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Spectrophotometric investigation of the interactions between cationic dye (C.I. Basic Yellow 2) and anionic surfactant (sodium dioctylsulfosuccinate) in the premicellar and micellar region

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ABSTRACT

The interactions between C.I. Basic Yellow 2 (BY2) and sodium dioctylsulfosuccinate (NaDS) have been investigated in the premicellar and micellar region using spectrophotometric method at 288.15, 298.15, 308.15 and 318.15 K. Absorption spectra of dye–surfactant mixtures show that dye–surfactant aggregates occur in the premicellar region. The addition of excess NaDS to the BY2 solution enhances the solubility of dye molecules due to the micelle formation. Moreover, the presence of excess surfactant micelles in the dye solution causes an increase in the absorption intensity of BY2 and leads to a bathochromic shift in the maximum absorption wavelength of dye. The values of binding constants, partition coefficients between the micellar and the bulk water phases and thermodynamic parameters have been calculated for the BY2–NaDS system at different temperatures. The binding of BY2 to surfactant micelles decreases with increasing temperature. The binding and partition processes are exothermic and occur spontaneously.

1. Introduction

Surfactant–dye interactions are very important in various dyeing processes such as textile dyeing, photography and printing ink as well as chemical researches such as analytical chemistry, biochemistry and photosensitization [1,2].

A certain amount of surfactant is introduced into the dyeing baths as an auxiliary agent for improving and accelerating dye adsorption onto the fibers during the dyeing process in textile industry [3]. The addition of ionic surfactant to the solution of an ionic dye with the opposite charge often causes the formation of insoluble or soluble complexes. These complexes formed in dyeing baths may reduce the dye adsorption onto the fibers. The precipitation of insoluble complexes in these baths can be prevented by adding the ionic surfactant at a concentration higher than the critical micelle concentration (CMC) to the solution [4]. Thus, it is necessary to understand the nature of interactions between surfactant and dye molecules and also the ratio of dye to surfactant concentration in dyeing baths in order to design desirable dye–surfactant systems [5].

CMC value is an important solution property [6]. Micelles can enhance the solubility of insoluble dye molecules in water. Solubilization of a dye aggregate by micelle formation is of great importance in various industrial processes. The interactions between dye molecules and micelles and dye and water molecules play a significant role in the partitioning process. The partitions of dyes between the micellar and the bulk water phases are described by partition coefficients. The investigation of partitioning process for a dye–surfactant system is useful to understand the solubilization of dye molecules [7–9]. The partition coefficients are also very important in the liquid chromatographic separations involving micellar phases and have considerable relevance in the interpretation of different chemical or biochemical processes. Unfortunately, there are not so many partition coefficient values for various dye–surfactant systems in the literature.

The interactions between dyes and surfactants have been studied by various techniques such as spectrophotometry [10–13], conductometry [14–17], potentiometry [18,19], and voltammetry [20]. Surfactants in the premicellar or micellar region affect the absorption spectra of many dyes [21]. Hence, the most widely used method to investigate these interactions is spectrophotometric method [22,23].

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Surfactants are of a great technological significance. Anionic surfactants are widely used in industrial cleaners, detergents, wetting, dispersing and leveling agents [24]. Sodium dioctylsulfosuccinate (NaDS), a double-chained anionic surfactant, exhibits good wetting and foam stabilizing abilities. It is widely used in textile, detergent, paper, paint and ink industries [25–27]. C.I. Basic Yellow 2 (BY2), a cationic dye, is also used commonly in textile, paper, leather and carpet industries [28]. Although NaDS and BY2 are widely used in the various industries, their interactions are not investigated yet.

The aim of the present study is to investigate the interactions between BY2 and NaDS at 288.15, 298.15, 308.15 and 318.15 K by spectrophotometric method and also calculate the values of binding constant, partition coefficient and thermodynamic parameters.

2. Experimental

2.1. Materials

C.I. Basic Yellow 2 (BY2) and sodium dioctylsulfosuccinate (NaDS) were purchased from Aldrich. The chemical structures of cationic dye and anionic surfactant are shown in Fig. 1. All chemicals were used as received. Solutions were prepared in deionized water.

