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THERMODYNAMIC STUDY OF BINARY MIXTURES OF P-CHLOROACETOPHENONE WITH N-ALKANES

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Abstract

The density (ρ), viscosity (η) and speeds of sound (u) of pure components and their binary liquids mixtures of p-chloroacetophenone (PCA) with n-pentane (PEN), n-hexane (HEX) and n-heptane (HEP) had been reported at T = 308.15 K over the whole range of composition under atmospheric pressure. Experimental data used to compute the excess molar volume (V_m^E), deviation in viscosity ($\Delta\eta$) and deviation in isentropic compressibility ($\Delta\kappa_s$), and these parameters interrelated to Redlich-Kister (R-K) equation using multi parametric non-linear regression analysis to evaluate the binary coefficients and standard deviations. Furthermore, excess Gibbs free energy of activation of viscous flow (G^{*E}) and Grunberg-Nissan interaction parameter (d') were also computed from the experimental data. These derived parameters used to understand the type of intermolecular forces presented between unlike molecules of investigated binary mixtures.

Keywords: Binary mixtures; p-chloroacetophenone; Redlich-Kister equation; intermolecular forces.

INTRODUCTION

The investigation of physical properties such as density (ρ), viscosity (η) and ultrasonic velocity (u) of solvents at different temperatures has an important role in the industry applications. They help to understanding the type of intermolecular interactions are present in the various solvent mixtures [1, 2]. The excess parameters were calculated from physical properties of solutions and which are mostly using in design calculations, mass transfer and fluid flow [3]. Volumetric and transport properties of solutions are compulsory in most of the engineering calculations [4]. The solution properties are used in process development, pollution control, etc. Commonly, on adding of two components into a mixture may result in deviation from the ideal axis, based on this

phenomenon we can determine the kind of intermolecular interactions in liquid mixtures [5, 6]. We are involved in the systematic examination of intermolecular interactions between dissimilar molecules [7-15].

The current work, study the physical properties of binary liquid systems of PCA with *n*-alkanes such as PEN, HEX and HEP at 308.15 K. The n-alkanes are non-polar organic liquids and these are used in chemical processes [16-18]. Earlier, several researchers' works on *n*-alkanes with various organic components of binary liquid mixtures at various temperatures and given in the literature [19-21]. The new data of density (ρ) viscosity (η) and speed of sound (u) for the binary liquid mixtures of PCA with PEN, HEX and HEP at T = 308.15 K and atmospheric pressure were described. Based on measured data, V_m^E, $\Delta\eta$ and $\Delta\kappa_s$ values were calculated and correlated to R-K equation and addition, G^{*E} and d' values were counted.

EXPERIMENTAL AND PROCEDURE

The components used in this study were PEN, HEX and HEP {mass fraction (0.99), purchased from Sigma–Aldrich USA}. PCA {mass fraction (0.97)} was procured from Sigma–Aldrich USA and further purified by the methods given in the literature [22]. The details of liquids were listed in Table 1. The density (ρ), viscosity (η) and speed of sound (u) of pure components with the corresponding published values at T = 298.15 K were stated in Table 2 [23-32].

The required samples of PCA with PEN, HEX and HEP were prepared by mass basis in airtight bottles. The mass of samples were determined using Dhona (100DS) India, with a precision of ($0.01\text{x}10^{-6}$ kg) and the uncertainty in mole fraction was predicted $\pm 1\text{x}10^{-4}$. Densities of studied liquid mixtures were determined by using a pycnometer of 25 cm^3 and calibrated with redistilled water [33].

Viscosity measurements were determined by using a Ubbelohde viscometer (0.8 - 0.9 mm) and it was calibrated with redistilled water and chromatographic grade ethanol [34]. The values of viscosity of liquid components were calculated by observing their flow times on applying the following equation [35]:

$$\eta = \rho\left(at - \frac{b}{t}\right) \qquad (1)$$

where ρ' is the density and t' represents the flow time of liquids/liquid mixtures; a, b are the characteristic constants of the viscometer.

