Effect of Temperature on the Surface Properties of Aqueous Solution of Arachidonic Acid

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The surface tension of aqueous solution of arachidonic acid (AA) was measured with a Du Nöuy tensiometer at pH 7.80 and various temperatures, and the effect of temperature on the surface properties of AA was investigated. The value of critical micelle concentration (cmc) of AA increased and the surface tension at the cmc, γ_{cmc}, decreased as the temperature was raised from 30 to 60 °C. The Langmuir constant k concerning the strength of adsorption decreased, the saturated adsorption amount of AA, Γ_∞, at the air–water interface decreased, and the area occupied by an AA molecule at the saturated adsorption, A_∞, increased as the temperature was raised. The standard free energy for the adsorption, ΔG_{ad}, was of negative value, and the absolute value $|\Delta G_{ad}|$ became larger as the temperature was raised because of the negative value of the standard enthalpy change ΔH_{ad} and the positive value of the standard entropy change ΔS_{ad}. The standard free energy of micellization, ΔG_m, was of negative value, and it was similarly found that $|\Delta G_m|$ became larger as the temperature was raised because of the negative value of the standard enthalpy change ΔH_m and the positive value of the standard entropy change ΔS_m.

Keywords arachidonic acid; surface tension; critical micelle concentration; micellization energy; adsorption; adsorption energy

Regarding the effect of temperature on the critical micelle concentration (cmc), it has been known that in general cmc increases as temperature is raised; the cmc of potassium alkanoate increases as the temperature is raised from 25 to 45 °C,[1)] and the cmc of sodium alkyl sulfonate increases as the temperature is raised from 40 to 50 °C.[1)] On the contrary, at lower temperature regions the cmc of sodium 10-undecenoate decreases as the temperature is raised from 10 to 30 °C.[2)] In addition, sodium dodecyl sulfate shows a minimum in the cmc–temperature profile at 25 °C.[3)]

Arachidonic acid (AA), [eicosa-*cis*-5,8,11,14-tetraenoic acid], is well known as a precursor of prostaglandins.[4)] Regarding the surface properties of AA, the surface pressure for the insoluble monomolecular film of AA has been measured at various temperatures, and the relationship between the limiting area of an AA molecule and temperature has been reported.[5)] However, the effect of temperature on the surface properties of an aqueous solution of AA has not yet been studied.

From these points of view, the surface tension of an aqueous solution of AA was measured at various temperatures and the effect of temperature on the surface properties of AA was investigated. Furthermore, the adsorption of AA at the air–water interface and the micellization of AA were discussed from the thermodynamic viewpoint.

Experimental

Materials AA was obtained from Wako Pure Chemicals. The purity was 99%, and was used without further purification. The peroxide value of AA was 0.02 m eq·kg^{-1}.

Preparation of Solution It is not easy to dissolve a large amount of AA directly in an aqueous solution of pH 7.80. So, a solution of AA was prepared as follows. To an aqueous solution containing an equivalent amount of NaOH, AA was added with stirring. After complete saponification, the pH 7.80 phosphate buffer, which contains 50 mol·m^{-3} of NaH_2PO_4 and 42.8 mol·m^{-3} of NaOH, was added.

Measurement of Surface Tension The surface tension was measured with a Du Nöuy tensiometer (Shimazu) with a platinum ring (d=23 mm) at pH 7.80 and ionic strength 93 mol·m^{-3}. This instrument was the same as that used previously[6)] and the method of handling was described in that paper. The measurement was carried out at 30.00, 40.02, 50.25 and 59.30±0.1 °C. For the calculation of the surface tension of aqueous solutions, the values of 71.15, 69.55, 67.90 and 66.17 mN·m^{-1} were used as the surface tension of pure water at those temperatures, respectively. The experimental determination of the surface tension was precise to ±0.1 mN·m^{-1}. Deionized and twice-distilled water was used throughout this study.

