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THERMODYNAMICS OF ANILINE + TOLUENE MIXTURES

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ABSTRACT

Heats of mixing, H^E, of aniline + toluene at 298.15 and 308.15 K and that of aniline + cyclohexane at 308.15 K have been measured over the entire composition range. The excess Gibbs free energies of mixing, G^E, for aniline + cyclohexane mixtures at 308.15 K have been obtained from the measured vapour pressure data. The H^E and G^E values are positive throughout the entire aniline concentration range and $H^E > G^E$. The results have been analysed in terms of the Barker and ideal associated model theory of non-electrolyte solutions. It has been observed that the ideal associated model approach which assumes the presence of AB, AB_2, A_2B_2 and B molecular species describes well (within ±40 J mole^{-1} at the worst) the general dependence of H^E on x_B (mole fraction of aniline) over the whole composition range for aniline + toluene mixtures. The equilibrium constants for the various association reactions, along with the enthalpies of formation of various molecular species have also been calculated.

INTRODUCTION

Aniline molecules are known to be associated [1—4] in the pure state through hydrogen bonding. IR [5] spectroscopic studies have indicated that while one of the hydrogens of the $—NH_2$ group of aniline forms a strong hydrogen bond, the second hydrogen in the $—NH_2$ group interacts weakly with the π-electrons of adjacent aniline molecules. Further, while Kreglewski and Wilhoit [6] have interpreted the thermodynamic data of aniline + benzene mixtures in terms of electron donor—acceptor interactions, Chowdary and Krishnan [7] have interpreted their excess volume data on aniline + cyclohexane mixtures as suggesting that the addition of cyclohexane causes a rupture of the hydrogen-bonded network in aniline. It would, therefore, be interesting to study the nature of aniline in its binary mixtures with toluene. The present work describes interactions in aniline + toluene mixtures.

EXPERIMENTAL

Aniline, cyclohexane and toluene (all B.D.H., A.R. grade) were purified as described earlier [8,9]. The purity of the final samples was checked by density determinations at 298.15 ± 0.01 K which agreed to within ±0.00005 g ml^{-1} with the corresponding literature values [10—14].

Heat of mixing measurements were made in an adiabatic calorimeter which has been described elsewhere [15]. The time was recorded by an electronic timer (type T01 SR No. 010) Systronic, Ahmedabad (India) which could be read to ±0.0001 sec. The temperature variations of the thermostatic bath were found to be within ±0.01 K. The uncertainty in H^E values is about 0.3%.

Vapour pressures of aniline + cyclohexane mixtures were determined by a static method described previously [16]. The apparatus (excluding the manometric part) was placed in a water thermostat which was placed in another water thermostat. The temperature of the outer thermostat was controlled to ±0.01 K and the temperature drift in the inner thermostat was found to be less than ±0.003 K. The mercury heights in the manometer were read by a cathetometer which could read to ±0.001 cm. All vapour pressure measurements are reproducible to better than ±0.02 Torr.

Our measured vapour pressure (150.7 Torr) for cyclohexane at 308.15 K agrees within 0.3% with the values evaluated at 308.15 K from the vapour pressure data reported by Willingham et al. [17] at various temperatures. We are unaware of any vapour pressure data for aniline at 308.15 K with which to compare our results.

RESULTS

The excess enthalpies of mixing for the system aniline + toluene at 298.15 K and 308.15 K and for the system aniline + cyclohexane at 308.15 K are recorded in Table 1. The data have been fitted to the equation

$$\frac{H^E}{x_B(1 - x_B)} = [h_0 + h_1(2x_B - 1) + h_2(2x_B - 1)^2] \tag{1}$$

where x_B is the mole fraction of aniline and h_0, h_1 and h_2 are disposable parameters. These parameters were evaluated by the method of least squares

