Colloid Polym Sci (2006) 284: 725–732
DOI 10.1007/s00396-005-1418-y

ORIGINAL CONTRIBUTION

Hiroshi Murata
Iwao Satake
Katumitu Hayakawa
Masaru Tanokura

Conformational phase diagram of poly(L-lysine) in sodium alkanesulfonate aqueous solutions at various temperatures

Received: 28 April 2005
Accepted: 27 August 2005
Published online: 10 February 2006

H. Murata
Kagoshima Prefectural Institute
of Industrial Technology,
1445-1 Oda, Hayato, Aira-gun,
Kagoshima 899-5105, Japan

I. Satake · K. Hayakawa (✉)
Department of Chemistry
and BioScience, Faculty of Science,
Kagoshima University,
1-21-35 Korimoto,
Kagoshima 890-0065, Japan
e-mail: hayak@sci.kagoshima-u.ac.jp
Tel.: +81-99-2858101
Fax: +81-99-2858117

M. Tanokura
Department of Applied Biological
Chemistry, Graduate School
of Agricultural and Life Sciences,
The University of Tokyo,
1-1-1 Yayoi, Bunkyo-ku,
Tokyo 113-8657, Japan

Abstract The conformation of poly (L-lysine) (PLL) was investigated in sodium alkanesulfonate C_nSO_3Na (n=8, 7) at various temperatures by circular dichroism spectrum measurements. C_8SO_3Na induced a double-step conformational change from a coil, to a β-sheet, and then to an α-helix, in which C_7SO_3Na induced a single-step coil-to-helix conformational change. Binding isotherms of C_8SO_3Na by PLL were constructed from the potentiometry of equilibrium concentration of the surfactant using a surfactant ion-selective electrode. The curves indicated the cooperative binding characteristic and were analyzed by a linear lattice model using the Bethe approximation. The thermodynamic parameters obtained from the model revealed that the binding of C_8SO_3Na by PLL was an entropy-driven process. The conformational change was observed at nearly full binding, presumably due to the surfactant clustering of the ordered conformation.

Keywords Conformation · Poly(L-lysine) · Anionic surfactant · CD spectra · Phase diagram

Introduction

Surfactants are effective denaturants of proteins because they induce a conformational change in native proteins [1–17]. To illustrate this change, much effort has been focused on the study of the conformation of polypeptides in surfactant solutions [18–30]. Ionic polypeptides are stable in the random coil conformation due to ionic repulsions. Induction of the ordered conformation is dependent on the charge density. Poly(L-lysine) (PLL), for example, induces formation of the β-sheet conformation in alkaline aqueous solutions, whereas poly(L-glutamic acid) induces formation of the α-helix conformation under aqueous acidic conditions [18, 29, 31–41]. Satake and Yang [31] systematically investigated the PLL conformation in anionic surfactant solutions with different alkyl chain lengths and identified the formation of the β-sheet conformation in sodium alkylsulfate solutions, C_nSO_4Na (n=10, 12, 16), and the formation of the α-helix in C_8SO_4Na solutions. Hayakawa et al. [33, 37] extended the work in solutions of sodium alkanesulfonates of different alkyl chain lengths. They found the stepwise conformational change from the coil-to-β-sheet and then to the α-helix as a function of surfactant concentration in C_8SO_3Na aqueous solutions and the single-step conformational change from the coil-to-β-sheet in C_nSO_3Na (n=9,10,11,12) aqueous solutions. Surfactants with an anionic group at each end, such as disodium α,ω-alkanedisulfates $Na_2C_nH_{2n}(SO_4)_2$, were also found to induce the single-step conformational change from the coil-to-β-sheet at n=16, and the double-step conformational change

from the coil-to-β-sheet and then to the α-helix at n=12 for PLL. However, no ordered conformation of PLL was observed in the surfactant solutions at n=10 [18]. The present work revisits the conformation of PLL in C_nSO_3Na (n=8 and 7) solutions and compares the circular dichroism (CD) spectra with the binding isotherms of surfactant at 10, 20, 30, and 40°C.

Experimental Section

Materials

Poly(L-lysine hydrobromide) (M.W. 37,000, Miles-Yeda, Rehovot, Israel) was used as the hydrochloride salt with dialysis in 0.1 M of HCl and water, and the small molecular components were removed. The molar concentration of the residue was determined using micro-Kjeldahl nitrogen analysis. The carbobenzoxyl group contained in commercial polypeptides was determined to be less than 2% by UV spectrometry at 258 nm. Sodium octanesulfonate and heptanesulfonate (C_8SO_3Na and C_7SO_3Na) were synthesized from the corresponding fractionally distilled alkyl bromides by reaction with saturated sodium sulfite, purified by fractional distillation by reaction with saturated sodium sulfite, purified by recrystallization with water once and butanol twice, and unreacted alkyl bromide was extracted with petroleum ether for 20 h [42]. The pure products were dried for 2 h before use.