Regarding the oxidation of unsaturated fatty acids by air, the following have been reported: (1) the surface pressures of the spreading monomolecular films of AA and the other unsaturated fatty acids with 1—6 double bonds are measured stably at least up to 40 °C without the influence of oxidation[5a)]; (2) the activation energy for the oxidation of ethyl eicosapentaenoate, which undergoes the oxidation reaction more easily than AA, is 57.7 kJ·mol^{-1};[7)] (3) the oxidation reaction has an induction period and then the oxidative product increases with time.[7)] Taking these reports into account, the oxidative product of AA seems to be not seriously produced during a short period even through at 59.30 °C.

Results

Surface Tension of the Aqueous Solution of AA The change in the surface tension, γ, of the aqueous solution of AA with time was shown in Fig. 1. As can be seen in Fig.

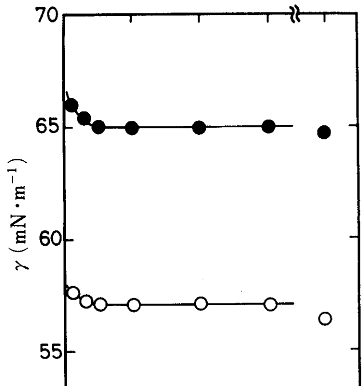

Fig. 1. Change in Surface Tension with Time

Concentration: ●, 9.5×10^{-4} mol·m^{-3}; ○, 9.5×10^{-3} mol·m^{-3}. Temperature: 60 °C.

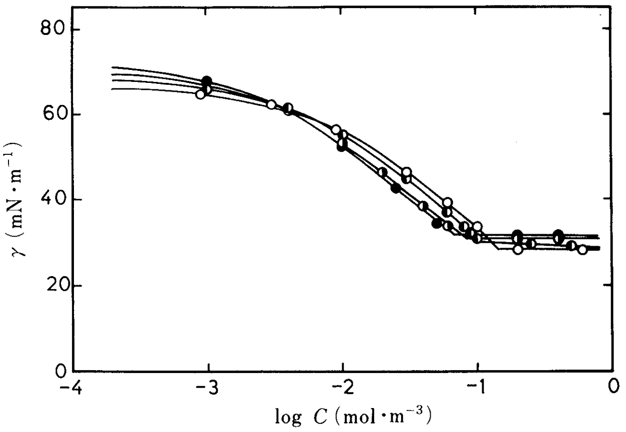

Fig. 2. Surface Tension of Aqueous Solution of AA at pH 7.80

Temperature: ●, 30 °C; ◑, 40 °C; ◐, 50 °C; ○, 60 °C.

Table I. Physicochemical Parameters for Micellization of AA

T (K)	cmc (mol·m^{-3})	γ_{cmc} (mN·m^{-1})	ΔG_{m} (kJ·mol^{-1})
303.15	6.8×10^{-2}	32.0	−54.2
313.17	8.4×10^{-2}	31.0	−55.1
323.40	1.1×10^{-1}	30.1	−55.7
332.45	1.4×10^{-1}	28.5	−56.2

1, the adsorption equilibrium has been attained at 5 min, although the time required for the equilibrium for the solution with a lower concentration is slightly longer than that with a higher concentration. The value of γ at the adsorption equilibrium was determined throughout this study. In addition, an evident decrease in γ, even at 60 °C and 240 min, was not found. So the effect of oxidation of AA should be negligible in this experiment. The change of γ of AA as a function of the logarithm of concentration is shown in Fig. 2. The values of cmc and the surface tension at the cmc, γ_{cmc}, were determined from the break points, and were listed in Table I. As can be seen in Table I, the value of cmc increased and the value or γ_{cmc} decreased as the temperature was raised.

Adsorption of AA at the Air–Water Interface The adsorption amount, Γ, is calculated by the Gibbs adsorption equation:

$$\Gamma = -\frac{C}{iRT}\left(\frac{\partial \gamma}{\partial C}\right) \tag{1}$$

where $\boldsymbol{R}$ is the gas constant, T is the absolute temperature, and C is the molar concentration of surface active substances. In addition,

$$i = \nu_+\left(\frac{\nu_+ C}{\nu_+ C + \nu_s C_S}\right) + \nu_- \tag{2}$$

where ν_+ and ν_- are the number of cation and anion, respectively, formed from a surface active molecule, and ν_s is the number of cation in a molecule of added salt, and C_S is the concentration of the surface-inactive salt. Under our experimental conditions, $i \fallingdotseq \nu_- = 1$. Therefore, Eq. 1 simply becomes:

$$\Gamma = -\frac{C}{RT}\cdot\frac{\partial \gamma}{\partial C} \tag{3}$$

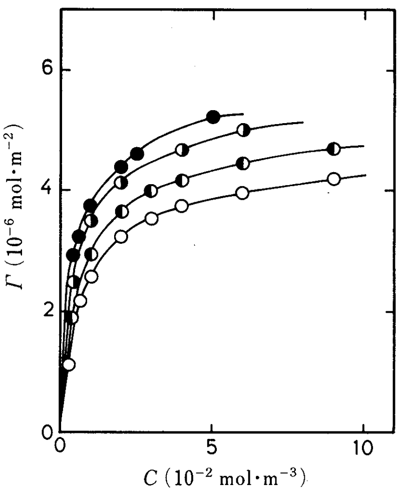

Fig. 3. Adsorption Amount of AA at Air–Water Interface

Symbols are the same as in Fig. 2.

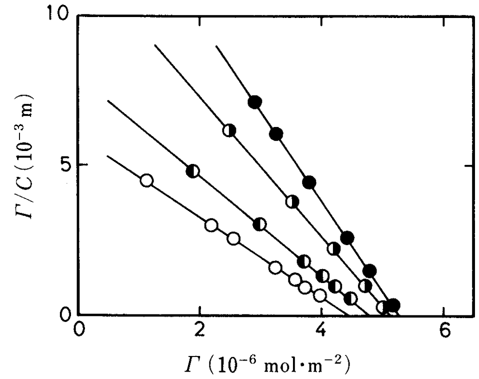

Fig. 4. Scatchard Plots

Symbols are the same as in Fig. 2.

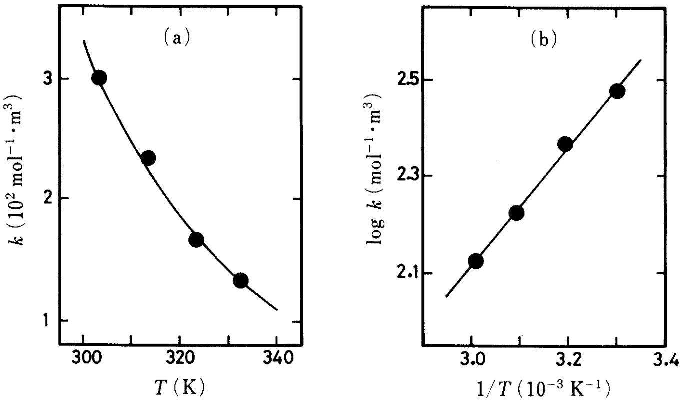

Fig. 5. Relationship between Langmuir Constant, k, and Temperature

Using Eq. 3, the adsorption amount, Γ, is calculated from the slope of curves in Fig. 1. The values of Γ thus obtained are plotted against C in Fig. 3. Since the curves in Fig. 3 are of the Langmuir type, the Langmuir equation is applied to these systems.

$$\frac{\Gamma}{C} = k(\Gamma_\infty - \Gamma) \tag{4}$$

where Γ_∞ is the saturated adsorption amount and k is the Langmuir constant concerning the strength of adsorption. The Scatchard plots of Γ/C against Γ are presented in Fig.

TABLE II. Physicochemical Parameters for Adsorption of AA at Air–Water Interface

T (K)	k ($\mathrm{mol^{-1}\cdot m^3}$)	Γ_∞ ($\mathrm{mol\cdot m^{-2}}$)	A_∞ ($\mathrm{\AA^2}$)	K ($\mathrm{mN\cdot m^{-1}}$)	α ($\mathrm{mN\cdot m^{-1}\cdot mol^{-1}\cdot m^3}$) from Fig. 6,	from Eq. 10	ΔG_{ad} ($\mathrm{kJ\cdot mol^{-1}}$)
303.15	3.00×10^2	5.27×10^{-6}	31.5	30.6	4.0×10^3	3.99×10^3	−38.3
313.17	2.33×10^2	5.13×10^{-6}	32.4	30.8	3.1×10^3	3.12×10^3	−38.9
323.40	1.67×10^2	4.78×10^{-6}	34.7	29.6	2.2×10^3	2.14×10^3	−39.2
332.45	1.33×10^2	4.45×10^{-6}	37.3	28.3	1.6×10^3	1.64×10^3	−39.6

4, indicating a good linear relationship. From the slope and intercept, k and Γ_∞ were obtained and were listed in Table II. The values of k and Γ_∞ decreased as the temperature was raised. Furthermore, the relationship between $\log k$ and $1/T$ indicated a linearity, as shown in Fig. 5.

