SPECTROPHOTOMETRIC STUDY OF THE EFFECT OF SODIUM DODECYL SULPHATE ON THE ACID-BASE BEHAVIOUR OF 4-(2-PYRIDYLAZO)-RESORCINOL

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The values of the apparent dissociation constants of the azo dye 4-(2-pyridylazo)-resorcinol (PAR) have been found in the presence of sub- and supercritical concentrations of the anionogenic tenside sodium dodecyl sulphate (SDS) and the effects of the strong electrolytes NaCl and NaNO$_3$ were also evaluated. In both electrolytes, the pK_{an} values initially are independent of the SDS concentration and then increase. Spectrophotometric and surface tension measurements were employed to determine the critical micelle concentration (cmc) of SDS alone and in the presence of PAR and the given electrolytes. The determined cmc values were in the range $0.20-3.02$ mmol . $. l^{-1}$ and depended on the type and concentration of the particular additive.
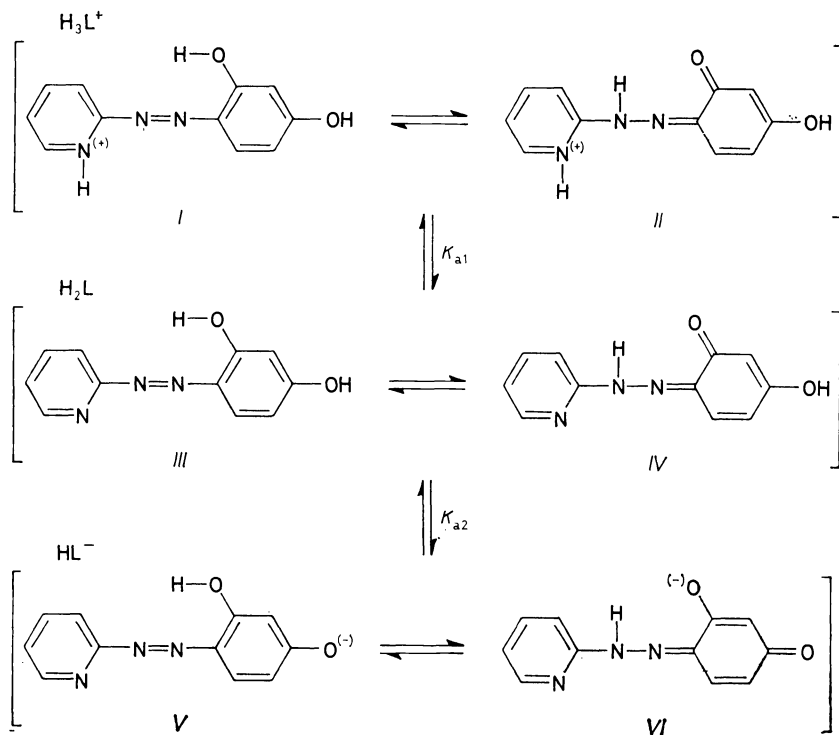
4-(2-pyridylazo)-resorcinol (PAR) is one of the best known and most widely used azo dyes and its acid-base and complexing properties have been studied by a number of authors[1-6]. The mechanism of PAR dissociation is depicted in Scheme 1 (ref.[6]).

The electronic absorption spectra of PAR exhibit characteristic maxima for the individual dissociated forms of the dye: at pH < 1.5 in aqueous PAR solutions, a yellow colour appears, corresponding to the tautomeric forms I and II (Scheme 1) of the protonated dye, H$_3$L$^+$ ($\lambda_{1,\text{max}} = 389$ nm, $\varepsilon_1 = 2.0 \cdot 10^4$ mol^{-1} l cm^{-1}; $\lambda_{2,\text{max}} = 455$ nm, $\varepsilon_2 = 1.59 \cdot 10^4$ mol^{-1} l cm^{-1}). It can be assumed, in agreement with the literature[7], that the short-wavelength maxima of the individual forms of the *o*- and *p*-hydroxazo dye can be assigned to the azo–enol tautomer and that the ketohydrazo form corresponds to the absorption maximum at longer wavelengths. Thus, form I corresponds to the band with $\lambda_{1,\text{max}}$ and form II to the maximum $\lambda_{2,\text{max}}$.