2.2. Method

A series of dye solution was prepared from stock solution of BY2 (1.0×10^{-4} M) to determine the molar absorptivity (ε_0) values of dye and the absorption spectra of these solutions were recorded by a Varian Cary 5000 model UV–vis-NIR spectrophotometer using a matched pair of quartz cells (1 cm path length) in a peltier thermostatted cell holder at 288.15, 298.15, 308.15 and 318.15 (±0.1 K). Then, the ε_0 values of BY2 in aqueous solutions were determined from the plots of absorbance versus dye concentration at the maximum absorption wavelengths.

The concentration of dye was kept constant as 2.0×10^{-5} M during the experimental investigation of interactions between dye and surfactant molecules. Firstly, 2.0×10^{-5} M dye solution was prepared from stock solution of BY2 and the absorption spectra of this solution were obtained at various temperatures. Then, a series of dye–surfactant solution containing different amount of NaDS was prepared and the absorption spectra of these solutions were recorded at different temperatures.

Binding constants and micelle/water partition coefficients for dye–surfactant system were determined from the absorbance changes occurred as a result of the interactions between dye and surfactant molecules.

Spectrophotometric method was used to determine the CMC values of the surfactant in the presence of 2.0×10^{-5} M BY2 at different temperatures. The CMC values were obtained from the inflection point, which corresponds to the CMC, of the plots of absorbance versus surfactant concentration. The absorbance value of BY2 initially decreased with increasing NaDS concentration, showed a minimum value and then increased with further surfactant concentration. The concentration at the minimum absorbance value was considered as the CMC of NaDS in the presence of 2.0×10^{-5} M BY2 [5].

3. Results and discussion

The absorption spectrum of 2.0×10^{-5} M BY2 in aqueous solution at 288.15 K is presented in Fig. 2. It is seen from Fig. 2(a) that BY2 is of two maximum absorption wavelengths (λ_{max}) at 432 nm and 369 nm. The ε_0 values of BY2 in water at 288.15, 298.15, 308.15 and 318.15 K are given in Table 1. As seen from Table 1 that the ε_0 values of BY2 decrease slightly with increasing temperature from 288.15 K to 318.15 K.

The absorption spectra of dye–surfactant solutions containing different NaDS concentrations were recorded to investigate the interactions between dye and surfactant molecules. The absorption spectra of 2.0×10^{-5} M BY2 obtained in the presence of various NaDS concentrations at 288.15 K are shown in Fig. 2. The presence of surfactant molecules within dye solution causes some changes in the absorption spectrum of BY2 depending on surfactant concentration. Firstly, the intensity of absorption of BY2 at 432 nm decreases until the surfactant concentration reaches to 0.80 mM within the dye–surfactant mixture at 288.15 K (Fig. 2(a)). After then, it starts to increase with the surfactant concentration (Fig. 2(b)). Finally, the absorption value of dye get become larger than that of dye solution without surfactant in the presence of excess surfactant molecules and the λ_{max} value of dye solution shifts from 432 nm to 437 nm (Fig. 2(c)). The λ_{max} value (at 437 nm) of dye–surfactant micelle system became constant at high micelle concentrations (>2.75 mM NaDS). This situation contributes to the solubilization of dye molecules into the micelles (Fig. 2(c)). The reason for this bathochromic shift in the spectrum may be explained with the entrance of dye particles into surfactant micelles and also the interaction between aromatic rings presented in the structure of the dye (π–π stacking) and formation of dye J-aggregates (head to tail) [29]. A similar result was also observed for anionic dye, C.I. Acid Blue 25, and cationic surfactant, hexadecyltrimethylammonium bromide, system [30].

Electrostatic interaction of BY2 with NaDS micelles takes place through the positively charged group of the dye ($=NH_2^+$). However, this interaction could not explain the spectral changes by itself. In the presence of anionic surfactant, dye molecules do not act simple

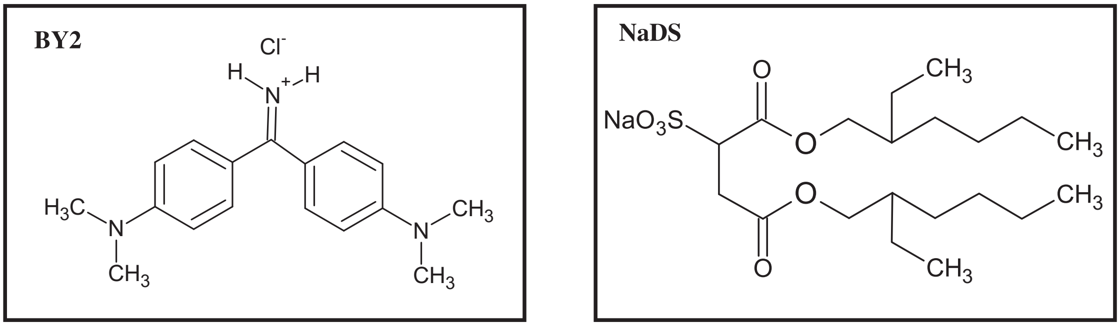

Fig. 1. Chemical structures of C.I. Basic Yellow 2 (BY2) and sodium dioctylsulfosuccinate (NaDS).

