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The flexibility of rodlike micelles in aqueous solutions and the crossover concentrations among dilute, semidilute, and concentrated regimes

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Abstract: A flexibility parameter, the persistence length, has been evaluated from the radii of gyration and the contour lengths for rodlike micelles of heptaoxyethylene alkyl ethers (C_nE_7, $n = 12, 14, 16$) and tetradecyldimethylammonium chloride (C_{14}DAC) and bromide (C_{14}DAB) at the observed crossover concentrations between dilute and semidilute regimes. The persistence length range is 43–73 nm, except for $C_{12}E_7$, for which it is 32 nm. The crossover concentrations between dilute and semidilute regimes for the semiflexible rodlike micelles calculated according to Ying and Chu as a function of the molecular weight, the contour length, and the persistence length are consistent with the observed values. The crossover concentration between semidilute and concentrated regimes was, on the other hand, calculated by using the same micelle parameters, including the value of thickness of cross-section of the rodlike micelles. The obtained values are at variance with the observed values. This means that rodlike micelles in semidilute and concentrated solutions might differ in size and/or flexibility from those in dilute solution.

Key words: Semiflexible rodlike micelle, persistence length, crossover concentration, dilute solution, semidilute solution, concentrated solution.

Introduction

Since the overlap and entanglement of rodlike micelles were first evidenced in semidilute solutions of tetradecylpyridinium *n*-heptanesulfonate [1] and hexadecylpyridinium salicylate [2], few researchers investigated aqueous surfactant solutions in semidilute regime. Candau et al. [3–5] applied the scaling laws [6] to aqueous salt solutions of alkyltrimethylammonium halides in semidilute regime, and Kato et al. [7] analyzed aqueous solutions of hexaoxyethylene dodecyl ether on the basis of the scaling laws. Zhou et al. [8] discussed the solution behavior in semidilute regime of rodlike micelles composed of metal soap solved in a polar organic solvent.

Imae et al. [9–15] measured the light scattering and the viscosity of aqueous sodium halide solutions of cationic and nonionic micelles, and discussed the size of semiflexible rodlike micelles and the intermicellar interaction in dilute regime and the scaling laws in semidilute regime. Moreover, they recognized first the existence of the concentrated regime for aqueous sodium halide solutions of $C_{14}E_7$ and $C_{16}E_7$ [12].

The classification of solutions into dilute, semidilute, and concentrated regimes originated from polymer solutions. Daoud et al. [16] suggested that solutions of randomly coiled polymer chains should be divided into three concentration regimes: the dilute regime where polymer chains disperse isolatedly, the semidilute regime where chains overlap one another, and the concentrated regime where chains behave as in the melt. They also proposed that the threshold concentration of overlap is scaled against the degree of polymerization and the radius of gyration.

Cotton et al. [17] and Graessley [18] defined the boundaries between dilute-semidilute solutions and semidilute-concentrated solutions of random coil chains in good solvent as the concentrations where polymer chains touch one another and behave like under the θ condition, respectively, although their definitions are slightly different from each other. The crossover concentrations for rigid thin rods were presented by Doi and Edwards [19]. Recently, Ying and Chu [20] developed the theoretical estimation of the crossover concentrations for polymer chains with different rigidity from random coils to rigid rods.

In this work the crossover concentrations among the dilute, semidilute, and concentrated regimes are estimated for aqueous sodium halide solutions of semiflexible rodlike micelles of C_nE_7, $C_{14}DAC$ and $C_{14}DAB$. The crossover concentrations depend on the flexibility of rodlike micelles. The flexibility of rodlike micelles was investigated by some workers, and the flexibility parameter, that is, the persistence length, was obtained for the semiflexible rodlike micelles [9, 21–25]. In the present paper, the persistence length of rodlike micelles of C_nE_7, $C_{14}DAC$ and $C_{14}DAB$ is evaluated, and the flexibilities or rodlike micelles from different types of surfactants are discussed.

