

## Solubilization of disperse dyes in cationic gemini surfactant micelles

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### Abstract

The solubilization of three disperse dyes, 1-(4-nitrophenylazo)-4-*N,N*-diethanolaminobenzene (NPDEAB), *p*-aminoazobenzene (4-PAA) and 1,4-diaminoanthraquinone (1,4-DAA) into the micelles formed by two cationic gemini surfactants, propanediyl- $\alpha,\omega$ -bis(dimethyldodecylammonium bromide) (DC3-12) and hexanediyl- $\alpha,\omega$ -bis(dimethyldodecylammonium bromide) (DC6-12), was investigated and compared with that achieved using two conventional surfactants, dodecyltrimethylammonium bromide (C12C1NBr), and ethyldodecyltrimethylammonium bromide (C12C2NBr). The solubilization power of the DC6-12 micelle was greater than that of the DC3-12 micelle, suggesting that the gemini surfactant micelle, which has a longer chain, enables greater solubilization power. The solubilization power of the gemini surfactant micelles was greater than that of the corresponding conventional surfactant micelles for NPDEAB and 1,4-DAA, while the opposite result was obtained for 4-PAA. The amounts of dye solubilized in the micelles increased in the order of NPDEAB < 1,4-DAA < 4-PAA, indicating that smaller dye molecules are solubilized more readily in micellar solutions. Thus, solubilization power is strongly dependent not only on the nature of the surfactant micelles but also on the structure of the solubilized dye molecules. The aggregation number of the surfactant micelles was determined using steady state fluorescence quenching. Although the aggregation number was different for the gemini and the conventional surfactant micelles, the number of the alkyl tail groups included in the gemini micelle was similar to that in a conventional one. The solubilization capacity was calculated from the solubilization power and the aggregation number. The solubilization capacity was significantly dependent on the dye structure. The visible absorption spectra of the disperse dyes revealed that the dye solubilized was not located in the inner part of the micelle core but in the outer regions. © 2000 Elsevier Science Ltd. All rights reserved.

**Keywords:** Gemini surfactant; Cationic surfactant; Solubilization; Disperse dye; Micelle

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### 1. Introduction

As the solubilization of dyes in surfactant micelles plays an important role in dyeing processes, the solubilization of organic compounds involving dyes in conventional surfactant micelles comprising one head and one tail group has been

studied extensively [1–23]. However, few studies on the solubilization of organic compounds into gemini surfactants with two head and two tail groups are available [24–25].

Devinsky et al. synthesized a homologous series of *N,N'*-bis(alkyldimethyl)- $\alpha,\omega$ -alkanediammonium dibromides, which has a terminal alkyl chain of 8–16 carbon atoms and a spacer chain of 2–12 carbon atoms. These workers examined the effect of the alkyl substituents as well as the connecting alkyl chain on the critical micelle concentration (CMC)

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and the solubilization properties of these compounds [24]. In this work, *trans*-azobenzene was used as a solubilize; the solubilization capacity linearly increased with an increase in alkyl chain length and showed a maximum when the spacer chain comprised six carbon atoms [24]. Dam et al. also synthesized similar gemini surfactants and examined their surface properties and oil solubilization capacity using *n*-hexane and toluene as solubilizes [25]. In the case of the gemini surfactants having dodecyl groups as the terminal groups, the solubilization capacity for toluene decreased with increasing spacer chain length. The solubilization capacity of the gemini surfactant micelles was much larger than that of conventional surfactants. However, the authors are unaware of a study of the solubilization of disperse dyes in gemini surfactant micelles.

To use the gemini surfactants as auxiliaries for disperse dyeing, it is necessary to obtain information on the dye bath. In this context, the solubilization of disperse dyes in gemini surfactant micelles was investigated.

In the present study, propanediyl- $\alpha,\omega$ -bis(dimethyldodecylammonium bromide) (DC3-12) and hexanediyl- $\alpha,\omega$ -bis(dimethyldodecylammonium bromide) (DC6-12) were synthesized and the solubilization of three disperse dyes, 1-(4-nitrophenylazo)-4-*N,N*-diethanolaminobenzene (NPDEAB), *p*-aminoazobenzene (4-PAA) and 1,4-diaminoanthraquinone (1,4-DAA), in their micelles was investigated and compared with that achieved using micelles formed by dodecyltrimethylammonium bromide (C12C1NBr), and ethyldodecyl dimethylammonium bromide (C12C2NBr). The solubilization power and capacity of the disperse dyes in both gemini and conventional surfactant micelles are discussed. The microenvironment of dyes solubilized in micelles is also discussed.

