

Filtration in Colloid Migration through Porous Media — Colloid Deposition Behavior onto Solid Surface —[†]

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Groundwater colloids which adsorb actinide ions play an important role in actinide migration. The existence of colloids causes an effect of facilitating the migration, while it also causes a possibility of retardation of colloid transport due to filtration effect. The filtration phenomenon in porous media is dominated by deposition of colloidal particles onto stationary grain surfaces from flowing suspensions. The deposition behavior depends on the chemical characteristics of the particle surfaces and grain surfaces and on the solution chemistry, while the mechanisms of the behavior are not understood enough under the subsurface aquatic environment. In this study, we carried out column and batch experiments to investigate effects of flow field and deposited particles on the filtration behavior in porous media. We clarified the importance of the effects in quantifying the filtration effect in the migration of colloids through porous media.

Keywords: colloid, filtration, deposition, DLVO theory, ionic strength, distribution coefficient, column experiment, batch experiment

アクチノイド元素イオンの移行において、これを吸着した地下水中のコロイドの挙動は重要な役割を果たす。コロイドの存在は核種の移行を促進する影響を与える一方で、フィルトレーションの効果によって、その移行を遅延させる可能性がある。多孔質媒体中でのフィルトレーションは、流れ場中から固相粒子表面へのコロイドの付着によって起こる。付着の挙動はコロイド・固相粒子の表面科学的性質と、溶媒の化学的性質に支配されるが、地下環境下ではこの現象について不明な点が多い。本研究では、フィルトレーションが多孔質媒体中でのコロイドの移行に与える影響を定量化する際に、流れ場と固相表面の付着コロイドの影響が重要となることを、カラムおよびバッチ実験から明らかにした。

Keywords: コロイド, フィルトレーション, 付着, DLVO 理論, イオン強度, 分配係数, カラム実験, バッチ実験

1 Introduction

Groundwater colloids which adsorb actinide ions play an important role in actinide migration from the viewpoint of the performance assessment of high-level radioactive waste disposal. The colloids act as a third phase which is neither liquid nor solid phase. This phase can increase the amount of actinides that can migrate in a natural aquifer system[1]. This possibility was found from overestimation of retardation which was predicted with distribution coefficients determined by batch adsorption experiments[2,3]. It was also pointed out that colloidal particles will generally travel at slightly higher velocities than the advecting fluid medium, known as hydrodynamic chromatography[4-6]. On the other hand, colloid transport can be retarded due to the filtration effect[1].

On the performance assessment of high-level radioactive waste disposal, the filtration effect on the migration of colloids in aquifer systems has been investigated. Saltelli et al.[7] studied filtration effect on migration of polydispersed colloids in porous media. They applied modified version of the filtration equations to describe their experimental results. Although their model shows good agreement, it has the problem of obtaining the parameters which were optimized in their simulation[8]. Hwang et al.[9] described a mathematical model of real colloids and solute considering colloid filtration in fractured porous media.

In this model, linear sorption of radionuclides is assumed on solid surfaces and on colloids. Experiments show, however, that it is not a linear sorption but a non-linear sorption which has considerable effects on the transport of radionuclides[10].

The mechanisms of the filtration have been investigated on subjects such as water treatment or transport of colloidal contaminants in groundwater[11-22]. To describe the removal of colloidal particles by solid surfaces, colloid deposition theories are introduced. Deposition is a removal process of colloids (described in detail in Chapter 2). A thorough understanding of colloid deposition is essential for prediction of colloidal transport in natural porous media[11]. The colloid deposition theories have been tested by conducting column experiments with model colloids and grains (collectors) under controlled physical and chemical conditions[11-19, 22]. Two types of colloid deposition are recognized. So-called “unfavorable” deposition involves colloid deposition in the presence of repulsive electrical double layer interaction between the particle and the collector, while that in the presence of attractive electrical double layer interaction is referred to as “favorable” deposition. Current deposition theories are adequate for predicting an initial particle deposition rate (see in detail in Chapter 2) under favorable conditions[12, 13]. The enhancement of the initial particle deposition rate is observed at the lower ionic strength of the suspension in this case. Under unfavorable conditions which is dominant in the groundwater system, however, these theories are not adequate for the prediction, although the enhancement of the initial particle deposition rate at the higher ionic strength of the suspension can be predicted qualitatively[14-16]. Several explanations for the discrepancy between theory and experiment under unfavorable conditions are as follows[17]: (1) distribution of surface

