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THERMODYNAMIC PARAMETERS OF IONIZATION OF 5-(2-SUBSTITUTED PHENYLAZO) BARBITURIC ACIDS

MAMDOUH S. MASOUD *, S.A. ABOU ALI, G.Y. ALI and I.M. ABED

Chemistry Department, Faculty of Science, Alexandria (Egypt)

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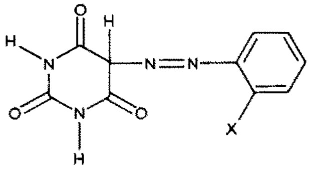
ABSTRACT

The acid dissociation constants of 5-(2-substituted phenylazo) barbituric acids were evaluated in 20–75% (v/v) dioxane–water mixtures potentiometrically. The acidity increased with decreasing dioxane percentage. The mechanism of dissociation was based on aquation and solvation properties. The ΔH values increased linearly with increase of the percentage of dioxane. The thermodynamic parameters were affected by the electronic character of the substituents and the nature of the hydrogen bonding.

INTRODUCTION

Azo compounds have attracted much attention because of their use as models of biological systems. The understanding of the biological action of a molecule requires a chemical and physicochemical knowledge of the molecule. In our laboratory, Masoud et al. [1–16] published a series of papers to throw light on the chemistry of azo compounds and their complexes. The series explored many important points but many questions still remain unanswered.

The new 5-(2-substituted phenylazo) barbituric acid compounds are the subject of this paper. The main purpose is three-fold: (i) evaluation of the acid dissociation constants potentiometrically in 20–75% (v/v) dioxane–water mixtures; (ii) explanation of the mechanism of dissociation in solvation and aquation; and (iii) evaluation of the thermodynamic parameters, based on the electronic character of the substituent and the nature of the hydrogen bond.



$-X = -H, -OH, -COOH, -CH_3, -OCH_3$ or $-NO_2$

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* Author to whom correspondence should be addressed.

EXPERIMENTAL

All the ligands were prepared in a similar manner. 0.1 mol of the amine (aniline, *o*-nitro aniline, *o*-anisidine, anthranilic acid and *o*-amino phenol) was dissolved in concentrated HCl (12 ml) and distilled water (34 ml). The amine hydrochloride was diazotized below 5 °C with a solution of $NaNO_2$ (7 g in 20 ml distilled water). The diazonium chloride was coupled with an alkaline solution of barbituric acid (12.8 g in 10% NaOH). The compound formed was collected by filtration from 50% acetic acid solution. The compound was crystallized from 75% (v/v) dioxane–water mixtures and dried under vacuum. The analytical data are shown in Table 1.

The dioxane solvent and the nitrogen gas and the standard KOH solution were prepared as usual [17]. The ligand solutions were prepared by dissolving the required weights in 75% (v/v) dioxane–water mixture at room temperature.

The electrometric titration runs were carried out by taking 50 ml of the ligand solution in 75% (v/v) dioxane–water mixture with an ionic strength of 0.1 M KNO_3 in a thermostatted cell under nitrogen atmosphere to exclude CO_2. The double-walled glass titration cell was provided with inlet and outlet tubes with a circulating water device at constant temperature (± 0.1 °C) from a Haake N2B ultrathermostat. After each addition of standard carbonate-free KOH titrant, the titrated solution was stirred using a stream of purified nitrogen gas and allowed to stand to attain equilibrium before recording the pH using a precision Beckman model 4500 digital pH meter reading to ± 0.002 pH unit, connected to a combined electrode. The pH meter was calibrated from time to time by means of standard buffer solutions of pH = 4 and pH = 7.

TABLE 1

Analytical data for 5-(2-substituted phenylazo) barbituric acid compounds

Substituent	Colour and appearance	% Calculated (% found)		
		C	H	N
-H	Pale yellow [a]	51.7 (51.5)	3.2 (3.1)	24.1 (23.9)
-OH	Orange [a]	47.8 (47.9)	2.9 (3.0)	20.3 (20.0)
-COOH	Pale brown [b]	48.4 48.1	3.2 (3.4)	22.6 (22.3)
$-NO_2$	Orange [a]	43.3 (43.0)	2.5 (2.8)	25.3 (25.0)
$-OCH_3$	Deep orange [a]	50.4 (50.3)	3.8 (3.9)	21.1 (21.5)
$-CH_3$	Orange [a]	53.6 (53.5)	4.1 (4.1)	22.8 (22.4)

[a] Microcrystalline. [b] Powder.

