Monatshefte für Chemie 121, 593–600 (1990)

Monatshefte für Chemie
Chemical Monthly
© by Springer-Verlag 1990

Correlation Between Topological Features and Surface Tension of Binary Liquid Mixtures

Raj K. Dewan and **Surinder K. Mehta***

Department of Chemistry, Panjab University, Chandigarh-160 014, India

Summary. The excess surface tension of a large number of binary liquid mixtures has been correlated with their topological features quantified in terms of the molecular connectivity indices. The agreement between the calculated and experimental σ^E values is reasonably well for all the mixtures. A simple correlation has also been proposed between σ^E and molar excess volume (V^E) of a binary mixture. The correlation is quite useful in correlating σ^E data even for the mixtures where either one or both the components are associated in the pure state and/or there is interaction between them.

Keywords. Surface tension; Binary mixtures; Molecular connectivity indices.

Korrelation zwischen topologischen Gegebenheiten und Oberflächenspannung von binären flüssigen Mischungen

Zusammenfassung. Die Exzeß-Oberflächenspannungen einer großen Anzahl von binären flüssigen Mischungen wurden mit der Topologie ihrer Komponenten in Form der molekularen Konnektivitätsindices korreliert. Die Übereinstimmung zwischen den σ^E-Werten ist für alle Mischungen relativ gut. Es wurde ebenfalls eine einfache Korrelation zwischen σ^E und den molaren Exzeß-Volumina (V^E) der binären Mischungen vorgeschlagen. Diese Korrelierung ist nützlich, um die σ^E-Werte sogar dann für die Korrelation von Mischungen verwenden zu können, wenn entweder eine oder beide Komponenten im Reinzustand assoziiert sind und/oder eine Wechselwirkung zwischen ihnen besteht.

Introduction

It has been recognised that surface tension plays an important role in interphase heat and mass transfer. It is of engineering and physico-chemical interest to develop equations for correlating data and predicting surface tensions of liquid mixtures. This has resulted in a number of equations which relate the mixture surface tension to properties of the pure components. The methods for computing surface tension of mixtures can be divided into two general categories: empirical relations and theoretical or semitheoretical equations. It has been found that most of the theoretically derived equations can only be applied to ideal systems while an empirical equation may correlate well for a few systems but fail on other systems. The former equations fail completely where one or more components are polar in nature. The problem becomes more complex for the class of mixtures where (i) one or both the components are associated in pure state and (ii) components interact with each other.

Recently much interest has concentrated around the utilization of the concept of molecular connectivity index of the third degree of graph theory to predict excess volumes [1] and heats of mixing [2] of binary mixtures. Graph theory has provided the chemist with a variety of very useful tools and one such tool is the topological index. A topological index is a numeric quantity that is mathematically derived in a direct manner from the structural graph of a molecule. Topological indices usually reflect both molecular size and shape. Trinajstic and coworkers [3] recently reported that 39 topological indices are presently available in literature. In 1975, Randic [4] proposed a topological index known as molecular connectivity index that has become one of the most widely used in both Quantitative structural activity relationship (QSAR) and Quantitative structural property relationship (QSPR) studies. The original work of Randic was expanded to broaden its application to compounds containing rings, multiple bonds, and heteroatoms by Kier and Hall [5].

An attempt has been made in the present work to correlate these connectivity parameters with surface tension (σ) of liquids and the excess surface tension (σ^E) of binary mixtures as no satisfactory approach, theoretical or empirical, is available to compute reasonably well the surface tension of liquids and their mixtures.

Theoretical

In the calculation of connectivity indices of any molecule, the first step is to draw a hydrogen suppressed graph (HSG) and this represents the topology of the non-hydrogen skeleton of the molecule. Each vertex is assigned a numerical values and the connectivity indices of the first, second and third degree viz. ${}^1\xi$, ${}^2\xi$ and ${}^3\xi$ respectively, can be calculated from the following equations

$$ {}^1\xi = \sum_{l<m} (\delta_l \delta_m)^{-0.5}, \tag{1} $$

$$ {}^2\xi = \sum_{l<m} \sum_{m<n} (\delta_l \delta_m \delta_n)^{-0.5}, \tag{2} $$

