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Biophysical Chemistry 117 (2005) 19 – 25

Biophysical Chemistry

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Model analysis of surfactant–polymer interaction as cooperative ligand binding to linear lattice

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Received 16 February 2005; accepted 16 March 2005
Available online 19 May 2005

Abstract

An improved model of the cooperative binding of monomeric ligands to a linear lattice is proposed for the analysis of surfactant association on the polymer. The interaction between bound ligands across an unoccupied site as well as the steric hindrance effect in consecutive bindings is taken into account here. Typical results of the model calculations are represented, and several least squares fittings of the binding isotherms of the ionic surfactant–polyelectrolyte systems are attempted. The characteristic binding behavior in those systems is interpretable by the feasible model of the interactions between surfactant molecules. The advantages and limitations of the analysis using this model also are discussed.
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Keywords: Cooperative binding of ligands; Lattice of linear polymer; Surfactant–polymer association; Interaction between ligands on the polymer; Binding isotherm; Matrix method

1. Introduction

The cooperative binding of small ligands to a linear polymer with a large number of binding sites is one of the interesting subjects to be studied in colloid and polymer science and in biophysics. In particular, the binding of ionic surfactants to the oppositely charged linear polyelectrolyte is the most attractive problem [1].

The binding profile is described by a binding isotherm where the binding fraction is plotted as a function of the equilibrium concentration of free ligands in most cases. In the experimental studies on the surfactant–polyelectrolyte association, various asymmetric and sometimes two-step (biphasic) binding isotherms are observed below a critical micelle concentration of the surfactant molecule [2]. These properties can be interpretable through a full understanding of the elementary binding processes.

Theoretical descriptions of the cooperative ligand binding to a polymeric array of the sites have been provided for the analysis of the binding properties in various systems [3–6]. Recently, several approaches, including computer simulation, were used to reveal the electrostatic effect in the surfactant–polyelectrolyte complex formation [7–9]. In the previous study, we treated the steric hindrance effect in the site-specific ligand binding, where the ligand size is slightly larger than the linear distance between neighboring sites [10].

An improved model for the ligand binding to the polymer lattice is presented here in order to understand the characteristic behavior of the surfactant binding mentioned above. Some typical results of the model calculations using the matrix method will be exhibited after the theoretical explanations. We also attempted to apply the new model to several cases of ionic surfactant binding to the polyelectrolyte with a stiff backbone. The present model has been developed so that sufficient curve fittings to the binding isotherms, which were not attained by the conventional treatment, are achieved.

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doi:10.1016/j.bpc.2005.03.011

In the present model, the following two additional assumptions are introduced to the simple cooperative binding model. One is the cooperative interaction between bound ligands across an unoccupied binding site, named 'skip-binding'. Furthermore, their affinity is assumed to be able to be modified by the existence of bound ligand next to them. The other one is the 'blocking' effect due to the steric hindrance against the ligand binding next to the two consecutively bound ligands. These assumptions are reasonable for the surfactant molecule which has a large ionic head group and a long hydrophobic tail. The curve fittings presented here will show that this model leads to the feasible interpretation of the interactions between the surfactants bound on a relatively stiff polymer chain, i.e., DNA or polysaccharide.

2. Theory

2.1. The binding model

We consider a linear polymer lattice consisting of a large number of equivalent binding sites. A monomeric ligand is assumed to be bound to one of the sites on the polymer. Here, a bound ligand is expected to interact with neighboring ligands. Interactions between them cause the cooperativity of ligand binding by making their binding affinity higher than the single binding.

Moreover, we assume that the interactions work even between the adjacent bound ligands separated by an unoccupied site (the cooperative 'skip-binding'). Their affinity can be modified indirectly by the second neighboring binding ligand at the third neighboring binding site. The incomplete 'blocking' due to steric hindrance effect is also assumed to occur at an unoccupied site neighboring two consecutive binding sites. We should note that this corresponds to the special case of $m=2$ in the previous model [10]. The model considered in this study is illustrated schematically in Fig. 1.