RESULTS AND DISCUSSION

The titration curves of barbituric acid, (phenylazo) barbituric acid and its o-NO_2 and o-OCH_3 compounds show one sharp inflection point at $a = 1$ (a is the number of mol of alkali added per one mol of the ligand), i.e. only one proton is dissociated. The curves of o-carboxy and o-hydroxy compounds show two separate inflections at $a = 1$ and $a = 2$ where two protons are available for ionization.

The dissociation of a ligand may be described in the following equation

$$H_nL = H^+ + H_{n-1}L^- \qquad K_1 = \frac{[H^+][H_{n-1}L]}{[H_nL]} \tag{1}$$

$$H_{n-1}L^- = H^+ + H_{n-2}L^{2-} \qquad K_2 = \frac{[H^+][H_{n-2}L^{2-}]}{[H_{n-1}L^-]} \tag{2}$$

The mass balance for the first step is

$$C^0_{H_nL} = [H_nL] + [H_{n-1}L^-] \tag{3}$$

Where: $C^0_{H_nL}$ is the initial concentration of the ligand while $[H_nL]$ and $[H_{n-1}L^-]$ are the concentrations of the non-dissociated ligand and its anion existing in equilibrium, respectively. The electroneutrality of the system implies that:

$$aC_{H_nL} + [H^+] = [H_{n-1}L^-] + [OH^-] \tag{4}$$

a is defined as the neutralization degree

$$a = \frac{V_{KOH}N_{KOH}}{V_{ligand}M_{ligand}}$$

N and M are the normality of KOH and molarity of the ligand, respectively. V_{KOH} is the volume of the added KOH and V_{ligand} is the initial volume of the titrated ligand. Subtracting eqn. (3) from eqn. (4)

$$[H_{n-1}L^-] = aC_{H_nL} + [H^+] + [OH^-] \tag{5}$$

$$[H_nL] = C_{H_nL}(1 - a) - [H^+] + [OH^-] \tag{6}$$

Substituting these concentrations in eqn. (2)

$$K_1 = \frac{[H^+](aC_{H_nL}) + [H^+] - [OH^-]}{C_{H_nL}(1 - a) - [H^+] + [OH^-]} \tag{7}$$

The second dissociation constant of H_2L may be determined from the following equilibrium

$$H_{n-1}L^- = H_{n-2}L^{2-} + H^+ \tag{8}$$

$$K_2 = \frac{[H_{n-2}L^{2-}][H^+]}{[H_{n-1}L^-]} \tag{9}$$

For the second buffer zone in the titration curve the following equations could express the material balance and electroneutrality

$$C_{H_{n-1}L^-} = [H_{n-1}L^-] + [H_{n-2}L^{2-}] \qquad (10)$$

$$aC_{H_{n-1}L^-} + [H^+] = [H_{n-1}L^-] + 2[H_{n-2}L^{2-}] + [OH^-] \qquad (11)$$

$$[H_{n-1}L^-] = (2-a)C_{H_{n-1}L^-} - [H^+] + [OH^-] \qquad (12)$$

$$[H_{n-2}L^{2-}] = (a-1)C_{H_{n-1}L^-} + [H^+] - [OH^-] \qquad (13)$$

$$K_2 = \frac{[H^+]\{(a-1)C_{H_{n-1}L^-} + [H^+] - [OH^-]\}}{(2-a)C_{H_{n-1}L^-} - [H^+] + [OH^-]} \qquad (14)$$

Most of the titrations were carried out in non-aqueous media (20, 40, 50, 55, 60, 70 and 75% (v/v) dioxane–water mixtures). The readings of the pH meter used for determination of $[H^+]$ were corrected based on the relation [18]

$$-\log[H^+] = B + \log \mu_H^0 - \log(1/\gamma^2) \qquad (15)$$

where B is the pH-meter reading, μ_H^0 is the correction factor at zero ionic strength for the solvent composition under consideration, and γ is the activity coefficient of the solvent composition.