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properties, (2) heterogeneity of surface charge, (3) surface roughness of particles and collectors, (4) dynamics of interaction, (5) deposition in secondary minimum of interaction energy, and (6) possibility of additional forces. These explanations, however, are not sufficient completely. For example, Elimelech and O'Melia[14] tested the effect of deposition in secondary minimum by conducting column experiments at two different approach velocities of the fluid towards the collector. They reported that significant deposition in secondary minima is not likely to occur, however, this conclusion is restricted to the condition under which they conducted the experiments. The effect of the flow field on the deposition is needed to be investigated further. On dynamic (transient) aspects of colloid transport (see in detail in Chapter 2), the effects of deposited particles on the blocking phenomenon during a single column experiment has been investigated[11, 18, 19]. A study on the effect of deposited particles by conducting sequential runs with back wash process would give us some information about the deposition behavior. Such study, however, has not been performed enough.

The purpose of the present paper is to investigate the effect of flow field on the deposition and the effect of deposited particles on the sequential deposition. We studied on these subjects to clarify the importance of the effects in quantifying the filtration effect in the migration of colloids through porous media. We carried out column and batch experiments to observe the effect of flow field by comparing the results of these experiments and performed column experiments with back wash process to study the effect of deposited particles.

2 Deposition Process in Filtration

2.1 Filtration Mechanisms in Porous Media

Filtration phenomenon in porous media is generally divided into three categories: surface (cake), straining, and physical-chemical (deep bed) filtration[20]. Surface filtration occurs when particles are comparable in size to or larger than the porous media grain size. In this case, particles can not penetrate into the media, resulting in formation of a filter cake or a surface mat above the media. In straining filtration, particles small enough to enter the porous media can be mechanically removed in smaller pore spaces. In the case of radionuclide migration in the groundwater, the physical-chemical filtration is the dominant mechanism which also dominates the transport of colloidal pollutants such as bacteria, viruses, and asbestos fibers. In this mechanism, very small particles relative to the porous media grain size (colloidal particles) can be removed from solution by deposition onto grain surfaces. To describe the removal of particles at the initial stage of filtration (the so-called "clean-bed removal"), filtration theories are introduced which determine the initial particle deposition rate[11]. In the filtration theories, the overall removal of suspended particles by porous media is divid-

ed into two sequential steps: transport and attachment.

2.2 Transport Step in Colloid Deposition

The transport step is a particle transport process from bulk fluid to the vicinity of the filter grains, which can be quantitatively described by the convective-diffusion and trajectory equations. Figure 1 shows three mechanisms (diffusion, interception, and sedimentation) which are responsible for the transport of particles from the bulk fluid to a vicinity of a single spherical filter grain (collector)[21]. Diffusion is a process in which a particle undergoing Brownian motion results in contact with the collector. Interception occurs when a particle moving with the streamline comes into contact with the collector because of its finite size. Sedimentation denotes a mechanism in which a particle having greater density than that of the fluid settles downwards, crosses the streamlines, and contacts the collector.

2.3 Attachment Step in Colloid Deposition

The attachment step is controlled by the chemical characteristics of the particle and grain surfaces and by the solution chemistry. The transported particle is attached when the surface force between the particle and the collector is attractive. Derjaguin-Landau[23] and Verwey-Overbeek[24] (DLVO) theory which accounts for the van der Waals and electrical double layer interactions describes the surface force. When the electrical double layer interaction is repulsive, the van der Waals force dominates

