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EMPIRICAL PARAMETERS OF LEWIS ACIDITY AND BASICITY FOR AQUEOUS BINARY SOLVENT MIXTURES

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Abstract - Empirical parameters of Lewis acidity, E_T^N, introduced by Reichardt et al., and Lewis basicity, B_{KT}, introduced by Kamlet and Taft, have been determined for mixtures of water with ten organic solvents. In the case of water/alcohol mixtures a distinct dependence between these acidity and basicity parameters have been found. For the other solvent mixtures the E_T^N on B_{KT} dependence is more complex even if these parameters are purified from non-specific solute/solvent interactions.

Various empirical solvent polarity parameters have often been used fairly effectively to describe the influence of solvents on physicochemical solute properties of different kinds.[1] One particularly useful set consists of complementary Lewis acid/base solvent parameters,[2-5] now known for most common organic solvents. Recently, more and more attention has been paid to binary solvent mixtures in this connexion.[6-8] It was found for example, by analysis of the variation of Kamlet-Tafts's B_{KT}-values (or β-values)[9] for mixtures of methanol and other organic solvents, that an important factor influencing the basicity of solvent mixtures is due to order/disorder processes; particularly in binary mixtures of protic with non-hydrogen bond donor (non-HBD) solvents.[8,10] The aim of this paper is to investigate the solvation behaviour of aqueous binary solvent mixtures by means of the normalized Lewis acidity parameter E_T^N [11] and the Lewis basicity parameter B_{KT} (resp. β).[9]

Results and Discussions

The E_T^N- and B_{KT}-parameters for ten mixtures, water and an organic solvent, are given in Table 1. The normalized, dimensionless Lewis acidity parameters E_T^N were calculated from E_T-values (in kcal/mol; determined by means of a solvatochromic pyridinium-N-phenoxide betaine dye[11]) according to equation (1):[11b]

$$E_T^N = [E_T(\text{solvent}) - E_T(\text{TMS})]/[E_T(\text{water}) - E_T(\text{TMS})] \qquad (1)$$

In those cases where a comparison was possible, the E_T^N-values of this paper are in reasonably good agreement with the previously published data.[11b] The Lewis basicity parameters B_{KT} will be published more detailed in another paper.[10]

SOLVENTS	MOL-% OF H_2O:	0	10	20	30	40	50	60	70	80	90	100
METHANOL (MEOH)												
E_T^N		0.77	0.77	0.78	0.79	0.80	0.81	0.82	0.84	0.88	0.92	1.00
B_{KT}		0.62	0.60	0.54	0.55	0.54	0.53	0.50	0.43	0.36	0.26	0.19
ETHANOL (ETOH)												
E_T^N		0.66	0.66	0.67	0.69	0.70	0.71	0.73	0.74	0.78	0.87	1.00
B_{KT}		0.77	0.76	0.71	0.68	0.66	0.63	0.60	0.57	0.51	0.31	0.19
1-PROPANOL (1-PROH)												
E_T^N		0.61	0.62	0.63	0.64	0.65	0.66	0.68	0.69	0.70	0.74	1.00
B_{KT}		0.86	0.78	0.77	0.75	0.71	0.68	0.66	0.64	0.60	0.53	0.19
2-PROPANOL (2-PROH)												
E_T^N		0.54	0.56	0.58	0.60	0.62	0.64	0.66	0.67	0.70	0.77	1.00
B_{KT}		0.88	0.89	0.83	0.79	0.75	0.72	0.69	0.65	0.60	0.49	0.19
ACETONITRILE (AN)												
E_T^N		0.46	0.64	0.70	0.73	0.75	0.77	0.79	0.81	0.83	0.89	1.00
B_{KT}		0.37	0.40	0.43	0.44	0.42	0.40	0.40	0.41	0.39	0.34	0.19
DIMETHYLSULFOXIDE (DMSO)												
E_T^N		0.44	0.46	0.48	0.52	0.55	0.58	0.63	0.69	0.75	0.85	1.00
B_{KT}		0.76	0.79	0.77	0.70	0.66	0.62	0.56	0.50	0.45	0.35	0.19
N,N-DIMETHYLFORMAMIDE (DMF)												
E_T^N		0.40	0.44	0.47	0.52	0.56	0.59	0.63	0.68	0.74	0.83	1.00
B_{KT}		0.72	0.70	0.69	0.69	0.68	0.64	0.61	0.58	0.50	0.41	0.19
ACETONE (A)												
E_T^N		0.36	0.45	0.53	0.59	0.62	0.65	0.68	0.71	0.74	0.82	1.00
B_{KT}		0.54	0.54	0.54	0.55	0.56	0.57	0.54	0.51	0.47	0.42	0.19
TETRAHYDROFURAN (THF)												
E_T^N		0.20	0.36	0.43	0.50	0.53	0.56	0.59	0.61	0.64	0.71	1.00
B_{KT}		0.54	0.49	0.45	0.43	0.43	0.41	0.43	0.47	0.52	0.55	0.19
1,4-DIOXANE (D)												
E_T^N		0.16	0.28	0.35	0.43	0.50	0.54	0.60	0.62	0.69	0.77	1.00
B_{KT}		0.39	0.45	0.50	0.55	0.54	0.51	0.50	0.48	0.45	0.36	0.19