The statistical weight of an occupied state of the binding site is defined relative to the unoccupied state, as follows:

s, relative statistical weight next to one occupied site;
σs, relative statistical weight next to two or more unoccupied sites;
$\beta_{1a}s$, relative statistical weight next to two or more consecutive occupied sites across one unoccupied site;
$\beta_{1b}s$, relative statistical weight next to a non-consecutive occupied site across one unoccupied site;
γs, relative statistical weight next to two or more consecutive occupied sites.

Here, σ is called the cooperative parameter. We assume individual values of β_{1a} and β_{1b} (the 'skip-binding' parameters) are equal to or above the value of σ. Furthermore, these are permitted to be larger than unity to represent the strong interaction across an unoccupied site. It is likely that the steric hindrance effect reduces the nearest-neighboring ligand interactions and makes the skip-binding parameter relatively large, depending on the polymer configuration and surfactant species. In the previous work, we defined β_{1a} only to take into account the steric hindrance [10]. Here, β_{1b} is newly introduced to represent the tendency of the alternate binding due to the sequence of skip-bindings.

We also adopted the 'blocking' parameter, γ (≤ 1), to represent the degree of blocking effect. When $\gamma=0$, there is complete blocking. However, this parameter is set equal to unity, namely non-blocking, in the most curve fittings in the present study. The roles of the parameter will be discussed later.

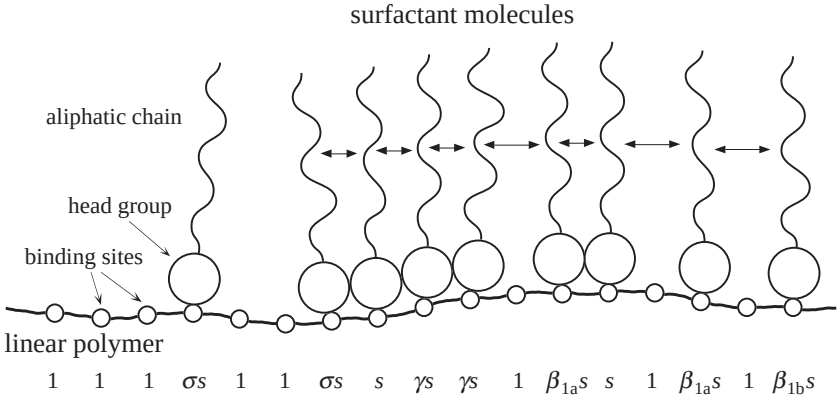

Fig. 1. Schematic representation of the binding of surfactant molecules to the linear polymer lattice. All possible attractive or repulsive interactions in the model are indicated by arrows between aliphatic chains. At the bottom of figure, the statistical weight of binding state in the model is added for each surfactant molecule.

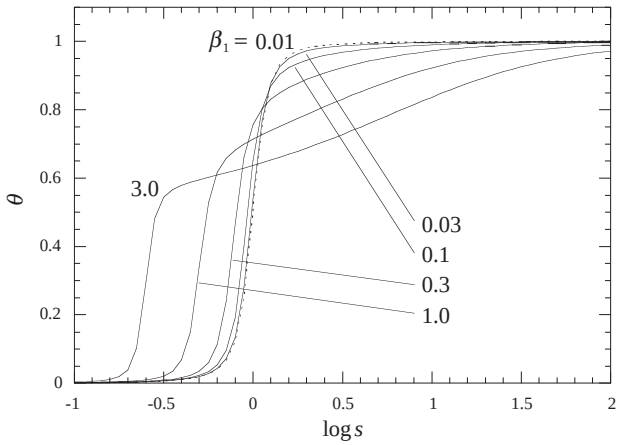

Fig. 2. Calculated binding isotherms (log s vs. θ) at various values of β_1 (given that $\beta_1=\beta_{1b}\equiv\beta_1$) at $\sigma=0.01$ and $\gamma=1$. $\beta_1=3.0$, 1.0, 0.3, 0.1, 0.03, and 0.01 (broken line).