The area occupied by a molecule at the saturated adsorption, A_∞, is obtained by the following equation:

$$A_\infty = \frac{1}{\Gamma_\infty \times N_{\mathrm{A}}} \tag{5}$$

where N_{A} is the Avogadro number. The values of A_∞ thus obtained are shown in Table II. The value of A_∞ increased as the temperature was raised. It was reported that the limiting area of AA in the spreading insoluble monomolecular film decreased slightly from 67 to 64 Å^2 molecule^{-1} when the temperature was raised from 5 to 20 °C.[5b)] This phenomenon was explained in that the interactions between AA molecules and/or between AA and water molecules became weaker with the temperature because of the thermal motion and that the AA molecule came to orient vertically.[5b)] In this paper, the area occupied by an AA molecule when AA was adsorbed at the air–water interface increased as the temperature was raised. The increase in the area occupied by a molecule is due to the decrease in the adsorption amount of AA at the air–water interface. The difference in the temperature dependence on the area occupied is considered to be caused by the difference between the spreading insoluble monolayer and the adsorption monolayer, namely the difference is caused in that the value of A_∞ obtained in this study is the value at the adsorption equilibrium. Furthermore, the value of A_∞ for AA at 30 °C was 31.5 Å^2, which is nearly equal to the area occupied by one molecule of normal fatty acid, 30.3 Å^2, at the adsorbed monolayer.[1)] It is, therefore, suggested that an AA molecule is oriented relatively vertically at the saturated adsorption.

Surface Pressure of the Adsorbed Monolayer Surface pressure, F, is expressed by the following equation:

$$F = \gamma_0 - \gamma \tag{6}$$

where γ_0 is the surface tension of pure water. The values of γ_0 are 71.15, 69.55, 67.90 and 66.17 mN·m^{-1} at 30, 40, 50 and 60 °C, respectively. Furthermore, the relationship between surface pressure and concentration can be expressed by the following equation proposed empirically by Szyszkowski and may be derived theoretically from Eqs. 3 and 4:

$$\gamma_0 - \gamma = F = K\log(1 + kC) \tag{7}$$

where K is the Szyszkowski constant and k is the Langmuir constant. When $kC \gg 1$, Eq. 7 is rewritten as follows:

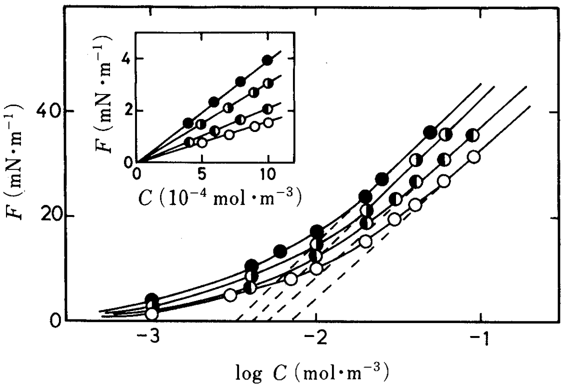

Fig. 6. Relationship between Surface Pressure and Concentration
Symbols are the same as in Fig. 2.