## 2. Experimental

### 2.1. Materials

The two gemini surfactants and two conventional surfactants shown in Fig. 1 were used. DC3-12 and DC6-12 were synthesized as follows. A

mixed solution of *n*-dodecyl bromide and the corresponding amine (*N,N,N',N'*-tetramethyl-1,3-diaminopropane or *N,N,N',N'*-tetramethyl-1,6-diaminohexane) in nitroethane was stirred at room temperature for 5 days. The products were then filtered, purified by repeated precipitation from ethanol into diethylether, and dried. The purity was confirmed by elemental analysis. (Calculated for DC3-12: C, 59.22; H, 10.90; N, 4.46; Br, 25.4%. Found: C, 57.76; H, 11.04; N, 4.38; Br, 27.0%. Calculated for DC6-12: C, 60.88; H, 11.12; N, 4.18; Br, 23.8%. Found: C, 60.36; H, 11.54; N, 4.08; Br, 24.7%.)

C12C1NBr was purchased from Tokyo Kasei Kogyo Co. (Tokyo, Japan) and used as received. C12C2NBr was synthesized as follows. A mixed solution of *N,N*-dimethyldodecylamine and ethyl bromide in nitroethane was stirred at room temperature for 3 days. After the solvent was removed by evaporation, the product was purified by repeated recrystallization from benzene, treated in diethylether for 1 day, and dried. The purity was confirmed

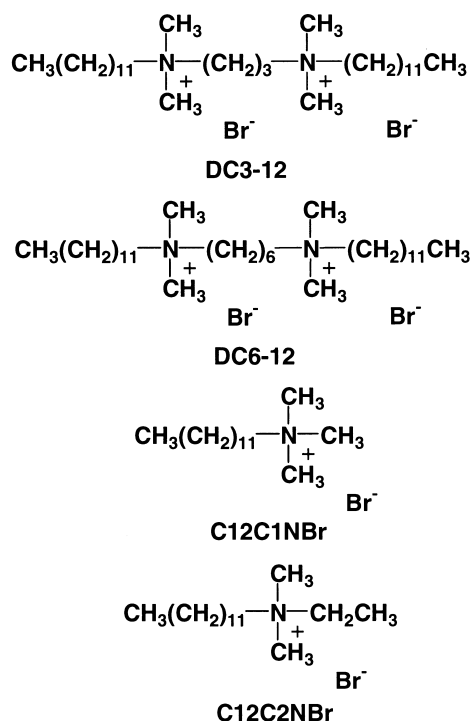
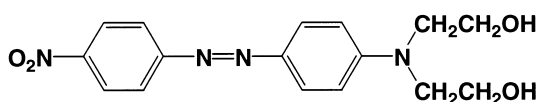


Fig. 1. Surfactants used.

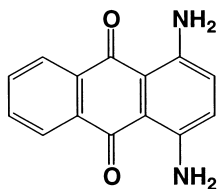
by elemental analysis. (Calculated for  $C_{12}C_2NBr$ : C, 59.61; H, 11.26; N, 4.34; Br, 24.8%. Found: C, 59.30; H, 11.18; N, 4.34; Br, 24.7%.)

The three disperse dyes shown in Fig. 2 were used. NPDEAB (C.I. Disperse Red 19) was synthesized by coupling diazotized *p*-nitroaniline with *N*-phenyldiethanolamine in an acidic solution. The dye thus obtained was purified by repeated precipitation from ethanol into water and dried. The purity was confirmed by elemental analysis. (Calculated for NPDEAB: C, 58.17; H, 5.49; N, 16.96%. Found: C, 58.07; H, 5.35; N, 16.89%.) 4-PAA purchased from Tokyo Kasei Kogyo Co. (Tokyo, Japan) was purified by repeated precipitation from 40% ethanol into water and dried. 1,4-DAA (C.I. Disperse Violet 1) was purchased from Tokyo Kasei Kogyo Co. (Tokyo, Japan) and used as received.