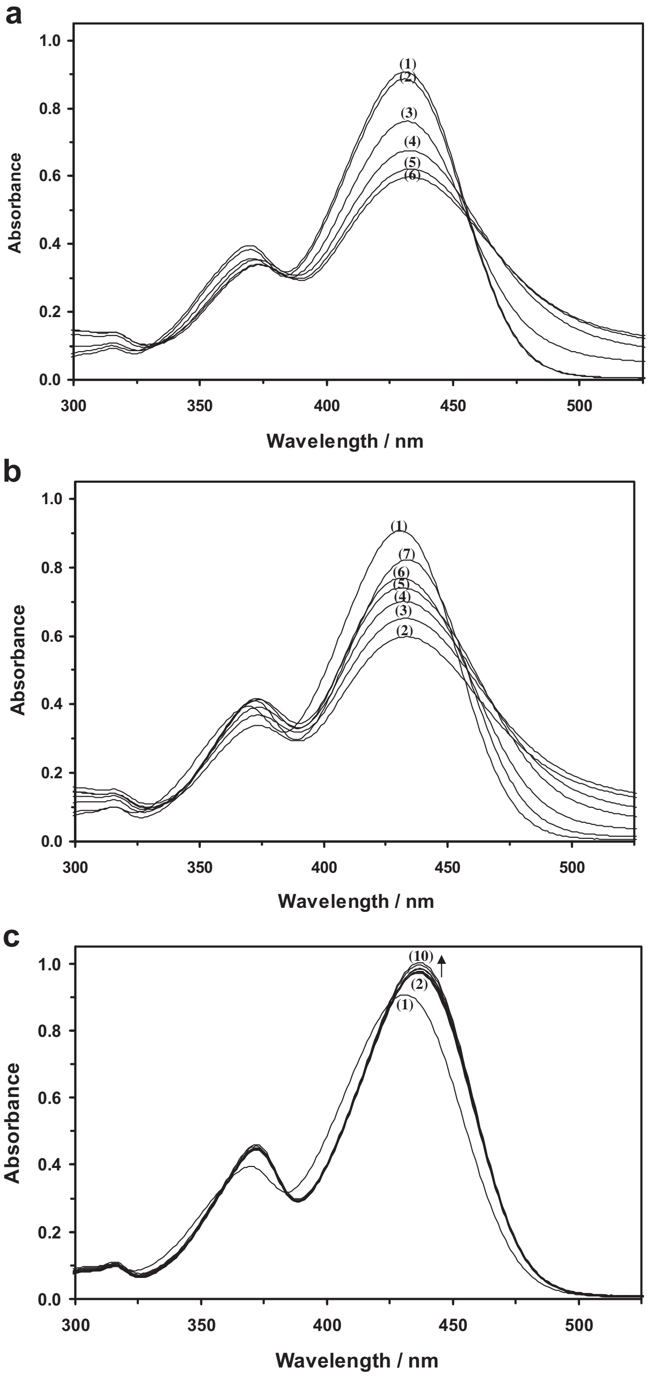

Fig. 2. Absorption spectra of BY2 (2.0×10^{-5} M) in the presence of various NaDS concentrations at 288.15 K. (a) [NaDS] = (1) 0, (2) 0.10, (3) 0.20, (4) 0.40, (5) 0.50 and (6) 0.80 mM, (b) [NaDS] = (1) 0, (2) 0.80, (3) 0.85, (4) 0.90, (5) 1.50, (6) 2.50 and (7) 2.75 mM, (c) [NaDS] = (1) 0, (2) 3.25, (3) 3.50, (4) 4.00, (5) 4.50, (6) 5.00, (7) 5.50, (8) 6.00, (9) 7.50 and (10) 9.50 mM.

as counter ions, but are incorporated into the water-rich Stern layer of the micelle in a sandwich arrangement. This permits not only the hydration of hydrophilic ($=NH_2^+$) group, but also the solution of the aromatic ring of the dye by the $-SO_3^-$ group and the participation of Van der Waals interactions between adjacent surfactant chains and the dye organic moiety (hydrophobic forces). In this situation, the microenvironment of the chromophore has changed and this change is the cause of the spectral shift observed [31].

