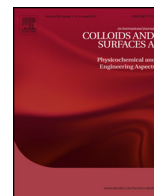




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Effect of anionic surfactants on the stability ratio and electrophoretic mobility of colloidal hematite particles

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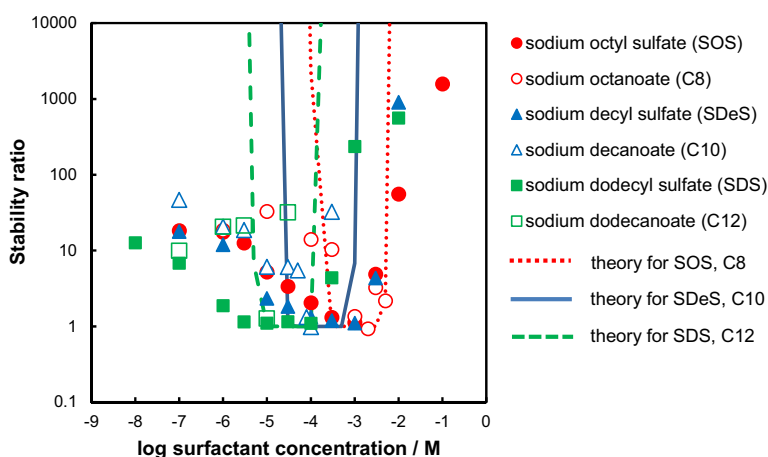
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HIGHLIGHTS

- The concentration at the zero mobility decreases with increasing the length of hydrophobic chain.
- The value of the intrinsic adsorption energy is around 0.8 k_BT per carbon.
- The value of the minimum stability ratio does not depend on the length of hydrophobic chain.
- The range of the minimum stability ratio was narrower for the suspension with fatty acids.

GRAPHICAL ABSTRACT



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ABSTRACT

The effect of anionic surfactants on the charging and aggregation of hematite particles was investigated. Charging behavior was studied by the measurement and analysis of the electrophoretic mobility. The stability ratio of hematite suspension was obtained from the temporal change of turbidity, and the stability ratio was analyzed by the DLVO theory. These experiments were performed as functions of types of surfactant, surfactant dosage, and surfactant chain length. Electrophoretic mobility of bare hematite was positive at pH below 7. The mobility went through zero and became negative with increasing surfactant dosage. Required surfactant concentrations reaching the isoelectric point decreased with increasing the chain length of surfactant. These results are reasonably described by a simple theoretical model. The stability ratio showed the minimum around the isoelectric point. That is, the suspension is destabilized by charge neutralization due to the adsorption of surfactants onto the hematite. Values of the minimum stability ratio in this work were almost unity irrespective of the chain length; the fastest aggregation rate with surfactants equaled the salt-induced rate. Therefore, in the case of ionic-surfactant, charge-patch attraction and steric hindrance do not significantly work around zero mobility concentration. The DLVO theory qualitatively describes the aggregation behavior. The range of the minimum stability ratio was narrower for the suspension with fatty acids. This result indicates that the suspension with fatty acids is more stable.

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1. Introduction

Surfactants are amphiphilic molecules having a hydrophobic tail and a hydrophilic head [1,2]. They are widely used in many industrial processes, because their adsorption on the solid-liquid interface of colloidal particles modifies the interfacial properties such as surface charging and wetting phenomena [1–7]. Such modifications affect surface interactions, and thus colloid stability and rheological behaviors of colloidal dispersions are controlled by the addition of surfactants. Therefore, detailed studies on surfactant adsorption, resulting change in surface charge, and colloid stability were carried out [6–14].

The addition of ionic-surfactants into a suspension containing oppositely charged particles causes changes in zeta potential and aggregation–dispersion of the particles [6–11]. In general, as the adsorbed amount of ionic surfactants to oppositely charged particles increases, the magnitude of zeta potential of the particle decreases and reaches zero at the isoelectric point, and the sign of zeta potential reverses [6–11]. The length of hydrophobic chain of surfactants is one of the key factors for determining the isoelectric point. The required surfactant concentration leading to the isoelectric point decreases with increasing the chain length [5–9]. Simple theoretical models were proposed to describe the dependence of isoelectric points on the chain length of surfactants [5,7]. From the analysis, it is suggested that the chemical/hydrophobic energy of adsorption per a carbon atom is close to the hydrophobic energy obtained from the analysis of critical micelle concentration [1,5,6]. However, theoretical models have not yet been sufficiently examined for the description of the zeta potential and stability ratio as a function of the concentration of surfactant.