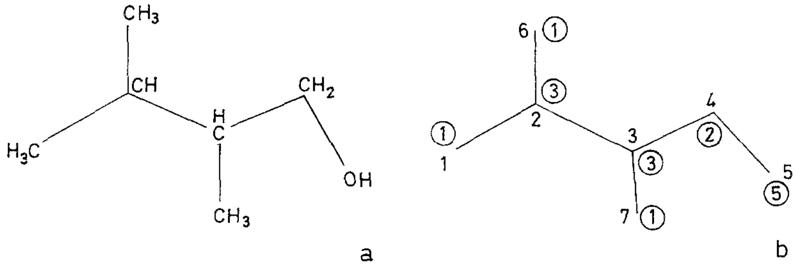
$$ {}^3\xi = \sum_{l<m} \sum_{m<n} \sum_{n<o} (\delta_l \delta_m \delta_n \delta_o)^{-0.5}, \tag{3} $$

where δ_l, δ_m, δ_n, and δ_o are the valencies of the lth, mth, nth and oth vertices in the molecular graph of a molecule.

In order to distinguish carbon vertices from those of heteroatoms, single bonds from multiple bonds, and aromatic systems from aliphatic cyclic systems, the topological indices are calculated in terms of δ values at the valence level. In this case δ for a vertex is defined by

$$ \delta_i^v = Z_i - h_i, \tag{4} $$

where Z_i is the number of valence electrons of ith vertex and h_i is the number of hydrogens attached to the ith vertex. The HSG (b), numbering and δ values



Scheme 1

of various vertices (shown encircled) for 2,3-dimethyl butanol are shown in Scheme 1.

$$^1\xi = \frac{1}{\sqrt{1.3}} + \frac{1}{\sqrt{1.3}} + \frac{1}{\sqrt{1.3}} + \frac{1}{\sqrt{1.3}} + \frac{1}{\sqrt{1.3}} + \frac{1}{\sqrt{1.3}} = 2.790,$$

$$^2\xi = \frac{1}{\sqrt{1.3.1}} + \frac{1}{\sqrt{1.3.3}} + \frac{1}{\sqrt{3.3.1}} + \frac{1}{\sqrt{3.3.2}} + \frac{1}{\sqrt{1.3.3}} + \frac{1}{\sqrt{3.2.5}} + \frac{1}{\sqrt{1.3.2}} = 1.996,$$

$$^3\xi = \frac{1}{\sqrt{1.3.3.2}} + \frac{1}{\sqrt{1.3.3.1}} + \frac{1}{\sqrt{3.3.2.5}} + \frac{1}{\sqrt{1.3.3.1}} + \frac{1}{\sqrt{1.3.3.2}} = 1.2435.$$

Basically a binary mixture (A + B) is formed by the replacement of like contacts in the pure A and B by unlike contacts A − B in the mixture. It implies that molar excess volume V^E and the excess surface tension (σ^E) of a binary mixture should have some correlation between them. Kier and Hall [5] and Jain and Gombar [1] have shown that the molar volumes of the non-electrolyte liquids depend directly on $^1\xi$ and $^3\xi$ parameters of these molecules.

Therefore, we can express σ of a liquid as

$$\sigma = \frac{\alpha}{^3\xi} + \beta\,{}^1\xi + \gamma. \tag{5}$$

Here α, β and γ are constants and $^1\xi$, $^3\xi$ are the connectivity parameters of the first and the third degree respectively. For alkanes, the values of α, β and γ are computed by taking known literature [6] values of surface tension of *n*-pentane, *n*-hexane and *n*-heptane at 298.15 K and Eq. (5) becomes

$$\sigma = \frac{1.3054}{^3\xi} + 4.985\,{}^1\xi + 1.6. \tag{6}$$

The above equation can be used to predict surface tension for any type of alkane. Table 1 lists such surface tensions for a number of alkanes and alkane isomers. The calculated surface tension values are within 6% of the literature values. For a binary mixture

$$\sigma_A = \frac{\alpha_A}{^3\xi_A} + \beta_A\,{}^1\xi_A + \gamma_A, \tag{7}$$

$$\sigma_B = \frac{\alpha_B}{^3\xi_B} + \beta_B\,{}^1\xi_B + \gamma_B. \tag{8}$$

Table 1. Comparison of calculated and experimental surface tension at 298.15 K

Component	σ/dyn cm^{-1} Calc.	σ/dyn cm^{-1} Lit.	${}^{1}\xi$	${}^{3}\xi$
n-Octane	22.00	21.18	3.914	1.457
2-Methylbutane	14.51	14.46	2.270	0.816
2-Methylpentane	16.91	16.87	2.770	0.866
3-Methylpentane	16.53	17.60	2.808	1.394
2,2-Dimethylbutane	15.60	15.81	2.561	1.061
2,3-Dimethylbutane	15.75	16.87	2.643	1.333
2-Methylhexane	18.37	18.80	3.270	2.808
3-Methylhexane	18.97	19.30	3.308	1.478
2,3-Dimethylpentane	18.36	19.47	3.181	1.449
2,4-Dimethylpentane	18.56	17.66	3.126	0.943