2.2. The partition function

The 8×8 statistical weight matrix, $\boldsymbol{W}$, is written for the present binding model as follows,

$$\boldsymbol{W}=\begin{bmatrix} \gamma s & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & \beta_{1a}s & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & s & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & \sigma s & 1 \\ \gamma s & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & \beta_{1b}s & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & s & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & \sigma s & 1 \end{bmatrix} \tag{1}$$

The partition function must be obtained to calculate the binding fraction of sites, θ. The partition function, Z, is expressed as a trace of a matrix of statistical weights, $\boldsymbol{W}$. For a chain long enough to eliminate the end effect, the partition function is approximately given by the maximum eigenvalue, λ_1. The eigenvalues of the matrix are obtained by solving the following characteristic equation:

$$\lambda^4-(1+\gamma s)\lambda^3+(\gamma-\beta_{1b})s\lambda^2+[\beta_{1b}-\sigma-(\beta_{1a}-\beta_{1b}\gamma)s]\times s\lambda+[\beta_{1a}-\beta_{1b}\gamma-\sigma(1-\gamma)]s^2=0. \tag{2}$$

The fraction of occupied sites is given as,

$$\theta=\frac{s}{\lambda_1}\frac{\partial\lambda_1}{\partial s}=s\frac{\left\{\gamma\lambda_1^3-(\gamma-\beta_{1b})\lambda_1^2-[\beta_{1b}-\sigma-2(\beta_{1a}-\beta_{1b}\gamma)s]\lambda_1-2[\beta_{1a}-\beta_{1b}\gamma-\sigma(1-\gamma)]s\right\}}{\{(1+\gamma s)\lambda_1^3-2(\gamma-\beta_{1b})s\lambda_1^2-3[\beta_{1b}-\sigma-(\beta_{1a}-\beta_{1b}\gamma)s]s\lambda_1-4[\beta_{1a}-\beta_{1b}\gamma-\sigma(1-\gamma)]s^2\}} \tag{3}$$

This is the binding equation which is used to calculate the fraction of occupied sites as a function of free ligand concentration. We note that β_{1a} appears only in the form of $\beta_{1a}-\beta_{1b}\gamma$ in Eqs. (2) and (3). Here, the parameter s is defined as

$$s=\frac{K}{\sigma}C_f, \tag{4}$$

given that K is the intrinsic binding constant and C_f is the free ligand concentration.

In the case of $\beta_{1b}=\sigma$, the characteristic equation, Eq. (2), is rewritten as

$$\lambda^4-(1+\gamma s)\lambda^3+(\gamma-\sigma)s\lambda^2+(\gamma\sigma-\beta_{1a})s^2\lambda+(\beta_{1a}-\sigma)s^2=0. \tag{5}$$

This is identical to the equation in the case of $m=2$ ($\beta_{1a}=\beta_{1b}$), Eq. (18) in our previous work [10].

In order to reduce the number of parameters for the practical curve fittings, additional hypotheses are introduced, such as non-blocking ($\gamma=1$) and/or two equal skip-binding parameters (i.e., $\beta_{1a}=\beta_{1b}\equiv\beta_1$).

In the case that $\beta_{1a}=\beta_{1b}=\beta_1=\sigma$ and $\gamma=1$, which means there is no special effects introduced in this model at all, we get a well-known characteristic equation and the fraction of occupied sites for simple cooperative binding model. These are exactly the same expressions as those for the usual case of cooperative phenomena [3,11].

2.3. On the procedure of the numerical calculation and data fitting

The method of Levenberg–Marquardt–Morrison algorithm was used for nonlinear least squares data fittings [12,13]. The procedure was carried out with iterative random sampling of parameters for the efficient fitting. Higher order polynomials are solved by the three-stage algorithm [14].

In some cases, it was difficult to determine the fitting parameters uniquely, and the uncertainty of parameter

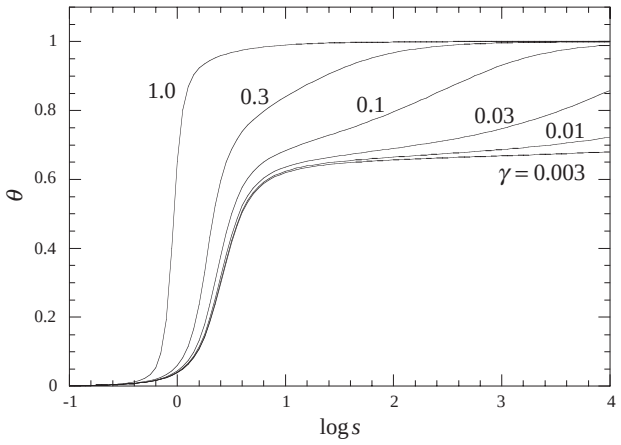

Fig. 3. The dependence of the isotherm on γ value in the case of $\sigma=0.01$ and $\beta_1=\beta_{1b}\equiv\beta_1=0.1$. $\gamma=1.0$, 0.3, 0.1, 0.03, 0.01, and 0.003, from top to bottom.

