, H^E values of mixtures depend upon the relative enthalpies of endothermic and exothermic effects that arise on mixing of the components. The factors that cause endothermic effect on mixing are:

- strong specific interactions, usually some type of chemical interactions,
- favourable geometrical fitting of component molecules due to occupation of void spaces of one component by the other when the molecular sizes of the unlike molecules differ by a large magnitude, and,
- ion-dipole interactions of solvent molecules with the DEC molecules.

The factors that cause exothermic effect on mixing of the components are:

- breakdown of the solvent self-associated molecules from each other,
- breakdown of the DEC ion pairs, and,
- formation of weaker DEC-solvent bonds than DEC-DEC and solvent-solvent bonds.

The negative H^E values for these mixtures are attributed to the fact that the chemical forces rather than the physical forces are the dominant interactions between the dissimilar molecules²⁴. Moreover, the excess enthalpies varying with the mole fraction of DEC are nearly symmetric, indicating that the minimum H^E values

occur at around the equal-molar composition. The negative H^E values for these three systems follow the order of: m-xylene ($-0.3705 \text{ kJ mol}^{-1}$) > p-xylene (-0.2928) > o-xylene (-0.1119).

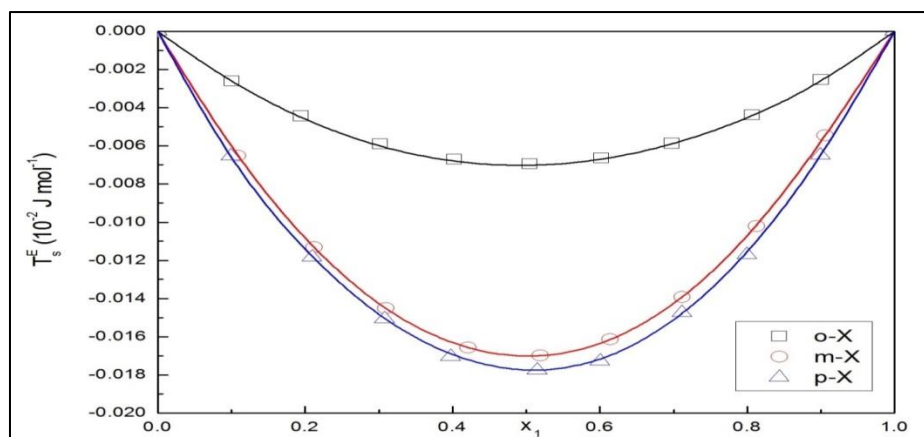


Figure 4: Variation of excess entropy, T_s^E with mole fraction of DEC for the three binary mixture DEC + o-xylene / + m-xylene / + p-xylene at 298.15K. Solid lines have been drawn from equation (10) using the coefficients given in Table 1

From Figure 4, the T_s^E values are observed to be negative for the three binary systems at temperature 298.15K. The negative T_s^E values of the mixtures further support the conclusions drawn from the π_i^E and V_f^E values, that the interaction between DEC ions and xylene molecules leads to closer packing of molecules, resulting in a contraction in volume. This leads to a decrease in the entropy of the mixture resulting in negative T_s^E values²⁵. Gibb's energy generally described this "available energy" as the greatest amount of mechanical work which can be obtained from a given quantity of a certain substance in a given initial state, without increasing its total volume or allowing heat to pass to or from external bodies, except such as at the close of the processes are left in their initial condition. The variation of excess free energy with mole fraction of DEC is shown in Figure 5. It is observed that G^E is negative for the three binary mixtures at temperatures 298.15K. A decrease in free energy favors the formation of intermediate complex called hemi-acetals followed by products from the reacting mixtures. The G^E values also found to decrease with a rise in temperature from 298.15 to 323.15 K with an interval of 5 K indicating the weakening of intermolecular interactions at elevated temperatures. The values of excess free energy are presented in Table S5. The negative values of G^E may be attributed to effective geometrical packing effect. The negative values of G^E are in the order of m-xylene > p-xylene > o-xylene.

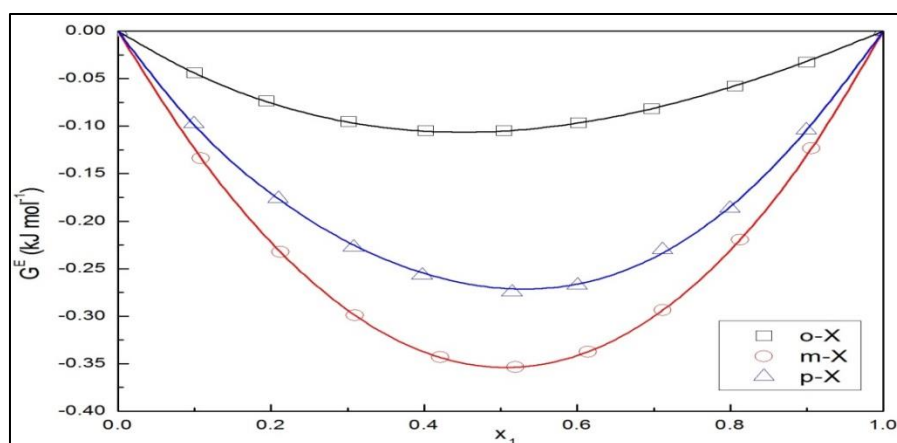


Figure 5: Variation of excess free energy, G^E with mole fraction of DEC for the three binary mixture DEC + o-xylene / + m-xylene / + p-xylene at 298.15K. Solid lines have been drawn from equation (10) using the coefficients given in Table 1

CONCLUSIONS

In this paper, values of the excess internal pressure (π_i^E), excess free volume (V_f^E), excess enthalpy (H^E), excess free energy (G^E) and excess entropy (T_s^E) were presented. These values were obtained from experimentally measured values of densities and speeds of sound. Values of V_f^E , H^E , G^E and T_s^E are negative and becomes less

negative with increase in temperature. The overall negative behaviour of V_f^E , H^E , G^E and T_s^E may be attributed to strong ion-ion/dipole-dipole interaction and easy accommodation of xylenes in the voids of DEC molecule.

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