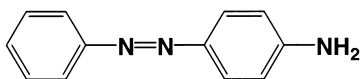
Pyrene and *N*-cetylpyridinium chloride (CPC) were used as fluorescent probe and quencher, respectively. The compounds were purchased from Wako Pure Chemical Industries, Ltd. (Osaka, Japan) and Tokyo Kasei Kogyo Co. (Tokyo, Japan), respectively and were used as received.



NPDEAB



1,4-DAA



4-PAA

Fig. 2. Dyes used.

## 2.2. Methods

The solubilization of disperse dyes in both the gemini and the conventional surfactant micelles was measured as follows. Aqueous solutions of a given surfactant covering a suitable range of concentration below and above its critical micelle concentration (CMC) were prepared using distilled water. After an excess of finely powdered disperse dyes were added to these solutions, they were shaken for 3 days at  $298 \pm 0.2$  K in a water bath incubator with Tokyo Rikakikai Thermistor Tempet T-80. The excess insoluble dye was filtered off using a glass fiber filter (Advantec GA55) of pore size of  $0.6 \mu\text{m}$ . The filtrate was diluted with an equal volume of ethanol and then an adequate amount of 50% aqueous ethanol solution was added to this solution to achieve a dilution at which the UV spectra could be accurately measured. The concentration of dye in 50% aqueous ethanol solution was determined spectrophotometrically from the absorbance of the solution at  $\lambda_{\text{max}}$ . The  $\lambda_{\text{max}}$  in the visible absorption spectra of the various dye solutions were determined using a Shimadzu UV-160A spectrophotometer at  $298 \pm 0.1$  K. The molar extinction coefficients,  $\epsilon$ , of the dyes in 50% aqueous ethanol solution at  $298 \pm 0.1$  K were  $2200 \text{ m}^2 \text{ mol}^{-1}$  at 381 nm for 4-PAA,  $3240 \text{ m}^2 \text{ mol}^{-1}$  at 494 nm for NPDEAB and  $1430 \text{ m}^2 \text{ mol}^{-1}$  at 550 nm for 1,4-DAA.

The aggregation number of both the gemini and conventional surfactant micelles was determined as follows. Aqueous solutions containing pyrene at a constant concentration below  $2 \times 10^{-6} \text{ mol dm}^{-3}$  and surfactant of constant concentration above its CMC were prepared so that  $[P]/[\text{micelle}] < 0.02$ , where  $[P]$  and  $[\text{micelle}]$  are the concentrations of pyrene and micelle, respectively. Furthermore, the surfactant concentration was high enough to ensure that pyrene and the quencher were completely solubilized in the micelle. CPC at different concentrations was incorporated in the above solution. The molar concentration ratio,  $R = [Q]/[\text{micelle}]$ , was varied between 0 and 1.5 by adjusting the concentration of CPC, where  $[Q]$  is the CPC (quencher) concentration. The fluorescence intensities of the aqueous solutions thus prepared were measured

using a Shimadzu RF-5000 spectrofluorophotometer equipped with a thermostated cell holder at  $298 \pm 0.1$  K; the excitation wavelength was 335 nm. The emission spectrum was recorded from 360 to 410 nm. At every value of  $[Q]$ , the intensity,  $I$ , of the first vibronic peak of pyrene was measured at 374 nm. Since the ratio of  $I_1/I_3$  did not change during the experiment, no problems were encountered regarding the sensitivity of  $I_1$  to the polarity of the environment that was sensed by the probe. Here,  $I_1$  and  $I_3$  are the intensity of the first and third vibronic peak of pyrene, respectively.

The microenvironment of the dyes solubilized in the micelles was investigated by measuring the visible absorption spectra. Aqueous solutions containing the dyes at constant concentration ( $3 \times 10^{-5}$  mol dm $^{-3}$ ) and the surfactants at constant concentration ( $3 \times 10^{-2}$  mol dm $^{-3}$ ) above CMC were prepared. Ethanol, benzene and *n*-hexane solutions containing the dyes at constant concentration ( $3 \times 10^{-5}$  mol dm $^{-3}$ ) were also prepared. The visible absorption spectra of the solutions thus prepared were recorded using a Shimadzu UV-160A spectrophotometer at  $298 \pm 0.1$  K.