The speed of sound were taken for liquid systems utilizing an ultrasonic interferometer [model M-82: Mittal Enterprise: India], working at a frequency of 2 MHz with a measuring cell [36]. The experimental values of density (ρ), viscosity (η) and speed of sound (u) were registered in Table 3 at temperature 308.15 K. The standard uncertainties in density, viscosity and speed of sound were ±0.020 $g.\,cm^{-3}$, ±0.028 m^2/s and ±0.5 $m.s^{-1}$.

A thermostatic water bath was used to maintain required temperature stability within ±0.01 K (INSREF model; IRI-016C, India).

THEORY AND DATA EVALUATION

The V_m^E values of PCA (1) + PEN (2), + HEX (2), + HEP (2) binary systems were calculated using equation 2 [37]:

$$V_m^E = \frac{\sum_{i=1}^{2}(x_i M_i)}{\rho_m} - \sum_{i=1}^{2}\frac{x_i M_i}{\rho_i} \qquad (2)$$

Where ρ_m is the density of the mixture, x_i, ρ_i and M_i represents the mole fraction, density and relative molar mass of pure components. The obtained V_m^E values were noted in Table 3 and fitted in Fig.1.

The $\Delta\eta$ data of above said binary solutions had been counted by the following relation [38]:

$$\Delta\eta = \eta_m - \sum_{i=1}^{2}(x_i\eta_i) \quad (3)$$

Where η_m and η_i indicates the viscosity of the liquid mixture, pure solvents. The $\Delta\eta$ data were tabulated in Table 3 and graphically shown in Fig.2.

The $\Delta\kappa_s$ values of like and unlike studied binaries were obtained according to the following equation 4, and using the measured ρ and u data [39]:

$$\Delta\kappa_s = \frac{1}{\rho u^2} - \sum_{i=1}^{2}\frac{x_i}{\rho_i u_i^2} \quad (4)$$

Where u and u_i designates the speed of sound of pure substances and liquid mixtures, respectively. The $\Delta\kappa_s$ values were listed in Table 3 and depicted in Fig.3.

The G^{*E} and d' values were computed by the following equations [35, 40]:

$$G^{*E} = RT[\ln(V_m\eta_m) - \sum_{i=1}^{2} x_i \ln(V_i\eta_i)\ ] \quad (5)$$

$$d' = \frac{ln\eta_m - (x_1 ln\eta_1 + x_2 ln\eta_1)}{x_1 x_2} \quad (6)$$

Where V_i and V_m shows the molar volume of pure components and binary solution, R and T indicates the ideal gas constant and absolute temperature, respectively.

The obtained G^{*E} and d' values were reported in Table 3,

The values of V_m^E, $\Delta\eta$ and $\Delta\kappa_s$can be correlated well with the Redlich-Kister (R-K) equation 7 [41];

$$Z = x_1 x_2 \sum_{i=0}^{4} A_i(2x_i - 1)^i \quad (7)$$

Where Z represents V_m^E, $\Delta\eta$ and $\Delta\kappa_s$ fitting functions and A_i represents the adjustable binary coefficient. The coefficients A_i is calculate using multi-parametric regression analysis based on a non-linear least-squares method.

The corresponding standard deviations, σ (Z) were computed using the following relation [42]:

$$\sigma(F(x)) = \left[\sum(Z_{exptl} - Z_{calcd})\right]^2 / (p-n)^{1/2} \quad (8)$$

Where p is the number of experimental data points and n is the number of parameters. The calculated parameters A_i values along with the standard deviations (σ) were given in Table 4.

RESULTS AND DISCUSSION

Excess Molar Volume

Fig.1 showed that the negative values had been observed for the binary mixtures of PCA with PEN and HEX, whereas positive values were noted for the system of PCA with HEP. The values of V_m^E in the binary systems of PCA with *n*-alkanes over the entire composition range can explain because of several effects that may be arbitrarily divided into physical and chemical forces [43].

(i) Interstitial accommodation of *n*-alkanes in the associated structure of PCA.
(ii) Dispersion forces between PCA and *n*-alkanes.