The $\log \mu_H^0$ and $\log(1/\gamma^2)$ values were evaluated as reported [18–20] (Table 2).

The $[OH^-]$ in eqns. (7) and (14) ought to be calculated using the values of pK_w in different percentage (v/v) dioxane–water solutions. The pK_w value of water in 50% (v/v) dioxane–water solution was 15.83 and had a higher value as the dioxane concentration was increased [18]. The higher pK_w value rendered the $[OH^-]$ very small and could be neglected compared to the other terms in eqns. (7) and (14).

TABLE 2

The correction factors for dioxane–water mixtures

Percentage (v/v) dioxane–water	$N_2^\star$	$\log \mu_H^0$	$\log(1/\gamma^2)$
20	0.049	0.020	0.145
40	0.123	0.090	0.235
50	0.174	0.180	0.325
55	0.2045	0.270	0.375
60	0.239	0.400	0.460
65	0.281	0.560	0.545
70	0.329	0.800	0.680
75	0.387	1.090	0.830

$N_2^\star$ is the mole fraction of the dioxane–water system.

Barbituric acid gives only one inflection point where only one proton is titrated. The mechanism of ionization of the acid is due to the active methylene group located at position 5 flanked by two carbonyl groups. Comparison of acidity for barbituric acid with that of phenylacetone (both possess the $CO-CH_2-CO$ radical) revealed that the acidity of the former (3.92) is much higher than the latter (8.95) [21]. In the case of barbituric acid, the presence of electron-attracting imino groups leads to a decrease of the charge density on the carbonyl groups, which leads to higher acidity. The reverse is true for the phenylacetone which contains an electron-donating methyl group.

*Effect of solvents on the p*K *values*

1. The pK values of barbituric acid in dioxane–water mixtures are higher than in pure water, and increase in a non-linear fashion with the increase in proportion of dioxane in the solvent. Increase of water percentage in the presence of dioxane decreases the extent of intramolecular hydrogen bonding, facilitating the ionization of H^+ and leading to a decrease in the pK value.

2. The pK values of *o*-(carboxy phenylazo) barbituric acid increase with the increase in solvent composition. The linear mode of pK_1–% solvent relationship may be attributed to the intramolecular hydrogen bonding existing between the *o*-COOH and the N=N groups, which increases the stability of the compound [5]. The non-linear mode for the pK_2–% solvent relationship is probably due to association.

3. The pK values of *o*-NO_2, *o*-OH, *o*-CH_3, *o*-OCH_3 and *o*-phenylazo barbituric acid compounds increase with the increase in the percentage of dioxane–water solvent composition in a linear fashion. The pK–% solvent relationships for all compounds are presented in Fig. 1 and collected in Tables 3 and 4.

In aqueous medium, the dissociation of a ligand may be described by the following equation

$$HL(aq) \rightleftharpoons H^+(aq) + L^-(aq) + JH_2O \tag{16}$$

The dissociation constant K' for eqn. (16) is given in the following [22]

$$K' = K[H_2O]^J \tag{17}$$

In mixed solvent the dissociation of a ligand may be described by the following equation

$$HL(xH_2O,\ bS) \rightleftharpoons H^+(yH_2O,\ aS) + L^-(zH_2O,\ tS) + dS + wH_2O \tag{18}$$