Table 1. Lewis acidity parameters, E_T^N, and Lewis basicity parameters, B_{KT}, for mixtures of water with ten other organic solvents.

When the measured values of E_T^N and B_{KT} were plotted one against another, the ten lines in Fig. 1 were obtained. It is immediately apparent that for five binary mixtures a reasonably good linear mutual dependence does in fact exist (correlation coefficient $|r| \geqslant 0.981$ for n = 11 data points; cf. Table 1).

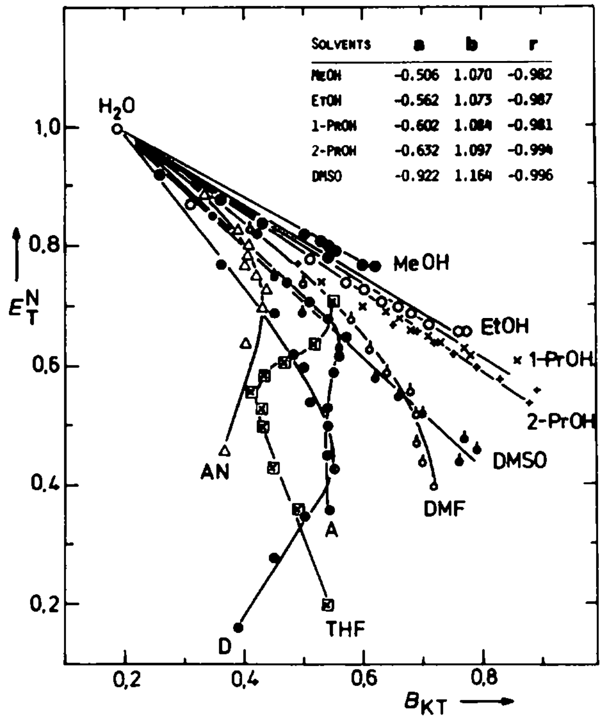

Fig. 1. Dependence of E_T^N-values on B_{KT}-parameters for binary mixtures of water with ten organic solvents (cf. Table 1): MeOH (●), EtOH (o), 1-PrOH (x), 2-PrOH (+), DMSO (♁), DMF (δ) A (⊕), D (⊗), THF (⊠), and AN (▲). The figures are correlation coefficients for those five solvent mixtures with a linear mutual dependence according to $E_T^N = a \cdot B_{KT} + b$. For solvent abbreviations cf. Table 1.

The negative slope of some lines in Fig. 1 indicates that some kind of a formal similarity with Brønsted acid/base equilibria exists: an increase of the Lewis basicity parameter B_{KT} is associated with a decrease of the Lewis acidity parameter E_T^N. However, this holds only for mixtures of water with solvents of high relative permittivity (ε_r>30). For other binary mixtures it is at best applicable only for a limited mole fraction range. These deviations from the „similarity" become even more pronounced for binary mixtures between methanol and other organic solvents.[12] In order to explain these phenomena, an analysis of the nonspecific solute/solvent contributions to E_T^N and B_{KT} is carried out.