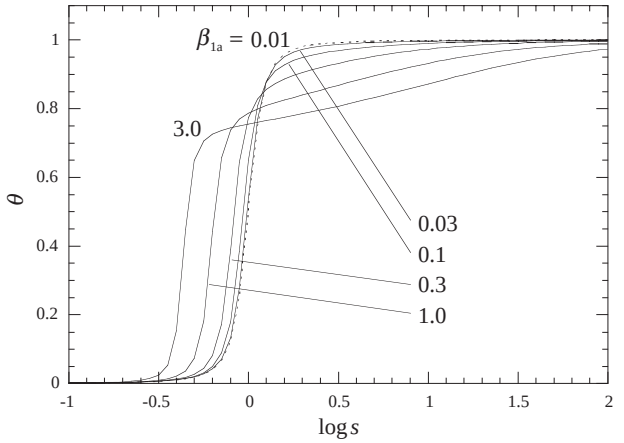

Fig. 4. The binding isotherms of various β_{1a} at $\beta_{1b}=\sigma=0.01$ and $\gamma=1$. $\beta_{1a}=3.0$, 1.0, 0.3, 0.1, 0.03, and 0.01 (broken line).

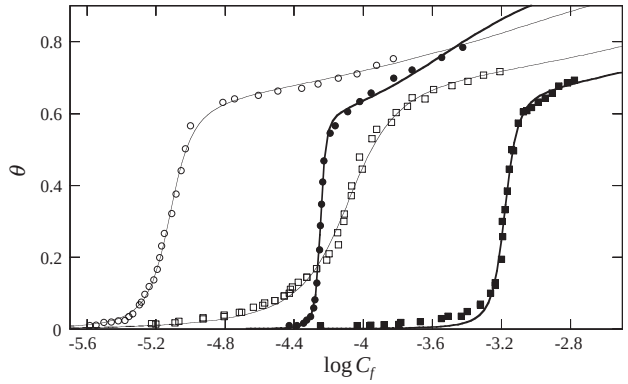

Fig. 6. Binding isotherms (log C_f vs. θ) of the experiments (symbols) of DTA^+ and TTA^+ to salmon sperm DNA at 30 °C [15] are represented with the present fitting results (lines). DTA^+ (squares) and TTA^+ (circles) in bromide salt; no added salt cases (open symbols, thin lines) and added 0.01 M NaCl cases (closed symbols, thick lines).

values depended greatly on the quality of experimental data. We tried to put restrictions appropriately, in those cases, on the ranges of parameter values as well as the number of adjustable parameters for the sufficient curve fitting based on the empirical judgements.

3. Results and discussion

3.1. Dependence of binding isotherm on the parameters

Typical results of the model calculations, i.e., parameter dependences of the binding isotherm (log s vs. θ curves), are given in Figs. 2–5.

Fig. 2 shows the dependence of binding isotherm on the β_1 value (given that $\beta_{1a}=\beta_{1b}=\beta_1$) in the case that $\sigma=0.01$ and $\gamma=1$. We can see clearly that the cooperative skip-binding makes the binding isotherm asymmetric. When the affinity of the skip-binding is high, the isotherm curve is deformed within high θ region and shifted to lower ligand concentration region due to the tendency to the skip-binding, even without the blocking effect.

In Fig. 3, the dependence of binding isotherm on the γ value is shown for $\sigma=0.01$ and $\beta_{1a}=\beta_{1b}=\beta_1=0.1$. The lower value of the blocking parameter makes the binding biphasic and the cooperativity in the first binding step is reduced. We can see that some sets of the parameter values containing the low γ one can give similar isotherm curves to those shown in Fig. 2. This means the two effects, i.e., skip-binding and blocking, are not completely independent. So, in the curve fittings, we should choose a suitable fitting type, which gives more relevant parameters with less uncertainty, even within the present model.