Table 1
The wavelength of maximum absorption (λ_{max}), molar absorptivity (ε_0) and regression coefficient (r) values of BY2 in water.

T/K	λ_{max}/nm	ε_0/M^{-1} cm^{-1}	r
288.15	432	45227 ± 78	0.9999
	369	19673 ± 84	0.9998
298.15	432	44844 ± 90	0.9999
	369	19620 ± 91	0.9998
308.15	432	44348 ± 91	0.9999
	369	19522 ± 95	0.9997
318.15	432	43881 ± 72	0.9999
	369	19481 ± 87	0.9997

At very low NaDS concentrations, formation of dye–surfactant aggregates occurs starting with the ion pairs ($D^+ \cdot S^-$) and continuing to a dye–surfactant aggregate represented by $(D^+ \cdot S^-)_n$. In the near and just below of the CMC $(D^+ \cdot S^-)_n$ aggregates reorganize into premicelle with a monomeric D^+ content, resulting in an increase in the absorbance of the dye in this premicellar region (just below the CMC). On further increase in surfactant concentration, the absorbance value reaches its limiting value and all dye molecules are accommodated into normal micelles as monomeric molecules (Fig. 2) [11,32].

The variation in the absorbance values of 2.0×10^{-5} M BY2 as a function of NaDS concentration for various temperatures is presented in Fig. 3. The λ_{max} value of BY2 in the monomer form is 432 nm, so this wavelength was used during the determination of CMC of surfactant in the presence of 2.0×10^{-5} M BY2 in Fig. 3. It is seen from this figure that the absorbance value of BY2 decreases until the CMC of surfactant, after then it increases. Absorbance of BY2 has a minimum value at the CMC of surfactant.

Dye–surfactant binding constant, partition coefficient of dye between aqueous and micellar phases and thermodynamic parameters can be calculated from the absorption spectra to obtain quantitative information about the interactions between BY2 and NaDS micelles at various temperatures.

3.1. Determination of binding constant

The equilibrium reaction for the binding of dye to the micelles can be written as [33,34]

$$D + S_m \overset{K_b}{\rightleftarrows} DS_m \tag{1}$$

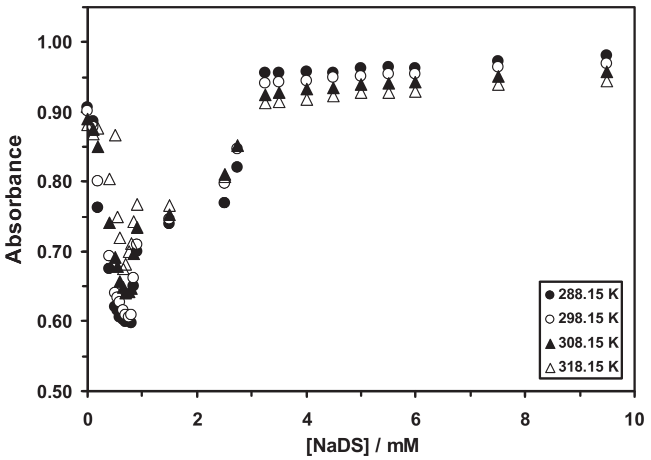

Fig. 3. The effect of NaDS concentration on the absorbance value of 2.0×10^{-5} M BY2 at various temperatures.

and

$$K_b = \frac{[DS_m]}{[D][S_m]} \tag{2}$$

where K_b is the binding constant, $[DS_m]$ and $[D]$ are the concentrations of bound and free dye, respectively, and $[S_m]$ is the concentration of micellized surfactant.

Binding constant can be determined using the Benesi–Hildebrand equation [35]. This equation is used in the following modified form for the dye–surfactant micelle system [36,37]:

$$\frac{d[D]_0}{A - A_0} = \frac{1}{\varepsilon_m - \varepsilon_0} + \frac{1}{(\varepsilon_m - \varepsilon_0)K_b[S_m]} \tag{3}$$

where $[D]_0$ is the total concentration of dye, d is the optical path length of the solution, A and A_0 are the absorbance values of dye in the presence and absence of surfactant, respectively, ε_m and ε_0 are the molar absorptivity of dye fully bound to micelles determined in large excess of the micelles and the molar absorptivity of dye in aqueous solution without surfactant, respectively. $[S_m]$ is equal to the difference between the total surfactant concentration and the CMC of surfactant solution containing dye.