At pH 4 to 5, it can be generally assumed that the electroneutral form H$_2$L exists in two tautomeric forms III and IV (Scheme 1). As H$_2$L has only a single strong absorption maximum, $\lambda_{\text{max}} = 383$ nm, in the visible spectral region, it can be assumed that one of the tautomeric forms will predominate. NMR studies have indicated that this absorption band corresponds to the azo form III (ref.[8]).

The HL$^-$ form of the dye is most important in complexing reactions; this form predominates in aqueous solutions at pH 6.5 to 11. HL$^-$ is assumed to exist as the

hydrazo form VI (Scheme 1) (ref.[9]) with $\lambda_{max} = 411$ nm and $\varepsilon_{max} = 3.45 . 10^4$ mol^{-1} . . l cm^{-1}.

H_3L^+

I   *II*

K_{a1}

H_2L

III   *IV*

K_{a2}

HL^-

V   *VI*

SCHEME 1

The unstable azo form L^{2-} ($\lambda = 488$ nm) appears in the alkaline region at pH > 13 (ref.[10]). The relationship between the absorption spectrum of the PAR dye and the pH of the solution is depicted in Fig. 1.

The dissociation constants of the individual forms of the dye pK_{an} in the absence of the tensides have been found by a number of authors[11-14]; for example, Russeva et al.[14] give $pK_{a1} = 3.02 \pm 0.02$, $pK_{a2} = 5.56 \pm 0.01$ and $pK_{a3} = 11.98 \pm 0.02$. The effect of some cationogenic tensides (CPB, Septonex) on these dissociation constant values was studied in an earlier work[15].

In this work, spectrophotometric methods are employed to study changes in the pK_{a1} and pK_{a2} values for the PAR dye in dependence on the concentration of the anionogenic tenside sodium dodecyl sulphate (SDS), $pK_{an} = f(c_T)$, below and above the critical micelle concentration (cmc) and the effect of the presence of various concentrations of strong electrolytes NaCl and NaNO$_3$. The results obtained are

then employed to determine the cmc values for tenside SDS and changes in this value in the presence of the given salts.

EXPERIMENTAL

Instruments and Chemicals

The spectrophotometric measurements were carried out on a PU 8800 recording spectrophotometer (Pye Unicam, Cambridge, Great Britain), with quartz cuvettes with an internal pathlength of 1·00 cm. The pH values were measured on a PHM 64 instrument (Radiometer, Copenhagen, Denmark) with a combined GK 2401B electrode. The surface tension of the tenside was measured using a tensiometer from the Lauda company (F.R.G.). All the measurements were carried out at a temperature of $(25 \pm 0.5)°C$.

The stock PAR solutions with a concentration of $c_{PAR} = 1·25 . 10^{-3}$ mol l^{-1} were prepared from the pure substance from Lachema (Brno, Czechoslovakia) by dissolving in 2 ml of 0·2 mol . $. l^{-1}$ NaOH and diluting with distilled water. The PAR purity was controlled by elemental analysis and thin-layer chromatography.

The stock SDS solutions with a concentration of $c_{SDS} = 5·0 . 10^{-2}$ mol l^{-1} was prepared by dissolving the pure substance from Sandoz (Switzerland). The purity of the substance was verified by elemental analysis; the height of the minimum on the curve of the dependence of the surface tension on the tenside concentration was not greater than $5 . 10^{-5}$ N m^{-1}.
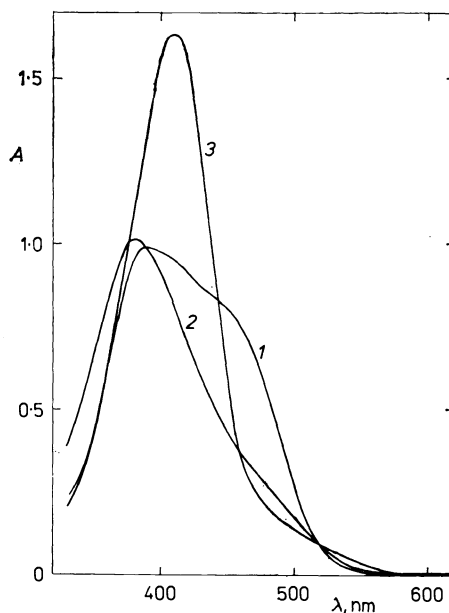