The carrier complex (CTA-C_8SO_3) used for the surfactant ion-selective liquid membrane electrode was prepared by mixing aqueous C_8SO_3Na solution with hexadecyltrimethylammonium bromide (CTAB) solid. The water used in the current work was distilled twice.

Measurements

Test solutions were prepared by adding PLL stock solution to a solution of sodium alkanesulfonate of a given concentration. The PLL concentration in the test solutions was fixed at 0.211 mM. The mixture sometimes shows a slow appearance of turbidity depending on the surfactant concentration and the temperature. In that case, CD intensity was obtained at the maximum, just before turbidity formation. CD spectra were recorded on a Jasco J-20A spectropolarimeter equipped with a quartz cell (1-cm light-path length) in a thermoregulated cell holder under nitrogen flush. The pH of the test solutions was 5.5–6.0 when PLL was present in the fully charged form in aqueous solution.

Binding isotherms of the C_8SO_3 anion by PLL were constructed from the potentiometric measurements of the equilibrium concentration of an unbound surfactant ion with a homemade liquid membrane electrode responsive to the surfactant ion [43]. The cell constitution is shown below:

Ag/AgCl electrode	Agar bridge	Reference solution C_0	Functional nitrobenzene membrane	Test solution C_X	Agar bridge	Ag/AgCl electrode

The electromotive force (emf, E) is given for this concentration cell constitution by Eq. 1.

$$E = \frac{RT}{F} \ln \frac{C_x}{C_0} \tag{1}$$

where C_X and C_0 are the concentration of the test and reference solutions, respectively; R is the gas constant and T is the temperature in Kelvin. The calibration curves were determined for the solution without PLL before each measurement of mixed solutions of PLL and C_8SO_3Na. The calibration curve afforded a straight line for the E vs log[C_8SO_3Na] plots at concentrations above 0.1 mM, in the absence and presence of NaCl. The slopes of the E vs log C_X plots were 55.3–60.8 mV/decade, depending on the temperature and the added NaCl concentration. They are reasonable from the Nernstian slopes (56.2–62.1 mV/decade at 10–40°).

Results and discussion

Circular dichroism spectra of poly(L-lysine) in C_nSO_3Na aqueous solutions

The ordered conformation formed in ionic polypeptides is dependent on the charge density. For example, PLL induces formation of the β-sheet conformation in alkaline solutions, whereas poly(L-glutamic acid) induces formation of the α-helix conformation in acidic solutions. Satake and Yang [31] identified formation of the β-sheet conformation of PLL in sodium alkylsulfate, C_nSO_4Na (n=10, 12, 16) solutions, and the α-helix conformation of PLL in C_8SO_4Na solutions. Hayakawa et al. [33] extended the work in sodium alkanesulfonates solutions and found the stepwise conformational change from the coil to the β-sheet conformation, and subsequently, to the α-helix as a function of surfactant concentration in C_8SO_3Na solutions. Figure 1 shows the CD spectra of PLL as a function of C_8SO_3Na concentrations at 10 and 30°C. Spectrum a in Fig. 1 represents a typical CD spectrum of the random coil conformation. At low C_8SO_3Na concentrations, spectrum b in Fig. 1 represents the random coil conformation of PLL. As the surfactant concentration further increases the CD spectrum of PLL changes to the

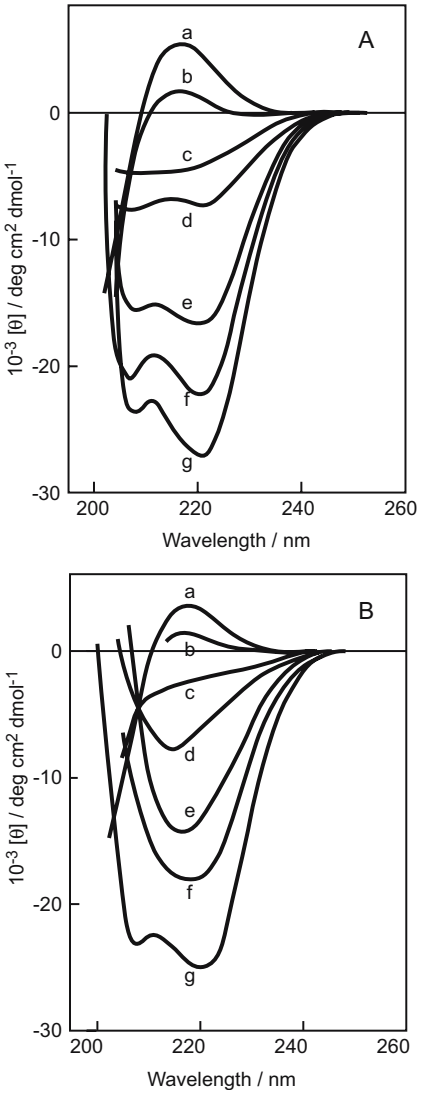

Fig. 1 CD spectra of PLL in C_8SO_3Na aqueous solutions at 10 (**a**) and 40°C (**b**). Salt-free, PLL: 0.211 mM, **a** C_8SO_3Na: *a* 0 mM, *b* 2.95 mM, *c* 3.93 mM, *d* 4.42 mM, *e* 4.92 mM, *f* 5.90 mM, *g* 8.85 mM. **b** C_8SO_3Na: *a* 0 mM, *b* 1.92 mM, *c* 2.46 mM, *d* 2.95 mM, *e* 4.92 mM, *f* 6.39 mM, *g* 8.85 mM