Colloidal particles are destabilized by the addition of optimum dosage of oppositely charged surfactants [4,6–11]. The range of the optimum aggregation concentration is around the isoelectric point, meaning that the colloid is destabilized by charge neutralization. Most of the studies on surfactant-induced aggregation were carried out by simple aggregation–sedimentation experiments. On the one hand, measurements of stability ratio W , which is the relative rate of aggregation, of well-characterized particles with ionic-surfactants are scarce [6–8,10]. It is difficult from the aggregation–sedimentation experiment to unveil the existence of additional inter-particle interaction such as steric repulsion and charge-patch attraction, which are found from the stability ratio in the presence of polymers [8,15–18]. As for the surfactant-induced aggregation, Liang and Morgan [6] showed the stability ratio of positively charged hematite as a function of the concentration of fatty acids. Their data demonstrated that the minimum value of stability ratio increases with increasing the chain length of fatty acids, implying the development of steric repulsion by the surfactants with longer hydrophobic chain [8]. On the one hand, the values of the minimum stability ratio of positively charged AgI particles with sodium alkyl sulfates were almost unity irrespective of the chain length [7], indicating that there is neither additional attraction nor repulsion at the optimum aggregation concentration. Leong et al. [12] claimed the appearance of additional hydrophobic attraction from the measurement of shear yield stress of concentrated ZrO_2 suspension with surfactants. Sakamoto et al. [14] suggested that the origin of hydrophobic attraction is due to the bridge formed by microscopic bubbles. These previous studies reported different effects of ionic-surfactants on the surface interactions and the behavior of colloidal suspensions.

While the minimum stability ratio W of hematite around isoelectric point increased with increasing the chain length of fatty acids and became larger than unity ($W > 1$) [6], the minimum stability ratio of positively charged AgI particles with sodium alkyl sulfates were almost unity ($W = 1$) irrespective of the chain length [7]. These two results are different in terms of the dependence

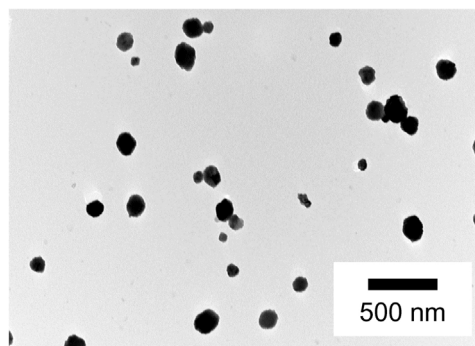


Fig. 1. Transmission electron microscope (TEM) picture of used hematite particles.

of the minimum stability ratio reflecting the existence of steric repulsion on the chain length, and one may explain that this inconsistency is because they used different particles and surfactants. In addition, the detailed comparison of theoretical model with experimental stability ratio has not yet fully been carried out for the ionic-surfactant-induced aggregation. To better understand the surfactant-induced aggregation, we need systematic measurements and analysis of stability ratios that examine the effect of type of oppositely charged surfactants on the stability ratio by using an identical colloidal suspension. By such systematic experiments with theoretical analysis, we can unveil the role of ionic surfactant on the aggregation–dispersion of oppositely charged particles in detail.

In this context, our goal was set to clarify the effect of both fatty acids and sodium alkyl sulfates on the stability ratio and electrophoresis of hematite particles, which were chosen since the minimum stability ratio larger than unity was reported for hematite with fatty acids [6]. Measured data were analyzed using theoretical models. From the comparison of the experiment and theory for the stability ratio and electrophoretic mobility, possible mechanisms are delineated.

2. Experiments

2.1. Materials

We used hematite particles, which were obtained from Chemirite Kogyo Industry, Japan. The hematite particles have 141 ± 35 nm in a number averaged diameter $2a$ determined from transmission electron microscopy (Fig. 1) and the specific surface area of 26.3 m²/g from N₂-BET measurement. The density of hematite particles is 5.24 g/cm³ [19]. The hematite particles were purified as follows: the particles were dispersed in 1 M NaCl with pH 4, then after sedimentation and decantation, the solution condition of hematite suspension was adjusted to 1 M NaCl with pH 10. These procedures were performed for the saturation of surface with Na⁺ and Cl[−] ions and for washing by acid and base. Finally, the suspension was extensively dialyzed in a Visking tube against pure water to remove excess ions. The purified hematite particles were freeze-dried and stored in a polystyrene vial. In the following experiment, hematite suspension was prepared by dispersing the dried powder in pure water. The prepared suspensions were used within two weeks after preparation to avoid the change of surface properties after the contact with water. Intensity-weighted average hydrodynamic diameters d_h of hematite are around 170 nm. The size is close to the corresponding size from TEM, $d_{h,TEM} = \langle d^6 \rangle / \langle d^5 \rangle = 174$ nm, where d is the diameter from TEM and $\langle \dots \rangle$ stands for averaging.

Six anionic surfactants with different lengths of hydrophobic chain and different charged groups were used as received. Three of

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