FIG. 1

Absorption spectra of the dye PAR. $c_{PAR} = = 5 . 10^{-5}$ mol l^{-1}; $I = 0·1$ mol l^{-1} (NaNO₃); pH values for the curves: 1 1·09 (the H_3L^+ form), 2 4·17 (the H_2L form), 3 9·50 (the HL^- form)

Procedure

The PAR dye was studied at a concentration, $c_{PAR} = 5 \cdot 0 \cdot 10^{-5}$ mol l^{-1}. The cmc value for a pure tenside solution is given in the literature as $6 \cdot 6$ mmol l^{-1}; values of $2 \cdot 24$ and $2 \cdot 43$ mmol . . l^{-1} are given in the presence of $c(NaCl) = 0 \cdot 05$ mol l^{-1} and $c(NaNO_3) = 0 \cdot 04$ mol l^{-1}, respectively ($t = 20°C$) (see ref.[16]). Consequently, the final SDS concentration in solution was in the range $c_{SDS} = 2 \cdot 10^{-4}$ to $6 \cdot 10^{-3}$ mol l^{-1}.

The conditional dissociation constants of PAR were found by the spectrophotometric method[17,18] with an increasing or decreasing section of the sigmoid dependence $A = f(pH)$. The pK_{a1} and pK_{a2} values were determined for $\lambda = 460$ nm and 411 nm, respectively. The experimental data was treated mathematically[19]. The standard deviation values for $pK_{a1,2}$ were not greater than $\pm 0 \cdot 04$ (found from 4 to 7 measurements).

The acidity of the solutions for pK_{an} measurements was adjusted by addition of $0 \cdot 2$M-HCl (HNO$_3$) or $0 \cdot 2$M-NaOH. The ionic strength of the solutions for the individual measurements in the whole pH range was maintained at a value of $I(NaCl)$ (or $I(NaNO_3)) = 0 \cdot 05$, $0 \cdot 1$, $0 \cdot 2$, $0 \cdot 3$ and $0 \cdot 5$ mol l^{-1} by addition of a $2 \cdot 0$ mol l^{-1} NaCl solution (or NaNO$_3$ solution). The spectrophotometric measurements were carried out within 30 min after mixing the solutions. Because of the low stability of the alkaline form L^{2-} in solution, which did not improve even after addition of the tenside, the pK_{a3} value was not determined.

The surface tension was found by a method involving tearing of a platinum ring from the liquid surface. The maximum strength acting on the ring during lifting from the liquid surface was found. The measured surface tension is then found from the relationship[20]: $\gamma = F/2o$, where γ is the surface tension, F is the maximum force acting on the ring and o is the circumference of the platinum ring. The tensiometer is calibrated directly in surface tension units, γ, with a precision of $\pm 3 \cdot 10^{-6}$ N m^{-1} and was corrected according to Harkins and Jordan[20] using the relationship: $\gamma' = f \cdot \gamma$, where γ is the measured surface tension, γ' is the corrected surface tension and f is the Harkins–Jordan coefficient corresponding to the weight of the column of liquid that is simultaneously drawn above the surface of the liquid with the ring. The coefficient value is found by linear interpolation of tabulated values for the liquid density and column height.

The critical micelle concentration of the tenside was found from these values for the surface tension of the solutions at various tenside concentrations, as the intercept of the two linear parts of the dependence $\gamma' = f(\log c_T)$.