CMC values of NaDS in the presence of 2.0×10^{-5} M BY2 were determined from the changes (inflection point) in the absorbance values of dye (Fig. 3) and found as 0.80, 0.75, 0.70 and 0.65 mM at 288.15, 298.15, 308.15 and 318.15 K, respectively. Chatterjee et al. [6] have measured the CMC values of NaDS in water at various temperatures by surface tension and microcalorimetric methods. CMC of anionic surfactant obtained from surface tension method was found as 2.85, 2.72 and 2.76 mM at 288.15, 298.15 and 308.15 K, respectively. CMC values of NaDS were reported as 2.75, 2.68, 2.95 and 3.12 mM at 288.15, 298.15, 308.15 and 318.15 K, respectively, by microcalorimetric method. When CMC values of NaDS in water and in the presence of 2.0×10^{-5} M BY2 are compared, it is seen that cationic dye molecules cause a reduction in the CMC values of anionic surfactant at all temperatures studied. The reduction of CMC values of surfactants with the addition of various organic molecules or anions was also reported for different dye–surfactant systems in the literature [31].

The $d[D]_0/(A - A_0)$ was plotted as a function of $1/[S_m]$ according to Eq. (3) using the absorbance values at the maximum absorption wavelength of the micelle-bound dye (at 437 nm) in order to find the K_b and the $(\varepsilon_m - \varepsilon_0)$ values at different temperatures [34]. The plot of $d[D]_0/(A - A_0)$ against $1/[S_m]$ is shown in Fig. 4. The data of Fig. 4 were linearly regressed according to the Eq. (3) and the K_b and the $(\varepsilon_m - \varepsilon_0)$ values were calculated from the slopes and the intercepts of plots and results were given in Table 2. The linear regression coefficients obtained for these plots are higher than 0.9690. It can be seen from Table 2 that the K_b values decrease with increasing temperature from 288.15 K to 318.15 K for the BY2–NaDS micelle systems. This means that high temperatures cause a reduction in the interactions between dye molecules and surfactant micelles. In other words, the strength of interactions between BY2 and NaDS micelles at 318.15 K is weaker than that of at 288.15 K. The positive value of $(\varepsilon_m - \varepsilon_0)$ indicates that molar absorptivity of dye within micelle is higher than that of within water. It seems from Table 2 that $(\varepsilon_m - \varepsilon_0)$ values decrease with temperature, but when the standard deviation values are taken into consideration, there is not any dependency between $(\varepsilon_m - \varepsilon_0)$ values and temperature.

Table 2
K_b, $\varepsilon_m - \varepsilon_0$ and regression coefficient (r) values for the interaction of BY2 with anionic surfactant at different temperatures.

T/K	$K_b/10^2$ M^{-1}	$\varepsilon_m - \varepsilon_0$/M^{-1} cm^{-1}	r
288.15	8.22 ± 0.83	6735 ± 212	0.9690
298.15	5.73 ± 0.27	6719 ± 134	0.9935
308.15	4.25 ± 0.21	6691 ± 178	0.9935
318.15	3.38 ± 0.19	6567 ± 226	0.9928

The interactions between BY2 and NaDS molecules are due to the electrostatic and hydrophobic forces and hydrogen bonding. Moreover, anionic surfactant used in this study does not have an aromatic ring in its structure. For this reason, the π–π interactions do not occur in BY2–NaDS system.

3.2. Determination of partition coefficient

Partition coefficient, K_c, is an important parameter to determine the partition of dye between the micellar and the bulk water phases. It can be calculated from the following equation [5,38,39]:

$$\frac{1}{A - A_0} = \frac{1}{K_c(A - A_0)^{\infty}([D]_0 + [S_m])} + \frac{1}{(A - A_0)^{\infty}} \tag{4}$$

where $(A - A_0)^{\infty}$ is differential absorbance at the infinity of surfactant concentration where all dye molecules are assumed to have formed aggregates with the surfactant.