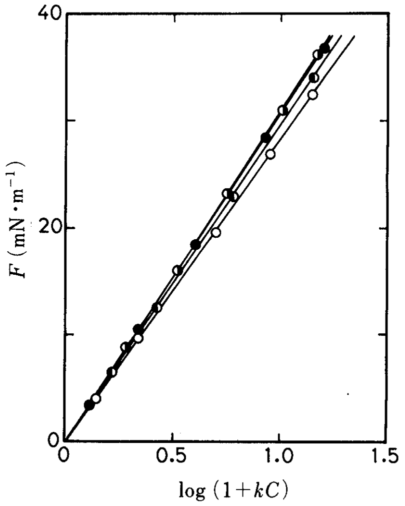

Fig. 7. Szyszkowski Plots
Symbols are the same as in Fig. 2.

$$F = K\log k + K\log C \qquad (kC \gg 1) \tag{8}$$

The relationship between F and $\log C$ is shown in Fig. 6. The relationship between F and C at lower concentration regions was also shown in Fig. 6 as an insertion figure. The Traube's constant, α, is expressed as follows[8)]:

$$\alpha = \lim_{C\to 0}\left(\frac{dF}{dC}\right) \tag{9}$$

A linear relationship was found between F and C at the lower concentrations, as can be seen in Fig. 6. The values of α were determined from the slopes of the straight lines, and were listed in Table II. Spitzer and Heerze[9)] reported that the value of α can be calculated by using the values of k and K as follows:

$$\alpha=\frac{kK}{2.303} \tag{10}$$

The values of α calculated from Eq. 10 were listed in Table II together with the values obtained graphically, and close agreement was obtained. The value of α decreased as the temperature was raised.

The value of K can be determined by Eq. 7 by plotting the relationship between F and $\log(1+kC)$; as is shown in Fig. 7. As can be seen in Fig. 7, good linearity was obtained. So the values of K were determined from the slopes, and were listed in Table II. The dotted straight lines in Fig. 6 were drawn according to Eq. 8 by using values of k and K shown in Table II. It is expected that the relationship between F and $\log C$ has a linearity at higher concentration regions. As can be seen in Fig. 6, a close agreement between the observed and calculated values of F was obtained at $kC \gg 1$. In addition, the value of K is related to Γ_∞ as follows[1]:

$$\Gamma_\infty=\frac{0.4343\,K}{RT} \tag{11}$$

The values of Γ_∞ calculated from Eq. 11 were in agreement with the values obtained from the Scatchard plots (Fig. 4).

***F–A* Curve** The area occupied by a molecule when Γ mol of AA was adsorbed at the air–water interface is expressed as follows:

$$A=\frac{1}{\Gamma\times N_A} \tag{12}$$

Furthermore, the relationship between F and A can be derived as follows by substituting Eqs. 4, 5 and 12 into Eq. 7:

$$F=K\log\left(\frac{A}{A-A_\infty}\right) \tag{13}$$

Equation 13 corresponds to the equation substituted zero for the energy of lateral interaction in the equation by Nakagaki *et al.*[10] with regard to the theory of the adsorption monolayer with lateral intermolecular interaction.

The values of A were calculated by Eq. 12 from the values of Γ shown in Fig. 3, and plotted in Fig. 8 with the F values obtained by Eq. 6. The plots for AA at four temperatures are compared with solid lines, which are the F–A curves calculated from Eq. 13 by using the values of K and A_∞ given in Table II. Close agreements between the plots and the solid curves were obtained.

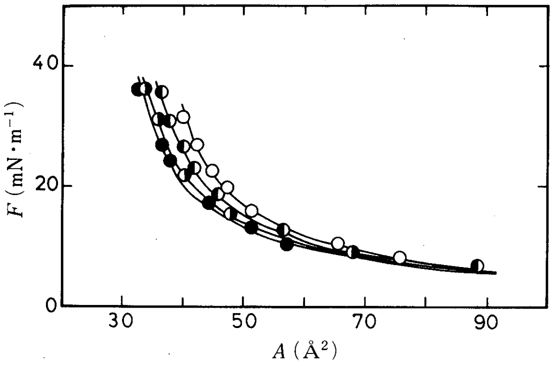

Fig. 8. *F–A* Curve

Symbols are the same as in Fig. 2.