In the following figures, the dependences on β_{1a} and β_{1b} are examined for the case of $\beta_{1a}\neq\beta_{1b}$ at $\gamma=1$. Fig. 4 represents the dependence of binding isotherm on β_{1a} for $\beta_{1b}=\sigma=0.01$ and $\gamma=1$, which is similar to that in Fig. 2. The shoulder of the isotherm curves of high is, however, higher than that in Fig. 2. This is attributable to the high tendency toward the skip-binding following two consecutive bindings in the low β_{1b} case. In Fig. 5, the dependence of binding isotherm on the β_{1b} value is shown in the case when $\sigma=0.01$, $\beta_{1a}=0.1$ and $\gamma=1$. For higher values of $\beta_{1b}(>1)$, the two-step binding behavior appears on the level of $\theta=0.5$ due to the tendency to the alternate binding of the ligands on the polymeric sites. In the range

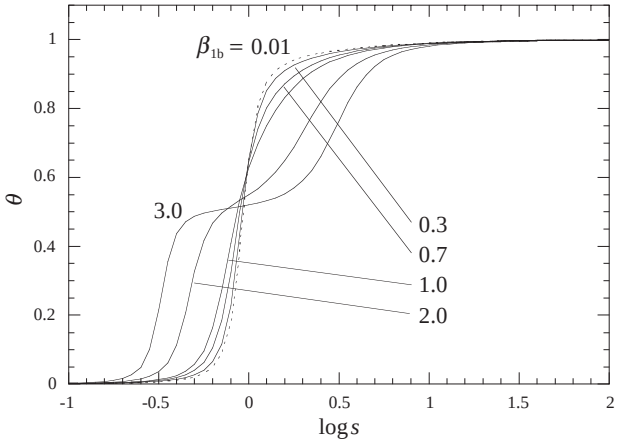

Fig. 5. The dependence of the isotherm β_{1b} on value at $\sigma=0.01$, $\beta_{1a}=0.1$ and $\gamma=1$. $\beta_{1b}=3.0$, 2.0, 1.0, 0.7, 0.3, and 0.01 (broken line).

Table 1
Binding parameters of DTA^+ and TTA^+ with DNA

Parameter	Salt-free		0.01 M NaCl	
	DTA^+	TTA^+	DTA^+	TTA^+
K [mol^{-1} kg]	1.70×10^3	2.77×10^3	10.4	11.0
σ	0.562	0.100	23.0×10^{-3}	1.45×10^{-3}
K/σ [mol^{-1} kg]	3.02×10^3	27.7×10^3	0.452×10^3	7.60×10^3
β_{1a}	12.6	9.83	4.84	0.793
β_{1b}	$=\sigma$	2.24	1.31	1.72

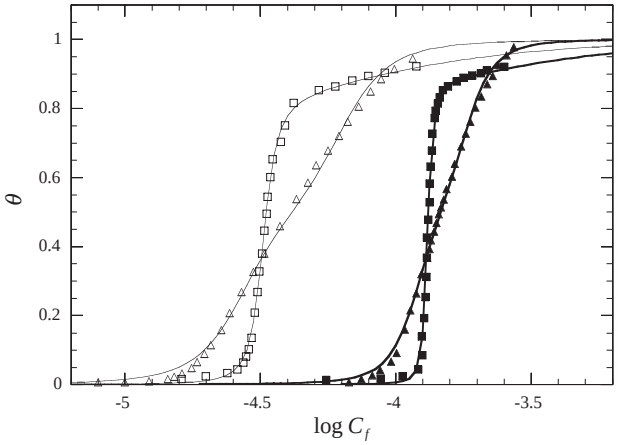

Fig. 7. Binding isotherms of the experiments (symbols) of TTA^+ to alginate (squares) and CMC (triangles) at 30 °C [18] are shown with the present fitting results (lines). No added salt cases (open symbols, thin lines) and added 0.01 mol kg^{-1} NaCl cases (closed symbols, thick lines).