Nonspecific contributions to E_T^N

The separation of the specific and nonspecific contributions to the empirical solvent parameter $E_T(30)$ (measured in kcal/mol[11]), carried out by Koppel and Pal'm[13], led to a definition of an electrophilicity parameter E [E = $E_T(30)$ - const. - nonspecific terms, according to equation (2)]

$$E = E_T(30) - \underset{(\pm 1.32)}{25.57} - \underset{(\pm 1.11)}{14.39} \cdot Y - \underset{(\pm 4.58)}{9.08} \cdot P \qquad (2)$$

(multiple regression coefficient R = 0.950 for n = 21 solvents;
Y and P are measures of polarization and polarizability)

which is assumed to be free from nonspecific solute/solvent interactions, reflecting only the Lewis acid/base interactions between solute and solvent. This electrophilicity parameter E is, however, closely correlated to the original $E_T(30)$-values, at least for solvents with an $E_T(30)$-value of >42 kcal/mol.[14] Reexamination of this analysis for all available data [using E_T^N instead of $E_T(30)$] leads to the results as presented in Fig. 2 where the dependence of Koppel and Pal'm's E, now called E^N, on normalized E_T^N [cf. equation (1)] is shown.

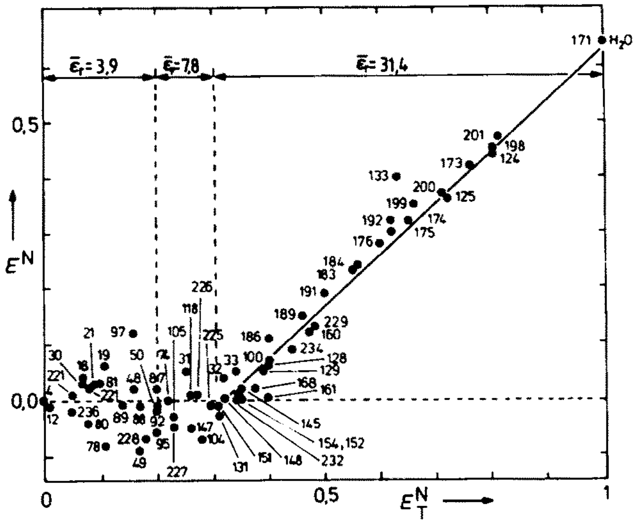

Fig. 2. Dependence of E^N-values on E_T^N-parameters[11b] for 66 organic solvents. The E^N-values are Lewis acidity parameters separated from nonspecific solute/solvent interactions according to Koppel and Pal'm[13]. Solvent numbering as in reference[13a]. Cf. equation (4).

It was assumed that equation (3) describes the contributions due to nonspecific solute/solvent interactions (R = 0.983):

$$E_T^N = \underset{(\pm 0.058)}{1.186} \cdot \frac{\varepsilon_r - 1}{2\varepsilon_r + 1} - \underset{(\pm 0.154)}{0.050} \cdot \frac{n^2 - 1}{n^2 + 2} + \underset{(\pm 0.046)}{0.212} \qquad (3)$$

Equation (3) was calculated with the data of twenty organic solvents (no. 4, 6, 12, 21, 30, 32, 50, 62, 74, 80, 87, 88, 89, 104, 105, 145, 147, 151, 152, and 168[x]) in the following way: at first, the dependence of E_T^N on $f(\varepsilon_r)$ and $f'(n^2)$ was calculated for solvents having E_T^N-values lower than 0.5. Next, the values of the recalculated E_T^N(calc.) were compared using the obtained $E_T^N \pm f(\varepsilon_r, n^2)$ relationship and the experimental E_T^N-values. Solvents for which the difference [E_T^N(calc.) - E_T^N(exp.)] was higher than 0.05 were omitted, and a new $E_T^N \pm f(\varepsilon_r, n^2)$ equation was calculated. This procedure was repeated until the difference [E_T^N(calc.) - E_T^N(exp.)] was less than 0.05 for all solvents.

From the results presented in Fig. 2 it is seen that for solvents with $E_T^N > 0.3$ according to equation (4) a good linear regression holds:

$$E^N = 0.985 \cdot E_T^N \pm 0.432 \tag{4}$$

A correlation coefficient of r = 0.988 for n = 30 data points was found. For the solvents on the regression line, the mean relative permittivity $\bar{\varepsilon}_r$ is equal to 31.4. The generally deviating points are for solvents with $\varepsilon_r \ll \bar{\varepsilon}_r$, e.g. for acetic acid (solvent no. 133; $\varepsilon_r = 6.15$). The other solvents which are not well described by equation (4) possess low ε_r-values with $\bar{\varepsilon}_r = 7.8$ and $\bar{\varepsilon}_r = 3.9$ (cf. Fig. 2).