TABLE 1

Experimentally measured heat of mixing values

Aniline (B) + toluene (A) Temperature 298.15 K		Aniline (B) + toluene (A) Temperature 308.15 K		Aniline (B) + cyclohexane (A) Temperature 308.15 K	
Mole fraction of B	H^E (J mole^{-1})	Mole fraction of B	H^E (J mole^{-1})	mole fraction of B	H^E (J mole^{-1})
0.2151	787.5	0.2322	695.3	0.1985	1086.3
0.3433	990.1	0.3658	901.8	0.2919	1312.9
0.4528	1022.5	0.4634	930.2	0.4125	1452.1
0.5136	998.7	0.5872	842.2	0.5105	1454.7
0.6255	896.4	0.6881	691.7	0.5529	1435.6
0.8852	357.6	0.7911	490.0	0.6765	1298.7

TABLE 2

Parameters of equation 1 along with the standard deviations $\delta(H^E)$ in J mole^{-1}

Mixture	Temp. (K)	h_0	h_1	h_2	δ (H^E)
Aniline + toluene	298.15	4047.38	−916.38	252.96	6.55
Aniline + toluene	308.15	3671.20	−860.70	−691.78	8.40
Aniline + cyclohexane	308.15	5844.39	−361.97	1977.39	7.47

and are given together with the standard deviations of molar heats of mixing, $\delta(H^E)$ in Table 2.

Vapour pressure data for aniline + cyclohexane mixtures at 308.15 K were used to evaluate the molar excess Gibbs free energies of mixing, G^E, by Barker's method [18]. The form of the function used for G^E following Redlich and Kister [19] is

$$\frac{G^E}{RT} = x_B(1 - x_B)[G_0 + G_1(2x_B - 1) + G_2(2x_B - 1)^2] \qquad (2)$$

where G_0, G_1 and G_2 are adjustable parameters. These parameters along with the G^E data at 308.15 K for aniline + cyclohexane mixtures are recorded in Table 3. In these calculations the required virial coefficients were obtained from Berthelot's equation [20] and the critical constant data were taken from the literature [21].

DISCUSSION

Deshpande et al. [22] have determined H^E for the aniline + toluene system at 298.15 K. Our results are in excellent agreement with those reported

TABLE 3

Measured total vapour pressure, P; activity coefficients, γ; residual vapour pressure, $R = P_{expt} - P_{calc}$; Gibbs excess free energy of mixing G^E and the parameters of eqn. 2 for different mole fractions x_B of aniline at 308.15 K

x_B	P (torr)	γ_A	γ_B	P_A (torr)	P_B (torr)	R (torr)	G^E (J mole^{-1})
0.1925	136.29	1.1116	3.7170	121.81	0.73	−1.83	713.99
0.2397	133.27	1.1711	3.0748	114.72	0.91	−3.88	846.34
0.3248	135.87	1.3118	2.3013	101.86	1.23	−0.58	1040.78
0.5183	134.70	1.8211	1.4577	72.67	1.96	−0.49	1254.89
0.5917	133.98	2.1030	1.2988	61.60	2.24	1.53	1244.32
0.6712	127.33	2.4828	1.1786	49.63	2.54	1.12	1168.81
0.6911	124.48	2.5925	1.1549	46.64	2.62	0.54	1138.79
0.7831	106.43	3.1949	1.0718	32.79	2.97	−1.51	937.70
0.8585	85.59	3.8397	1.0296	21.42	3.26	−0.10	690.56
0.9480	41.44	4.8617	1.0040	7.90	3.62	−0.06	290.49

$G_0 = 1.9505$, $G_1 = 0.3108$ and $G_2 = 0.0888$.

by those workers for $0 < x_B \leqslant 0.3$ and $x_B \geqslant 0.7$. However, in the intermediate range our results differ from their results by as much as +70 J mole^{-1} at $x_B = 0.45$. We are, however, unaware of any G^E data for aniline + cyclohexane mixtures at 308.15 K with which to compare our results.

Heats of mixing for aniline + toluene are positive throughout the concentration range studied here. Further, H^E data at 308.15 K when combined with G^E data [22] at 308.15 K gave TS^E values that are positive at all aniline mole fractions and the curve of TS^E against x_B is unsymmetrical about x_B (Fig. 1).