RESULTS

The addition of anionogenic tenside SDS to a solution of reagent PAR at various pH values leads to interactions between these substances, affecting the electronic structures of the individual dissociated forms of the dye, especially the H_3L^+ and H_2L forms, with changes in the absorption in the visible spectral region. It follows from Figs 2 and 3 that the presence of SDS at concentrations of $c_{SDS} <$ cmc has no basic effect on the character of the spectra of the protonated (H_3L^+, Fig. 2) and neutral (H_2L, Fig. 3) forms of the reagent PAR. However, the presence of the micellar forms of SDS ($c_{SDS} >$ cmc) leads to an increase in the absorption maxima of both dye forms at $\lambda \approx 460$ nm (also corresponding to a bathochromic shift in the presence of the tenside micelles of about 5 nm), with a simultaneous decrease in the shorter wavelength maximum at $\lambda = 389$ nm for H_3L^+ or $\lambda = 383$ nm for H_2L. The indivi-

dual absorption curves intercept in the isosbestic point at $\lambda_{ip} = 421$ nm (Fig. 2) or 412 nm (Fig. 3).

The absorption spectra of the dissociated form HL^- are practically not affected by the tenside over the whole concentration interval studied, c_T, with an absorption decrease of about 4 to 6% overall.

Figure 4 depicts the effect of the addition of a strong electrolyte on the absorption spectra of the H_3L^+ for in the presence of SDS; an increase in the ionic strength of the $NaNO_3$ solution in the PAR–SDS system at $c_T \approx cmc$ (practically no effect on the PAR spectra was observed at $c_T < cmc$) leads to an increase in the longer wavelength maximum corresponding to micelle solubilization of the hydrazo form II $(\lambda_{max} \approx 460$ nm). The absorption curves intercept in the isosbestic point at $\lambda_{ip} = = 421$ nm.

Figure 5 depicts an example of the effect of addition of an $NaNO_3$ solution on the spectra of the H_2L form in the presence of SDS. This effect is not obvious at $c_T < cmc$ (similarly as for H_3L^+); however in the micelle region of SDS concentrations a gradual increase in $I(NaNO_3)$ leads to suppression of the solubilization of the H_2L form by the strong electrolyte, i.e. a decrease is observed in the absorbance at $\lambda_{max} \approx \approx 460$ nm with a simultaneous increase in the absorption at $\lambda = 383$ nm.

Analogous effects have also been observed for solutions of the PAR–SDS system whose ionic strength was adjusted by addition of an NaCl solution.
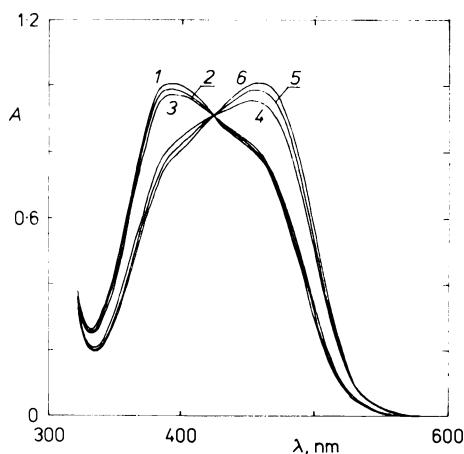

FIG. 2

The effect of SDS on the absorption spectra of PAR. pH 1·51; $I = 0·1$ mol l^{-1} ($NaNO_3$); $c_{PAR} = 5 . 10^{-5}$ mol l^{-1}; c_{SDS} (mmol l^{-1}) for curves: 1 0, 2 0·2, 3 1, 4 2, 5 3, 6 6
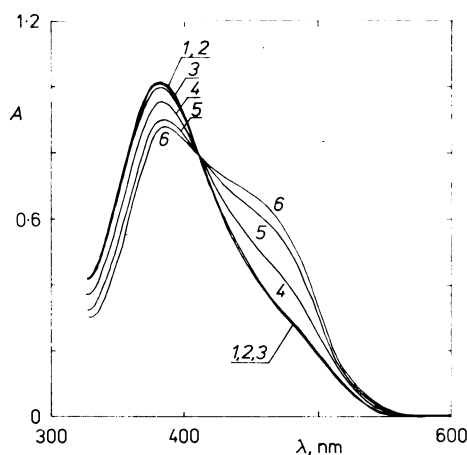
FIG. 3

The effect of SDS on the absorption spectra of PAR. pH 4·20; $I = 0·1$ mol l^{-1} ($NaNO_3$); $c_{PAR} = 5 . 10^{-5}$ mol l^{-1}; c_{SDS} (mmol l^{-1}) for curves: 1 0, 2 0·2, 3 1, 4 2, 5 3, 6 6

The degree of the interaction between PAR and SDS is reflected in changes in the pK_{an} values, which are also affected by the presence of increasing concentrations of both of the strong electrolytes.