The graphical evidence which shown in Fig.1 supports that both the forces are operating between unlike molecules of the systems under study, Liu *et al*. [44] also observed the similar results for the binary mixture of butyl cyclohexane with n-alkanes. The observed positive values of V_m^E for PCA + HEP are attributed to the disruption of the associated structure of the PCA during the formation of mixtures leads to the increase in the volume. The negative values of V_m^E for PCA + PEN, + HEX mixtures may be due to interstitial accommodation of PEN and HEX molecules in the voids which are created by PCA molecules [29]. The algebraic values at the equimolar composition of all the binary systems with PCA fall in the order: PEN < HEX < 0 < HEP.

Deviation in Viscosity

The isotherm that was presented in Fig. 2 indicates the negative values of $\Delta\eta$ for binary mixtures of PCA with PEN, HEX and HEP. A correlation between the sign of $\Delta\eta$ and V_m^E had been observed for several binary solvent systems. V_m^E is negative where $\Delta\eta$ being positive or vice versa [45]

In true sense, the values of V_m^E indicate the specific interactions like complexes complex formation, while $\Delta\eta$ values indicate the dispersion forces (no complex formation) between unlike molecules. According to Fort and Moore [46], for the same mixture, it cannot be true. Therefore, the strength of the specific interactions/dispersion forces is not the only factor influencing the values of $\Delta\eta$ for liquid mixtures. The size and shape of the components are equally important factors. Hence, to elucidate the deviation in viscosity, it has resorted to G^{*E} and d' parameters.

According to Reed *et al*. [47] and Mayer *et al.* [48] the positive G^{*E} values indicate the specific interactions, while the negative values show the dominance of dispersion forces between unlike molecules. From the Table 3, it is clear that the negative values of G^{*E} for (PCA + HEP) indicates the presence of dispersion forces while positive values for (PCA + PEN and HEX) confirming the presence of specific interactions. The positive values of d' indicates the presence of specific interactions, while the negative values of d' suggest the dominance of dispersion forces between unlike molecules [49]. Table 3 reveals that the negative values of d' for (PCA + HEP) signifying the presence of dispersion forces, while the positive values of d' for (PCA + PEN and HEX) confirming the existence of specific interactions. Rastogi *et al*. [50] suggested that the deviation in viscosity is a combination of an interaction part and non-interaction part, further the non-interaction part was depending on the size and shape of the molecules [51].

$$Y^{E}_{(\text{observed})} = Y^{E}_{(\text{interaction})} + Y^{E}_{(\text{size effect})}$$

Where Y^E refers to the deviation in the property. Based on this assumption, the observed anomaly in the isotherms may be attributed to the size effect. The reported values of deviation in viscosity fall in the order: HEP < HEX < PEN < 0

Deviation in Isentropic Compressibility

From Fig.3, it can be observed that the negative curves from the ideal axis were observed for the systems (PCA+ PEN, HEX and HEP) over the entire range of composition. This behavior can be explained in terms of contraction of volume due to interstitial accommodation of *n*-alkanes in the associated structure of PCA which in turn decreases the inter-molecular path lengths leading to a negative deviation in isentropic compressibility [52]**.** From the Table 3, it is clear that the less negative values of $\Delta\kappa_s$ supports weak dispersion type interactions in the system (PCA + HEP)**.** The observed values of ($\Delta\kappa_s$) of the mixtures line up in the following order:

PEN < HEX < HEP < 0

It can be pointed out that the influence of the structure of the homologous series of *n*- alkanes on the values of $\Delta\kappa_s$ is observed. On the other hand, the $\Delta\kappa_s$ values become more negative with decreasing of the size of *n*-alkane.

CONCLUSIONS

In this work, the new data of density (ρ) viscosity (η) and speed of sound (u) for binary liquid mixtures of PCA with PEN, HEX and HEP at T = 308.15 K were reported as a function of mole fraction of PCA. Using these data, the V_m^E, $\Delta\eta$ and $\Delta\kappa_s$ have been calculated. The present study indicates the curves of V^E, $\Delta\eta$ and $\Delta\kappa_s$ are negative for the binary mixtures of PCA with *n*-alkanes except *n*-heptane which is observed as positive values of V^E. The excess functions of these systems give the

evidence of interstitial accommodation of PEN and HEX in the associated structure of PCA, whereas dispersion forces acting between PCA and HEP. The effect of chain length of *n*-alkanes on the physicochemical properties of the binary mixtures (PCA + PEN, HEX and HEP) had been investigated.