Discussion

Thermodynamic Quantities for the Adsorption The standard free energy for the adsorption, ΔG_{ad}, is defined as follows[11]:

$$\Delta G_{ad}=-RT\ln\alpha \tag{14}$$

where α is the Traube's constant. According to Eq. 14, the values of ΔG_{ad} at each temperature were calculated, and were listed in Table II. The negative value of ΔG_{ad} became larger as the temperature was raised, although the value of k, which is a parameter concerning the strength of adsorption, decreased as the temperature was raised, indicating that the adsorption of AA at the air–water interface became favorable as the temperature was raised from the viewpoint of thermodynamics. So, the contributions of the standard enthalpy change, ΔH_{ad}, and the standard entropy change, ΔS_{ad}, for the adsorption of AA will be investigated. ΔG_{ad} is related to ΔH_{ad} and ΔS_{ad} as follows:

$$\Delta G_{ad}=\Delta H_{ad}-T\Delta S_{ad} \tag{15}$$

$$\frac{\Delta G_{ad}}{T}=\frac{\Delta H_{ad}}{T}-\Delta S_{ad} \tag{16}$$

According to Eqs. 15 and 16, the plots of ΔG_{ad} *vs.* T and $\Delta G_{ad}/T$ *vs.* $1/T$ were shown in Fig. 9. Good linearities were found, so the values of ΔH_{ad} and ΔS_{ad} were obtained from the linear relationship by using Eq. 16, and were summarized in Table III. The values of ΔH_{ad} and ΔS_{ad} are negative and positive, respectively. It was found that both ΔH_{ad} and ΔS_{ad} contribute to the negative value of ΔG_{ad}; the adsorption of AA at the air–water interface was favorable not only from enthalpic, but also from entropic, viewpoints. The phenomenon that the adsorption becomes favorable from

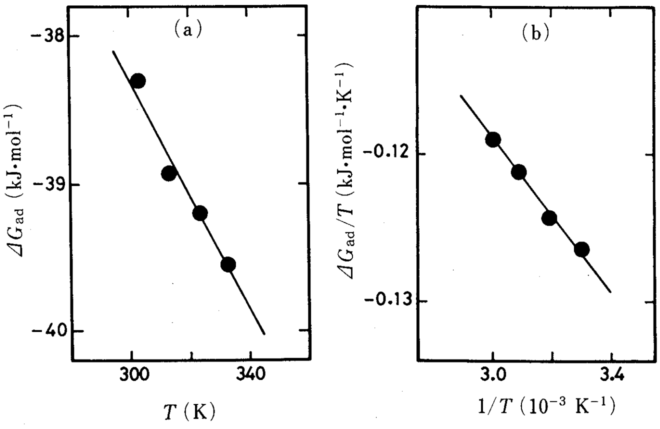

Fig. 9. Relationship between Standard Free Energy for Adsorption, ΔG_{ad}, and Temperature

(a) ΔG_{ad} *vs.* T, (b) $\Delta G_{ad}/T$ *vs.* $1/T$.

Table III. Thermodynamic Quantities for Adsorption of AA at Air–Water Interface

ΔH_{ad} (kJ·mol^{-1})	ΔS_{ad} (J·mol^{-1}·K^{-1})
−27	39

the viewpoint of free energy although the value of k decreases as the temperature is raised can be explained by the positive value of ΔS_{ad}. In addition, it is considered that the positive value of ΔS_{ad} is caused in that the iceberg of water surrounding the hydrophobic moieties of AA is liberated when the hydrophobic moieties are transferred from the aqueous phase to the air–water interface and that the entropy of the water molecule is recovered.

The value of ΔG_{ad} of disodium dodecyl phosphate, whose k value is $4.5\times10^{2}\,\mathrm{mol^{-1}\cdot m^{3}}$, is calculated as $-37.3\,\mathrm{kJ\cdot mol^{-1}}$ at 25 °C by using the values[12] of k and Γ_{∞} and Eqs. 10, 11 and 14. The value of k for AA at 30 °C was $3.0\times10^{2}\,\mathrm{mol^{-1}\cdot m^{3}}$, and the value of ΔG_{ad} for AA at 30 °C was $-38.3\,\mathrm{kJ\cdot mol^{-1}}$. The adsorption energy of the double bond, ΔG_{ad} (-double bond-), is a positive value (approximately 2.5 kJ (-double bond-)$^{-1}$),[13] so that the negative value of the adsorption energy for the molecule with double bonds becomes smaller than that without a double bond when the comparison is made between the molecules whose carbon numbers in the alkyl chain is the same. So it seems to be adequate that the value of ΔG_{ad} for AA, which has four double bonds, is similar to that for disodium dodecyl phosphate.