The dissociation constant is given by the following equations

$$K^{\star\prime} = \frac{[H^+][L^-]}{[HL]}[S]^d[H_2O]_s^w \tag{19}$$

$$K^{\star\prime} = K^\star[S]^d[H_2O]_s^w \tag{20}$$

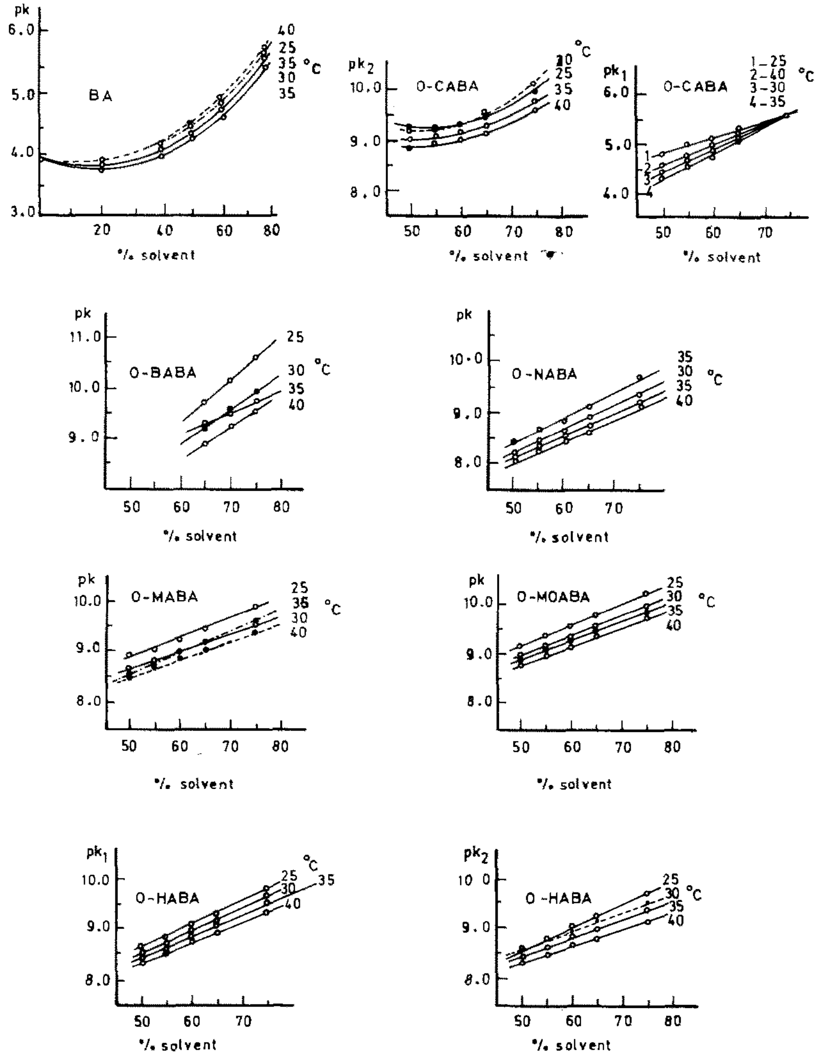

Fig. 1. pK–% solvent relationship for the free ligands at different temperatures (25–40 °C).

where

$$K^{\star} = \frac{[\mathrm{H^+}][\mathrm{L^-}]}{[\mathrm{HL}]} \quad \text{in co-solvent} \tag{21}$$

In aqueous solution b, a, t and d are all zero, and will be represented in this case by a different symbol, J.

The stoichiometric equilibrium constants (concentration quotients) K and K' in mixed solvent and water, respectively, can be related to the standard free energy of transfer and to the apparent equilibrium constants as follows

$$\log K^{\star\prime} - \log K' = -\Delta G^{t}_{\mathrm{R}}/2.3RT \tag{22}$$

TABLE 3

Thermodynamic parameters of barbituric acid (pK ± 0.007)

Percentage (v/v) dioxane–water	Barbituric acid						
	pK				$\Delta G_{25^\circ\mathrm{C}}$ (kcal mol^{-1})	ΔH (kcal mol^{-1})	ΔS (e.u.)
	25 °C	30 °C	35 °C	40 °C			
0	3.924	3.869	3.942	3.959	5.332	−1.824	−24.013
20	3.902	3.857	3.712	3.942	5.302	8.755	11.587
40	4.247	4.215	4.023	4.204	5.771	10.639	16.335
50	4.498	4.349	4.276	4.517	6.112	10.943	16.211
60	4.876	4.787	4.632	4.892	6.626	11.340	15.818
75	5.733	5.669	5.454	5.720	7.790	15.199	24.862

$$\log K^{\star\prime} - \log K' = \log K^\star - \log K + \log \frac{[\mathrm{H_2O}]_s^w [\mathrm{S}]^a}{[\mathrm{H_2O}]^j} \tag{23}$$

$$\frac{-\Delta G_\mathrm{R}^t}{2.3RT} = \log K^\star - \log K + \log \frac{[\mathrm{H_2O}]_s^w [\mathrm{S}]^d}{[\mathrm{H_2O}]^j} \tag{24}$$