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TABLES

Table 1 Specifications of the chemicals used**.**

Table 2 Comparison of experimental density (ρ), viscosity (η) and speed of sound (u) with literature values at temperature 308.15 K.

Table 3 Values of density (ρ), excess molar volume (V_m^E), viscosity (η), speed of sound (u), deviation in viscosity ($\Delta\eta$), deviation in isentropic compressibility ($\Delta\kappa_s$), excess Gibbs free energy of activation of viscous flow (G^{*E}) and Grunberg-Nissan interaction parameter (d') for the binary solvent systems at T = 308.15 K.

Table 4 Adjustable parameters (A_i) and standard deviations (σ) for the binary mixtures of p-chloroacetophenone (1) + *n*-alkanes (2) at T = 308.15 K.

Table 1

Component	Assay	CAS.NO	Company
p-chloroacetophenone	97%	99-91-2	Sigma-Aldrich
n-pentane	99%	109-66-0	Sigma-Aldrich
n-hexane	99%	110-54-3	Sigma-Aldrich
n-heptane	99%	142-82-5	Sigma-Aldrich

Table 2

Liquids	ρ x10^{-3}/kg.m^{-3}		η x10^{3}/kg.m^{-1}. s^{-1}		u / m.s^{-1}	
p-chloro acetophenone	1.1888	1.1889 [a]	2.6142	2.6150 [b]	1362.0	---
	1.1814	1.1813 [a]	2.3550 [*]	2.3553 [b]	1398.2 [*]	1395.0 [c]
n-Pentane	0.6210	0.6212 [d]	0.2152	0.2160 [d]	1016.6	---
	0.6114 [*]	0.6110 [e]	0.2077 [*]	---	964.9 [*]	---
n-Hexane	0.6548	0.6548 [f]	0.2979	0.2980 [g]	1081.2	1081.0 [h]
	0.6459 [*]	0.6457 [h]	0.2702 [*]	0.2730 [g]	1027.0 [*]	1025.0 [i]
n-Heptane	0.6796	0.6796 [h]	0.4043	0.4040 [j]	1129.2	1128.0 [k]
	0.6704 [*]	0.6705 [j]	0.3490 [*]	0.3490 [g]	1092.5 [*]	1090.0 [i]

* values at 308.15 K; a =23, b =13, c = 24, d = 25, *e* = 26, *f* = 27, *g* = 28, *h* =29, i = 30, j = 31 and k = 32.

The standard uncertainty components u for each variable are u(T)= ±0.01 K, u(x) = ±1×10^{-4}, u(p) = ± 1.0 kPa, u(ρ) = ±0.02 g.cm^{-3}, u(η) = ±0.028 m^2/s and u(u) = 0.5 m.s^{-1}.