Thermodynamic Quantities for Micellization The standard free energy of micellization, ΔG_m, is defined as follows[14]:

$$\Delta G_m = 2.303RT(1+\beta)(\log \mathrm{cmc} - \log \omega) \tag{17}$$

where β is the ratio of counter ion bound to micelle, the value of β is reported as 0.58 for carboxylic acid,[14] and ω is the molar concentration of water, the values of ω calculated from the values of the density of water[15] are 55.3×10^{3}, 55.1×10^{3}, 54.8×10^{3} and $54.6\times10^{3}\,\mathrm{mol\cdot m^{-3}}$ at 30, 40, 50 and 60 °C, respectively. The values of ΔG_m calculated by Eq. 17 were listed in the righthand column in Table I. The negative value of ΔG_m becomes larger as the temperature is raised, indicating that the micellization of AA becomes favorable from the viewpoint of thermodynamics although the value of cmc becomes larger as the temperature is raised. So the contribution of the standard enthalpy change, ΔH_m, and the standard entropy change, ΔS_m, for the micellization were investigated. ΔG_m is related to ΔH_m and ΔS_m as follows:

$$\Delta G_m = \Delta H_m - T\Delta S_m \tag{18}$$

$$\frac{\Delta G_m}{T} = \frac{\Delta H_m}{T} - \Delta S_m \tag{19}$$

The plots of ΔG_m *vs.* T and $\Delta G_m/T$ *vs.* $1/T$ were shown in Figs. 10a and 10b, respectively. As can be seen in Fig. 10, good linear relationships were obtained, so the values of ΔH_m and ΔS_m were obtained from the linear relationship by using Eq. 19 and were summarized in Table IV. The values of ΔH_m and ΔS_m are negative and positive, respectively, and both ΔH_m and ΔS_m contribute to the negative value of ΔG_m; the micellization of AA was favorable enthalpically and entropically. The negative value of ΔH_m and the positive value of ΔS_m are considered to be due to that the iceberg of water surrounding the hydrophobic moieties of AA is liberated when the hydrophobic moieties are transferred from the aqueous bulk phase to the micellar phase.

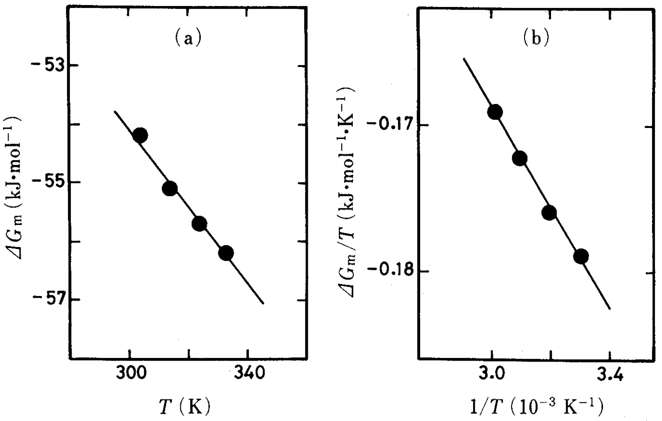

Fig. 10. Relationship between Standard Free Energy for Micellization, ΔG_m, and Temperature

(a) ΔG_m *vs.* T, (b) $\Delta G_m/T$ *vs.* $1/T$.

TABLE IV. Thermodynamic Quantities for Micellization of AA

ΔH_m (kJ·mol^{-1})	ΔS_m (J·mol^{-1}·K^{-1})
−35	65

The values of ΔG_m for sodium 11-(N-methyl acrylamido) undecanoate (MAAU), whose cmc values are $4.7\,\mathrm{mol\cdot m^{-3}}$ at 298 K and $5.1\,\mathrm{mol\cdot m^{-3}}$ at 303 K, are $-43\,\mathrm{kJ\cdot mol^{-1}}$ at 298 K and $-44\,\mathrm{kJ\cdot mol^{-1}}$ at 303 K, and the negative value of ΔG_m becomes slightly larger as the temperature is raised.[16] The absolute value of negative ΔG_m for AA is larger than that for MAAU, and this seems to be adequate taking into account the cmc values. In addition, it was reported that the values of ΔH_m and ΔS_m for MAAU were $-36\,\mathrm{kJ\cdot mol^{-1}}$ and $25\,\mathrm{J\,mol^{-1}\cdot K^{-1}}$, respectively.[16] As compared with these values, the values of ΔH_m and ΔS_m for AA are also considered to be adequate.

