

Brownian flocculation of negatively charged latex particles with low charge density polycation at various ionic strengths



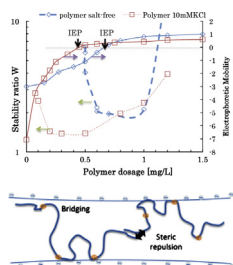
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HIGHLIGHTS

- Bridging flocculation without mixing flow enabled colloid stability analysis.
- The maximum rate of flocculation i.e., the optimum dosage, was confirmed at IEP.
- Steric effect is competing with bridging even at the optimum dosage.
- The IEP shifts to lower polymer dosage with increasing ionic strength.

GRAPHICAL ABSTRACT



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ABSTRACT

Rate of Brownian flocculation of polystyrene latex particles with low charge density polyelectrolyte ($\sigma = 4\%$, $M_w = 5.2 \times 10^6$ g/mol) was measured by Coulter Counter as a function of the ionic strength. As reported in the case of high charge density polyelectrolyte [L. Feng, Y. Adachi, A. Kobayashi, *Colloid Surf. A. 440* (2014) 155–160], the rate of flocculation goes through a maximum against polyelectrolyte dosage, and a maximum rate of flocculation was observed near the isoelectric point. However, several important trends are different from the results obtained with high charge density polyelectrolyte. (1) The rate of flocculation is always slower than that of salt-induced rapid coagulation, even at the isoelectric point. (2) The optimum polyelectrolyte dosage corresponding to the isoelectric point appears several times higher. (3) The shift of isoelectric point was also observed with an increase of ionic strength but toward a lower polyelectrolyte dosage. (4) The thickness of adsorbed polyelectrolyte layer was found to be detectable (about 15 nm) at the optimum dosage of flocculation where the fastest rate was observed. These results suggest a picture of bridging flocculation that competes with steric effects of adsorbed polyelectrolytes.

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1. Introduction

Water-soluble polyelectrolytes with an ability of adsorption onto the surface of colloidal particles have been widely used as flocculants as well as stabilizer in a wide range of industries, such as paper making, minerals processing and water/wastewater treatment. As for the flocculant, the charge neutralization of

colloidal particles by the opposite charge of adsorbed chains and the formation of bridges [1–6] between colloidal particles by simultaneous adsorption of polyelectrolyte chains onto more than two particles are recognized as the main flocculation mechanism. The mechanism of charge neutralization is enforced by the concept of “patch” [7–13] which suggests an additional electric attraction. The mechanism of stabilization induced by adsorbed polyelectrolytes is sometimes called electro-steric effect which implies a coupled interaction of the electrostatic effect interacting between charge reversed surfaces adsorbing excess oppositely charged chains and the steric effect interacting between adsorbed polyelectrolytes of

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approaching surfaces. The former can be expected to be long-distance interaction under low ionic strength, and the latter can be expected to be operative when the electrostatic interaction is weak. However, clear distinction of the dominance of these effects; electrostatic interaction, bridging or steric effects, has not been conducted so far.

In our previous study [14], we have studied the Brownian flocculation of polystyrene particles with high charge density polyelectrolyte and confirmed the predominant electrostatic interaction for salt free and KCl 1 mM. In this case, polyelectrolytes are flatly adsorbed on the surface, and the electrostatic force dominates the interaction between colloidal particles. The electrostatic stabilization is induced between charge reversed particles adsorbing excess polyelectrolytes. And the electrostatic interaction was replaced by the steric effect of adsorbed polyelectrolytes under sufficient high ionic strength. The formation of bridging by polyelectrolyte chains, even though it is predominant in the mixing flow condition [15], was not definitely detected in the Brownian flocculation with polyelectrolyte of high charge density.

However, the importance of the formation of bridging and steric effects can be expected by the application of low charge density polyelectrolytes. In the series of flocculation experiment using jar tester, Audebert [16] found that polyelectrolytes with charge density less than 5% induce the flocculation of silica suspension through an inter-particulate bridging mechanism and the steric stabilization of suspension upon addition of excess polyelectrolytes. Polymer induced bridge and steric stabilization were confirmed between silica surfaces adsorbing with low charge density (15%) polyelectrolytes by Abraham et al. [17]. However, in the experiment of jar tester, flocculation is induced by shear flow where two surfaces are forced to approach. As far as we know, these effects have not been confirmed in the Brownian motion and no quantitative data is available for the stability ratio.