negative direction and reaches spectra g, with double minima at 209 and 222 nm. This double minima was ascribed to the α-helix conformation of PLL. No isosbestic point is observed in Fig. 1A at 10°C. This suggests that the spectrum change reflects a complex change including various intermediate conformations of less ordered conformations rather than a simple coil-to-helix conformational change. Figure 1B also indicates a similar spectral change from a random coil (a) to an α-helix (g). However, spectrum d in Fig. 1B indicates a single minimum at 217 nm. This minima is ascribed to the typical β-sheet conformation. Therefore, this spectral change was ascribed to a double-step conformational change from a random coil to a β-sheet and then to an α-helix as the surfactant concentration increases [18, 33, 37]. The isoelliptic point appears in the spectra shown in Fig. 1c, suggesting a simple conformational change between a typical random coil and a typical β-sheet conformation in the first step at low surfactant concentrations. The CD intensity at 222 nm was plotted against the surfactant concentration in Fig. 2 (top). At low surfactant concentrations, the molar ellipticity changes slowly and decreases quickly above a critical concentration. The curves show a plateau at 3–6 mM at 30 and 40°C, corresponding to the β-sheet conformation. No plateau region was observed at 20°C; however, the spectrum for a β-sheet conformation was observed in the 3.93-mM surfactant solution.

Surfactants with an anionic group at each end, such as disodium α,ω-alkanedisulfates $Na_2C_nH_{2n}(SO_4)_2$, also induced the single-step conformational change from the coil-to-β-sheet at n=16 and the double-step conformational change from coil-to-β-sheet and then to α-helix at n=12 for PLL. However, no ordered conformation of PLL was found in the surfactant solutions at n=10. The effect of C_7SO_3Na surfactant on the PLL conformations was also examined. Figure 3 shows the typical change in the CD spectrum with respect to C_7SO_3Na concentrations at 20°C. Spectrum f clearly indicates the α-helix conformation of PLL; however, no isoelliptic point was observed between spectra a and f, suggesting a complex conformational change including var-

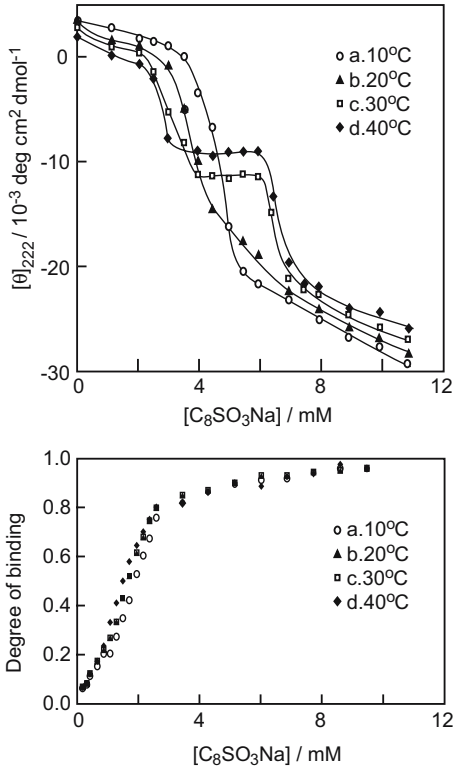

Fig. 2 Dependence of CD intensity at 222 nm (**top**) and the degree of binding (**bottom**) on the total concentration of C_8SO_3Na at various temperatures. Salt-free, PLL: 0.211 mM, Temperature: *a* 10°C, *b* 20°C, *c* 30°C, *d* 40°C

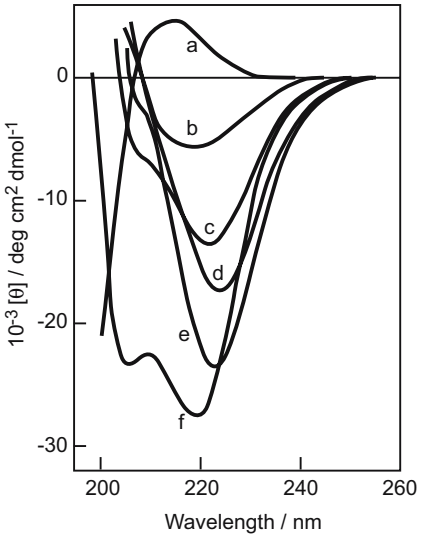

Fig. 3 CD spectra of PLL in C_7SO_3Na solutions at 20°C. Salt-free, PLL: 0.211 mM, C_7SO_3Na: *a* 0 mM, *b* 13.3 mM, *c* 17.0 mM, *d* 21.3 mM, *e* 22.2 mM, *f* 28.6 mM

ious ordered states. The CD intensity at 222 nm was plotted against surfactant concentration in Fig. 4. A plateau was observed at the 16–22 mM C_7SO_3Na concentration range, but the spectra did not show typical ordered conformations such as α-helices or β-sheets at these concentrations.