Next, log cmc is expressed as follows from Eqs. 17 and 18:

$$\log \mathrm{cmc} = \frac{\Delta H_m}{2.303RT(1+\beta)} - \frac{\Delta S_m}{2.303R(1+\beta)} + \log \omega \tag{20}$$

As can be seen in Table I, the value of cmc increased as the temperature was raised. This is realized from Eq. 20; the first term at the right side, which depends on temperature, becomes of negatively smaller value as the temperature is raised, leading to a larger value of cmc.

As described above, the absolute value of negative ΔG_m increased (namely, the micelle formation became favorable from the viewpoint of thermodynamics) as the temperature was raised, although the value of cmc increased. This was caused by the negative value of ΔH_m and the positive value of ΔS_m.

Conclusion

The value of cmc increased and the values of k and Γ_{∞} decreased as the temperature was raised, while the negative values of ΔG_m and ΔG_{ad} became larger as the temperature was raised. These phenomenon were explained by the negative values of ΔH_m and ΔH_{ad} and the positive values of ΔS_m and ΔS_{ad}.

References

1) M. Nakagaki, "Surface State and Colloid State," Tokyo Kagaku Dohjin, Tokyo, 1979, pp. 102, 103, 282.

2) E. D. Sprague, D. C. Duecker, and C. E. Larrabee, *J. Colloid Interface Sci.*, **92**, 416 (1983).
3) M. J. Schick, *J. Phys. Chem.*, **67**, 1796 (1963).
4) T. O. Oesterling, W. Morozowich, and T. J. Roseman, *J. Pharm. Sci.*, **61**, 1861 (1972).
5) *a*) T. Funada, Y. Tanaka, J. Hirano, E. Mizuguchi, Y. Goto, M. Abe, and K. Ogino, *J. Jpn. Oil Chem. Soc.*, **38**, 432 (1989); *b*) K. Ogino, M. Abe, Y. Goto, M. Goto, Y. Tanaka, T. Funada, and J. Hirano, *Yukagaku,* **39**, 398 (1990).
6) M. Nakagaki and S. Yokoyama, *Bull. Chem. Soc. Jpn.*, **58**, 753 (1985).
7) R. Teraoka, M. Otsuka, and Y. Matsuda, Abstracts of Papers, 111th Annual Meeting of the Pharmaceutical Society of Japan, Tokyo, March 1991, No. 4, p. 118.
8) M. Ueno, Y. Takasawa, H. Miyashige, Y. Tabata, and K. Meguro, *Colloid Polym. Sci.*, **259**, 761 (1981).
9) J. J. Spitzer and L. D. Heerze, *Can. J. Chem.*, **61**, 1067 (1983).
10) M. Nakagaki and M. Yamamoto, *Bull. Chem. Soc. Jpn.*, **50**, 873 (1977).
11) N. D. Weiner and G. Zografi, *J. Pharm. Sci.*, **54**, 436 (1965).
12) M. Nakagaki and T. Handa, *Bull. Chem. Soc. Jpn.*, **48**, 630 (1975).
13) C. Tanford, "The Hydrophobic Effect," 2nd ed., Wiley, New York, 1980, pp. 8, 9.
14) P. Molyneux, C. T. Rhodes and J. Swarbrick, *Trans. Faraday Soc.*, **61**, 1043 (1965).
15) The Chemical Society of Japan (ed.), "Handbook of Chemistry II," Maruzen, Tokyo, 1980, p. 667.
16) L. M. Gan, C. H. Chew, K. W. Yeoh, and L. L. Koh, *J. Colloid Interface Sci.*, **137**, 597 (1990).