The K_c values were obtained from intercepts and slopes of the plots of $1/(A - A_0)$ versus $1/([D]_0 + [S_m])$ in Fig. 5. The dimensionless partition coefficient, K_x, defined as the ratio of mole fraction of dye in micellar phase to mole fraction of dye in aqueous phase is related

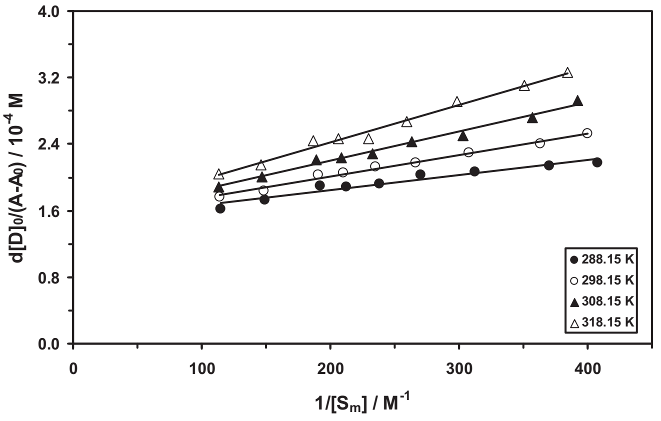

Fig. 4. Plot of $d[D]_0/(A - A_0)$ versus $1/[S_m]$ for the interaction of BY2 with NaDS at various temperatures.

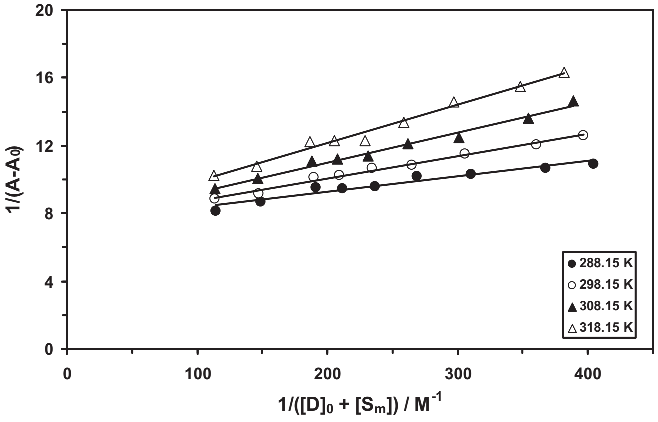

Fig. 5. Plot of $1/(A - A_0)$ versus $1/([D]_0 + [S_m])$ for the interaction of BY2 with NaDS at various temperatures.

Table 3
K_c, regression coefficient (r) and K_x values for the interaction of BY2 with anionic surfactant at different temperatures.

T/K	$K_c/10^2$ M^{-1}	r	$K_x/10^4$
288.15	8.11 ± 0.08	0.9694	4.51 ± 0.04
298.15	5.66 ± 0.20	0.9936	3.14 ± 0.15
308.15	4.20 ± 0.21	0.9934	2.33 ± 0.12
318.15	3.34 ± 0.19	0.9928	1.86 ± 0.11

with K_c as $K_x = K_c \cdot n_w$, where n_w is the molarity of water. The calculated K_c and K_x values are shown in Table 3. It can be seen from Table 3 that if the temperature of the dye–surfactant micelle system is increased, the value of both partition coefficients decreases. These values are comparable with literature values reported previously for some cationic dye molecules bound to anionic or nonionic surfactant micelles. The K_c and K_x values for the interactions of cationic thiazine dye (thionine) with anionic surfactant (sodium dodecylsulfate) were reported as 8.18×10^2 M^{-1} and 4.54×10^5 at 298.15 K, respectively, by Göktürk and Talman [5]. Sarkar and Poddar [11] reported the K_c values of cationic dye (methyl violet) and nonionic surfactant (polyethylene sorbitonmonooleate) system as 4.46×10^2 M^{-1}, 3.62×10^2 M^{-1} and 2.98×10^2 M^{-1} at 298.15 K, 303.15 K and 308.15 K, respectively.