A conformational phase diagram of PLL in C_8SO_3Na and C_7SO_3Na solutions was constructed from Figs. 2 and 4, and is shown in Fig. 5. The β-sheet conformation was observed at the intermediate concentrations of C_8SO_3Na at temperatures above 20°C, but no β-sheet conformation was induced at 10–40°C in C_7SO_3Na solutions.

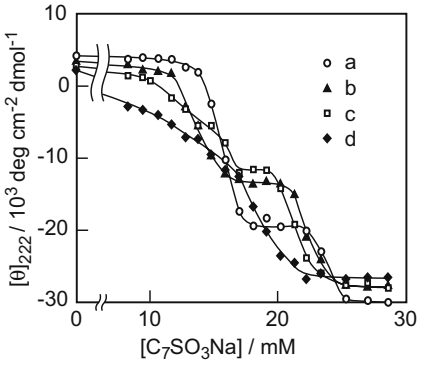

Fig. 4 Dependence of CD intensity at 222 nm of PLL on C_7SO_3Na concentration at various temperatures. Salt-free, Temperature: *a* 10°C, *b* 20°C, *c* 30°C, *d* 40°C

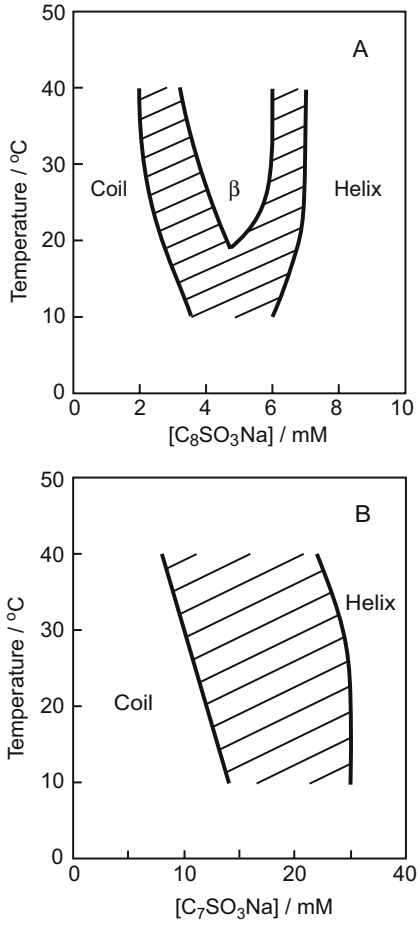

Fig. 5 Conformational phase diagram of PLL in C_8SO_3Na (**a**) and C_7SO_3Na (**b**) aqueous solutions. Salt-free, PLL: 0.211 mM

Binding isotherms of C_8SO_3Na

The equilibrium concentration of surfactant for C_8SO_3Na–PLL systems in the absence and presence of NaCl was determined with potentiometry using a surfactant ion-selective electrode. The calibrations were performed at surfactant concentrations low enough to sufficiently allow accurate determination of the monomer concentration at all temperatures and indicate their linear response as a function of the logarithm of the concentration. The nitrobenzene liquid membrane electrode did not show a reasonable straight line for the calibration of C_7SO_3Na due to the low selectivity for the surfactant ion. Therefore, the binding isotherms for the C_7SO_3Na–PLL system were not measured.

Deviation from the calibration curves indicates surfactant binding by PLL and reduced concentrations of surfactant ions in the bulk solution. No deviation at surfactant concentrations less than log C=−2.4 was observed in the

presence of 21.3 mM NaCl, whereas deviation was observed already at log C=−3.6 in the absence of NaCl. This finding indicates that NaCl interferes with surfactant binding. The comparison of the emf values to the calibration enables determination of the equilibrium surfactant concentration C_F. The degree of binding, y, was calculated using Eq. 2.

$$y = (C_T - C_F)/C_P \tag{2}$$

where C_T is the total surfactant concentration and C_P the residue concentration of PLL. The binding degree of surfactant, y, was plotted against the total concentration of surfactant in the absence of NaCl in Fig. 2 (bottom). A large deviation in total surfactant concentration was observed between the surfactant binding (Fig. 2 (bottom)) and the conformational change (Fig. 2 (top)), suggesting that the conformational change occurs only at a nearly full binding.