### 3. Results and discussion

#### 3.1. Solubilization power

The dependence of the amounts of dye solubilized in micelles,  $C_{\text{solubilized}}$ , on surfactant concentration,  $C_s$ , for NPDEAB, 4-PAA and 1,4-DAA is shown in Figs. 3–5, respectively. In the case of NPDEAB and 4-PAA, the amounts of dye solubilized in the micelles increased linearly with increasing surfactant concentration for all the surfactants used. However, in the case of 1,4-DAA, the amounts of dye solubilized increased sigmoidally with increasing surfactant concentration for DC3-12 and DC6-12, while those increased linearly with increasing surfactant concentration for C12C1NBr and C12C2NBr. This suggests that the solubilization mechanism of the gemini surfactant micelles for 1,4-DAA differs to that of the other two dyes.

It can be assumed that the slope of the linear plots expresses the solubilization power,  $S_e$ , which

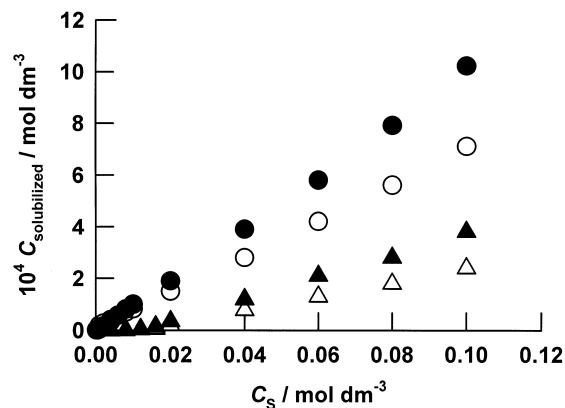


Fig. 3. Dependence of amounts of dye solubilized,  $C_{\text{solubilized}}$ , on the surfactant concentration,  $C_s$ , for NPDEAB at 298 K:  $\circ$ , DC3-12;  $\bullet$ , DC6-12;  $\triangle$ , C12C1NBr;  $\blacktriangle$ , C12C2NBr.

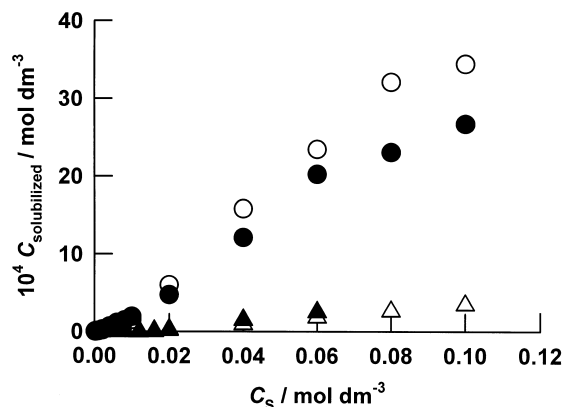


Fig. 4. Dependence of amounts of dye solubilized,  $C_{\text{solubilized}}$ , on the surfactant concentration,  $C_s$ , for 1,4-DAA at 298 K:  $\circ$ , DC3-12;  $\bullet$ , DC6-12;  $\triangle$ , C12C1NBr;  $\blacktriangle$ , C12C2NBr.

gives the amount of dye solubilized per mol of micellar surfactant:

$$S_e = C_{\text{solubilized}} / (C_s - \text{CMC}) \quad (1)$$

These values are given in Table 1.