At the simplest qualitative level, the observed H^E data for this mixture may be accounted for if we assume that (i) aniline is self associated and there is a decrease in self association when it is mixed with toluene, and (ii) there is a weak interaction between aniline and toluene.

The decrease of self association of aniline due to factor (i) would increase the orientational freedom of the aniline molecules, so that TS^E should be strongly positive. On the other hand, since the measured H^E is due to the cumulative effects of factors (i) and (ii) mentioned above, the positive H^E for this mixture may be accounted for if it is assumed that the effects due to factor (i) outweigh those due to factor (ii).

We examined our results using Barker's theory [23] which allows a molecule of type A occupying r_A sites on a Z co-ordinated lattice to have $q^Z_A = r_A Z - 2r_A + 2$ neighbouring contact sites, each site having an interaction energy with the neighbouring part of the surface of another molecule. The contact sites are further sub-divided into classes, the number in the μth class of the molecule A denoted by Q^A_μ. It was assumed that aniline + toluene have the following geometrical parameters.

Lattice $Z = 4$
Aniline molecule = B: $r_B = 2$, $Q^B_H = 1$, $Q^B_N = 1$, $Q^B_R = 4$,
Toluene molecule = A: $r_A = 2$, $Q^A_{R'} = 6$,

where H, N and R represent respectively, the hydrogen, nitrogen and hydro-

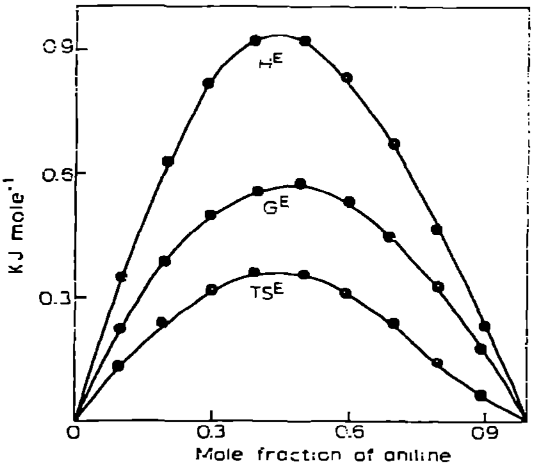

Fig. 1. Aniline + toluene system at 308.15 K.

carbon surface of aniline while R′ denotes the hydrocarbon surface of toluene (since molar volumes of aniline and toluene at 298.15 K are nearly the same, we have taken $r_A = r_B = 2$).

The interactions first considered were a specific (N ... H) of strength U_2 between the nitrogen and hydrogen of aniline, an interaction (R ... R′) of strength U_3 between the aromatic ring of aniline and the aromatic ring of toluene and non-specific interactions for all the remaining contact points (these interactions correspond to a model that takes into account the self association of aniline and also dipole—induced dipole interactions between aniline and toluene). For the sake of simplicity, these non-specific interactions were considered to have the same strength U_1. Excess energies of mixing at constant volume (U_V^E) were then calculated. From the expression

$$U_V^E = -2RT[\{X_R(X_N + X_H + X_{R'}) + X_{R'}(X_N + X_H)\}\eta_1 \ln \eta_1 + X_R X_{R'}\eta_3 \ln \eta_3 + (X_N X_H - x_B X'_N X'_H)\eta_2 \ln \eta_2]$$

where the various parameters X depend upon the number of interactions of various contact sites of different classes in molecules A and B in the manner described in ref. 23. These parameters were evaluated in the manner suggested by Barker [23] and the calculated U_V^E values at $X_B = 0.3$, 0.5 and 0.7 are recorded in Table 4. It is evident from Table 4 that the theoretical H^E values fail to reproduce the corresponding experimental H^E values very well. (It is normally customary to convert U_V^E values to H^E values using the relation $U_V^E = H^E - TV^E\alpha_m/(K_T)m$ where α_m, $(K_T)m$ and V^E are the expansivity, isothermal-compressibility and excess volume of the mixture. As $V^E \approx 0$, therefore, $U_V^E \approx H^E$.)