The ideal surface tension for a mixture σ_m(id) would be given by

$$\begin{aligned}\sigma_m(\mathrm{id}) &= x\sigma_{\mathrm{A}} + (1-x)\sigma_{\mathrm{B}}\\ &= x\left[\frac{\alpha_{\mathrm{A}}}{{}^{3}\xi_{\mathrm{A}}} + \beta_{\mathrm{A}}\,{}^{1}\xi_{\mathrm{A}} + \gamma_{\mathrm{A}}\right] + (1-x)\left[\frac{\alpha_{\mathrm{B}}}{{}^{3}\xi_{\mathrm{B}}} + \beta_{\mathrm{B}}\,{}^{1}\xi_{\mathrm{B}} + \gamma_{\mathrm{B}}\right]. \qquad (9)\end{aligned}$$

The surface tension σ_m of the mixture should also be expressible by an expression analogous to Eq. (5),

$$\sigma_m = \frac{\alpha_m}{{}^{3}\xi_m} + \beta_m\,{}^{1}\xi_m + \gamma_m\,. \qquad (10)$$

If it is assumed that

$${}^{3}\xi_m = x\,{}^{3}\xi_{\mathrm{A}} + (1-x)\,{}^{3}\xi_{\mathrm{B}}\,, \qquad (11)$$

$$\beta_m({}^{1}\xi_m) = x\,\beta_{\mathrm{A}}({}^{1}\xi_{\mathrm{A}}) + (1-x)\,\beta_{\mathrm{B}}({}^{1}\xi_{\mathrm{B}}), \qquad (12)$$

$$\gamma_m = x\gamma_{\mathrm{A}} + (1-x)\gamma_{\mathrm{B}}\,, \qquad (13)$$

then the excess surface tension (σ^E) for a mixture is given by

$$\sigma^E = \alpha_{\mathrm{AB}}\left[\frac{1}{x\,{}^{3}\xi_{\mathrm{A}} + (1-x)\,{}^{3}\xi_{\mathrm{B}}} - \frac{x}{{}^{3}\xi_{\mathrm{A}}} - \frac{(1-x)}{{}^{3}\xi_{\mathrm{B}}}\right], \qquad (14)$$

where $\alpha_{\mathrm{AB}} = \alpha_{\mathrm{A}} = \alpha_{\mathrm{B}} = \alpha$.

This approach, however, cannot be used for the mixtures of the type alkanol (A) + alkanol (B) since alkanols are known to be associated in the pure state, and their degree of association is not known with certainty. The ${}^{3}\xi$ parameter of alkanol, therefore, cannot be evaluated in the pure state. The calculation of ${}^{3}\xi$ values in (A + B) mixtures is all the more difficult. In one of our earlier V^E studies [7] on quinoline + alkanols ($C_1 - C_{10}$), it has been shown that methanol exists mainly as a cyclic dimer and ethanol as an open chain trimer in the pure state. In these mixtures ${}^{3}\xi$ values of alkanols are quite different in the pure and in the mixed state.

Table 2. Experimental and calculated values of excess surface tension σ^E/dyn cm^{-1} at $T = 298.15$ K