The values of pK_{a1} obtained in the presence of SDS and strong electrolytes NaCl and $NaNO_3$ are given in Tables I and II. It follows from these values that the pK_{a1} values in the PAR–SDS system for both strong electrolytes decrease slightly with increasing tenside concentration to a certain limiting value and that the pK_{a1} value then rapidly increases again. The tenside concentration range where the character of the $pK_{a1} = f(c_T)$ dependence changes depends greatly on the ionic strength of the solution and on the type of strong electrolyte. As the I value increases in solutions of both electrolytes, this region decreases to lower c_T values. In the PAR–SDS system whose ionic strength was adjusted by addition of a NaCl solution, this effect is greater than in the presence of $NaNO_3$.

Tables III and IV give the determined values of pK_{a2} for the PAR–SDS system in the presence of both electrolytes NaCl and $NaNO_3$. The effect of the anionogenic tenside on pK_{a2} for the dye PAR is analogous to the effect of this tenside on pK_{a1}. An increase in the tenside concentration up to a certain limiting value has no great effect on the pK_{a2} values; an increase in c_T above this value leads to an increase in pK_{a2} values. The concentration region c_T where marked changes occur in the pK_{a2}

Fig. 4

The effect of the ionic strength on the absorption spectra of PAR in the presence of SDS. pH 1·51, $c_{PAR} = 5 . 10^{-5}$ mol l^{-1}, $c_T = 1 . 10^{-3}$ mol l^{-1}; $I(NaNO_3)$ (mol l^{-1}) for curves: 1 0·05, 2 0·1, 3 0·2, 4 0·3, 5 0·5
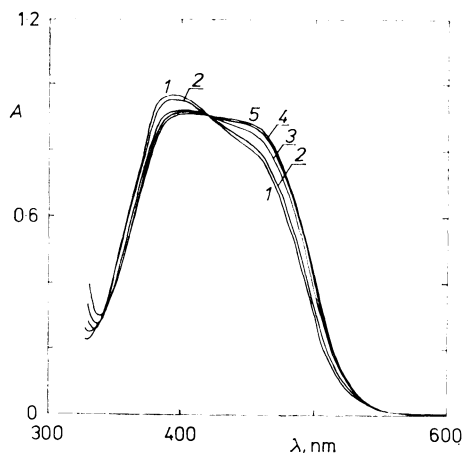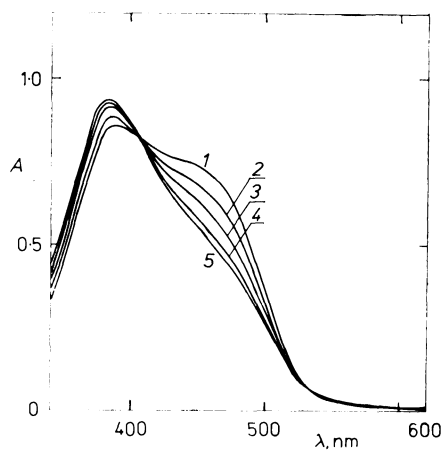
Fig. 5

The effect of the ionic strength on the absorption spectra of PAR in the presence of SDS. pH 4·20; $c_{PAR} = 5 . 10^{-5}$ mol l^{-1}, $c_T = 6 . 10^{-3}$ mol l^{-1}; $I(NaNO_3)$ (mol l^{-1}) for curves: 1 0·05, 2 0·1, 3 0·2, 4 0·3, 4 0·5

value is higher than for the effect of SDS on pK_{a1}. These concentration regions were found for various experimental conditions from the relationship of pK_{a1} or pK_{a2} vs $\log c_T$. These values depend strongly on the amount of strong electrolyte in the solution.

As these concentration regions can be considered to correspond to the cmc values for the given conditions, Table V simultaneously lists the cmc values for tenside SDS found from measurements of the surface tension of the PAR–SDS system solutions (pH 1·12).