The characteristics of semiflexible rodlike micelles of C_nE_7, $C_{14}DAC$ and $C_{14}DAB$ have already been evaluated at the micelle concentrations where the reciprocal scattering intensities are minimum and, therefore, the apparent micelle molecular weights are maximum [12, 15]. These concentrations correspond to the threshold micelle concentrations above which rodlike micelles overlap one another, that is, the observed cross-over concentrations between dilute and semidilute regimes. For such analysis, the present writer developed a procedure where the data of static and dynamic light scattering observed at only one micelle concentration are used: at a finite micelle concentration, micelle parameters such as the molecular weight, the radius of gyration, and the hydrodynamic radius of rodlike micelles were evaluated with the intermicellar interaction parameters such as the virial coefficients.

Evaluation of the persistence length of rodlike micelles

If semiflexible rodlike micelles behave like wormlike chains defined by Kratky and Porod [26], they are characterized by the contour length L_c and the persistence length a. Then the mean-square radius of gyration of rodlike micelles $\langle R_G^2 \rangle_0$ can be represented by the contour length and the persistence length as follows [27]:

$$\frac{\langle R_G^2 \rangle_0}{a^2} = \frac{L_c}{3a} - 1 + \frac{2a}{L_c}\left[1 - \frac{a}{L_c}\left(1 - e^{-L_c/a}\right)\right]. \quad (1)$$

The radius of gyration for an unperturbed chain without the excluded volume effect $\langle R_G^2 \rangle_0^{1/2}$ can be replaced by the radius of gyration R_G, which is observable by the static light scattering method.

The contour length of a linear chain is defined as

$$L_c \approx M/M_L, \quad (2)$$

where M is the molecular weight and M_L is the molecular weight per unit contour length. For the present rodlike micelles of C_nE_7, $C_{14}DAC$, and $C_{14}DAB$ the numerical values of M_L were calculated according to the method described previously [9, 28]. The results are listed in Table 1 together with the numerical values of the radius of cross section r, the ratio ϱ of the radius of gyration to the hydrodynamic radius of the rodlike micelles, and the characteristic exponent ν of the scaling laws defined for the radii vs. micelle aggregation numbers [12, 15].

The r values for C_nE_7 micelles were assumed to be equivalent to the radii of spherical micelles or corresponding surfactants [28], and the r values of $C_{14}DAC$ and $C_{14}DAB$ were estimated from the radii of cross

Table 1. Characteristics of semiflexible rodlike micelles

	r nm	M_L nm^{-1}	a nm	ϱ ^a)	ν ^a)	ν_{conc} ^a)
$C_{12}E_7$	3.0_3	7280	32	1.4–1.5		
$C_{14}E_7$	3.5_8	7810	46–59	1.7–2.0	0.54–0.75	0.53
$C_{16}E_7$	4.1_3	8320	54–73	1.8–1.9	0.61–0.79	0.49
$C_{14}DAC$	2.1_2	5190	48–61	1.8	0.53–0.58	
$C_{14}DAB$	2.3_3	6840	43–66	1.6–1.8	0.58–0.63	

^a) From [12, 15].

section for rodlike micelles of homologous surfactants [9]. Rodlike micelles of C_nE_7 have a thicker cross-section than those of C_{14}DAC and C_{14}DAB, because C_nE_7 micelles maintain the thick hydrophilic layer of oligooxyethylene. In proportion to the thick radius, the molecular weight per unit contour length of the micelle is large for C_nE_7.

The contour length and the persistence length for semiflexible rodlike micelles of C_nE_7, C_{14}DAC, and C_{14}DAB were evaluated from the observed values of the molecular weight and the radius of gyration [12, 15] according to Eqs. (1) and (2). Table 2 lists the utilized parameters and the obtained contour lengths and persistence lengths. The persistence lengths a_{LS} were evaluated as 32, 46–59, and 54–73 nm for $C_{12}E_7$, $C_{14}E_7$, and $C_{16}E_7$, respectively. The corresponding values for C_{14}DAC and C_{14}DAB were 48–61 and 43–66 nm, respectively. The obtained values are independent of the salt concentration C_s and the temperature T, and indicate the semiflexibility of these rodlike micelles.