S. No.	Mixture	σ^E	x_1				Ref.
			0.2	0.3	0.7	0.8	
1.	Cyclopentane	Calc.	−0.07	−0.09	−0.08	−0.06	
	+ CCl_4	Expt.	−0.03	−0.05	−0.12	−0.10	[8]
		(Calc.)^a	−0.05	−0.08	−0.08	−0.06	
[$^3\xi_A = 1.25$, $^3\xi_B = 1.0$, $\alpha_{AB} = 9.00$, $K = 2.849$]							
2.	Cyclopentane +	Calc.	−0.78	−0.96	−0.75	−0.54	
	Benzene	Expt.	−0.83	−0.98	−0.79	−0.61	[8]
		(Calc.)^a	−0.65	−0.84	−0.84	−0.64	
[$^3\xi_A = 1.25$, $^3\xi_B = 0.667$, $\alpha_{AB} = 9.40$, $K = -3.3256$]							
3.	Cyclopentane +	Calc.	−0.33	−0.42	−0.38	−0.28	
	Toluene	Expt.	−0.35	−0.44	−0.36	−0.26	[8]
		(Calc.)^a	−0.32	−0.40	−0.39	−0.30	
[$^3\xi_A = 1.25$, $^3\xi_B = 0.94$, $\alpha_{AB} = 25.30$, $K = -6.225$]							
4.	Cyclohexane +	Calc.	−0.63	−0.83	−0.82	−0.62	
	Tetrachloro-	Expt.	−0.74	−0.92	−0.73	−0.52	[8]
	ethylene	(Calc.)^a	−0.63	−0.82	−0.82	−0.63	
[$^3\xi_A = 1.5$, $^3\xi_B = 1.449$, $\alpha_{AB} = 4817.6$, $K = -1.89886$]							
5.	Cyclohexane +	Calc.	−0.82	−0.98	−0.72	−0.52	
	Benzene	Expt.	−0.77	−0.95	−0.71	−0.50	[8]
		(Calc.)^a	−0.71	−0.93	−0.93	−0.70	
[$^3\xi_A = 1.5$, $^3\xi_B = 0.667$, $\alpha_{AB} = 6.92$, $K = -1.73654$]							
6.	Cyclohexane +	Calc.	−0.58	−0.72	−0.60	−0.44	
	Toluene	Expt.	−0.53	−0.69	−0.62	−0.46	[8]
		(Calc.)^a	−0.51	−0.66	−0.65	−0.50	
[$^3\xi_A = 1.5$, $^3\xi_B = 0.940$, $\alpha_{AB} = 17.08$, $K = -1.36962$]							
7.	Cyclohexane +	Calc.	0.05	0.07	0.09	0.07	
	cis-Decalin	Expt.	0.06	0.08	0.08	0.06	[8]
		(Calc.)^a	0.06	0.08	0.08	0.06	
[$^3\xi_A = 1.5$, $^3\xi_B = 2.966$, $\alpha_{AB} = -1.68$, $K = -0.49451$]							
8.	Cyclohexane +	Calc.	0.06	0.09	0.12	0.10	
	trans-Decalin	Expt.	0.08	0.10	0.10	0.08	[8]
		(Calc.)^a	0.10	0.11	0.11	0.08	
[$^3\xi_A = 1.5$, $^3\xi_B = 2.966$, $\alpha_{AB} = -2.24$, $K = -1.61074$]							
9.	Benzene + *o*-Xylene	Calc.	−0.07	−0.09	−0.08	−0.06	
		Expt.	−0.03	−0.05	−0.12	−0.10	[8]
		(Calc.)^a	−0.07	−0.08	−0.09	−0.07	
[$^3\xi_A = 0.667$, $^3\xi_B = 0.94$, $\alpha_{AB} = 2.97$, $K = 1.0$]							
10.	Methanol + Decanol	Calc.	1.27	1.77	2.31	1.91	
		Expt.	1.20	1.76	2.08	1.57	[9]
		(Calc.)^a	1.96	2.22	2.06	1.44	
[$^3\xi_A = 1.0$, $^3\xi_B = 2.012$, $\alpha_{AB} = -28.22$, $K = 13.344$]							

Table 2 (continued)