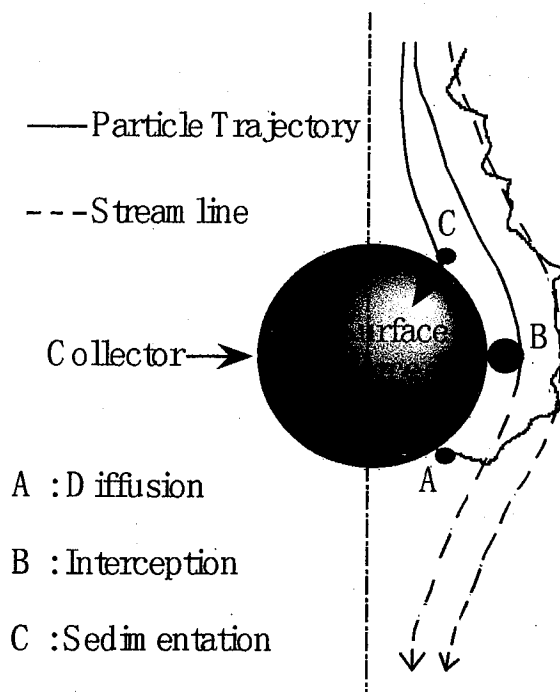


Fig. 1 Particle transport mechanisms

at short and long distances between the particle and the collector and the interaction is attractive while at intermediate distances the interaction is repulsive[25]. In this case, the curve of the interaction energy as a function of the distance has two energy minima. The minimum at short distance is referred to as the primary minimum and the minimum at long distance is referred to as the secondary minimum. The depth of the primary minimum is larger than the secondary minimum. Between the two minima, an energy maximum exists. The depth of the minima and the height of the maximum are varied by varying either physical and chemical conditions such as surface potentials of the colloid and the collector, ionic strength of the solution, and colloid size.

2.4 Dynamic (Transient) Aspects of Colloid Transport

It should be noted that filtration theories mentioned above are applicable only to the initial stage of filtration where collectors are devoid of deposited particles. The initial stage corresponds to the breakthrough of the solution front. In experimental studies, dynamic (transient) particle deposition processes were observed as either a decreasing[11, 18, 19] or increasing[22] deposition rate, which is referred to as "blocking" and "ripening" respectively. When collectors have net opposite charge to particles, deposition occurs onto major area that favors particle deposition. On the other hand, when collectors have net similar charge to particles, deposition is considered to occur onto minor patches on collector surfaces that favor particle deposition. For both situation stated above, "ripening" occurs when favorable particle-particle interaction exists, while "blocking" occurs when unfavorable particle-particle interaction exists[11]. Blocking prohibits interparticle contact, and restricts particle density on collector surfaces to a single monolayer. On the other hand, ripening enhances the rate of deposition, because deposited particles act as additional collectors. Ripening leads to a rather complex multilayer coverage of collector surfaces.

3 Experimental

3.1 Materials

Spherical soda lime glass beads (Toshin Riko) with diameter of 0.35-0.40 mm were used as packing material in column experiments and as solid phase in batch experiments. The glass beads were cleaned by soaking for 30 minutes in an ultrasonic bath with distilled deionized water (Barnstead; Fistream II, E-pure) and 1M HNO₃ solution successively, then rinsed with distilled deionized water repeatedly, and dried in an oven at 60 °C. Distilled deionized water was used for preparing the all solutions used in this work.

Monodispersed suspensions of polystyrene latex particles having mean diameters of 55 nm and 102 nm (Magsphere Inc. for 55 nm particles and Seradyn Inc. for 102 nm particles) were

used as model colloids in column and batch experiments. The particles contain surface functional groups of sulfate (SO₄⁻), and their density, as reported by the manufacturer, are 1.05 g/mL for both. Aqueous suspensions of latex particles (100 mg/L and 10³ mg/L for the particles of 55 nm and 102 nm in diameter, respectively) were used in column and batch experiments. The ionic strength *I* in the suspensions was adjusted by the addition of NaNO₃. The concentrations of NaNO₃ were 0 M, 10⁻³ M, 10⁻² M and 10⁻¹ M.

3.2 Column Experiments

Before each run, exactly 6.05 g of the prepared dry glass beads was wet packed with distilled deionized water in a cylindrical Teflon column of 50 mm length and 10 mm inner diameter. This procedure resulted in a saturated column with porosity of 0.384, based on a glass beads density of 2.5 g/cm³. A low flow rate plunger pump (Nihon Seimitsu Kagaku; NP-FX series) was used in feeding solutions into a column. The pH of the effluent is initially larger than that of influent. This increase in the pH is caused by the contact of the fed solution with the glass beads; it has been postulated that the soda lime glass surface consume protons from the solution while releasing sodium ions[26]. To minimize the changes in the pH of the effluent during column experiments, particle-free solution have been fed until the increase in the pH became less than 0.1 pH unit.