The persistence length a_η was also estimated from the intrinsic viscosity $[\eta]$, by using the equation

$$[\eta] = \Phi(2a_\eta L_c)^{3/2}/M\,. \tag{3}$$

The function Φ, which depends on the contour length, the thickness, and the persistence length of the semiflexible chain involved has been numerically given by Yamakawa and Fujii [29]. The intrinsic viscosity for semiflexible rodlike micelles of $C_{14}E_7$ in 1 M NaCl at 35 °C was obtained as $0.743 \times 10^2\ \mathrm{cm^3\ g^{-1}}$ [13]. Then the persistence length a_η is claculated as 46.8 nm, which is consistent with 55.3 nm from the light scattering.

Evaluation of the crossover concentration between dilute and semidilute regimes

The crossover concentration between dilute and semidilute regimes c^* is defined for randomly coiled polymer chains in good solvent as [17]

$$c^*_{rc} = M/[(4/3)\pi R_G^3 N_A]\,, \tag{4}$$

where N_A is Avogadro's constant. Recently, Ying and Chu [20] presented the crossover concentrations between dilute and semidilute regimes as follows

$$c^*_w = 2^{3/2} M/(L_c^{*3} N_A) \tag{5}$$

for wormlike chains with an effective length of

$$L_c^* = (aL_c)^{1/2} \tag{6}$$

and

$$c^*_{\mathrm{rod}} = 2^{2/3} M/(L_c^3 N_A) \tag{7}$$

Table 2. Characteristics of semiflexible rodlike micelles

	C_s (M)	T (°C)	M[a]) ($10^4\ \mathrm{g mol^{-1}}$)	R_G[a]) (nm)	L_c (nm)	a_{LS} (nm)	a_η (nm)	$L_c/2a_{LS}$ (nm)	$D_{0,\,\mathrm{obs}}$[c]) ($10^{-7}\ \mathrm{cm^2\ sec^{-1}}$)	$D_{0,\,\mathrm{calc}}$
$C_{12}E_7$	4	25	1450	142	2000	31.7		31.5	0.17	0.15
$C_{14}E_7$	1	25	54.1	27.1-	69.2	53.4[b])		0.64	1.62	1.77
	1	35	164	44.9	210	55.3	46.8	1.9	1.13	1.12
	1	40	334	69.4	427	45.7		4.7	0.87	0.44
	2	25	170	55.8	217	53.4[b])		2.0	0.64	0.74
	3	25	661	117	846	59.2		7.2	0.28	0.18
$C_{16}E_7$	0	25	78.8	28.0	94.7	63.6[b])		0.74	1.64	1.53
	0.1	25	92.1	32.4	111	63.6[b])		0.87	1.40	1.35
	1	25	327	69.7	392	54.4		3.6	0.57	0.28
	2	25	811	138	974	72.9		6.7	0.27	0.16
C_{14}DAC	2.6	20	1020	191	1980	60.9		16.3	0.15	0.10
	2.6	30	1870	237	3610	48.4		37.3	0.16	0.12
C_{14}DAB	4.3	25	1710	226	2500	66.1		18.9	0.13	0.09
	4.3	30	2300	232	3360	50.2		33.5	0.13	0.10
	4.3	35	3810	278	5580	42.6		65.5	0.11	0.10

[a]) From [12,15]; [b]) Estimated values; [c]) From dynamic scattering hydrodynamic radii data [12,15].

for rigid rods with

$$L_c^2 = 12R_G^2 \,. \tag{8}$$

They [20] proved that Eqs. (4) and (5) were consistent with the experimental values for randomly coiled and wormlike polymer chains, respectively.

When any one of the Eqs. (4), (5) or (7) is applied to rodlike micelles, the concentration c, must be replaced by the micelle concentration $(c - c_o)$, where c and c_o are the total surfactant concentration and the critical micelle concentration, respectively. Then the crossover concentration between dilute and semidilute regimes for the aqueous sodium halide solutions of rodlike micelles were evaluated by using the appropriate micelle parameters listed in Table 2.