The binding isotherms, y vs C_F plots, thus obtained are shown in Fig. 6. All binding isotherms show sigmoidal curves, indicating cooperative binding of the surfactant ion by PLL [44–46]. A steep rise in the curves was observed at a lower equilibrium concentration of C_8SO_3 anions at higher temperatures. This is considered to indicate an endothermic process for surfactant binding. In the presence of 21.3 mM NaCl, the binding isotherms shift to a higher equilibrium concentration of surfactant than those of salt-free mixed solutions. This interference by NaCl is caused by the weakened electrostatic attraction of anionic surfactant with cationic PLL. The cooperative binding model with electrostatic interaction was used to analyze the binding isotherms.

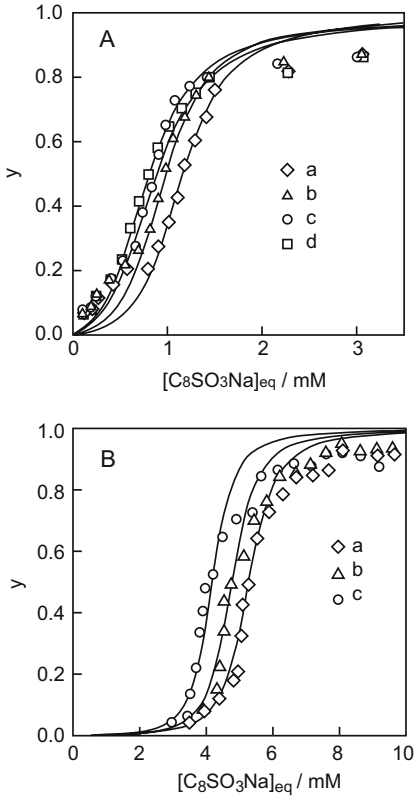

Fig. 6 Binding isotherms of C_8SO_3Na by PLL at various temperatures. **a** Salt-free, *a* 10°C, *b* 20°C, *c* 30°C, *d* 40°C. **b** 21.3 mM NaCl, *a* 10°C, *b* 20°C, *c* 35°C

The binding of surfactant by a PLL binding site is given by Eq. 3.

$$\underset{1}{-NH_3^+} + \underset{2}{S^-} \rightleftarrows \underset{3}{NH_3^+ \cdot S^-} \tag{3}$$

where a PLL binding site is noted as 1, the surfactant ion in the bulk phase is noted as 2, and the ionic complex as 3. At equilibrium,

$$\mu_1 + \mu_2 = \mu_3 \tag{4}$$

Considering the PLL site as a mixture of empty site 1 and the surfactant bound site 3 on a linear lattice and applying the linear lattice model with the Bethe approximation for the chemical potentials of sites 1 and 3, the chemical potentials of ionic site 1 and ion-pair site 3 may be represented by Eqs. 5 and 6, respectively.

$$\mu_1 = \mu_1^0 + kT \ln \frac{\beta + 1 - 2y}{\beta + 1} + e\varphi \tag{5}$$

$$\mu_3 = \mu_3^0 + kT \ln \frac{\beta - 1 + y}{\beta + 1} \tag{6}$$

Where the $e\varphi$ term denotes the electric energy of the ionic site 1 by the electric field φ of PLL and

$$\beta = \sqrt{1 + 4y(1-y)(u-1)} \tag{7}$$

and

$$u = \exp\left(\frac{2E_{13} - E_{11} - E_{33}}{kT}\right) \tag{8}$$

where E_{ij} is the pair interaction energy of neighboring sites, i–j pair. The parameter u is a measure of cooperativity of the surfactant binding. For the chemical potential of surfactant represented by the mole fraction concentration

in bulk solution, the ideal solution was assumed at a low concentration.

$$\mu_2 = \mu_2^0 + kT \ln (C_F/55.5) \tag{9}$$

When $\Delta G^0 = \mu_3^0 - \mu_1^0 - \mu_2^0$ and K is represented as

$$K = \frac{1}{55.5} \exp\left(-\frac{\Delta G^0}{kT}\right) \exp\left(\frac{e\varphi}{kT}\right), \tag{10}$$

the degree of binding y is given by Eq. 11.

$$y = \frac{1}{2}\left\{1 + \frac{KC_F - 1}{\sqrt{(KC_F - 1)^2 + 4KC_F/u}}\right\} \tag{11}$$

This is equivalent to the equation derived from the linear lattice model by Schwarz [47] for a dye-stacking on linear polymer and by Satake and Yang [32] for surfactant binding. From Eq. 11 the value of K is calculated from the C_F at $y = 0.5$ as $1/C_F$.

Applying Eq. 11 for the binding isotherms in Fig. 6, the calculated curves (solid lines) are obtained, giving the best fit to the experimental data for the lower part of the binding curves. The parameter values are listed in Table 1.