S. No.	Mixture	σ^E	x_1				Ref.
			0.2	0.3	0.7	0.8	
11.	Ethanol + Decanol	Calc.	1.00	1.39	1.82	1.50	
		Expt.	1.06	1.45	1.70	1.34	[9]
		(Calc.)^a	1.21	1.57	1.57	1.19	
$[{}^3\xi_A = 1.0,\ {}^3\xi_B = 2.012,\ \alpha_{AB} = -22.17,\ K = 21.124]$							
12.	Propanol + Decanol	Calc.	0.67	0.93	1.22	1.01	
		Expt.	0.73	0.99	1.12	0.88	[9]
		(Calc.)^a	0.88	1.08	1.06	0.80	
$[{}^3\xi_A = 1.0,\ {}^3\xi_B = 2.012,\ \alpha_{AB} = -14.84,\ K = 14.049]$							
13.	Isopropanol + Decanol	Calc.	0.76	1.06	1.39	1.15	
		Expt.	0.80	1.10	1.31	1.03	[9]
		(Calc.)^a	0.94	1.22	1.21	0.91	
$[{}^3\xi_A = 1.0,\ {}^3\xi_B = 2.012,\ \alpha_{AB} = -16.96,\ K = 54.1667]$							
14.	Butanol + Decanol	Calc.	0.36	0.52	0.84	0.76	
		Expt.	0.49	0.64	0.64	0.49	[9]
		(Calc.)^a	0.52	0.66	0.65	0.49	
$[{}^3\xi_A = 0.512,\ {}^3\xi_B = 2.012,\ \alpha_{AB} = -1.77,\ K = 9.8972]$							
15.	Methanol + Butanol	Calc.	0.23	0.28	0.22	0.16	
		Expt.	0.19	0.25	0.25	0.19	[9]
		(Calc.)^a	0.21	0.26	0.25	0.19	
$[{}^3\xi_A = 1.0,\ {}^3\xi_B = 0.512,\ \alpha_{AB} = -1.90,\ K = 3.9295]$							
16.	Benzene + Ethanol	Calc.	−0.10	−0.13	−0.16	−0.12	
		Expt.	−0.08	−0.13	−0.10	−0.09	[10]
		(Calc.)^a	−0.05	−0.08	−0.08	−0.06	
$[{}^3\xi_A = 0.667,\ {}^3\xi_B = 1.0,\ \alpha_{AB} = 3.44,\ K = -4.9708]$							
17.	Cyclohexane + Ethanol	Calc.	−0.51	−0.64	−0.55	−0.40	
		Expt.	−0.56	−0.68	−0.54	−0.41	[10]
		(Calc.)^a	−0.61	−0.68	−0.62	−0.47	
$[{}^3\xi_A = 1.5,\ {}^3\xi_B = 1.0,\ \alpha_{AB} = 21.15,\ K = -1.2994]$							
18.	Methylcyclohexane + Ethanol	Calc.	−0.30	−0.36	−0.28	−0.26	
		Expt.	−0.23	−0.30	−0.33	−0.26	[10]
		(Calc.)^a	−0.31	−0.35	−0.33	−0.26	
$[{}^3\xi_A = 1.894,\ {}^3\xi_B = 1.0,\ \alpha_{AB} = 5.20,\ K = 0.9124]$							
19.	Benzene + *t*-Pentylalcohol	Calc.	−0.57	−0.77	−0.85	−0.67	
		Expt.	−0.47	−0.70	−1.12	−0.97	[10]
		(Calc.)^a	−0.73	−0.86	−0.84	−0.68	
$[{}^3\xi_A = 0.667,\ {}^3\xi_B = 0.865,\ \alpha_{AB} = 43.43,\ K = -1.6326]$							
20.	Toluene + *t*-Pentylalcohol	Calc.	−0.40	−0.52	−0.51	−0.38	
		Expt.	−0.47	−0.56	−0.45	−0.28	[10]
		(Calc.)^a	−0.50	−0.56	−0.53	−0.45	
$[{}^3\xi_A = 0.94,\ {}^3\xi_B = 0.865,\ \alpha_{AB} = 320.65,\ K = -1.3889]$							

Table 2 (continued)

S. No.	Mixture	σ^E	x_1				Ref.
			0.2	0.3	0.7	0.8	
21.	Cyclohexane +	Calc.	0.37	0.46	0.37	0.27	
	t-Pentylalcohol	Expt.	0.27	0.39	0.48	0.40	[10]
		(Calc.)[a]	0.32	0.42	0.41	0.29	
$[{}^3\xi_A = 1.5,\ {}^3\xi_B = 0.865,\ \alpha_{AB} = -7.28,\ K = 0.86957]$							
22.	Methylcyclohexane +	Calc.	−0.22	−0.26	−0.19	−0.14	
	t-Pentylalcohol	Expt.	−0.18	−0.25	−0.15	−0.10	[10]
		(Calc.)[a]	−0.20	−0.25	−0.22	−0.16	
$[{}^3\xi_A = 1.894,\ {}^3\xi_B = 0.865,\ \alpha_{AB} = -2.26,\ K = 0.64436]$							
23.	Cyclohexane +	Calc.	−3.04	−3.20	−1.79	−1.23	
	Nitroethane	Expt.	−3.05	−3.44	−1.67	−1.10	[11]
		(Calc.)[a]	−1.37	−2.10	−2.21	−1.66	
$[{}^3\xi_A = 1.5,\ {}^3\xi_B = 0.258,\ \alpha_{AB} = 2.42,\ K = -3.3423]$							
24.	Methylcyclohexane +	Calc.	−4.00	−4.10	−2.19	−1.49	
	Nitroethane	Expt.	−4.05	−4.46	−2.00	−1.21	[11]
		(Calc.)[a]	−2.36	−3.00	−2.89	−2.10	
$[{}^3\xi_A = 1.894,\ {}^3\xi_B = 0.258,\ \alpha_{AB} = 2.67,\ K = -6.4639]$							
25.	Benzene +	Calc.	−0.45	−0.53	−0.37	−0.26	
	Nitroethane	Expt.	−0.70	−0.59	−0.55	−0.42	[11]
		(Calc.)[a]	−0.61	−0.58	−0.50	−0.40	
$[{}^3\xi_A = 0.667,\ {}^3\xi_B = 0.258,\ \alpha_{AB} = 0.984,\ K = 17.627]$							