Table 3

x_1	ρx10^{-3} kg.m^{-3}	V_m^Ex10^6 m^3.mol^{-1}	ηx10^3 kg.m^{-1}. s^{-1}	u m.s^{-1}	$\Delta\eta$x10^3 kg.m^{-1}. s^{-1}	$\Delta\kappa_s$x10^{11} m^2.N^{-1}	G^{*E}x10^3 N.mol^{-1}	d'
PCA (1) + PEN (2)								
0.0000	0.6114	0.0000	0.2077	964.9	0.0000	0.0000	0.00	----
0.0229	0.6390	-2.4366	0.2164	979.8	-0.0404	-9.25914	24.40	0.6368
0.1353	0.7260	-5.0011	0.2897	1036.2	-0.2085	-27.5465	23.01	0.0380
0.2508	0.8138	-7.1637	0.3907	1082.5	-0.3555	-34.5528	22.62	0.1222
0.3722	0.9018	-8.7558	0.5443	1140.2	-0.4626	-37.3621	7.95	0.2557
0.4754	0.9684	-9.1361	0.7181	1193.2	-0.5104	-36.2678	9.77	0.3456
0.5791	1.0245	-8.4219	0.9316	1245.4	-0.5196	-32.271	16.23	0.3886
0.6785	1.0690	-6.9047	1.1989	1289.6	-0.4657	-26.2327	31.05	0.4841
0.8487	1.1267	-2.7694	1.7550	1365.6	-0.2751	-13.7872	32.03	0.5712
0.9775	1.1728	-0.3274	2.2780	1405.0	-0.0287	-2.75121	11.64	0.9729
1.0000	1.1814	0.0000	2.3550	1398.2	0.0000	0.0000	0.00	----
PCA (1) + HEX (2)								
0.0000	0.6459	0.0000	0.2727	1027.0	0.0000	0.0000	0.00	-----
0.0238	0.6645	-1.2231	0.2846	1031.2	-0.0377	-3.6645	11.02	0.3778
0.1544	0.7391	-2.1359	0.3522	1089.2	-0.2421	-21.8140	57.18	0.5914
0.2807	0.8080	-2.2788	0.4551	1133.2	-0.4022	-29.5581	67.65	0.4615
0.4013	0.8735	-2.3033	0.5874	1175.5	-0.5209	-32.6434	70.66	0.4074
0.4962	0.9245	-2.2132	0.7222	1213.3	-0.5838	-33.0427	69.05	0.3837
0.6026	0.9805	-1.9370	0.9167	1260.0	-0.6108	-31.3096	62.21	0.3626
0.7307	1.0471	-1.5031	1.2351	1312.8	-0.5592	-25.4517	46.74	0.3297
0.8529	1.1090	-0.9564	1.7115	1352.0	-0.3372	-15.7522	5.73	0.0165
0.9799	1.1719	-0.1642	2.2638	1381.4	-0.0493	-1.7581	1.58	0.1948
1.0000	1.1814	0.0000	2.3550	1398.2	0.0000	0.0000	0.00	----
PCA (1) + HEP (2)								
0.0000	0.6704	0.0000	0.3490	1092.5	0.0000	0.0000	0.00	----
0.0264	0.6808	0.3172	0.3620	1094.7	-0.0399	-1.14545	-7.02	-0.5371
0.1576	0.7372	1.0169	0.4002	1123.9	-0.2650	-9.62256	-95.53	-1.2357
0.2929	0.7990	1.3325	0.4856	1154.9	-0.4510	-15.3764	-133.44	-1.1053
0.4215	0.8605	1.4663	0.6195	1192.0	-0.5750	-19.0010	-133.79	-0.9470
0.5282	0.9133	1.5342	0.7666	1227.3	-0.6420	-20.2249	-127.64	-0.8892
0.6307	0.9675	1.2957	0.9535	1264.7	-0.6607	-19.8093	-114.91	-0.8548
0.7491	1.0326	0.9710	1.2805	1302.0	-0.5712	-16.1620	-74.37	-0.6932
0.8669	1.1001	0.6126	1.7324	1343.1	-0.3356	-10.0673	-28.96	-0.4587
0.9810	1.1692	0.1279	2.2515	1386.9	-0.0654	-1.48691	-4.61	-0.4651
1.0000	1.1814	0.0000	2.3550	1398.2	0.0000	0.0000	0.00	----

The standard uncertainty components u for each variable are u(T)= ±0.01 K, u(x) = ±1×10^{-4}, u(p) = ± 1.0 kPa, u(ρ) = ±0.02 g.cm^{-3}, u(η) = ±0.028 m^2/s and u(u) = 0.5 m.s^{-1}.

Table 4

	Temp.	Property	A_0	A_1	A_2	A_3	A_4	σ
PCA (1) + PEN (2)	308.15 K	V_m^Ex10^6 (m^3. mol^{-1})	-0.88	0.34	0.10	-0.03	-0.32	0.007
	308.15 K	$\Delta\eta$x10^3 (kg.m^{-1}. s^{-1})	-0.17	-0.09	-0.08	-0.07	0.15	0.011
	308.15 K	$\Delta\kappa_s$x10^{11} (m^2.N^{-1})	-8.18	1.57	-0.33	-0.68	-0.35	0.025
PCA (1) +HEX (2)	308.15 K	V_m^Ex10^6 (m^3. mol^{-1})	-0.11	-1.36	6.92	-0.77	1.67	0.003
	308.15 K	$\Delta\eta$x10^3 (kg.m^{-1}.s^{-1})	-3.09	2.17	-2.79	4.83	-3.01	0.051
	308.15 K	$\Delta\kappa_s$x10^{11} (m^2.N^{-1})	-5.48	-7.50	1.64	-1.05	2.42	0.073
PCA (1) + HEP (2)	308.15 K	V_m^Ex10^6 (m^3.mol^{-1})	4.28	-2.38	-0.25	-3.13	-1.05	0.005
	308.15 K	$\Delta\eta$x10^3 (kg. m^{-1}.s^{-1})	-1.75	-0.88	5.81	-8.37	2.54	0.084
	308.15 K	$\Delta\kappa_s$x10^{11} (m^2.N^{-1})	-2.60	-6.52	-7.37	4.38	-0.11	0.059