The main conclusion of this analysis is in agreement with our former results[14] but better confirmed by the much larger group of solvents taken into account. It means, that for solvents of lower relative permittivity ($\varepsilon_r \leqslant 10$) the specific contributions to the solute/solvent interactions are of negligible importance. The reverse is true for solvents with $\varepsilon_r \geqslant 10$, for which the nonspecific contributions seem to be negligible. This conclusion is in agreement with the correlation analysis using the Born function ($1/\varepsilon_r$) or the Kirkwood function [$(\varepsilon_r-1)/(2\varepsilon_r+1)$] as solvent parameters[14,15].

Nonspecific contributions to B_{KT}

In principle, a similar procedure can be applied to the Lewis basicity parameter B_{KT}.[9] For solvents with $B_{KT} < 0.45$ (the selection of solvents was carried out as mentioned before), plotting their B_{KT}-values against $f(\varepsilon_r)$ and $f'(n^2)$, equation (5) is obtained:

$$B_{KT} = \underset{(\pm 0.04)}{1.80} \cdot \frac{\varepsilon_r - 1}{2\varepsilon_r + 1} - \underset{(\pm 0.13)}{1.28} \cdot \frac{n^2 - 1}{n^2 + 2} + \underset{(\pm 0.03)}{0.04} \tag{5}$$

A correlation coefficient of R = 0.997 for n = 17 data points (solvent no. 3, 4, 6, 8, 9, 12, 13, 30, 88, 89, 103, 104, 145, 146, 147, 168, and 232 [x]) was found.

A Lewis basicity parameter B^N, free from nonspecific solute/solvent interactions, can now be defined as in equation (6):

$$B^N = B_{KT} - 1.80 \cdot \frac{\varepsilon_r - 1}{2\varepsilon_r + 1} + 1.28 \cdot \frac{n^2 - 1}{n^2 + 1} + 0.04 \tag{6}$$

---

[x] The numbering of solvents is the same as in the review of Koppel and Pal'm.[13a]

The B_{KT}-values have been plotted against the B^N-values (Fig. 3) in the same way as for the Lewis acidity parameters. The picture obtained is evidently different from that for the acidity parameters.

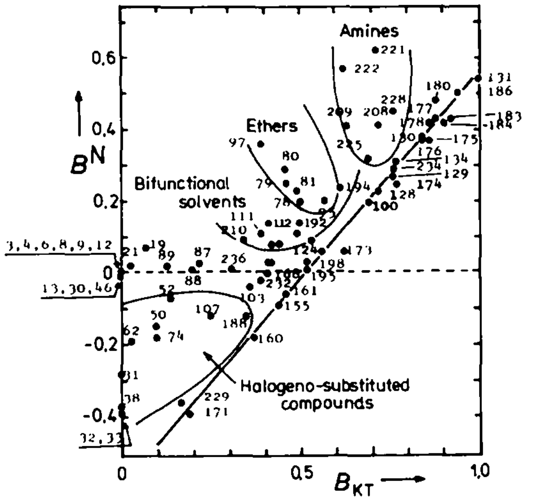

Fig. 3. Dependence of B^N-values on B_{KT}-parameters[9] for 78 organic solvents. The B^N-values are Lewis basicity parameters separated from nonspecific solute/solvent interactions according to equation (6). Solvent numbering as in reference[13a].

The solvents chosen by their basicity properties as weakly interacting do in fact lie closely to $B^N = 0 \pm 0.05$. The other solvents, however, are distributed in a complex way (cf. Fig. 3): some of them form clusters located above the line with positive slope (amines, aliphatic ethers, and bifunctional compounds such as aromatic esters, ketones, and alcohols), some of these solvents lie even below $B^N = 0$ (mainly halogeno alkanes and halogeno arenes). Other solvents are located closely to the slanting line, for example some aliphatic alcohols (e.g. no. 173: methanol; no. 174: ethanol; no. 198: glycol).

For a selected set of solvents, the slanting line of Fig. 3 refers to equation (7), the correlation coefficient of which is r = 0.994 for n = 26 pure

$$B^N = 1.147 \cdot B_{KT} - 0.589 \qquad (7)$$

organic solvents. Two facts should be noted: First of all, the solvents on the line given by equation (7) are mainly alcohols. For alcohols and some other solvents, mainly due to the fact that $\varepsilon_r > 10$ for these solvents, the correction calculated by equation (6) is approximately constant and equal to 0.470 (standard deviation s = 0.055). Secondly, the remarkably good B^N on B_{KT} dependence for the selected set of solvents, would suggest that, the basicity of these solvents is not only a function of ε_r and n^2.