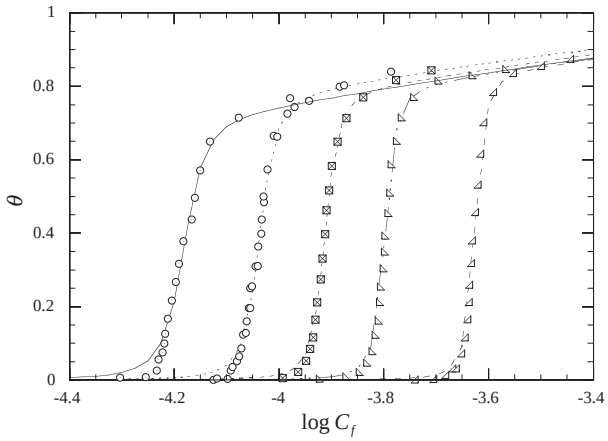

Fig. 8. Binding isotherms of the experiments (symbols) of DTA^+ to DxS at various concentrations of $MgCl_2$ at 30 °C [20] are represented with the fitting results (lines). $MgCl_2$ concentration (10^{-3} mol kg^{-1}); 1.00, 2.00, 4.00, 8.06, and 20.0, from left to right.

of lower β_{1b} value (≤ 0.3), the curve stays almost unchanged.

3.2. The case of cationic surfactant binding to DNA

Typical two-step ligand bindings have been observed for the cationic surfactant–DNA systems with a potentiometric method [15,16]. In particular, the binding isotherms level off at a binding fraction of about 0.6 in the cases of dodecyltrimethylammonium ion (DTA^+) and tetradecyltrimethylammonium ion (TTA^+) in bromide salt as ligands. This is considered to be due to the steric hindrance effect of large head group of surfactant molecule [15].

For these binding isotherms, we obtained satisfying curve fittings on the supposition of non-blocking with $\gamma = 1$, as shown in Fig. 6. Determined values of fitting parameters are listed in Table 1. Addition of the salt decreases the values of K, σ, and β_{1a}. The salt dependence of β_{1b} is not so significant. The decreases of σ, in particular, are larger than those of simple cooperative binding model [15]. The decrease of the value K/σ moves the isotherm toward the low C_f (right) with the increase of the salt. It seems that the weak dependence of the hydrophobic interaction between the surfactant molecules on salt concentration makes the binding cooperativity high in the added salt systems.

These results indicate that the characteristic binding curves can be interpreted by attractive interactions between surfactant molecules across an unoccupied site with the relatively high affinities, even without apparent steric hindrance effect. Large values of skip-binding parameter above unity may suggest some interferences in the ligand binding next to an occupied site.

Equivalent curve fittings can also be achieved adopting variable γ on the assumption that $\beta_{1a} = \beta_{1b} \equiv \beta_1$, where added salt increases the values of both β_1 and γ (data not shown). The blocking with low γ value makes the curves biphasic in the fittings. The results differ for each type in this model system, as mentioned above.

DNA is one of the typical linear polyelectrolytes with high charge density (the charge density parameter, $\xi = 4.2$) which have been studied on simple ion binding [17]. The study of two kinds of surfactant binding to DNA indicates that the short-range interactions between the surfactant molecules are of great importance for the binding process.

3.3. Cases of binding to polysaccharides

The bindings of DTA^+ and TTA^+ to some carboxylic polysaccharides were also measured by Hayakawa et al [18]. The data fittings for alginate, pectate, and (carboxymethyl)cellulose (CMC) in sodium form are attempted by

Table 2
Binding parameters of TTA^+ with alginate, pectate, and CMC

Parameter	Alginate ($\xi = 1.43$)		Pectate ($\xi = 1.61$)		CMC ($\xi = 1.32$)	
	Salt-free	0.01 M NaCl	Salt-free	0.01 M NaCl	Salt-free	0.01 M NaCl
K [mol^{-1} kg]	90.4	3.43	378	2.67	475	25.5
σ	3.79×10^{-3}	0.534×10^{-3}	14.5×10^{-3}	0.445×10^{-3}	19.6×10^{-3}	3.75×10^{-3}
K/σ [mol^{-1} kg]	23.9×10^3	6.43×10^3	26.1×10^3	5.99×10^3	24.2×10^3	6.80×10^3
β_{1a}	0.298	0.185	0.164	0.211	$=\sigma$	$=\sigma$
β_{1b}	$=\beta_{1a}$	$=\beta_{1a}$	$=\beta_{1a}$	$=\beta_{1a}$	1.31	1.10

Values of linear charge density parameter are from Ref. [18].