As described above, a solubilization power that was independent of surfactant concentration was found to be the solubilization of NPDEAB and 4-PAA in both the gemini and conventional surfactant micelles and also for the solubilization of 1,4-DAA in the conventional surfactants. Similar results were observed in previous studies of insoluble dyes [12–23]. Solubilization power that

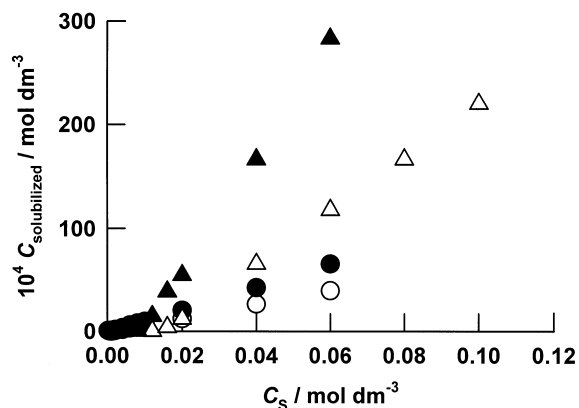


Fig. 5. Dependence of amounts of dye solubilized,  $C_{\text{solubilized}}$ , on the surfactant concentration,  $C_s$ , for 4-PAA at 298 K: ○, DC3-12; ●, DC6-12; △, C12C1NBr; ▲, C12C2NBr.

was dependent upon surfactant concentration was found for 1,4-DAA in DC3-12 and DC6-12; such behavior was also observed previously for some water-soluble dyes [20–23].

If the micelle size and shape does not change with micelle concentration and the intermicellar interaction does not perturb the solubilization then solubilization power should be independent of the micelle concentration (the amount of dye solubilized per mol of micellar surfactant is constant in wide concentration region). However, in the present study, solubilization behavior was dependent on the dye structure in the gemini surfactant micelles. These results show that the solubilization behavior itself is strongly dependent not

only on the nature of the surfactant micelles but also on dye structure.

Solubilization power increased in the order: C12C1NBr < C12C2NBr < DC3-12 < DC6-12 for NPDEAB and 1,4-DAA, whereas it increased in the order of DC3-12 < DC6-12 < C12C1NBr < C12C2NBr for 4-PAA. This finding reflects the location of the dyes as described in the latter section. The solubilization power of the three disperse dyes into the micelles formed by the gemini surfactant having the spacer group of six carbon atoms was greater than that for the gemini surfactant which carried the propyl spacer group, reflecting the almost linear increase of hydrophobic pseudo-phase volume with spacer chain length. A similar result was previously obtained for the solubilization of *trans*-azobenzene into alkanediyl- $\alpha,\omega$ -bis(dimethyldecylammonium bromide) micelles, where the solubilization capacity was a maximum for the gemini surfactant having hexyl spacer group [24]. This particular result was interpreted in terms of spacer flexibility and micelle structure. A different result was obtained for the solubilization of toluene and *n*-hexane into alkanediyl- $\alpha,\omega$ -bis(dimethyldodecylammonium bromide) micelles, where the solubilization capacity decreased with spacer chain length [25]. Thus the solubilization structure plays an important role in the solubilization the gemini surfactant micelles.

The amounts of dye solubilized in each micelle increased in the order of NPDEAB < 1,4-DAA < 4-PAA. This was consistent with that of the dye molecular size, suggesting that smaller

Table 1

Solubilization power of the disperse dyes into the surfactant micelles (mole of solubilized dye/mole of surfactant)

	NPDEAB		1,4-DAA		4-PAA	
DC3-12	0.0069	(2.6) <sup>c</sup>	0.0146 <sup>a</sup>	(5.4) <sup>c</sup>	0.0676	(25) <sup>c</sup>
			0.0434 <sup>b</sup>	(16) <sup>c</sup>		
DC6-12	0.0100	(3.7) <sup>c</sup>	0.0204 <sup>a</sup>	(7.6) <sup>c</sup>	0.1102	(41) <sup>c</sup>
			0.0320 <sup>b</sup>	(12) <sup>c</sup>		
C12C1NBr	0.0027	(1.0) <sup>c</sup>	0.0040	(1.5) <sup>c</sup>	0.2566	(95) <sup>c</sup>
C12C2NBr	0.0043	(1.6) <sup>c</sup>	0.0056	(2.1) <sup>c</sup>	0.5564	(206) <sup>c</sup>

<sup>a</sup> The solubilization power when the surfactant concentration is less than  $1 \times 10^{-2}$  mol dm<sup>-3</sup>.

<sup>b</sup> The solubilization power when the surfactant concentration is greater than  $1 \times 10^{-2}$  mol dm<sup>-3</sup>.