where ΔG_R^t is the standard molar free energy of transfer of the reactive species from water to the mixed solvent. It is commonly held that water molecules are preferentially solvated in mixed solvent [23]. If the co-solvent is essentially inactive with respect to solvation in reaction (18), $d = 0$, so

$$\log K^\star - \log K = -\Delta G_\mathrm{R}^t/2.3RT - W - \log[\mathrm{H_2O}]_s + J \log[\mathrm{H_2O}]$$

and if we assume in the case that $W = J$

$$\log K^\star - \log K = -\Delta G_\mathrm{R}^t/2.3RT - \log\{[\mathrm{H_2O}]_s \mathrm{H_2O}\} \tag{25}$$

Suppose that J represents a solvent transfer number characteristic to a reaction, but part of this number should be attributed to the transfer of the co-solvent, thus $J = w + d$

$$\log K^\star - \log K' = \log K^\star - \log K + \log \frac{[\mathrm{H_2O}]_s^w}{[\mathrm{H_2O}]^s} \cdot \frac{[\mathrm{S}]^j}{[\mathrm{S}]^w} \tag{26}$$

$$\log K^{\star'} - \log K' = \log K^\star - \log K + \log \left[\frac{[\mathrm{S}]}{[\mathrm{H_2O}]}\right]^j \cdot \left[\frac{[\mathrm{H_2O}]_s}{[\mathrm{S}]}\right]^w \tag{27}$$

$$(J \log[\mathrm{S}] + \log K^\star) - \log K = -\frac{\Delta G_\mathrm{R}^t}{2.3RT} - w \log \frac{[\mathrm{H_2O}]_s}{[\mathrm{S}]} + J \log[\mathrm{H_2O}] \tag{28}$$

Trial values of $J = 1, 2, 3, 4$ are used to find the values of w. For simplicity put $(J \log[\mathrm{S}] + \log K^\star) = Y$ and $\log([\mathrm{H_2O}]_s/[\mathrm{S}]) = X$, then plot Y vs. X

(Fig. 2). From the straight line, W values are evaluated. The data are collected in Table 5.

The data obtained may throw light on the role of aquation and solvation during the course of dissociation. In the case of o-NO_2(phenylazo) barbituric acid, the W values are higher than that of J to pinpoint more aquation. In the cases of o-CH_3, o-OCH_3 and o-OH, the W values are slightly smaller than that of J, which suggests more solvation.

Thermodynamic parameters (Tables 3 and 4)

This could serve as a measure of the strength of the hydrogen bonding that exists in the ligands.

TABLE 4

Thermodynamic parameters of the azo ligands

Percentage (v/v) dioxane–water	H						
	pK				$\Delta G_{25\,°C}$ (kcal mol^{-1})	ΔH (kcal mol^{-1})	ΔS (e.u.)
	25 °C	30 °C	35 °C	40 °C			
65	9.715	9.450	9.272	8.844	13.201	22.08	+29.79
70	10.130	9.553	9.501	9.282	13.765	24.84	+37.16
75	10.607	9.906	9.763	9.547	14.413	30.36	+63.51

Percentage (v/v) dioxane–water	o-OH						
	pK ^{a}				$\Delta G_{25\,°C}$ (kcal mol^{-1})	ΔH (kcal mol^{-1})	ΔS (e.u.)
	25 °C	30 °C	35 °C	40 °C			
50	8.679	8.545	8.454	8.304	11.793	8.28	−11.78
	(10.557)	(10.611)	(10.287)	(10.287)	14.345	5.59	−29.70
55	8.777	8.725	8.635	8.526	11.927	9.20	−9.15
	(10.741)	(10.739)	(10.612)	(10.470)	14.596	8.28	−21.19
60	9.075	8.984	8.847	8.785	12.332	11.50	−2.82
	(11.066)	(10.988)	(10.841)	(10.718)	15.037	11.01	−13.41
65	9.277	9.168	9.087	8.892	12.606	13.80	+4.01
	(11.215)	(11.173)	(11.004)	(10.777)	15.239	13.80	−4.83
75	9.803	9.623	9.475	9.279	13.321	16.56	+10.87
	(11.662)	(11.485)	(11.292)	(11.089)	15.847	15.64	−0.69