Following the preparation of the column, an aqueous suspension of latex particles was pumped through the vertical column at the constant upward flow rate of 1.025 ± 0.045 mL/min. Particle concentrations in the effluent leaving from the columns were continuously monitored with a UV-VIS spectrophotometer (Shimadzu; UV-1200) equipping a flow-through quartz cell at wavelength of 260 nm for both particles. Prior to the column experiments, reproducibility of the characteristics of the columns was checked on several runs by feeding 0.3 M CoCl₂ solution as a conservative tracer, which flows at the same velocity as that of advective fluid. We had previously checked that CoCl₂ does not sorb on the glass beads used in this work by batch experiment. CoCl₂ concentration was determined by using the same spectrophotometer at wavelength of 510 nm. During column experiments, the pH values were maintained at 4.3-4.5, 5.5-5.7, 4.7-4.9 for smaller particles, larger particles, and CoCl₂ solution respectively. The pH values of smaller particles and larger particles were different, but the pH were not adjusted further in the present work.

A column was prepared for every single run, except for experiments on the effect of back wash process. In the back wash experiments, after finishing the first run at *I*=10⁻¹ M, the column used was back washed by pure 10⁻¹ M NaNO₃ solution at relatively high flow rate (several times higher than the breakthrough runs) to remove the latex particles retained in the column. After confirming that the latex particle concentration in the effluent

dropped to zero, the second run at $I=10^{-1}$ M was performed. These experiments were carried out for both particles at $I=10^{-1}$ M. All column experiments were conducted at room temperature ($20 \pm 2^\circ\text{C}$) under aerobic conditions.

We observed latex particles deposited at $I=10^{-1}$ M on fibers of a $5 \mu\text{m}$ membrane filter by scanning electron microscopy (SEM). This observation revealed that there was no large aggregate which was large enough to cause surface or straining filtration. We also observed the surfaces of glass beads and latex particles deposited on them by SEM to certify whether the deposited latex particles remained on glass beads or not after back wash process.

3.3 Batch Experiments

Exactly 5.00 g of prepared glass beads were weighed into centrifuge tubes, and then the tubes containing the glass beads were weighed. The glass beads in the tubes were washed with distilled deionized water 15-20 times until there was no further increase in pH of the water. The water was carefully drained without loss of the glass beads. The tubes containing the glass beads and remaining water were weighed again. The amounts of the remaining water were determined from the obtained total weights of the tubes before and after washing the glass beads. The same amount of distilled deionized water as the remaining water was weighed into centrifuge tubes for blank samples. These procedures were conducted to minimize the effects of changes in pH of liquid phases.

5.00 mL of aqueous suspensions were put into the tubes, and the tubes were shaken by hand moderately for 1 minute. Particle concentrations in the liquid phase were obtained with a UV-VIS spectrophotometer (Hitachi; U-3300) at wavelength of 260 nm for both particles. Measurements were made in quartz cells of 1.0 cm path length. All runs were conducted at room temperature ($20 \pm 2^\circ\text{C}$) under aerobic conditions.

4 Results and Discussion

4.1 Particle Breakthrough Curves

Breakthrough curves within two pore volumes of effluent for the conservative tracer (CoCl_2) and latex particles of both sizes at various NaNO_3 concentrations are shown in Figs. 2 and 3. The earlier arrival of latex particles with respect to the tracer was observed for lower ionic strength conditions. This enhancement of particle velocity is a result of hydrodynamic chromatography[4-6]. Because of a particle's finite dimensions, it is prohibited from contacting the slower moving fluid near the pore boundaries. For higher ionic strength, however, the particle deposition onto glass bead surfaces increases, resulting in a smaller fraction of particles being eluted from the column. This is often ascribed to reduced electrical double layer repulsion forces caused by compression of the layer[14].