3.3. Thermodynamic parameters for the binding and partition processes

The free energy changes, ΔG^0, the enthalpy changes, ΔH^0, and the entropy changes, ΔS^0, belong to the binding and partition processes for the interactions of BY2 with NaDS at different temperatures can be calculated using Eqs. (5)–(7), respectively [40], from the K_b and K_x values in Tables 2 and 3.

$$\Delta G^0 = -RT \ln K \tag{5}$$

$$\Delta H^0 = \left[\frac{\partial(\Delta G^0/T)}{\partial(1/T)}\right]_P \tag{6}$$

$$\Delta S^0 = \frac{\Delta H^0 - \Delta G^0}{T} \tag{7}$$

The calculated values of ΔG^0 are given in Table 4 for the binding and partition processes at various temperatures. As seen from Table 4, all ΔG^0 values are negative. The negative ΔG^0 values indicate that the binding process of BY2 to the NaDS micelles and the partition process of BY2 between the micellar and the bulk water phases occur spontaneously. Moreover, the ΔG^0 values increase with increasing of temperature for binding process. This means that binding of dye to surfactant micelles is highly spontaneous at lower temperatures. As seen from Table 4 that a significant variation in the ΔG^0 values of partition process was not observed with temperature. The ΔG^0 values belong to the partition process are lower than those obtained for the BY2–NaDS binding process.

Table 4
Thermodynamic parameters for the interaction of BY2 with anionic surfactant at 288.15, 298.15, 308.15 and 318.15 K.

Process	T/K	$\Delta G°$/kJ mol^{-1}	$\Delta H°$/kJ mol^{-1}	$\Delta S°$/J mol^{-1} K^{-1}
Binding	288.15	−16.08 ± 0.10	−22.57 ± 1.15	−22.52 ± 0.01
	298.15	−15.74 ± 0.05		−22.91 ± 0.01
	308.15	−15.51 ± 0.05		−22.91 ± 0.01
	318.15	−15.41 ± 0.06		−22.51 ± 0.01
Partition	288.15	−25.67 ± 0.10	−22.60 ± 1.15	10.65 ± 0.00
	298.15	−25.67 ± 0.05		10.30 ± 0.01
	308.15	−25.76 ± 0.05		10.25 ± 0.01
	318.15	−26.00 ± 0.06		10.69 ± 0.01

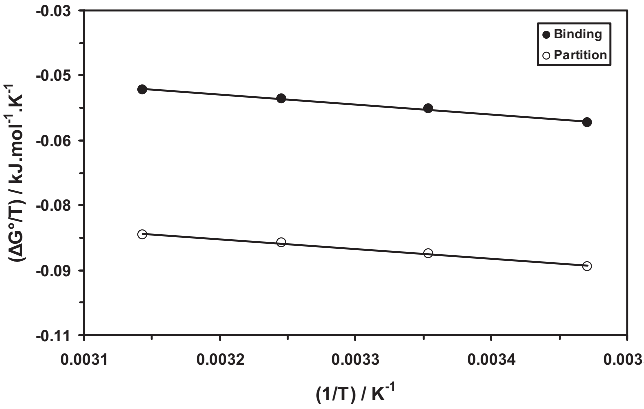

Fig. 6. Plot of $\Delta G°/T$ versus $1/T$ for the binding and partition of BY2 to NaDS micelles.

According to Eq. (6), a plot of $\Delta G^0/T$ versus $1/T$ gives a straight line (Fig. 6) and the slope of this line is equal to ΔH^0. The calculated ΔH^0 values are given in Table 4. The negative ΔH^0 values indicate that the binding and partition processes are exothermic. Binding of BY2 to the NaDS micelles is accompanied by the negative ΔS^0 values (Table 4). This means that the binding of dye molecules to the surfactant micelles causes an increase in the order of the process [41]. On the other hand, the partition of BY2 molecules between the micellar and the bulk water phases is accompanied by the positive ΔS^0 values (Table 4). The reason of positive ΔS^0 values obtained can be explained by considering ΔG^0 and ΔH^0 values. Although the ΔH^0 values for the binding and partition processes are of similar values, ΔG^0 values of partition process have more negative values than that of binding process as seen in Table 4.

4. Conclusions

The interactions between BY2 and NaDS have been investigated by following the absorbance changes in the absorption spectra of dye molecules as a function of surfactant concentration and as a function of temperature. The results show that the absorbance value of dye decreases with the addition of surfactant until the CMC and then starts to increase with the further addition of surfactant. The increase in the temperature of solution causes a reduction in the binding constant and partition coefficient values of the BY2–NaDS system due to the weakening of interactions between dye and surfactant molecules at high temperatures. The negative ΔG^0 and ΔH^0 values point out that the binding of BY2 to NaDS micelles and the partition of BY2 between the micellar and the bulk water phases occur spontaneously and both processes are exothermic.

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