The binding constant K at the half-bound point is larger at a higher temperature, indicating that surfactant binding is an endothermic process. The cooperative parameter, u, decreases with temperature for the salt-free system and is independent of the temperature in the presence of 21.3 mM NaCl. The binding of the dodecyltrimethylammonium ion by dextran sulfate at 10 mM NaCl further indicated the temperature dependence of K and u [48]. However, u showed an irregular variation between 4.1 and 7.1 at 5 to 50°C. In the current system, maximum K was observed at 30°C and the enthalpy change was obtained as −9.9 and 9.3 kJ/mol at temperatures above and below 30°C, respectively. However, the present surfactant binding system shows no maximum in K.

Table 1 Binding parameters of C_8SO_3Na binding by PLL

	T (°C)	C_F at y=0.5 (mM)	K=1/C_F at y=0.5	u
Non	10	1.16	860	20
	20	0.98	1,000	15
	30	0.89	1,100	9
	40	0.80	1,240	6
23.1 mM NaCl	10	5.3	190	150
	20	4.8	210	150
	35	4.1	240	150

The thermodynamic parameters were estimated considering the electrostatic interaction term at y=0.5.

$$\frac{d \ln K}{d(1/T)} = -\frac{\Delta H^0}{R} + \frac{d\left(e\varphi_{0.5}/kT\right)}{d(1/T)} \tag{12}$$

$$T\Delta S^0 = \Delta H^0 - \Delta G^0 \tag{13}$$

where $\varphi_{0.5}$ is the electrostatic potential at $y = 0.5$, ΔH^0 is the enthalpy change, and ΔS^0 is the entropy change for the binding of C_8SO_3Na by PLL. To calculate for $\varphi_{0.5}$, the smeared charge model was utilized in which the following assumptions were introduced: (1) the charge density is smeared, (2) the counterion condensation occurs at the charge density parameter $\xi > 1$ given by $e^2/DkTd$ and the effective ξ_{eff} is unity, (3) the interaction energy between polyions is neglected, and (4) the Debye–Hückel approximation is applied for uncondensed ions. D denotes the dielectric constant and d the average charge separation, which is equal to $2d_0$ at y=0.5. d_0 is the intrinsic charge separation determined from the polyion chemical structure. Eq. 14 for $\varphi_{0.5}$ was derived based on these assumptions

$$\frac{e\varphi_{0.5}}{kT} = \frac{e^2}{DkTd_0}(0.116 - \ln \kappa a) \tag{14}$$

where κ is the inverse of the Debye length calculated from the ionic strength, a is the distance from polyion rod center set as 0.63 nm from the lysine side chain length, and d_0 was set as 0.38 nm.

The plots of ln K vs $1/T$ and $e\varphi_{0.5}/kT$ vs $1/T$ are given in Fig. 7. ΔH^0 was calculated from the slopes of the two plots using Eq. 12, and ΔG^0 and ΔS^0 were calculated using Eqs. 10 and 13, respectively. The values obtained are given in Table 2. In the presence of 21.3 mM NaCl, the electrostatic term showed lower values than the salt-free system due to the shielding effect by added salt. A weak temperature dependence was also observed due to the minor change of ionic strength, which resulted in an increased ΔH^0. Table 2 also indicates that the Gibbs energy change is mostly introduced by the entropy change ($-T\Delta S^0$). This finding indicates that the surfactant binding by PLL at y=0.5 is a result of the hydrophobic interaction of bound surfactants, which leads to a clustering of bound surfactant. The molar thermodynamic parameters of micelle formation of C_8SO_4Na are −15 kJ for ΔG^0, 6.3 kJ for ΔH^0, and −21.4 kJ for $-T\Delta S^0$ at 25°C [49]. These values are close to those of the present system, and thus, may support the formation of a surfactant cluster on PLL.

The conformational phase diagram was depicted as a function of the degree of binding of C_8SO_3Na in Fig. 8.

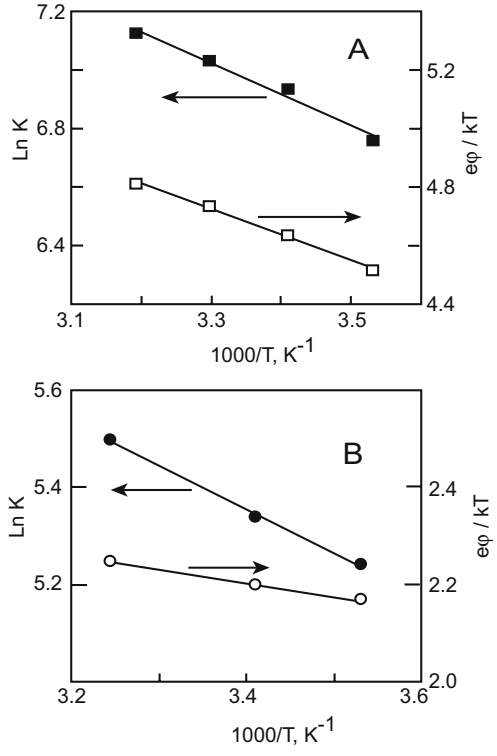

Fig. 7 Temperature dependence of K (*solid marks*) and the electrostatic energy term (*open marks*) in the absence (a) and presence of 21.3 mM NaCl (b)