FIGURES

Fig.1 variation of V_m^E of the binary liquid mixtures of p-chloroacetophenone (1) with ●; *n*- pentane (2), ■; *n*-hexane (2), ▲; *n*-heptane (2) at 308.15 K.

Fig.2 variation of $\Delta\eta$ the binary liquid mixtures of p-chloroacetophenone (1) with ●; *n*-pentane (2), ■; *n*-hexane (2), ▲; *n*-heptane (2) at 308.15 K.

Fig.3 variation of $\Delta\kappa_s$ the binary liquid mixtures of p-chloroacetophenone (1) with ●; *n*-pentane (2), ■; *n*-hexane (2), ▲; *n*-heptane (2) at 308.15 K.

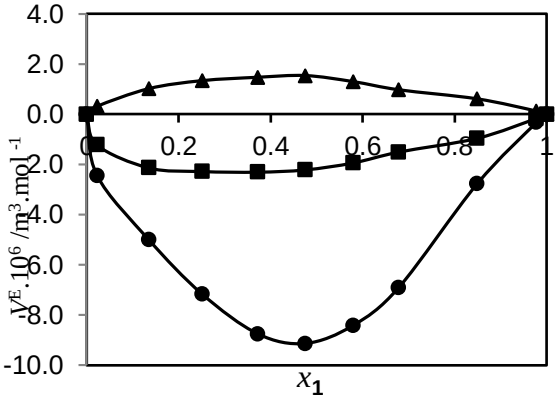

Fig.1

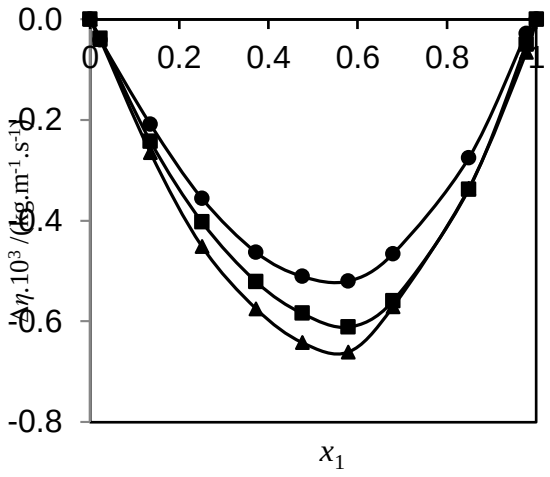

Fig. 2

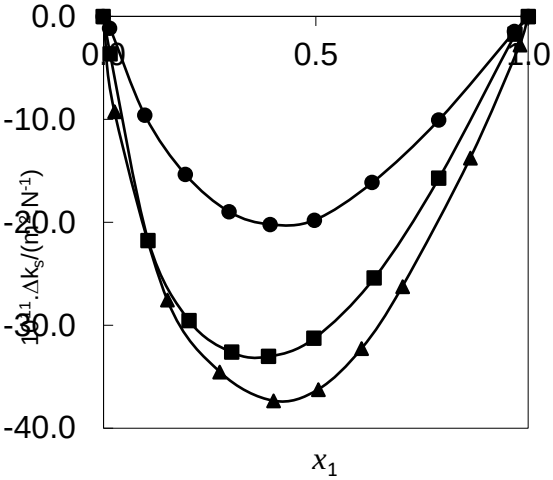

Fig. 3

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