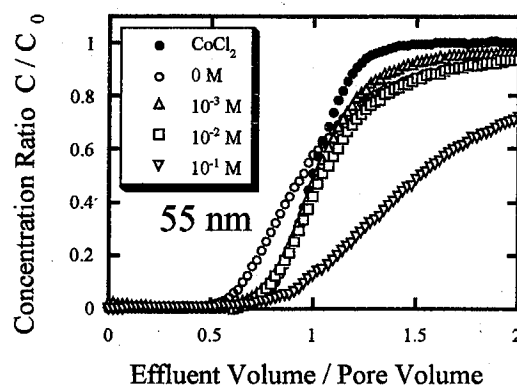


Fig. 2 Breakthrough curves of the 55 nm latex particles as a function of NaNO_3 concentration. Inlet concentration: 100 mg/L

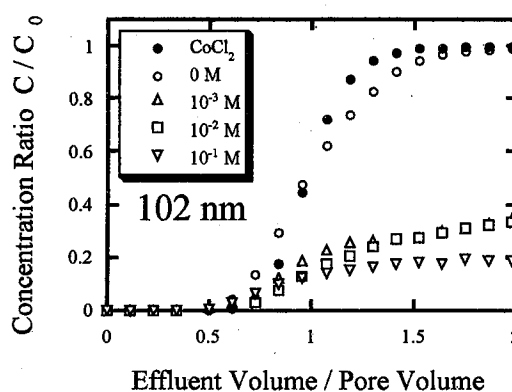


Fig. 3 Breakthrough curves of the 102 nm latex particles as a function of NaNO_3 concentration. Inlet concentration: 10 mg/L

Breakthrough curves over the entire period of each run are presented in Figs. 4 and 5. As mentioned above, the dependence of ionic strength on particle deposition can be clearly seen in these figures. It was revealed that total breakthrough ($C/C_0=1$) was accomplished within several pore volumes of effluent for the smaller particles, and the approach rate to total breakthrough after one pore volume of effluent for the larger particles was much smaller than that of smaller particles. This can be ascribed to the effect of particle size. Larger colloidal particles experience enhanced deposition onto collector surfaces due to the interception effect[21]. As we explained before, the pH conditions were not the same in these experiment series. However, since the higher the pH value is, the less the deposition is[11], our conclusion was not influenced by this pH difference.

The approach rate to total breakthrough after one pore volume

of effluent is an indication of the relative area blocked by a deposited particle, with steeper breakthrough curve slopes corresponding to larger excluded areas[18, 19, 22]. The lower the ionic strength is, the steeper the slope is. This can be explained as exclusion of succeeding particles from deposition due to expanded electrical double layer of deposited particles. For the particles of 102 nm size at $I=10^{-1}$ M, the breakthrough curve slope changed dramatically after one pore volume of effluent. This is because breakthrough of the solution front occurred. After that, the breakthrough curve slope was gentle. This indicates that the blocking phenomenon was not effective and that the collector almost maintained its initial removal efficiency. Total breakthrough was not accomplished in this case, just because the run was finished on the way to the total breakthrough (The run was finished at 40 pore volumes of effluent). When ripening occurs, total breakthrough will not be accomplished, and the breakthrough curve slope after one pore volume of effluent should be negative. A transition from blocking to ripening will be observed, for example, when the concentration of an electrolyte containing divalent counterions is gradually increased[22].

4.2 Effect of Deposited Particles

Figures 6 and 7 show the breakthrough curves for the first and the second runs (before and after back wash process) for particles of both sizes at $I=10^{-1}$ M. The back wash process between these runs was conducted by feeding the particle-free solutions at relatively high flow rate (several times higher than the runs). For the particles of 55 nm size, blocking phenomenon occurred in the first run (total breakthrough was accomplished). In the second run, the blocked area was larger than that in the first run (the slope was steeper), which indicates that all particles deposited at the first run were not detached by back wash process. For the particles of 102 nm, the value of C/C_0 at one pore volume of effluent for the second run was larger than that of the first run, because of the deposited particles remaining on the glass

