

Electrophoretic mobility of carboxyl latex particles in the mixed solution of 1:1 and 2:1 electrolytes or 1:1 and 3:1 electrolytes: Experiments and modeling



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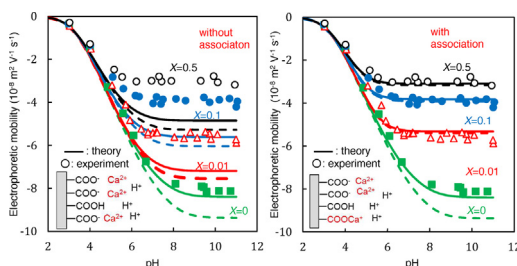
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HIGHLIGHTS

- We measured and calculated electrophoretic mobility of carboxyl latex particles in mixed solution of KCl and CaCl₂ or KCl and LaCl₃.
- Used theoretical models consider the deprotonation, the binding of Ca²⁺/La³⁺, diffuse double layer model, and the double layer relaxation. Experimental data are reasonably described by the theoretical models except the case with La³⁺ at high pH.
- The surface charge reduces due to the binding of Ca²⁺/La³⁺ to deprotonated carboxyl groups.
- The model with the binding of La³⁺ is unable to describe charge reversals in EPM at high pH.

GRAPHICAL ABSTRACT



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1. Introduction

Colloidal particles aggregate when the solution condition is close to the isoelectric point of the particles and/or the ionic strength is high. Also, the critical coagulation concentration, which is the minimum electrolyte concentration inducing fast coagula-

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tion, decreases with increasing the valence of counter-ion. These trends found experimentally are theoretically explained by the Derjaguin, Landau, Verwey, and Overbeek (DLVO) theory [1,2]. The DLVO theory assumes that the interparticle interaction is given by the sum of electrostatic double layer force and the van der Waals attractive force. The former force is a function of surface electric potential, electrolyte concentration, and valence of ions. Therefore, to discuss the aggregation-dispersion behavior, we need to quantitatively evaluate charging behaviors of the particle in solutions having different ionic composition.

Measurement of electrophoretic mobility (EPM) is widely used to characterize the charging behavior of colloidal particles. While the EPM reflects the sign and magnitude of surface potential, proper theories must be used to evaluate EPM from surface charge of the particle. EPM of monodisperse latex spheres as model colloidal particles has been extensively researched over the years [3–6]. For the latex particles bearing a constant surface charge density in 1:1 electrolyte solution, the dependence of EPM on salt concentration can be described by the O'Brien-White (OW) theory or the Ohshima equation that calculates EPM from the surface or electrokinetic potential, which is obtained from the charge density with the Gouy-Chapman (GC) theory derived from the Poisson-Boltzmann equation [4]. Such methodology can be successfully applied to EPM in a solution containing divalent ions [7,8]. For carboxyl latex particles in KCl solution, measured values of pH-dependent EPM were compared with theoretical ones calculated using 1- pK_H GC model and OW theory [9]. The 1- pK_H GC model is applied for the evaluation of surface charge and potential as a function of pH. Their result demonstrated that theoretical EPM reasonably agrees with experimental data. A similar result was obtained by Sugimoto *et al.* [10]. Ottewill and Shaw measured the EPM of carboxyl latex particles with various diameters and surface charge densities as a function of $\text{La}(\text{NO}_3)_3$ and $\text{Nd}(\text{NO}_3)_3$ concentration [11], and they showed that the charge reversal occurred in a solution containing trivalent ions. James and Healy also observed the charge reversal for SiO_2 in $\text{La}(\text{NO}_3)_3$ at high pH [12]. However, these researchers did not sufficiently focus on the effect of surface charge density and did not report on a comparison with the theoretical calculations assuming the presence of multivalent ions.