The obtained values are collected in Table 3, which includes the observed crossover concentrations $(c - c_o)^*_{\mathrm{obs}}$ [12,15]. The crossover concentration evaluated for a wormlike chain model $(c - c_o)^*_w$ is more consistent with the observed one than that for a random coil model $(c - c_o)^*_{rc}$. That for a rigid rod model $(c - c_o)^*_{\mathrm{rod}}$ is far too small as compared with the observed value for the rodlike micelles. These results confirm the semiflexibility of rodlike micelles.

Evalution of the crossover concentration between semidilute and concentrated regimes

It was recognized that, with the transition from semidilute to concentrated regimes, randomly coiled polymer chains in good solvent are constricted to the limit of the unperturbed dimension, that is, the θ condition [17,18]. Therefore, the constriction of the hydrodynamic volume is a measure of the transition from semidilute to concentrated regimes.

Ying and Chu [20] defined the crossover concentration between semidilute and concentrated regimes c^{**} for wormlike chains as

$$c_w^{**} = 0.243\, M/(d^* L_c^{*2} N_A) \,, \tag{9}$$

$$d^* = (L_c/a)^{1/4} d \tag{10}$$

and

$$L_c^* = (aL_c)^{1/2} \,, \tag{6}$$

where d^* is the effective diameter of the chain and $d = 2r$. The corresponding crossover concentration for rigid rods is given by

$$c_{\mathrm{rod}}^{**} = 0.243\, M/(dL_c{}^2 N_A) \,, \tag{11}$$

where L_c may be replaced by $12^{1/2}\, R_G$. It was verified that Eq. (9) coincided with the observed cross-over concentration for wormlike polymer chains [20].

For aqueous micellar solutions, the boundary between semidilute and concentrated regimes was observed as the micelle concentration where the deviation at high micelle concentrations from the straight

Table 3. Crossover concentrations among dilute, semidilute, and concentrated regimes of rodlike micelles

	C_s (M)	T (°C)	$(c-c_o)^*_{\mathrm{obs}}$ [a])	$(c-c_o)^*_{rc}$	$(c-c_o)^*_w$	$(c-c_o)^*_{\mathrm{rod}}$	$(c-c_o)^{**}_{\mathrm{obs}}$ [a])	$(c-c_o)^{**}_w$	$(c-c_o)^{**}_{\mathrm{rod}}$
				$(10^{-2}\,\mathrm{g\,cm^{-3}})$			$(10^{-2}\,\mathrm{g\,cm^{-3}})$		
$C_{12}E_7$	4	25	0.80	0.20	0.43	0.06			
$C_{14}E_7$	1	25	1.26	1.08	1.13	0.31			
	1	35	0.54	0.70	0.62	0.21			
	1	40	0.54	0.37	0.57	0.11			
	2	25	0.40	0.39	0.64	0.11	5.6	0.58	0.26
	3	25	0.42	0.16	0.28	0.05	4.3	0.34	0.23
$C_{16}E_7$	0	25	0.79	1.42	0.79	0.41			
	0.1	25	0.60	1.08	0.73	0.21			
	1	25	0.20	0.38	0.49	0.11			
	2	25	0.21	0.12	0.20	0.03	6.1	0.29	0.17
C_{14}DAC	2.6	20	0.20	0.06	0.12	0.02			
	2.6	30	0.20	0.06	0.12	0.02			
C_{14}DAB	4.3	25	0.19	0.06	0.12	0.02			
	4.3	30	0.19	0.07	0.16	0.02			
	4.3	35	0.19	0.07	0.15	0.02			

[a]) From [12,15]

line occurred, when the reciprocal scattering intensity was plotted in a double logarithmic scale against the micelle concentration [12]. Such boundary was obtained as 5.6 and 4.3 × 10^{-2} g cm^{-3} for $C_{14}E_7$ in 2 and 3 M NaCl, respectively, and 6.1 × 10^{-2} g cm^{-3} for $C_{16}E_7$ in 2 M NaCl.

The characteristic exponents ν_{conc} for the scaling law obtained in concentrated regime were 0.53 for $C_{14}E_7$ in 2 and 3 M NaCl and 0.49 for $C_{16}E_7$ in 2 M NaCl, as listed in Table 1 [12]. These ν_{conc} values are smaller than the ν values obtained for the rodlike micelles in semidilute regime and are close to the value of 0.5 for polymer chains in the θ condition, as seen in Table 1. This indicates that rodlike micelles in concentrated regime constrict into the limit of the unperturbed dimension.