The difference between Figs. 2 and 3 lies in the following: For E_T^N-values >0.3 there is a very good linear relationship between E^N and E_T^N [cf. equation (4)] irrespective of the nature of the solvents. In the case of the B^N vs. B_{KT} plot, a good mutual dependence exists only for a selected set of solvents, whereas other solvents (aliphatic and aromatic amines, ethers, as well as aromatic esters, ketones, and alcohols) deviate considerably from the regression line given by equation (7). Below $B^N = 0$ but still on the line, there are the solvents water (no. 171), acetonitrile (no. 160), and nitromethane (no. 229).

For solvents with $B^N = o$, the variation in B_{KT} depends mainly on the variation in $f(\varepsilon_r)$ and $f'(n^2)$ [cf. equation (6)]. These solvents involve principally nonspecific solute/solvent interactions. For solvents fulfilling equation (7), i.e. solvents for which $B^N = 1.15 \cdot B_{KT} - 0.59$ operates, one would expect both nonspecific and specific contributions to B_{KT}, with the nonspecific term being nearly constant.

For solvents deviating from the line described by equation (7) and given in Fig. 3, additional specific interactions causing an increase in basicity can be assumed. Frequently, these solvents are either bifunctional compounds, aromatic compounds or ethers.

The solvents deviating from the line and lieing below $B^N = 0$ (cf. Fig. 3), are halogeno-substituted alkanes and arenes and form a separate group. For them, the nonspecific solute/solvent interactions have - according to equation (6)- large and negative contributions, whereas B_{KT} is approximately zero.

The location of water in Fig. 3 (solvent no. 171) is more unexpected: its B_{KT}-value is with 0.19 very low and hence a negative deviation from the line defined by $B^N = 0$ results.

However, one should realize that this complicated behaviour was only obtained for the B_{KT}-parameters. For other Lewis basicity parameters the situation may be different. A similar correlation analysis for the B-parameter of Koppel and Pal'm[13a] (B_{KP}; based on the solvent-induced shift from $\tilde{\nu}_{OD}$ of CH_3OD) does not give the same picture, since there are not enough data points available to set up an equation such as $B_{KP} = f(\varepsilon_r, n^2)$ with an acceptable precision.

Notwithstanding, it was interesting to compare the results presented in Fig. 3 with a corresponding $B_{KP} = f(B_{KT})$ dependence.

In both cases, the solvents which deviate most, are amines, halogeno-substituted alkanes and arenes, and ethers. In all these cases, the B_{KT}-values are lower than the corresponding B_{KP}-values. Thus, except for solvents which fulfill equation (7), the position of the other solvents may differ depending on the Lewis basicity parameter used. This can result from the structural differences of the solute molecules involved in establishing the Lewis basicity scales. Due to their great structural variety (e.g. B_{KT}: 4-nitroaniline and N,N-diethyl-4-nitroaniline; B_{KP}: CH_3OD), the possible variation in the $f(n^2)$-term may be different.

The differences between the Lewis acidity and basicity parameters, E_T^N and B_{KT}, as described by equations (3) and (5) lies in the much greater contribution of the $f'(n^2)$-term in explaining the B_{KT}-values. A possible explanation is: The acidity parameters are determined by means of standard substrates which itself are Lewis bases (e.g. E_T^N: a pyridinium-N-phenoxide betaine dye). Thus, their interactions with the acidic centers of the surrounding solvent molecules occur with solvent sites which are by definition scarce in electron density. Hence, the polarizability term, $f'(n^2)$, is less important for the solute/solvent interaction.

The situation is quite different for the determination of Lewis basicity parameters. In this case, the standard solute molecules are Lewis acids (e.g. B_{KT}: the NH_2-group of 4-nitroaniline; B_{KP}: the OD-group of CH_3OD; Gutmann's donor number: $SbCl_5$) and they are sensitive to changes in the electron density of those solvent sites which are rich in electrons, i.e. more or less negatively charged. Solvent molecules with such basic properties i.e. rich in electrons are easier polarizable. Thus, the Lewis basicity parameters describing their solvation properties are more strongly dependent on their polarizability.

The E^N on B^N dependence

Fig. 4 exhibits the mutual dependence of the Lewis acid/base parameters E^N (as defined by Koppel and Pal'm[13]) and B^N [as defined by equation (6)], respectively, for all analyzed organic solvents.

