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# Deposition and subsequent release of Na-kaolinite particles by adjusting pH in the column packed with Toyoura sand

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#### Abstract

Deposition of kaolinite onto Toyoura sand packed in the column and subsequent release by switching influent to higher pH solution were studied to reveal the relation between the state of deposition and release behavior of colloidal particle. Deposition experiments were performed as a function of solution pH based on the difference in colloidal stability of kaolinite. Deposition was continued until the sum of deposited particles reaches predetermined value. In the case of low pH condition, ripening behavior of deposition was observed. The rate of deposition decreases with an increase of pH. At higher pH condition, where kaolinite particles do not coagulate, however, deposition takes place at constant rate. By microscope observation, flocs deposited on sand surfaces can be easily found for lower pH condition. On the other hand, few particles deposited on sand surfaces were detected at high pH condition implying the dominancy of straining. Release experiment was started by stepwise increase of solution pH by means of switching the influent to the aqueous solution of  $1.0 \times 10^{-4}$  M NaOH. In the column with the kaolinite deposited at lower pH, numerous kaolinite particles were flowed out immediately corresponding to the increase of solution pH. While, in the column with the kaolinite deposited at higher pH, few particles were detected in the effluent. The difference of release behavior means that the release of colloidal particles strongly depends on the dynamics of deposition via state of deposited particle. Our results imply the significance of colloid straining in the migration of colloidal particle through porous media.

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Keywords: Colloid-facilitated transport; Colloid release; Colloid deposition; Kaolinite; Sand column; Straining

## 1. Introduction

Evaluation of colloid release from porous matrix is essential for analysis of colloid-facilitated transport of contaminants in soil and groundwater. Colloid release is related to colloid generation and colloid transport, which are most important stages of colloid-facilitated transport. Release of colloidal particles in soil has been studied by various column experiments consisting of model collector grains and colloidal particles [1–17]. Many of them use sand and natural colloidal particles in soil or aquifer [7,9,10,12,14–16]. These researchers clarified that decrease of ionic strength [9,10,12,14] and increase of pH [7,10,16] will induce colloid release. Decrease of ionic strength results in the expansion of electrical double layer. Increase of pH enlarges negative charge on variable charge of colloidal par-

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ticle. These effects increase repulsive force between particles and collector surface, thus the release of deposited particles is enhanced.

Recently, Hahn et al. performed release experiment of latex particles deposited on glass beads in the aqueous solution of NaCl and CaCl<sub>2</sub> [11]. They detected that little release of the particles deposited in the solution of 0.2 M CaCl<sub>2</sub>, however, it is not the case for the solution of NaCl. The reason was ascribed to the stronger adhesive bond between colloidal particle and the surface of collector grain is formed in CaCl<sub>2</sub> solution. They also reported similar result for the release of hematite deposited onto quartz sand at low pH [17]. Lenhart and Saiers carried out release experiments using silica colloids and column packed with quartz sand grains [13]. They demonstrated that the release behavior varies according to the ionic strength of deposition step. This result agreed well with their model that explains the relation between release behavior heterogeneity and distribution of energy barrier assigned to each particle deposited on grain surface.