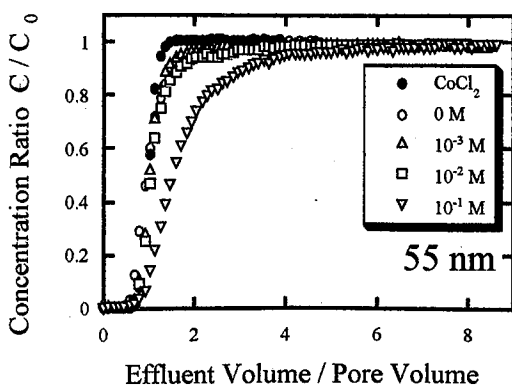


Fig. 4 Breakthrough curves of the 55 nm latex particle as a function of NaNO_3 concentration. Inlet concentration: 100 mg/L

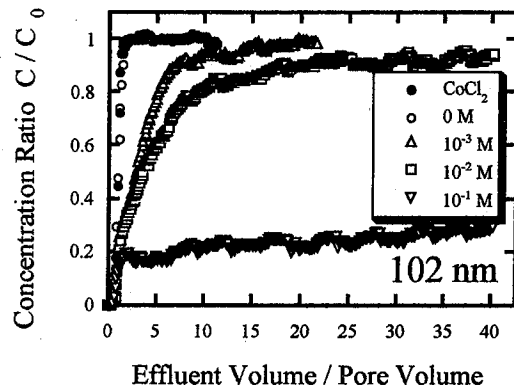


Fig.5 Breakthrough curves of the 102 nm latex particles as a function of NaNO_3 concentration. Inlet concentration: 10 mg/L

beads. It should be also noted that the value of C/C_0 of the second run at one pore volume of effluent was lower than that at the end of the first run. The lower value of C/C_0 indicates higher removal efficiency. The increase in removal efficiency can be ascribed to detachment of the deposited particles by which excluded areas blocked by deposited particles are reduced.

To certify whether the deposited latex particles remained on glass beads or not, we observed surfaces of glass beads by SEM after back wash process. This observation revealed the existence of deposited particles remaining after back wash process although the number of particles deposited decreased after back wash process. These particles were inclined to be observed at cavernous sites on the glass beads. This inclination indicates that surface roughness of the collector and maybe heterogeneity of surface charge of the collector also cause difference in magnitude of the surface force between the particle and the collector. Particles deposited in primary minimum of interaction energy diagram are not likely to be detached. This is because these particles have to surmount the energy barrier to be detached. For particles deposited in secondary minimum, it can be possible to detach. Further studies concerning detachment of deposited particles will be needed to reveal the behavior of deposition.

4.3 Distribution Coefficient

The distribution coefficient K_d (mL/g) was obtained in three ways: batch method, fitting method, and integration method to find out characteristics of colloid deposition during its transport process in a porous media.

In the batch method, K_d was obtained as a ratio of amount of particles deposited on collector surfaces to that in the liquid-phase. In the fitting method, K_d was obtained by fitting the column experiment data to the analytical solution of the convective-diffusion equation. In the integration method, the retarda-

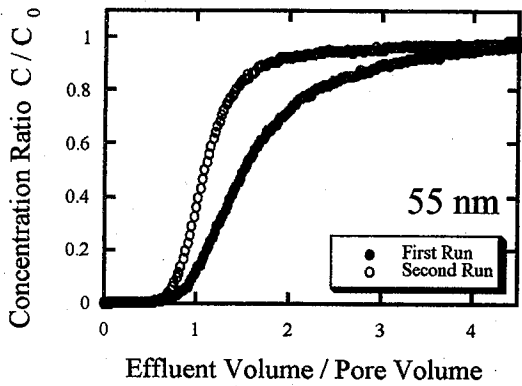


Fig.6 Breakthrough curves of the 55 nm latex particles at NaNO_3 concentration of 10^{-1} M for the first and the second runs. Inlet concentration: 100 mg/L

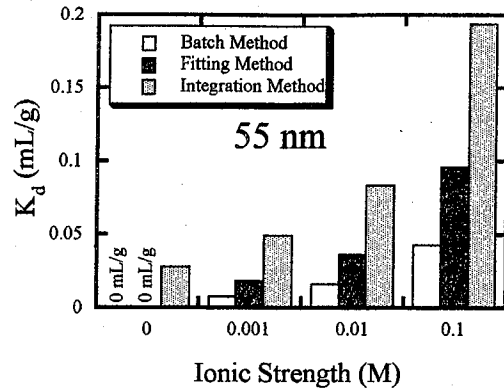


Fig.8 Distribution coefficient of 55 nm latex particles for spherical soda lime glass beads determined by batch, fitting, and integration methods.