Percentage (v/v) dioxane–water	o-NO_2						
	pK				$\Delta G_{25\,°C}$ (kcal mol^{-1})	ΔH (kcal mol^{-1})	ΔS (e.u.)
	25 °C	30 °C	35 °C	40 °C			
50	8.496	8.247	8.233	8.099	11.545	9.20	−7.87
55	8.696	8.478	8.382	8.288	11.817	11.04	−2.61
60	8.848	8.608	8.488	8.480	12.023	13.80	+5.96
65	9.095	8.906	8.751	8.626	12.359	16.56	+14.10
75	9.799	9.375	9.250	9.150	13.315	23.00	+32.50

TABLE 4 (continued)

Percentage (v/v) dioxane–water	o-CH_3						
	pK				$\Delta G_{25\,°C}$ (kcal mol^{-1}	ΔH (kcal mol^{-1})	ΔS (e.u.)
	25 °C	30 °C	35 °C	40 °C			
50	8.953	8.655	8.587	8.586	12.166	2.76	−31.50
55	9.058	8.765	8.743	8.749	12.309	4.60	−25.87
60	9.206	8.858	8.815	8.862	12.509	6.44	−20.36
65	9.477	9.018	8.846	9.030	12.878	9.20	−12.34
75	9.976	9.563	9.603	9.415	13.556	12.88	−2.27

Percentage (v/v) dioxane–water	o-COOH						
	pK				$\Delta G_{25\,°C}$ (kcal mol^{-1})	ΔH (kcal mol^{-1})	ΔS (e.u.)
	25 °C	30 °C	35 °C	40 °C			
50	4.767	4.372	4.438	4.484	6.477	–	−21.737
	(9.204)	(9.246)	(9.036)	(8.838)	12.507	6.670	−19.587
55	4.965	4.708	4.639	4.668	6.747	–	−22.640
	(9.362)	(9.304)	(8.147)	(8.943)	12.722	8.280	−14.906
60	5.046	4.782	4.867	4.942	6.897	–	−23.009
	(9.306)	(9.281)	(9.211)	(9.036)	12.645	9.246	−11.406
65	5.368	5.054	5.011	5.160	7.294	–	−24.477
	(9.552)	(9.433)	(9.281)	(9.112)	12.979	11.040	−6.507
75	5.466	5.577	5.627	5.586	7.427	−3.218	−35.732
	(10.460)	(9.981)	(9.786)	(9.597)	14.214	28.520	+14.306

Percentage (v/v) dioxane–water	o-OCH_3						
	pK				$\Delta G_{25\,°C}$ (kcal mol^{-1})	ΔH (kcal mol^{-1})	ΔS (e.u.)
	25 °C	30 °C	35 °C	40 °C			
50	9.090	8.854	8.866	8.818	12.352	7.36	−16.75
55	9.358	9.067	8.086	8.995	12.716	10.12	−8.71
60	9.475	9.279	9.135	9.068	12.875	11.96	−3.07
65	9.734	9.547	9.390	9.250	13.227	13.80	+1.92
75	10.145	9.834	9.733	9.703	13.785	16.56	+9.31

[a] Values in parentheses are pK_2 values.

1. The ΔH values increase with the increase in the percentage of dioxane, in linear functions in the order H $<$ o-NO_2 $<$ o-OH $<$ o-OCH_3 $<$ o-CH_3. This could be explained on the basis that the ortho effect and the steric hindrance of the groups produce this trend. The o-CH_3 group is larger than the H which hinders the hydrogen bond strength leading to more acidity (small pK) and greater ΔH values. The trend for the ΔH value for the o-NO_2 group is higher than that of the o-CH_3 group, but still less than the H, again due to the size factor. The intramolecular hydrogen bonding gives rise to a positive entropy change whereas the hydrogen bonding to solvent leads to a higher degree of solvent ordering with a negative entropy change