We next considered two different models α and β in which, in addition to the non-specific interactions mentioned above, the interactions assumed were:

Model α: (i) specific (R ... R′) interactions of strength U_3 between the aromatic ring of aniline and the aromatic ring of toluene. (ii) no self association of aniline. This model is based on the observations of Bellamy and Williams [24] that there is no self association in aniline and only non specific interaction play the predominant role.

Model β: (i) specific (N ... H) interactions of strength U_2 between aniline—aniline molecules. (ii) specific (H ... R′) interactions of strength U_4 between the hydrogen of the —NH_2 group and the aromatic ring of the hydrocarbon.

It was found that H^E values calculated from Barker's theory for Model β (which is based on spectroscopic studies of aniline [5]) at $x_B = 0.3$ and 0.5 reproduce the experimental data reasonably well but the agreement was not that impressive for $x_B > 0.5$ (this in sharp contrast with the analysis [22] of H^E data for this mixture in terms of the Balescu's theory which fails to predict even the sign of H^E). The failure of Model β at $x_B > 0.5$ may be due to the simplicity of the model assumed or to the presence of associated complexes of general formula $A_m B_x$ and B_l. H^E values (designated as $H^{E(1)}$, $H^{E(\alpha)}$ and $H^{E(\beta)}$) for the three models given above, along with the experimental H^E values at 298.15 K, and H^E_{expt} values together with H^E_{calc} values at 308.15 K (from Model β) at $x_B = 0.3$, 0.5 and 0.7 for aniline + toluene mixtures are

TABLE 4

Comparison of H^E values calculated according to Barker's theory with the corresponding H^E experimental values at x_B = 0.3, 0.5 and 0.7 at 298.15 and 308.15 K (for aniline (B) + toluene (A) mixtures)

H^E (J mole^{-1})	Temperature (K)	Mole fraction of component B			Interaction energies (J mole^{-1})			
		0.3	0.5	0.7	U_1	U_2	U_3	U_4
Experimental	298.15	941.54	1058.20	813.75	—	—	—	—
H^E (1)	298.15	591.71	857.88	908.35	615.86	−1005.03	−120.94	—
H^E (α)	298.15	611.50	829.68	904.39	615.86	—	−120.94	—
H^E (β)	298.15	780.15	1019.91	1050.24	615.86	−1005.03	—	−120.94
Experimental	308.15	820.01	917.80	675.40	—	—	—	—
H^E (β)	308.15	695.79	909.45	917.80	539.83	−672.13	—	−75.73

reported in Table 4 along with their interaction energies U_i ($i = 1$—4). The H^E and activity coefficient data for this mixture were then analysed in terms of an ideal associated model [25,26].

It is assumed that in a binary solution of aniline + toluene (aniline designated as B_n since aniline is self associated) mutual equilibrium of the species A_mB_x and B_l where $l = 1, 2, 3, \ldots l$, $m = 1, 2, 3, \ldots m$, $x = 1, 2, 3, \ldots x$ and $n = 1, 2, 3, \ldots n$, exists according to the reactions

$$mA + B_n \rightleftharpoons A_mB_x + B_l \tag{3}$$

so that equilibrium constants for the various association reactions represented by eqn. (3) are

$$K_{m,x/n} = a_{A_mB_x}/a_A^m a_{B_n}^{x/n} \tag{4}$$

and

$$K_{l/n} = \frac{a_{B_l}}{a_{B_n}^{l/n}} \tag{5}$$

where a denotes activities. If the activity coefficients of the various species represented in eqn. (3) are assumed to be unity [25,28], the material balance equation for the system can be written as

$$a_A + a_{B_n} + \sum_x K_{m,x/n} a_A^m a_{B_n}^{x/n} + \sum_l K_{l/n} a_{B_n}^{l/n} = 1 \tag{6}$$

We next considered the two simple cases

case (i) $l = m = 1, x = 1, 2$ and $n = 2$.

case (ii) $l = 1, n = 2; m = 1, x = 1, 2$ and $m = x = 2$.