Table 3
Binding parameters of DTA^+ with DxS at various concentration of $MgCl_2$

$MgCl_2$ [10^{-3} mol kg^{-1}]	Parameter			
	K [mol kg^{-1}]	σ	K/σ [mol kg^{-1}]	β_1
1.00	25.0	2.76×10^{-3}	9.05×10^{3}	0.680
2.00	10.7	1.40×10^{-3}	7.63×10^{3}	0.428
4.00	5.23	0.893×10^{-3}	5.86×10^{3}	0.396
8.06	3.22	0.694×10^{-3}	4.64×10^{3}	0.354
20.0	1.78	0.531×10^{-3}	3.35×10^{3}	0.260

applying the present model. In these cases, it is also practicable to fix the γ value to unity in the fittings. Fig. 7 shows the fitting results to the binding isotherms of TTA^+ to alginate and CMC without and with added salt. Their values of fitting parameter with those for pectate are evaluated as shown in Table 2.

In the cases of alginate and pectate, an additional assumption of $\beta_{1a}=\beta_{1b}\equiv\beta$ is adopted. The effects of salt on K, σ, and K/σ are also similar to those of DNA system, while the dependence of β_1 on added salt is not significant. Although their linear charge densities as well as fitting parameters are different, the binding curves of pectate system look similar to those of alginate (data not shown). This suggests the high sensitivity of the fitting parameters to the quality of the experimental data.

In the case of CMC, whose degree of substitution (DS) is 0.96, another assumption, $\beta_{1a}=\sigma$, is appropriate for the sufficient fittings. The biphasic curve with an obscure boundary between two binding steps makes the gentle slope of the binding isotherms in this system. The dependence of β_{1b} on added salt is not also significant in this case.

The difference in binding isotherm among these stiff polymers seems to be due to not only the difference of linear charge density but also that of polymer configuration. The choice of the appropriate fitting type is required for the reasonable interpretation.

The salt dependence of the bindings of DTA^+ to dextran sulfate (DxS, $\xi=2.8$) was investigated in detail by Hayakawa et al. [19,20]. For this polysaccharide derivative, the assumptions of $\gamma=1$ and $\beta_{1a}=\beta_{1b}\equiv\beta$ are practicable for the respectable fittings. In Fig. 8 and Table 3, the results are presented for the experiments in $MgCl_2$. Fittings are sufficient, and the salt concentration dependences of the parameter are well exhibited, including that of β_1.

4. Conclusions

The problem of the cooperative binding of surfactant molecules to a linear polymer is solved in novel model. It is shown that the one-dimensional lattice model and its formulation are applicable to the analysis of several measurements of the binding isotherms of the surfactant–polymer systems. Unsymmetrical binding isotherms and two-step binding, which are sometimes observed in this kind of systems, can be interpreted semi-quantitatively by the model.

Most isotherm curves can be fitted only with the assumption of the cooperative skip-binding effect. Although this fact does not rule out the blocking effect, the skip-binding effect seems to be necessary for the explanation of extremely high cooperativity in the unsymmetrical binding isotherm. In some cases, much better fittings can be achieved by introducing two kinds of the skip-binding parameter. This indicates the modification of the affinity by a bound ligand next to the skip-binding ligands.

Our results are important to understand the molecular process of the surfactant association depending on the polymer configuration. Fitting of the experimental binding isotherms based on this site-specific model must be practicable for the characterization of the binding properties.

Electrostatic interactions between the charged groups must play an important role in the system. Those effects can be considered in the dependence of the parameters on the binding fraction. Our treatment should be extended in this way for more precise understanding of the electrostatic interactions.

Acknowledgments

The authors are most grateful to Ms. E. Kamofuji for her skillful technical support. The calculations in this study were carried out using the workstations in the information processing center of the Hamamatsu University School of Medicine.

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