These values confirm that an increase in the ionic strength of the solution leads to a decrease in the cmc value for tenside SDS, both for solutions of SDS alone and

TABLE I

Dissociation constants pK_{a1} for the binary PAR–SDS system; the effect of $NaNO_3$

c_T mmol l^{-1}	pK_{a1} at $I(NaNO_3)$ (mol l^{-1})				
	0·05	0·1	0·2	0·3	0·5
0	3·00	3·02	3·09	3·10	3·11
0·2	2·90	2·93	2·96	2·95	3·07
0·5	2·96	2·92	2·92	2·80	3·03
1	2·90	2·88	2·99	3·13	3·20
2	2·83	3·47	3·53	3·40	3·26
3	3·51	3·73	3·63	3·48	3·34
6	4·23	4·01	3·73	3·59	3·40

TABLE II

Dissociation constants pK_{a1} for the binary PAR–SDS system; the effect of NaCl

c_T mmol l^{-1}	pK_{a1} at $I(NaCl)$ (mol l^{-1})				
	0·05	0·1	0·2	0·3	0·5
0	3·01	3·02	3·00	3·01	3·01
0·2	2·84	2·96	2·99	2·97	3·00
0·5	2·93	2·93	2·97	3·01	3·09
1	2·83	2·85	3·13	3·22	3·25
2	3·14	3·52	3·46	3·39	3·32
3	3·80	3·74	3·62	3·48	3·35
6	4·17	3·92	3·68	3·58	3·43

for the PAR–SDS system. These results are in agreement with spectrophotometric studies.

Comparison of the cmc values obtained from the graphs of γ' vs log c_T and of pK_{a1} vs log c_T for the binary PAR–SDS system $(I(NaCl))$ reveals that these values agree within experimental error $(5\cdot0$ to $12\cdot0$ rel. %). The cmc values found from the pK_{a2} vs log c_T graph indicate a shift to higher c_T values, especially for $I(NaCl) = 0\cdot3$ and $0\cdot5$. It thus follows that the two methods are comparable only under identical conditions (including the pH of the solution).

TABLE III

Dissociation constants pK_{a2} for the binary PAR–SDS system; the effect of NaNO$_3$

c_T mmol l^{-1}	pK_{a2} at I(NaNO$_3$) (mol l^{-1})				
	0·05	0·1	0·2	0·3	0·5
0	5·58	5·51	5·55	5·53	5·53
0·2	5·50	5·46	5·45	5·47	5·45
0·5	5·51	5·46	5·41	5·47	5·43
1	5·51	5·47	5·48	5·49	5·51
2	5·55	5·65	5·72	5·71	5·70
3	5·60	5·72	5·80	5·83	5·82
6	6·11	6·12	6·11	6·11	6·06

TABLE IV

Dissociation constants pK_{a2} for the PAR–SDS binary system; the effect of NaCl

c_T mmol l^{-1}	pK_{a2} at I(NaCl) (mol l^{-1})				
	0·05	0·1	0·2	0·3	0·5
0	5·50	5·52	5·52	5·53	5·54
0·2	5·51	5·47	5·46	5·40	5·39
0·5	5·52	5·50	5·45	5·48	5·48
1	5·53	5·48	5·47	5·57	5·48
2	5·54	5·64	4·68	5·69	5·74
3	5·77	5·80	5·86	5·85	5·81
6	6·15	6·10	6·08	6·09	6·09

It follows from the evaluation of the dependence of pK_{a1} and pK_{a2} values on c_T that the decrease in the surface tension is greater at pH values where the H_3L^+ form predominates $(pH < 2)$ than at higher pH values. This difference indicates that the individual dissociated forms of the PAR dye interact differently with the tenside SDS. The determined effect of the individual studied strong electrolyte anions (Cl^-, NO_3^-) indicates that the studied changes are analogous in both cases; the cmc values are generally higher in the presence of NO_3^- anions than in the presence of Cl^-.