In order to evaluate the various K's in eqn. (6) for the two cases (i) and (ii) mentioned above, the observed activities of the components of these binary mixtures were corrected [29—31] for dispersion contributions by expressing $a_A = \gamma_A X_A/\gamma_A^*$ and $a_{B_2} = \gamma_{B_2} x_{B_2}/\gamma_{B_2}^*$ where γ_A^* and $\gamma_{B_2}^*$ are the activity coefficients of a reference mixture. Since cyclohexane is inert in nature and has nearly the same volume as that of toluene, cyclohexane (A^*) + aniline (B_2^*) was taken as the reference system for the present analysis. A series of values were next assumed for the various K's in eqn. (6) and the process was repeated till a set of K values was obtained which yielded

$$\frac{(1 - a_A - a_{B_2})}{a_{B_2}^{1/2}} = D$$

values that corresponded very closely with those obtained from the experimental a_A and a_{B_2} values. It was observed that eqn. (6) with the following set of K values

$K_{1,0.5} = 0.58$; $K_{1,1} = 0.06$; $K_{0.5} = 0.06$ for case (i)

and eq. (6) with the following set of K values

$K_{1,0.5} = 0.57$; $K_{1,1} = 0.06$; $K_{0.5} = 0.06$; $K_{2,1} = 0.8$ for case (ii) yielded D values that reproduce equally well the corresponding values obtained from experimental values for this system. The criterion of effectiveness was the variance of the fit σ^2D defined by

$$\sigma^2D = \frac{\sum_q (D_{\text{calc}} - D_{\text{expt}})^2}{(q-p)} \tag{7}$$

where q is the number of points used in the fit and p is the number of adjustable parameters.

$$\text{Since} \quad \underset{\text{case (i)}}{\sigma^2D = 0.0057} \quad \text{and} \quad \underset{\text{case (ii)}}{\sigma^2D = 0.0051}$$

are nearly the same, therefore, the analysis of the activity coefficient data described above suggests that these mixtures may be assumed to have either AB, AB_2 and B, or AB, AB_2, A_2B_2 and B molecular species in solution.

We next considered our H^E data. Examination of H^E data of this mixture in terms of models that involved consideration of AB, AB_2 and B and AB, AB_2, A_2B_2 and B molecular species in solutions required that H^E be expressed as

$$H^E = \frac{n_{AB}\Delta H_{AB} + n_{AB_2}\Delta H_{AB_2} + n_B\Delta H_B}{(N_A + N_{B_2})} \tag{8}$$

$$H^E = \frac{n_{AB}\Delta H_{AB} + n_{AB_2}\Delta H_{AB_2} + n_{A_2B_2}\Delta H_{A_2B_2} + n_B\Delta H_B}{(N_A + N_{B_2})} \tag{9}$$

where n_{AB}, n_{AB_2}, $n_{A_2B_2}$ and n_B are the amounts of species AB, AB_2, A_2B_2 and B present at equilibrium in solution. N_A and N_{B_2} are the stoichiometric amounts of A and B_2, and ΔH represent the enthalpy of formation of the various molecular species present in this mixture.

If the equilibrium mole fraction of A, B_2, AB, AB_2 and B are represented by Z_A, Z_{B_2}, Z_{AB_2} and Z_B then for an ideal associated mixture A + B_2, containing AB, AB_2 and B molecular species

$$Z_A + Z_{B_2} + Z_{AB} + Z_{AB_2} + Z_B = 1 \tag{10}$$

where

$$Z_{A_mB_x} = K_{m,x/m} Z_A^m / Z_{B_n}^{x/n} \quad (m = 1, x = 1 \text{ and } 2, n = 2)$$

$$Z_B = K_{0.5} Z_{B_2}^{1/2}$$

The experimental H^E values were again corrected for dispersion contributions by subtracting from H^E_{expt} the H^E values for the cyclohexane + aniline system at 308.15 K.