Assuming rodlike micelles in semidilute and concentrated solutions behave like polymer chains in the corresponding regimes, we replaced the concentration c in Eq. (9) or (11) by the micelle concentration $(c - c_o)$ and then calculated the crossover concentration between semidilute and concentrated regimes $(c - c_o)^{**}_{w}$ or $(c - c_o)^{**}_{\mathrm{rod}}$. The obtained values are listed in Table 3 with the observed values.

The observed crossover concentrations between semidilute and concentrated regimes $(c - c_o)^{**}_{\mathrm{obs}}$, are 9–30 times higher than those between dilute and semidilute regimes $(c - c_o)^{*}_{\mathrm{obs}}$. However, the $(c - c_o)^{**}$ values calculated for wormlike chain and thin rod models are both fairly small and are comparable to the $(c - c_o)^{*}$ values.

Discussion

A method to evaluate the persistence length of semiflexible rodlike micelles was described, and the persistence lengths were evaluated for rodlike micelles of C_nE_7, $C_{14}DAC$, and $C_{14}DAB$ in aqueous sodium halide solutions. The persistence lengths range 43–73 nm, except for $C_{12}E_7$, which has 32 nm.

The same procedure was applied to rodlike micelles of tetradecyltrimethylammonium bromide and hexadecyltrimethylammonium chloride and bromide [9,10]. Their persistence lengths ranged 37–60 nm. Rodlike micelles of dodecyldimethylammonium chloride and bromide exhibited the shorter persistence lengths of 35 and 31 nm, respectively, on the average [9].

The persistence lengths were also evaluated for rodlike micelles of hexadecylpyridinium bromide in aqueous NaBr solutions [21], dodecyldimethylammonium chloride in aqueous NaCl solutions [22,24], dodecyldimethylammonium bromide in aqueous NaBr solutions [24], and dimethyloleylamine oxide in aqueous NaCl solutions with added HCl [23,25]. Since the evaluation of the persistance length depended on the utilized parameters [30], the correct evaluation of the parameters characterizing rodlike micelles must always be undertaken.

In order to check the adequacy of the persistence lengths evaluated in this paper, the translational diffusion coefficients were evaluated by using the calculated persistence lengths. Yamakawa and Fujii [31] gave a theoretical equation of the translational diffusion coefficient for wormlike chains as

$$\begin{aligned} D_o = {} & (k_BT/3\pi\eta_oL_c)[1.843(L_c/2a)^{1/2} - 1.0561 \\ & + 0.1382(L_c/2\mathrm{a})^{-1/2} - 0.3301(L_c/2a)^{-1} \\ & - 0.030(L_c/2a)^{-3/2}] \quad \text{at } L_c/2a \geq 2.278 \qquad (12) \end{aligned}$$

and

$$\begin{aligned} D_o = {} & (k_BT/3\pi\eta_oL_c)[\ln(L_c/2r) \\ & + 0.3863 + 0.1667(L_c/2a) + 0.01883(L_c/2a)^2 \\ & - 0.002039(L_c/2a)^3] \quad \text{at } L_c/2a \leq 2.278 \qquad (13) \end{aligned}$$

where k_B is the Boltzmann constant and η_o is the viscosity of the solvent.

Applying these equations to the L_c and data in Table 2 for the semiflexible rodlike micelles, we calculated the translational diffusion coefficients $D_{0,\mathrm{calc}}$, and compared them with the observed values $D_{0,\mathrm{obs}}$, which were derived by using the Einstein-Stokes equation from dynamic scattering hydrodynamic radii data reported previously [12,15]. The translational diffusion coefficients and the $L_c/2a_{LS}$ values are listed in Table 2. The calculated translational diffusion coefficients are reasonably consistent with the observed values.