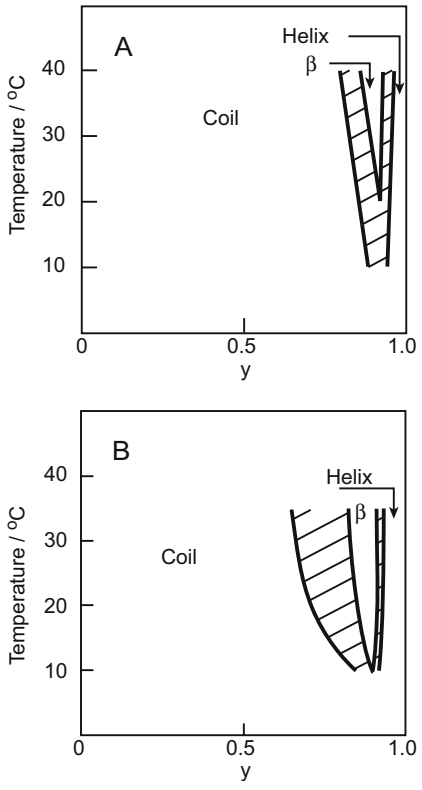

Fig. 8 The conformational phase diagram of PLL as a function of the degree of surfactant binding

The conformational change begins at a high degree of binding, and the helix conformation is observed at nearly full binding (y>0.9) in the absence and presence of NaCl. Satake and Yang proposed a model in which the ordered structure was stable inside a surfactant cluster and calculated the minimum cluster size of 7 or 8 for helix stabilization. Hayakawa and coworkers [18, 50] qualitatively interpreted the temperature dependence of conformational change of PLL and determined that this dependence was induced by the cluster size change.

Table 2 Molar thermodynamic parameters for the binding of C_8SO_3Na by PLL

	T (°C)	$e\varphi$ (kJ mol^{-1})	ΔH^0 (kJ)	ΔG^0 (kJ)	$-T\Delta S^0$ (kJ)	ΔS^0 (J K^{-1})
Salt-free	10	10.6	1.59	−14.7	−16.3	58
	20	11.3		−15.4	−17.0	
	30	11.9		−15.9	−17.5	
	40	12.5		−16.5	−18.1	
21.3 mM NaCl	10	5.1	5.17	−16.7	−21.9	77
	20	5.4		−17.4	−22.6	
	35	5.8		−18.6	−23.8	

C_8SO_3Na-binding by PLL provided a less cooperative parameter u than C_nSO_3Na (n>8) binding, leading to the formation of small cluster followed by a lack of an ordered conformation at a low binding degree [37]. In the present system, the cooperative parameter u was larger for the added salt system than the salt-free system. The ordered conformation appears at a lower degree of binding for the added salt system than the salt-free system, which corresponds to the interpretation by cluster size dependence.

Conclusions

Conformational phase diagrams of PLL were constructed based on CD spectrometry data. PLL indicated the double-step conformational change from the random coil to the β-sheet and then an α-helix as a function of the C_8SO_3Na concentration at 20–40°C. On the other hand, a single-step conformational change from the random coil to an α-helix was observed at 10°C. C_7SO_3Na induced the single-step conformational change from random coil to an α-helix at 10–40°C. The binding isotherms of C_8SO_3Na by PLL were determined to indicate cooperative binding characteristics. Data analysis revealed the entropy-driven process of the binding of C_8SO_3Na by PLL, and the electrostatic term was

reduced in the presence of 21.3 mM NaCl. The ordered conformation was determined to appear at nearly complete binding, suggesting that the ordered conformation is stable in a surfactant cluster on PLL. This stability is thought to be due to the formation of a hydrophobic environment as well as charge neutralization.