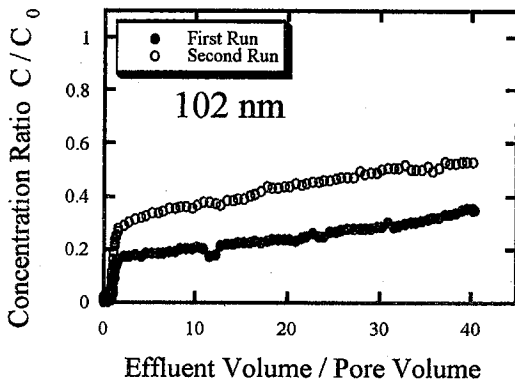


Fig.7 Breakthrough curves of the 102 nm latex particles at NaNO_3 concentration of 10^{-1} M for the first and the second runs. Inlet concentration: 10 mg/L

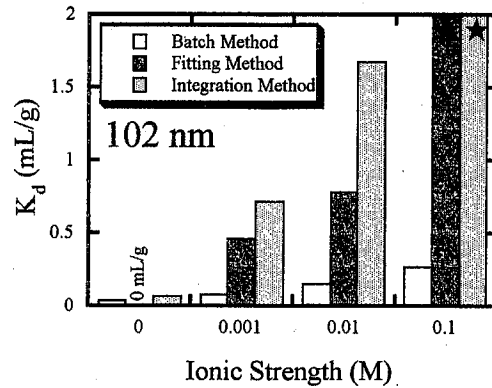


Fig.9 Distribution coefficient of 102 nm latex particles for spherical soda lime glass beads determined by batch, fitting, and integration methods. '★' indicates the value which is larger than determinable limit

tion factor R of each run was calculated from the results of column experiments[27, 28]. R was obtained by numerical integration of the area between the breakthrough curve and a horizontal line of $C/C_0=1$. Since the retardation factor R is related to K_d , porosity ϵ , and the collector density ρ , and described as $R = 1 + K_d \rho (1 - \epsilon) / \epsilon$, K_d was obtained from this equation.

K_d obtained by the three methods are presented in Figs. 8 and 9. The results indicate that all K_d increased with the ionic strength, which is ascribed to the compression of the diffuse electrical double layer. K_d for the larger particles was larger than that for the smaller particles due to increase in interception effect. The transport mechanism of interception may still operative in a batch system because of the random flow field during the shaking process. K_d obtained by fitting method and those obtained by integration method were different. This difference can be explained as follows. In the fitting method, some errors may be included in fitting of dispersivity of particles. The integration method is valid for a phenomenon which has a linear relation be-

tween concentration and amount of deposition (sigmoidal breakthrough curve). In the column experiments, this condition was not realized, resulting in including some errors. Nevertheless, it should be noted that the values obtained from the batch method were smaller than those obtained by the other two methods in which column experiment data were utilized. This implies that the effect of flow velocity on the deposition should be clarified. In the column experiments, the particles which have higher kinetic energy than those in the batch experiments can surmount the interaction potential energy barrier, resulting in higher deposition rate. The other reason for the difference may be the existence of trapped particles in cavern sites. We will investigate the effect of flow velocity and physical-chemical interactions near the interface on the colloid deposition in detail in near future.

5 Conclusions

We investigated the deposition behavior in colloid transport through porous media by conducting column experiments and batch experiments using polystyrene latex particles and spherical glass beads. The conclusions of the present work are summarized below.

- (1) The comparison between the results of the batch and the column experiments indicated that the deposition was enhanced in the column experiments compared with the batch experiments due to particles trapped by the effect of flow field.
- (2) The results of the column experiments revealed that breakthrough behavior of colloidal particles of the second run after back wash process is affected by remaining particles on collector surfaces.

The results of the column experiments and the batch experiments implies necessity of the further investigation on the colloid filtration with respect to the behavior of deposition, especially, the effect of flow field and characteristics of surfaces.

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