[a] Values calculated using Eq. (17)

In view of the above limitations Eq. (14) may be written as

$$\sigma^E = \alpha_{AB}\left[\frac{1}{{}^3\xi_m} - \frac{x}{{}^3\xi_A} - \frac{(1-x)}{{}^3\xi_B}\right], \tag{15}$$

where ${}^3\xi_m$ is the ${}^3\xi$ of the mixture and is not given by Eq. (11). Since no method is available for the evaluation of α_{AB} and ${}^3\xi_m$ for these mixtures from the ${}^3\xi$ values of pure components, we have tried an alternative approach.

It has earlier [7] been shown that V^E can be expressed as

$$V^E = \alpha\left[\frac{1}{{}^3\xi_m} - \frac{x}{{}^3\xi_A} - \frac{(1-x)}{{}^3\xi_B}\right], \tag{16}$$

which gives

$$\frac{\sigma^E}{V^E} = \frac{\alpha_{AB}}{\alpha} = K$$

or

$$\sigma^E = K\,V^E. \tag{17}$$

Eq. (17) provides a very simple relationship between σ^E and V^E values of a binary mixture.

Results and Discussion

It is clear from Eq. (14) that computation of σ^E requires a knowledge of α_{AB} for the mixture. This was calculated by fitting $\sigma^E(x = 0.5)$ data to Eq. (14) and this value was then employed to compute $\sigma^E(x = 0.2$, 0.3 and 0.7, 0.8) for the same mixtures. The computation of σ^E has been done for several binary mixtures. Table 2 lists the comparison between calculated and experimental values (taken from literature) for these very binary mixtures. A close scrutiny of Table 2 reveals that the agreement between experimental and calculated σ^E values is reasonably well.

The prediciton of σ^E values of a binary mixture from Eq. (17) requires a knowledge of the K parameter. In the present study, K was computed by substituting σ^E and V^E values at $x = 0.5$. The computed value of K was then used alongwith V^E data at any mole fraction to calculate σ^E values for the mixtures at the same mole fraction. Such σ^E values at $x = 0.2, 0.3, 0.7, 0.8$ are presented (in parenthesis) in Table 2. The required V^E data were taken from the literature [7–10]. It is clear from Table 2 that Eq. (17) does describe satisfactorily σ^E values of all the listed binary mixtures.

It may be concluded that in mixtures where computation of $^3\xi$ of both the components is possible, Eq. (14) can be used to evaluate σ^E of the mixtures. On the other hand Eq. (17) can be used universally for computing σ^E of binary mixtures at any composition provided V^E data at that composition is available, the plus point being that it can be applied also in cases where Eq. (14) is not applicable.

References

[1] Jain D. V. S., Gombar V. K. (1975) J. Chem. Soc. Faraday I **75**: 1132
[2] Singh P. P. (1983) Thermochim. Acta **66**: 37
[3] Barysz M., Plavsic D., Trinajstic N. (1986) Math. **19**: 89
[4] Randić M. (1975) J. Am. Chem. Soc. **97**: 6609
[5] Kier L. B., Hall L. H. (1975) Molecular Connectivity Chemistry and Drug Research. Academic Press, New York
[6] Riddick J. A., Bunger W. B. (1970) Organic Solvents: Physical Properties and Methods of Purification, 3rd edn. Wiley-Interscience, New York
[7] Dewan R. K., Madaan S., Mehta S. K. (in press) J. Soln. Chem.
[8] Benson G. C., Lam V. T. (1970) Can. J. Chem. **48**: 3773
[9] Benson G. C., Lam V. T. (1972) J. Colloid. Interface Sci. **38**: 294
[10] Myers R. S., Clever H. L. (1974) J. Chem. Thermodyn. **6**: 949
[11] Myers R. S., Angel G. P., Clever H. L. (1976) J. Colloid. Interface Sci. **3**: 443

Received November 2, 1989. Accepted February 28, 1990