References

1. Lapanje S (1978) Physicochemical aspects of protein denaturation. Wiley, New York
2. Inoue Y, Sase S, Chujo R, Nagaoka S, Sogami M (1979) Biopolymers 18:273–382
3. Fukushima K, Murata Y, Nishikido N (1981) Bull Chem Soc Jpn 54:3122–3127
4. Fukushima K, Murata Y, Sugihara G, Tanaka M (1982) Bull Chem Soc Jpn 55:1376–1378
5. Guo XH, Zhao NM, Chem SH, Teixeira J (1990) Biopolymers 29:335–346
6. Ananthapadmanabhan KP (1992) In: Goddard ED Ananthapadmanabhan KP (eds) Interactions of surfactants with polymers and proteins. CRC, Boca Raton, FL, pp 319–365
7. Tsuji E, Maeda H (1992) Colloid Polym Sci 270:894–900
8. Turro NJ, Lei X-G, Ananthapadmanabhan KP, Aronson M (1995) Langmuir 11:2525–2533
9. Takeda K, Moriyama Y (1997) Curr Top Colloid Interface Sci 1:109–135
10. Fainerman VB, Lucassen-Reynders EH, Miller R (1998) Colloid Surf A Physicochem Eng Asp 143:141–165
11. Moriyama Y, Takeda K (1999) Langmuir 15:2003–2008
12. Murakami K (1999) Langmuir 15:4270–4275
13. Savelli G, Spreti N, Di Profio P (2000) Curr Opin Colloid Interface Sci 5:111–117
14. Miksik I, Eckhardt A, Forgacs E, Cserhati T, Deyl Z (2002) Electrophoresis 23:1882–1886
15. Diaz X, Abuin E, Lissi E (2003) J Photochem Photobiol A Chem 155:157–162
16. Gohon Y, Popot J-L (2003) Curr Opin Colloid Interface Sci 8:15–22
17. Santos SF, Zanette D, Fischer H, Itri R (2003) J Colloid Interface Sci 262: 400–408
18. Hayakawa K, Fujita M, Yokoi S, Satake I (1991) J Bioact Compat Polym 6:36–51
19. Takeda K, Wada A, Sato Y, Hamada S (1991) J Colloid Interface Sci 147:51–56
20. Satake I, Uchino K, Hayakawa K, Maeda T (1992) Bull Chem Soc Jpn 65:1146–1148
21. Hayakawa K, Satake I, Kwak JCT (1995). Conformation of cationic polypeptides in solutions of anionic surfactants. In: International symposium on polyelectrolytes and international bunsen-discussion–meeting, Potsdam, Germany, 18–20 September 1995
22. Ponomarenko EA, Waddon AJ, Tirrell DA, MacKnight WJ (1996) Langmuir 12:2169–2172
23. Kurawaki J, Sameshima Y, Kusumoto Y (1997) J Phys Chem B 101:10548–10553
24. Liu J, Takisawa N, Kodama H, Shirahama K (1998) Langmuir 14:4489–4494
25. Liu J, Takisawa N, Shirahama K (1998) J Phys Chem B 102:6696–6701
26. Kurawaki J, Kusumoto Y (2000) J Colloid Interface Sci 225:265–272
27. Lezov AV, Mel'nikov AB, Polushina GE, Ryumtsev EI, Lysenko EA, Zezin AB, Kabanov VA (2001) Polym Sci Ser A 43:915–920
28. Kamio K, Sakai M, Kaminaga A, Satake I, Hayakawa K (2002) Colloid Polym Sci 280:78–84
29. Kamande MW, Zhu X, Kapnissi-Christodoulou C, Warner IM (2004) Anal Chem 76:6681–6692
30. Polushina GE, Ryumtsev EI, Lezov AV, Lysenko EA (2004) Polym Sci Ser A 46:491–497
31. Satake I, Yang JT (1973) Biochem Biophys Res Commun 54:930–936
32. Satake I, Yang JT (1976) Biopolymers 15:2263–2275
33. Hayakawa K, Ohara K, Satake I (1980) Chem Lett:647–650
34. Takeda K, Iba A, Shirahama K (1981) Bull Chem Soc Jpn 54:1793–1796
35. Takeda K, Iba A, Shirahama K (1982) Bull Chem Soc Jpn 55:985–989
36. Takeda K (1985) Bull Chem Soc Jpn 58:1210–1214
37. Hayakawa K, Murata H, Satake I (1990) Colloid Polym Sci 268: 1044–1051
38. Aoyagi M, Maeda T, Hayakawa K, Satake I (1991) Bull Chem Soc Jpn 64:1378–1380
39. Satake I, Gondo T, Kimizuka H (1979) Bull Chem Soc Jpn 52:361–364
40. Maeda H, Kato H, Ikeda S (1984) Biopolymers 23:1333–1346
41. Hayakawa K, Nagahama T, Satake I (1994) Bull Chem Soc Jpn 67:1232–1237
42. Zuffanti S (1940) J Am Chem Soc 62:1044
43. Satake I, Noda S, Maeda T (1979) Bull Chem Soc Jpn 52:361–364
44. Hayakawa K, Kwak JCT (1982) J Phys Chem 86:3866–3870
45. Hayakawa K, Kwak JCT (1983) J Phys Chem 87:506–509
46. Hayakawa K, Kwak JCT (1991) In: Rubingh DNHolland PM (eds) Cationic surfactants: physical chemistry, Marcel Dekker, New York, pp 189–248
47. Schwarz G (1970) Eur J Biochem 12:442–453
48. Santerre JP, Hayakawa K, Kwak JCT (1985) Colloids Surf 13:35–45
49. Sarker PK, Doty P (1966) Proc Natl Acad Sci USA 55:981
50. Satake I, Hayakawa K (1990) Chem Lett:1051–1054