Consequently, in eqns. (8) and (9)

$$H^E = H^E_{\text{expt}} - H^E \text{ (cyclohexane + aniline)} \tag{11}$$

Algebraic manipulation of eqns. (8) and (10) and material balance equations

$$N_A = n_A + n_{AB} + n_{AB_2} \tag{12}$$

$$N_{B_2} = n_{B_2} + \tfrac{1}{2}n_{AB} + \tfrac{1}{2}n_B + n_{AB_2} \tag{13}$$

TABLE 5

Equilibrium constants, K, and the enthalpies of formation ΔH, of the various molecular species present in aniline + toluene mixtures at 308.15 K

	AB	A_2B_2	AB_2	B	Reaction
K	0.58		0.06	0.06	$A + B_2 \rightarrow AB + AB_2 + B$
ΔH (kJ mole^{-1})	−4.24		−0.05	2.10	
K	0.57	0.08	0.06	0.06	$A + B_2 \rightarrow AB + AB_2 + A_2B_2 + B$
ΔH (kJ mole^{-1})	−4.00	−1.02	−0.05	2.10	

leads to

$$JH^E = K_{1,0.5}Z_A\Delta H_{AB} + K_{1,1}Z_A\Delta H_{AB_2} + K_{0.5}Z_{B_2}^{1/2}\Delta H_B \qquad (14)$$

where

$$J = \frac{Z_{B_2}^{1/2} + 0.5K_{0.5} + 0.5K_{1,0.5}Z_A + K_{1,1}Z_AZ_{B_2}^{1/2}}{x_{B_2}}$$

In order to evaluate the various ΔH in eqn. (14), we assumed some values of Z_A and determined the corresponding Z_{B_2} values from eqn. (10) using the K values described above for a solution containing AB, AB_2 and B molecular species. With these Z_A and Z_{B_2} values we calculated x_{B_2} (mole fraction of aniline) by suitable manipulation of eqns. (10), (12) and (13) and then assigned various values to ΔH_{AB}, ΔH_{AB_2} and ΔH_B till they gave H^E values [from eqns. (11) and (14)] that compared well with the corresponding experimental H^E values. The various ΔH values are recorded in Table 5 and the calculated H^E values are compared with the corresponding H^E experimental values in Fig. 2. It is evident from this figure that the theoretical H^E values describe well the general behaviour of H^E for $0 \leqslant x_{B_2} \leqslant 0.5$ for this mixture. However, for $x_{B_2} > 0.5$, the theoretical values exceed the corre-

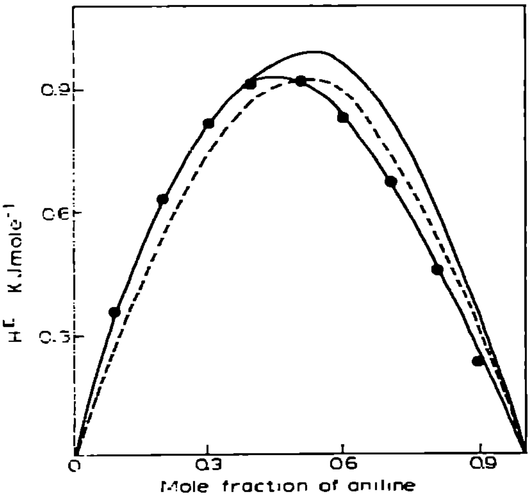

Fig. 2. Aniline + toluene system at 308.15 K. ●, Experimental values; ———, 1st case; - - - - - -, 2nd case.

sponding H^E_{expt} values as much as 140 J mole^{-1}.