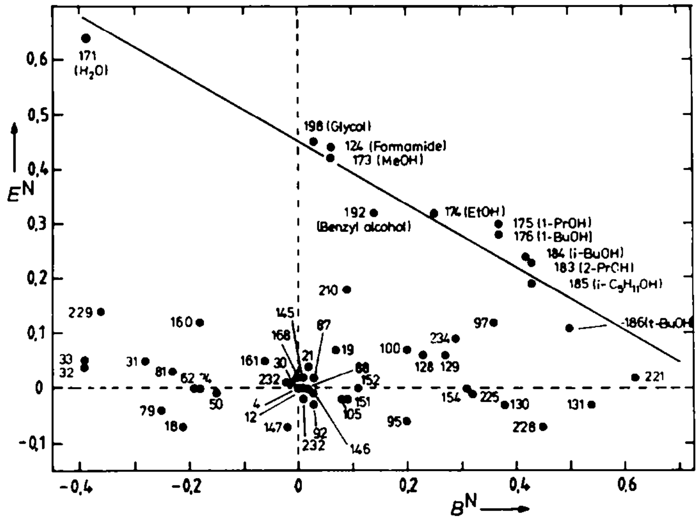

Fig. 4. Dependence of E^N-values on B^N-values for 56 organic solvents. The regression line for a selected set of twelve solvents is described by equation (8). Solvent numbering as in reference [13a].

It follows from the pattern presented in Fig. 4 that there is a group of solvents for which the acidity and the basicity parameters, separated from the nonspecific solute/solvent interactions, are mutually related. For twelve solvents (i.e. water, glycol, formamide, methanol, benzyl alcohol, ethanol, 1- and 2-propanol, 1-butanol, i-butanol, i-pentanol, and t-butanol) equation (8) with a correlation coefficient of r = -0.969 was observed. According to this equation, an increase in solvent basicity is correlated with a decrease in solvent acidity

$$E^N = -0.537 \cdot B^N - 0.448 \qquad (8)$$

for this particular set of solvents.

The position of two other solvent groups of Fig. 4 is not clear: (i) generally halogeno-substituted compounds with $B^N<0$, and (ii) generally amines, amides, and solvents with similar structures, shifted towards lower B^N-values. For another group of fifteen solvents, the E^N- and B^N-values are close to zero. It is possible that for this solvent group no specific solute/solvent interactions can be expected.

In conclusion, the mutual Lewis acidity/basicity dependence clearly holds, in fact, only for alcohols as solvents. For this solvent group, the nonspecific solute/solvent contributions to E_T^N and B_{KT} are both constant in practice. The similar mutual Lewis acidity/basicity observed for mixtures of alcohols with water (cf. Fig. 1) seems to be now understandable.

The interpretation of the E_T^N on B_{KT} dependence found for the mixtures of other organic solvents with water requires further investigations.

Conclusions

It was found, that, the Lewis basicity parameter B_{KT} [9] and the Lewis acidity parameter E_T^N [11], both separated from nonspecific solute/solvent contributions and now called B^N and E^N respectively, make it possible to divide organic solvents into four different groups: (a) a group for which the nonspecific solute/solvent contributions are nearly constant ($\varepsilon_r > 10$). For this solvent group a clear mutual Lewis acidity/basicity dependence was found, suggesting that these solvents behave similar to the Brønsted type properties for aqueous solutions; (b) a group of solvents for which both the B^N and E^N parameter are mainly dependent on nonspecific solute/solvent interactions; (c) a group of mainly halogeno-substituted compounds with $B^N < 0$; and (d) a group of solvents including anilines, amides, and dimethylsulfoxide. The mutual B^N and E^N dependence for the two latter groups is not certain. The physical interpretation of this grouping of organic solvents is not clear at present.

EXPERIMENTAL

The E_T(30-values have been determined by means of the standard betaine dye 2,6-diphenyl-4-(2,4,6-triphenyl-1-pyridino)phenoxide, the synthesis of which is given in reference [11a].

The B_{KT}-values were measured with commercially available 4-nitroaniline (Fluka AG, Buchs) and N,N-diethyl-4-nitroaniline, the latter synthesized according to Vogel.[16]

The solvents used were commercially products purified by standard procedures.[17] These involved thorough drying and fractional distillation.

The UV/Vis spectra were recorded with an UV/Vis Specord spectrophotometer (Carl Zeiss, Jena, GDR) with an approximate precision of 40 cm^{-1}.

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