The present study focuses on the Brownian flocculation of polystyrene particles with low charge density polyelectrolyte ($\sigma = 4\%$, $M_w = 5.2 \times 10^6$ g/mol), and aims to clarify the predominant interaction of flocculation or stabilization of colloidal suspension at various ionic strengths. Absolute aggregation rate was measured by Coulter Counter as a function of the polyelectrolyte dosage. In order to obtain information on the effect of the state of adsorbed polyelectrolytes on the stability of suspension, the electrophoretic mobility was analyzed as well as the thickness of adsorbed polyelectrolyte layer on the basis of diffusion constant of Brownian motion.

2. Materials and methods

2.1. Materials

The monodispersed negatively charged polystyrene latex (PSL) was prepared by the standard emulsion polymerization of styrene in the absence of surfactant [18]. The diameter of particle was determined by electron microscopy. The surface charge density was calculated from the measurement of electrophoretic mobility as a function of the ionic strength on the basis of Ohshima's theory [19]. All information was summarized in Table 1. The PSL suspension was sonicated for 20 min to eliminate the aggregates before each experiment. The linear cationic polyelectrolyte, acrylamide-2-(dimethylamino)ethyl methacrylates methyl chloride, was employed as the flocculant (kindly supplied by Kaya Flocc Co. Ltd.). The charge density, i.e. the ratio of the number of charged monomers to the total number of monomers, is 4%. The molecular weight is 5.2×10^6 g/mol. Structural formula of polyelectrolyte is shown in Fig. 1. The ionic strength was adjusted by the potassium chloride solution and pure water ($0.7 \mu\text{s/cm}$). Electrolyte solution was filtered through $0.2 \mu\text{m}$ millipore filters prior to use.

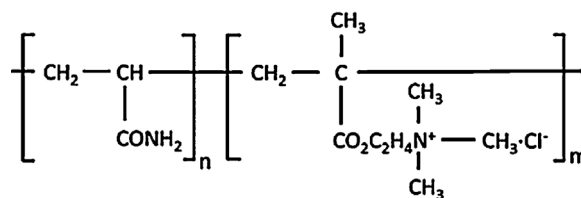


Fig. 1. Structural formula of polyelectrolyte used.

2.2. Electrophoretic mobility

Charging state of the particle was investigated by electrophoretic mobility. The experiments were carried out with a laser velocimetry setup Zetasizer Nano-ZS (Malvern Instruments, UK). The sample was prepared as follow. As depicted in Fig. 2, initially, one side of the forked flask was filled with 5 mL colloidal dispersion and the other one with an equal volume polyelectrolyte solution. Mixing starts by pouring the colloidal dispersion into the electrolyte or polymer solution. The mixed dispersion was then poured back into the empty side. The mixing procedure was repeated periodically at a fixed frequency of 1 Hz for 5 min. More detailed information on the rotation apparatus can be found in Ref. [1]. The measurements were carried out in plastic capillary cells rinsed with pure water. Each sample was measured 3 times with 12 sub-runs for each measurement.

2.3. Particle aggregation by Coulter Counter

For a monodisperse suspension which undergoes the rapid Brownian aggregation, the aggregation rate obeys the following equation [20],

$$\frac{dN(t)}{dt} = -\alpha \frac{4kT}{3\mu} N(t)^2 \quad (1)$$

where μ , k , T and t denote viscosity, Boltzmann constant, temperature and elapsed time, respectively. α represents the correction factor of capture efficiency of aggregation. It should be noted that the factor is less than unity. The value of 0.3–0.7 was reported for the salt-induced rapid coagulation of polystyrene particles starting from a monodisperse suspension [21].

The total number concentration of clusters, $N(t)$, is expressed as

$$\frac{1}{N(t)} - \frac{1}{N(0)} = kt, \quad k = \alpha \frac{4kT}{3\mu} \quad (2)$$

The aggregation rate, k , was directly obtained from the slope of the plot of $1/N(t)$ vs. t . In our study, the home-made shaker was also employed for mixing. 5 mL colloidal dispersion was mixed with equal volume KCl or polyelectrolyte solution end-over-end at the frequency of 1 Hz for 5 s, and then the mixture was left to stand for Brownian coagulation or flocculation. The aggregation process was monitored every 10 min by Coulter Counter (Multisizer 3). The

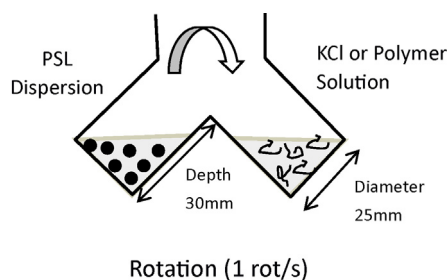


Fig. 2. Schematic drawing of the end-over-end rotation shaker.

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