<sup>c</sup> The values in the bracket are reference values when the solubilization power of NPDEAB into C12C1NBr is supposed to be 1.

molecules are solubilized more readily in micellar solutions.

### 3.2. Micelle aggregation number and solubilization capacity

If the micelle aggregation number,  $N$ , is not influenced by the solubilization of dye, the average number of dye molecules solubilized in a micelle,  $\Sigma$ , which is designated as the solubilization capacity, is given by Eq. (2)

$$\Sigma = N\Sigma_e \quad (2)$$

The mean aggregation number of the micelles,  $N$ , was determined by means of steady-state fluorescence quenching. The basis for determining the aggregation number in this procedure is that in a system containing both fluorescent probe and quencher molecules, solubilized in an excess of micelles, the extent of quenching will decrease with an increasing number of micelles because of a reduced probability of finding both probe and quencher in the same micelle [26]. In the following equation,  $N$  could be calculated from the slope of the linear plots of  $\ln(I_0/I)$  against  $[Q]$ .

$$\ln(I_0/I) = [Q]N/([C] - \text{CMC}) \quad (3)$$

where  $I_0$  and  $I$  are the fluorescence intensity in the absence and presence of quencher,  $[Q]$  the quencher concentration, and  $[C]$  the surfactant concentration. A plot of  $\ln(I_0/I)$  against  $[Q]$  is shown in Fig. 6. For all the systems measured, the plots showed good linearity and the  $N$  values were determined from the slope of the plots (Table 2). Here the CMC values determined by means of the conductivity measurements in a previous paper [27] were used for the calculation of  $N$ .

The aggregation number of the DC3-12 micelle obtained using steady-state fluorescence quenching (SSFQ) in the present study was quite different from that secured from time-resolved fluorescence quenching (TRFQ) [28]: the aggregation number obtained of DC3-12 micelle by TRFQ for DC3-12 increased with an increase in surfactant concentration, while that obtained by SSFQ is independent of surfactant concentration. Alternatively, the  $N$

values of DC6-12, C12C1NBr and C12C2NBr micelle obtained by SSFQ had a similar tendency to those obtained by TRFQ [28,29]. This result may be concerned with the assumption of SSFQ.

The aggregation numbers of the gemini surfactants were smaller than those of the conventional surfactants. It is worthwhile to discuss the number of alkyl tail chains included in a micelle,  $N_{\text{Alkyl}}$ , because the gemini surfactants have two alkyl terminal groups. The  $N_{\text{Alkyl}}$  values were calculated as 49–57, 31–41, 41–50 and 45–49 for DC3-12, DC6-12, C12C1NBr and C12C2NBr, respectively,

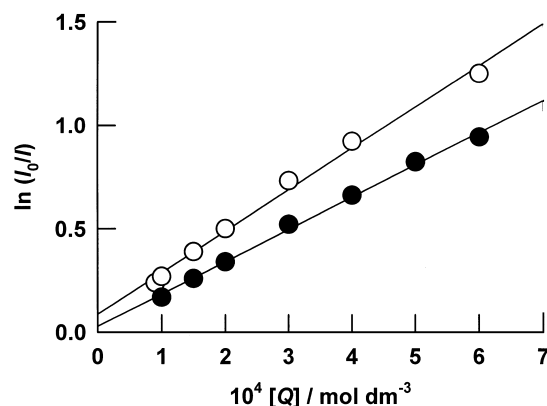


Fig. 6. Plots of  $\ln(I_0/I)$  against  $[Q]$  for the gemini surfactant micelles at 298 K:  $\circ$ , DC3-12;  $\bullet$ , DC6-12.

Table 2  
Solubilization capacity of the disperse dyes into the surfactant micelles

Surfactant	C (M)	N	$\Sigma$		
			NPDEAB	1,4-DAA	4-PAA
DC3-12	0.01	25.9	0.24	0.43	1.7
	0.02	26.5	0.21	0.83	1.7
	0.04	24.6	0.18	1.0	1.7
	0.06	28.2	0.20	1.1	1.9
DC6-12	0.01	15.3	0.17	0.33	1.7
	0.04	18.1	0.18	0.57	2.0
	0.06	20.1	0.20	0.69	2.2
C12C1NBr	0.04	40.7	0.13	0.15	11
	0.06	48.5	0.14	0.20	13
	0.08	49.5	0.14	0.20	13
C12C2NBr	0.04	45.2	0.21	0.26	29
	0.06	48.5	0.22	0.27	30

suggesting that the micelle formed by the gemini surfactant having the shorter spacer group includes the similar number of the alkyl tail groups to the corresponding conventional surfactant micelle.