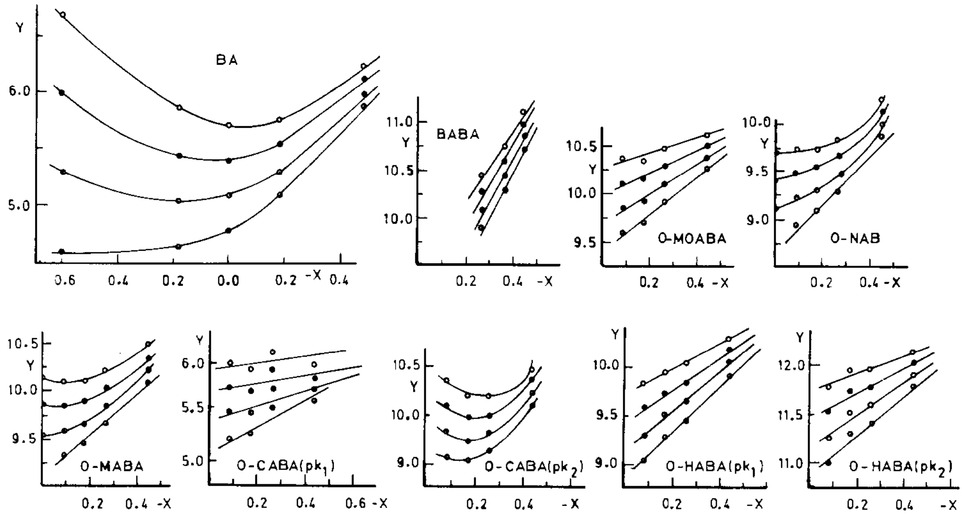

Fig. 2. X–Y relationship for the free ligands at different percentages of dioxane–water mixtures. Symbols: ○———○, $J=4$; ●———●, $J=3$; ⊙———⊙, $J=2$; ◒———◒, $J=1$.

[24]. The hydrogen bonding property is marked in the case of the unsubstituted azo compound (H) leading to a positive entropy change. The negative ΔS values for *o*-OH and *o*-CH_3 compounds suggest that the dioxane solvent affects their behaviour; thus, solute–solvent interaction in both cases is important. In the case of the *o*-NO_2 and *o*-OCH_3 compounds both interactions coexist. Again, the difficulty in the solubility of the H compound and its higher positive ΔS value support arguments concerning

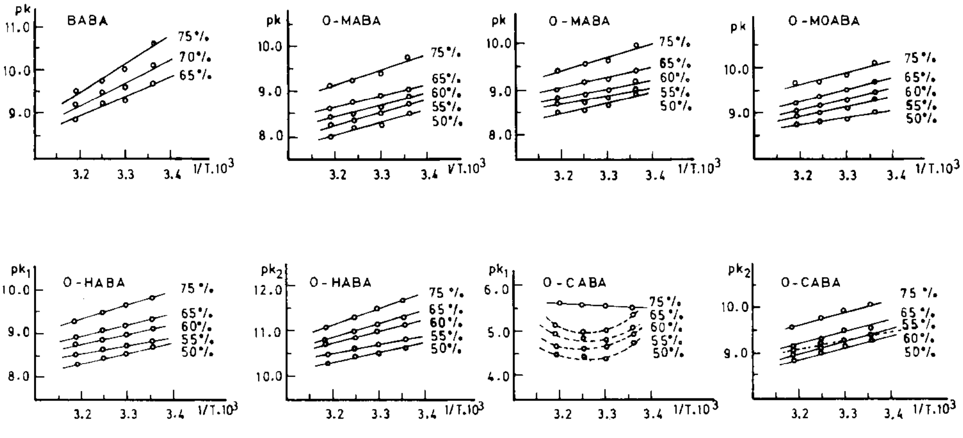

Fig. 3. pK–$1/T$ relationship for the free ligands at different dioxane–water mixtures.

TABLE 5

The values of W at different values of J

Ligand	J	W [a]
o-OH	1	1.5 (2.0)
	2	1.75 (1.5)
	3	2.0 (1.25)
	4	2.08 (1.0)
o-COOH	1	1.32
	2	0.66
	3	0.5
	4	0.38
o-OCH_3	1	2.0
	2	1.5
	3	1.3
	4	0.75
H	1	4.75
	2	4.50
	3	4.00
	4	3.75
o-NO_2	1	2.75

[a] Values in parentheses are pK_2 values.

the effect of the hydrogen bonding [26]. Figure 3 shows the pK–$1/T$ relationship for all compounds.

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