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Not only the magnitude or distribution of interaction energy between deposited particle and surface of collector grain, but the structure and state of deposited particle can be expected to affect the release of colloid. The structure of deposited particles is influenced by the change of solution chemistry of deposition. Yao et al. confirmed that deposition rate increases in accordance with an increase of deposited colloidal particles under rapid coagulation condition [18]. This is called ripening. In ripening, the deposited particles can be new sites for further particle coming to the surface of grain. And the deposition is enhanced sequentially forming some surface aggregate of deposited particles. Cerda observed "surface aggregation" of kaolinite fines formed on sand surface at favorable condition of kaolinite-kaolinite attachment by microscopy [7]. Yamashita and Adachi confirmed that large flocs of polystyrene latex formed on upper part of column at rapid coagulation condition [21]. On the contrary, Rajagopalan and Chu confirmed that deposition rate decreases with an increase of amounts of deposited colloidal particles at slow coagulation condition (when colloid-colloid attachment is unfavorable due to repulsive force) [19]. This is the phenomenon called blocking that deposited particles prevent the approach of new particles in the neighbor to form thin monolayer. Kuhnen et al. confirmed that structure of hematite deposition layer changes denser with ionic strength increasing [20]. Physical capture mechanism of colloidal particle, straining, also has to be considered. Straining is the trapping of colloidal particles in down-gradient pore throats that are too small to allow particle passage. Tufenkji et al. suggested the importance of straining in the deposition mechanism of colloidal particles [22]. They indicated that straining is become important capture mechanism when physicochemical attachment is not dominant and is not influenced by solution chemistry. It can be easily hypothesized that release of colloidal particles is affected by the state and structure of deposition.

The aim of present study is to analyze colloid release in accordance with the history of deposition. Kaolinite; abundant and representative colloid in soil with relatively clear characteristic, was used as colloid material. Toyoura sand; conventionally used as standard material in the field of soil mechanics in Japan, was used as collector grains packed in column. Deposition experiments were performed for different pH condition in accordance with the change of colloidal stability. Then release was induced by flowing  $1.0 \times 10^{-4}$  M NaOH solution. The deposition state was characterized by the analysis of breakthrough curve, observation of collector surface with microscopy and coagulation properties of kaolinite.

## 2. Experiment

### 2.1. Materials

Iriki kaolinite purchased from Iwamoto Kousan Co. Ltd. was used after the pretreatment. The procedure of the pretreatment is as follows. (1) A portion of the kaolinite (150 g) was washed with deionized distilled water. (2) The kaolinite was washed with 1.0 M NaCl and  $1.0 \times 10^{-4} \text{ M}$  NaOH solution for three times to exchange adsorbed cation with Na<sup>+</sup> and keep the colloidal



Fig. 1. Electrophoretic mobility of kaolinite in  $1.0 \times 10^{-4}$  M NaCl solution.

suspension stable by adjusting solution pH to sufficiently high value. In each washing step, the kaolinite suspension was stirred for at least 1 day. (3) The suspension was dialyzed until the electric conductivity decreases to less than  $2 \mu$ S/cm. (4) The size fraction less than  $2 \mu$ m in Stokes diameter was collected by sedimentation.

Colloidal chemical properties of prepared sample were analyzed by electrophoresis and coagulation experiments. Electrophoretic mobility was measured as a function of pH under the ultramicroscopy. Concentration of the suspension was diluted by  $1.0 \times 10^{-4}$  M NaCl solution to 500 mg/l. pH of the suspension was adjusted by  $1.0 \times 10^{-4}$  M HCl and NaOH. The measurement was performed using commercial apparatus (Model502, Nihon Rufuto Inc.). Electrophoretic mobility was measured with the electric field of 11.3 V/cm. The obtained data of electrophoretic mobility of kaolinite in the  $1.0 \times 10^{-4}$  M NaCl solution is plotted as a function of pH in Fig. 1. The rate of Brownian coagulation was measured as a function of pH by turbidity method using spectrophotometer (UV-1650PC, Shimadzu) at a wavelength of 632.8 nm [23]. Increase of absorbance was plotted with elapse time in Fig. 2.

Toyoura sand (Toyoura Silicate Mineral Corp.), which is widely used as a standard material of sand especially in the field of civil engineering in Japan, were used as collector grain. It is a quartz dominative sand and its particle size is distributed between 150 and 300  $\mu$ m. A portion of the sand (300 g) was washed several times by deionized distilled water to remove the



Fig. 2. Temporal variation of light absorbance of kaolinite suspension under stationary condition. Concentration and ionic strength of kaolinite suspension was adjusted 510 mg/l and  $1.0\times10^{-4}$  M NaCl.

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