The solubilization capacity,  $\Sigma$ , was calculated from the solubilization power,  $S_e$ , and the aggregation number,  $N$ , using Eq. (3) and is given in Table 2. The behavior of the solubilization capacity was significantly dependent on the dye structure. The  $\Sigma$  values for NPDEAB were almost the same for both the gemini and conventional surfactant micelles. In the case of 1,4-DAA, the solubilization capacity for the gemini surfactant micelles was larger than that for the conventional one. The solubilization capacity for 4-PAA showed the opposite tendency to that for 1,4-DAA: the  $S_e$  values for the conventional surfactant micelles were much larger than those for the Gemini surfactant. This indicates that less than one dye molecule is incorporated in one micelle for NPDEAB and 1,4-DAA, while more than one dye molecule is present in one micelle for 4-PAA; in the case of the 4-PAA/conventional surfactant systems, many dye molecules are present in a micelle. Such differences in the nature of the solubilization, probably reflects the location of the dyes in the micelles as described later.

### 3.3. Visible absorption spectra and location of dye in micelle

To elucidate the location of the dyes in the micelles, visible absorption spectra are useful. Fig. 7 shows the spectra of 1,4-DAA in aqueous DC6-12 solution, ethanol and benzene, from which  $\lambda_{\max}$  were determined (Table 3).

The results for water, *n*-hexane and aqueous gemini surfactant solution at a concentration below the CMC might contain large errors owing to the poor solubility of the dyes. Furthermore, in water and aqueous surfactant solution below CMC, the dyes are believed to form aggregates, which shifts the  $\lambda_{\max}$  values to lower wavelength. Thus, the real  $\lambda_{\max}$  values in water and aqueous surfactant solution below CMC are estimated to occur at higher values than 505, 595/554 and 395 for NPDEAB, 1,4-DAA and 4-PAA, respectively.

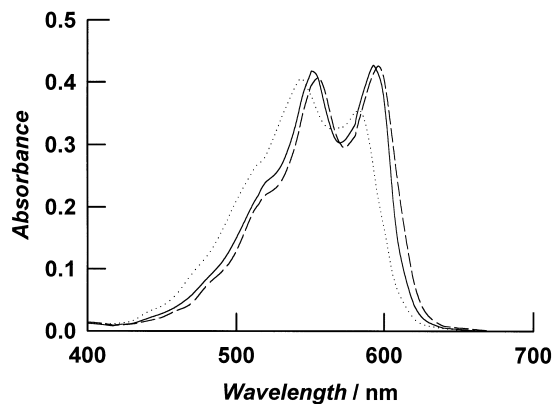


Fig. 7. Visible absorption spectra of 1,4-DAA ( $3 \times 10^{-5}$  mol  $\text{dm}^{-3}$ ) in ethanol (—), benzene (···) and aqueous DC6-12 solution with  $3 \times 10^{-2}$  mol  $\text{dm}^{-3}$  (---).

Table 3  
Maximum absorption wavelength

	NPDEAB	1,4-DAA	4-PAA
Water	493	584/546	375
DC3-12 below CMC	494	585/546	376
DC3-12 above CMC	505	595/554	395
DC6-12 below CMC	—	—	375
DC6-12 above CMC	507	596/555	398
Ethanol 100%	478	593/551	387
Ethanol 50%	495	591/550	381
Benzene	464	581/545	375
<i>n</i> -Hexane	440	564/536	362

The nature of the visible absorption spectra in aqueous surfactant solutions above CMC were very similar to those in ethanol solution, but they were different to those in the non-polar solvents, benzene and *n*-hexane. As shown in Fig. 7, the spectrum of an aqueous 1,4-DAA solution above CMC has two peaks; the ratio of these two peaks can be used as a measure of comparing spectra. The ratio for the spectra in the aqueous surfactant solutions above CMC was similar to that in ethanol, suggesting that the dye is located in a micro-environment that is similar in character to ethanol. Hence, we can deduce that these three disperse dyes were solubilized in the hydrophilic region rather than the hydrophobic region of the micelles; consequently, the dyes were located not in the inner part of the micelle core but in the outer part.