The persistence length, which is a flexibility parameter, can be approximated as a sum of two terms [32–34]:

$$a = a_i + a_e \,. \qquad (14)$$

a_i is the intrinsic persistence length and depends on the thickness of a chain and the degree of hydration; a_e is an electrostatic term and occurs from the electrostatic interaction between charges on a chain. According to Odijk's theory [34], a_e is large if the total charge number is large or if the ionic strength or the contour length is small. Since the total persistence length is a sum of these two terms it is very difficult to estimate and compare the persistence lengths of rodlike micelles constituted of different types of surfactants. In this connection it can be realized that the persistence lengths evaluated for rodlike micelles of alkyltrimethylammonium halides, alkyldimethylammonium halides, and heptaoxyethylene alkyl ethers happen to be very close.

The flexibility of rodlike micelles can also be discussed from the dimensionless ratio ϱ and the characteristic exponent ν. The numerical values of ϱ and ν obtained for the present micellar solutions are cited from literatures [12, 15] and collected in Table 1 with the values of the persistence length a. When the flexibility of a linear chain varies from random coil to rigid rod, the a value changes from 0 to ∞, whereas the ϱ and ν values change from 1.3 to 3 and from 0.5 to 1, respectively. The persistence length, the dimensionless ratio, and the characteristic exponent of micelles are independent of the kind of surfactants, that is, $C_{14}E_7$, $C_{16}E_7$, C_{14}DAC and C_{14}DAB, except micelles of $C_{12}E_7$, indicating that these parameters are reasonable measures for the semiflexibility of rodlike micelles.

The aqueous solutions of semiflexible rodlike micelles were divided into four regimes, that is, dilute, semidilute but nonentangled, semidilute and entangled, and concentrated regimes, depending on the situation or rodlike micelles in solutions [12, 13, 15]. Figure 1 represents schematically the relation between the situation of rodlike micelles and the concentration regimes.

The crossover concentrations among dilute, semidilute, and concentrated regimes for aqueous sodium halide solutions of rodlike micelles were described as a function of the molecular weight, the diameter of cross-section, the contour length, and the persistence length or the radius of gyration. The calculated values based on a wormlike chain model for the crossover concentration between dilute and semidilute regimes are reproducible the observed values, more so than those based on radom coil and rigid rod models. On the other hand, the theoretical equation (9) of the crossover concentration between semidilute and concentrated regimes is not proper to the aqueous sodium halide solutions of rodlike micelles.

Equation (9) can be expressed as

$$c_w^{**} = 0.243\, M_L^{5/4}/(d a^{3/4} M^{1/4} N_A)\,, \qquad (15)$$

since $L_c \approx M/M_L$. It may be interpreted from Eq. (15) that the deviation between the observed and calculated $(c - c_0)^{**}$ values originates from the overestimated values of the molecular weight and/or the persistence length. Then the molecular weight and/or the persistence length of rodlike micelles are assumed to become smaller in semidilute and concentrated regimes than in dilute regime. It is possible that micelles change their size and/or flexibility, because micelles are molecular assemblies formed through self-association; they differ from polymer chains in which the units are covalently bonded.

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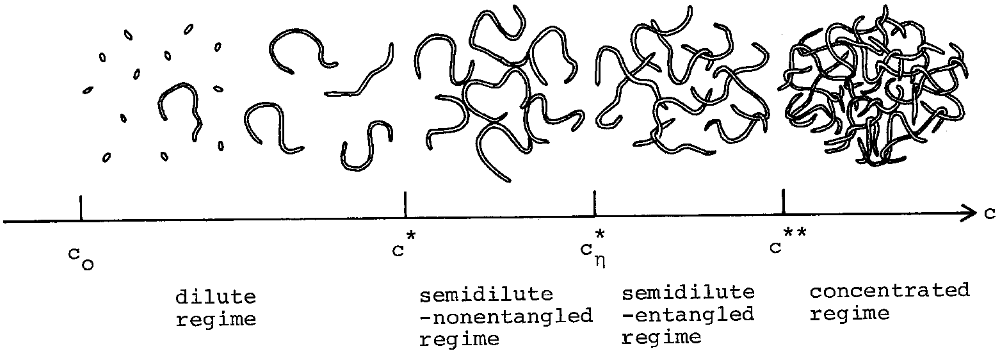

Fig. 1. Schematic of the relation between the situation of rodlike micelles and the concentration regimes

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