A similar process was applied to the case when the mixture contained AB, AB_2, A_2B_2 and B molecular species and the various ΔH values were evaluated in a manner similar to the one described above when the solution was assumed to contain the AB, AB_2 and B molecular species. These ΔH values are given in Table 5 and the calculated H^E values are plotted in Fig. 2. The theoretical H^E curve reproduces reasonably well (within ±40 J mole^{-1} at the worst) the corresponding experimental results.

The analysis of H^E and activity coefficient data for (toluene + aniline) mixtures thus suggests that this mixture is characterized by the presence of AB, AB_2, A_2B_2 and B molecular species in solution.

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REFERENCES

1 W. Godry, J. Chem. Phys., 7 (1939) 600.
2 L.J. Bellamy and R.L. Williams, Spectrochim. Acta, 9 (1957) 341.
3 I. Yamaguchi, Bull. Chem. Soc. Jpn., 34 (1961) 1606.
4 J. Feeney and L.H. Sutcliffe, J. Chem. Soc., (1962) 1123.
5 H. Wolff and D. Mathias, J. Phys. Chem.. 77 (1973) 2081.
6 A. Kreglewski and R.C. Wilhoit, J. Phys. Chem. 79 (1975) 449.
7 M.C. Chowdary and V.R. Krishnan, Indian J. Chem., 14 (1976) 377.
8 R.K. Nigam and B.S. Mahl, Indian J. Chem., 9 (1971) 1255.
9 B.S. Mahl, R.K. Nigam, S.L. Chopra and P.P. Singh, J. Chem. Thermodyn., 3 (1971) 363.
10 J.C. Smith, N.J. Foecking and W.P. Barber, Ind. Eng. Chem., 41 (1949) 2289.
11 A.F. Forziati, A.R. Glasgow Jr., C.B. Willingham and F.D. Rossini, J. Res. Natl. Bur. Stand., 36 (1946) 120.
12 A.V. Few and J.W. Smith, J. Chem. Soc., (1949) 753.
13 J.W. William and I.J. Krchma, J. Am. Chem. Soc., 49 (1927) 1676.
14 H. Perkin, J. Prakt. Chem., 69 (1896) 1025: quoted from Physico-Chemical Constants of Pure Organic Compounds, Elsevier, Amsterdam, 1950, p. 165.
15 R.K. Nigam and B.S. Mahl, J. Chem. Soc., Faraday Trans. 1, 68 (1972) 1508.
16 R.K. Nigam and B.S. Mahl, Indian J. Chem., 9 (1971) 1250.
17 C.B. Willingham, W.J. Taylor, J.M. Pignocco and F.D. Rossini, J. Res. Natl. Bur. Stand., 35 (1945) 219.
18 J.A. Barker, Aust. J. Chem., 6 (1953) 207.
19 O. Redlich and A.I. Kister, Ind. Eng. Chem.. 40 (1948) 345.
20 A. Scatchard and L.B. Tichnor, J. Am. Chem. Soc. 74 (1952) 3724.
21 R.C. Weast, Handbook of Chemistry and Physics, 58th ed.. C.R.C. Press, Cleveland, Ohio, p. f-89.
22 D.D. Deshpande and M.V. Pandya, Trans. Faraday Soc., 65 (1969) 1456.
23 J.A. Barker, J. Chem. Phys., 20 (1952) 1526.
24 L.J. Bellamy and R.L. Williams, Spectrochim. Acta, 9 (1957) 341.
25 M.L. McGlashan and R.P. Rastogi, Trans. Faraday Soc., 54 (1958) 496.
26 D.V. Fenby and L.G. Hpler, J. Chem. Thermodyn., 6 (1974) 185.
27 L. Sarolea-Mathot, Trans. Faraday Soc., 8 (1953) 49.
28 A. Alexander, J. Phys. Chem., 74 (1970) 2214.
29 W.J. Gaw and F.L. Swinton, Trans. Faraday Soc., 64 (1968) 2023.
30 A.G. Willianson, Trans. Faraday Soc., 64 (1968) 1763.
31 D.D. Deshpande and S.L. Oswal, J. Chem. Soc., Faraday Trans. 1, 68 (1972) 1059.