DISCUSSION

In spite of the ionic character of the components present, i.e. the protonized form H_3L^+ and the negatively charged anion DS^-, in acidic medium, an ion associate

TABLE V

The effect of the presence of strong electrolytes (NaCl, NaNO$_3$) on the cmc values for SDS alone and for the SDS–PAR system found tensiometrically ($\gamma = f(\log c_T)$) and spectrophotometrically ($pK_{an} = f(\log c_T)$)

System	I mol l^{-1}	cmc, mmol l^{-1}		
		γ' vs log c_T	pK_{a1} vs log c_T	pK_{a2} vs log c_T
SDS	not adjusted	3·02	—	—
effect of NaCl				
SDS	0·05	2·58	—	—
SDS	0·1	1·99	—	—
SDS	0·2	1·47	—	—
SDS	0·5	0·61	—	—
SDS–PAR	0·05	1·46	1·34	1·99
SDS–PAR	0·1	1·09	1·00	1·44
SDS–PAR	0·2	0·75	0·66	1·35
SDS–PAR	0·3	—	0·47	1·26
SDS–PAR	0·5	0·21	0·20	1·12
effect of NaNO$_3$				
SDS–PAR	0·05	—	1·83	2·68
SDS–PAR	0·1	—	1·12	2·23
SDS–PAR	0·2	—	0·97	1·98
SDS–PAR	0·3	—	0·56	1·12
SDS–PAR	0·5	—	0·44	1·02

is not formed in the subcritical tenside concentration region, as the presence of SDS in solution has no marked effect on the absorption spectrum or the pK_{a1} values for the dye PAR.

Changes in the absorption curves for the H_3L^+ form and pK_{a1} values in the binary PAR–SDS system for c_T values above the cmc value indicate that there is strong interaction between the two components. As c_T increases, the absorbance value at $\lambda_{max} = 389$ nm decreases, with simultaneous increase in the absorbance at $\lambda_{max} \approx$ ≈ 460 nm. If tautomeric equilibrium is assumed between forms I and II, then micellar solubilization leads to disturbance of the azo–hydrazo tautomeric equilibrium ·between forms I and II in favour of the latter. The charge distribution of the hydrazo form of the protonated dye H_3L^+ and the negative charge on the SDS micelles suggest that there will be strong interaction between the two species primarily as a result of electrostatic effects, leading to an increase in the pK_{a1} value for $c_T \geqq$ cmc.

If the dissociation form H_2L predominates in aqueous PAR solutions, then there is no marked change in the character of the spectra of the PAR–SDS system at $c_T <$ cmc, similarly as in the previous case, also corresponding to the small changes in the pK_{a2} values for the PAR dye. At concentrations of $c_T >$ cmc, there is a clear increase in the absorption maximum at $\lambda_{max} \approx 460$ nm. This fact can be explained in terms of a shift in the dissociation equilibrium between forms H_3L^+ and H_2L in favour of the former. This is also obvious from changes in the apparent dissociation constants pK_{a2} for the PAR–SDS binary system. These values indicate that, in general, the association constant between the protonated form H_3L^+ and the alkyl sulphate anion is greater than the analogous association constant for the adduct between the dissociation form H_2L and SDS.

The action of strong electrolytes ($NaNO_3$, NaCl) on the binary PAR–SDS system can be considered from two aspects: a) the effect of the electrolyte on the parameters of the micelles of SDS alone (aggregation micelle number, cmc, etc.); b) the competitive equilibrium between Na^+ ions and the individual forms of the PAR dye on the charged micelle surface. It follows from measurements of the surface tension of the solutions of the tenside alone and of the tenside in the presence of PAR that the presence of the strong electrolyte and of the dye affect the cmc value. The addition of $NaNO_3$, NaCl, or any electrolyte in general affects the charge density on the micelle surface, where the charge on the SDS micelle surface is about 70% neutralized for $c_{NaCl} = 0-0.8$ mol l^{-1} (see refs[21,22]). This effect explains the strong effect on the pK_{a1} and pK_{a2} values above the cmc for various $I(NaNO_3)$ and $I(NaCl)$, where, for example, for pK_{a1} the H_3L^+ form (i.e. the hydrazo form II) is competitively replaced on the micelle surface by sodium ions as the $NaNO_3$ (NaCl) concentration increases.

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