The larger solubilization capacity of 4-PAA in the conventional surfactants micelles suggests that most of the dyes are located at the micelle–water interface and not in the interior of the micelle. Similar results were obtained for the systems containing hydrophobic compounds (Orange OT, azobenzene and naphthalene) and ionic surfactants (alkylpyridinium bromide, alkylcarboxylic acid and alkylbetaine) [2]. It can thus be concluded that the location of the dyes is one of the most important factors in the solubilization behavior, which reflects both the dye and micelle structures.

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### References

- [1] Ogino K, Abe M, Takesita N. *Bull Chem Soc Jpn* 1976; 49:3679.
- [2] Birdi KS, Magnusson T. *Colloid Polym Sci* 1976; 254:1059.
- [3] Chaiko MA, Nagarajan R, Ruckenstein E. *J Colloid Interface Sci* 1984;99:168.
- [4] Hoffmann H, Ulbricht W. *J Colloid Interface Sci* 1989; 129:388.
- [5] Shah SS, Khan MS, Ullah H, Awan MA. *J Colloid Interface Sci* 1997;186:382.
- [6] Takeuchi M, Moroi Y. *J Colloid Interface Sci* 1998; 197:230.
- [7] Mukerjee P, Cardinal JR. *J Phys Chem* 1978;82:1620.
- [8] Moroi Y, Saito K, Matsuura R. *J Phys Chem* 1982; 86:2463.
- [9] Moroi Y, Noma H, Matsuura R. *J Phys Chem* 1983; 87:872.
- [10] Takeuchi M, Moroi Y. *Langmuir* 1995;11:4719.
- [11] Eda Y, Takisawa N, Shirahama K. *Langmuir* 1997;13:2432.
- [12] Abe A, Imae T, Ikeda S. *Colloid Polym Sci* 1987;265:637.
- [13] Sinha S, Dogra A, Jain N, Bahadur P. *Indian J Chem* 1998;37A:118.
- [14] Anacker EW, Gerry HE, Jacobs PT, Petrariu I. *J Colloid Interface Sci* 1977;60:514.
- [15] Gerry HE, Jacobs PT, Anacker EW. *J Colloid Interface Sci* 1977;62:556.
- [16] Abe M, Suzuki N, Ogino K. *J Colloid Interface Sci* 1984;99:226.
- [17] Imae T, Abe A, Taguchi Y, Ikeda S. *J Colloid Interface Sci* 1986;109:567.
- [18] Ikeda S, Maruyama Y. *J Colloid Interface Sci* 1994;166:1.
- [19] Ozeki S, Ikeda S. *J Phys Chem* 1985;89:5088.
- [20] Tokiwa F. *Bull Chem Soc Jpn* 1963;36:222.
- [21] Tokiwa F. *Bull Chem Soc Jpn* 1963;36:1589.
- [22] Kolthoff IM, Stricks W. *J Phys Chem* 1948;52:915.
- [23] Tokiwa F. *J Phys Chem* 1968;72:1214.
- [24] Devinsky F, Lacko I, Imam T. *J Colloid Interface Sci* 1991;143:336.
- [25] Dam T, Engberts JBFN, Karthaus J, Karaborni S, van Os NM. *Colloid Surf A: Physicochem Eng Aspects* 1996;118:41.
- [26] Turro NJ, Yekta A. *J Am Chem Soc* 1978;100:5951.
- [27] Choi TS, Ichikawa H, Qian J, Shimizu Y, Shirai H, Hamada K. *Sen'i Gakkaishi* 1999;55:597.
- [28] Danino D, Talmon Y, Zana R. *Langmuir* 1995;11:1448.
- [29] Lianos P, Lang J, Zana R. *J Colloid Interface Sci* 1983;91:276.