In soil and aquatic environments, colloidal dispersions contain multiple ionic species with different valences. The ionic composition significantly affects the charging and aggregation of colloidal particles [13,14]. Therefore, it is important to understand the charging properties of colloidal particles in a mixed solution of multiple ionic species. Chow and Takamura measured EPMs of bitumen-in-water emulsion particles in mixed solutions of NaCl and CaCl_2 or MgCl_2 [15]. Their experimental values were compared with theoretical ones calculated using the OW theory and a charging model taking account of the counter-ion binding to the surface charge group. However, they did not obtain quantitative agreement between the experimental and theoretical values. Kobayashi and Sasaki showed the results of measurements and analysis of the EPM of sulfate latex in mixed solutions of KCl and MgCl_2 [16]. The experiments were carried out as functions of the concentration ratio of Mg^{2+} to K^+ and of the total concentration of electrolytes. From the comparison of theoretical and experimental data, they suggested that the position of the slipping plane, where the zeta or electrokinetic potentials are defined, was needed to shift from the particle surface in the presence of MgCl_2 , indicating that the presence of multivalent ions significantly affect EPM compared to monovalent ions. Several studies pointed out the occurrence of counter-ion adsorption to the surface [5,6], counter-ion association on the surface groups [17], reduction of the surface charge [7], and ion-ion correlation [18,19]. However, systematic measurements and modeling of EPMs of colloidal particles in mixed electrolyte solutions containing divalent and trivalent counter-ions are still lacking.

In this study, the EPM of carboxyl latex particles was measured as functions of pH, the ionic strength, and the mixing molar ratio of Ca^{2+} to K^+ ($X = [\text{Ca}^{2+}]/[\text{K}^+]$) or La^{3+} to K^+ ($X = [\text{La}^{3+}]/[\text{K}^+]$). The obtained experimental data were compared with theoretical ones calculated by using 1- pK_H model with or without Ca^{2+} or La^{3+} association, the diffuse double layer model, and the Ohshima equation and the CellMobility program including the relaxation effect or the Smoluchowski equation neglecting the relaxation effect. From comparisons of experiments and the theory, we demonstrate the significance and limitation of the theoretical model assuming the binding of multivalent counter-ion to the particle surface.

2. Material and methods

2.1. Materials

Carboxylated polystyrene latex particles (Interfacial Dynamics Corporation) were used as colloidal particles. Carboxyl latex particles bear pH-dependent negative charge due to the deprotonation of the carboxyl groups on the particle surface. The manufacturer reports that the maximum surface charge density is -182 mC/m^2 , the diameter is $2a = 1.5 \pm 0.03 \mu\text{m}$, and the density is 1.055 g/cm^3 , respectively. KCl, $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ and $\text{LaCl}_3 \cdot 7\text{H}_2\text{O}$ (JIS special grade, Wako Pure Chemical Industries) were used to prepare the electrolyte solutions. The pH was adjusted by the addition of HCl (JIS special grade, Wako Pure Chemical Industries) and KOH solutions. Carbonate free KOH solution was prepared by following the method described in the literature [20]. Before sample preparation, all solutions were filtered with a $0.20 \mu\text{m}$ pore filter (Millex-LG, MILLIPORE). All solutions and suspensions were prepared from pure water (Elix, MILLIPORE) and degassed before use.

2.2. Experimental methods

EPM was measured by electrophoretic light scattering using Zetasizer NANO-ZS (Malvern). Measurements were carried out as a function of solution pH at different mixed molar ratio $X = [\text{Ca}^{2+}]/[\text{K}^+]$ or $X = [\text{La}^{3+}]/[\text{K}^+]$ with fixed ionic strength. The pH was adjusted in the range from 3 to 11 with HCl and KOH solutions. In mixed electrolyte solutions containing Ca^{2+} , ionic strengths were 1 mM, 10 mM, and 50 mM, and mixed molar ratios X were 0, 0.01, 0.1, and 0.5. In mixed electrolyte solutions containing La^{3+} , ionic strengths were 1 mM and 10 mM, and mixed molar ratios X were 0, 0.001, 0.01, 0.1, and 0.5. The particle concentration was 50 mg/L. The samples were prepared by mixing the suspension of latex particles, KCl solution, CaCl_2 solution or LaCl_3 solution, pH adjuster, and degassed water. The pH was measured with a combination electrode (ELP-035, TOA-DKK). All experiments were carried out at 20°C .

3. Theoretical analysis

1- pK_H model with or without $\text{Ca}^{2+}/\text{La}^{3+}$ association and the Ohshima equation were used to calculate theoretical EPM.

3.1. 1- pK model

In the 1- pK_H model without $\text{Ca}^{2+}/\text{La}^{3+}$ association, the surface charge arises from the deprotonation of carboxyl groups. On the one hand, in the 1- pK_H model with $\text{Ca}^{2+}/\text{La}^{3+}$ association, the surface charge is developed by the deprotonation of carboxyl groups and the binding of $\text{Ca}^{2+}/\text{La}^{3+}$ to the deprotonated carboxyl groups. That is,



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