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Foam-enhanced removal of adsorbed metal ions from packed sands with biosurfactant solution flushing



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ABSTRACT

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Keywords: Biosurfactant Foam-enhanced solution flushing Surfactant solution flushing This study demonstrated the abilities of negatively charged biosurfactants, surfactin and rhamnolipid, to remove adsorbed copper and cadmium ions from sand surfaces with the foam-enhanced solution flushing technique. A popular anionic surfactant, sodium dodecylsulfate (SDS), was used for the purpose of removal efficiency comparison. The role of surfactant foaming ability in the flushing approach was then identified. It was found that the surfactant solution flushing could only result in limited removal efficiency of 3–10% and 13–36% for copper ion and cadmium ion, respectively, after 24-pore volume (PV) flushing due to the channeling effect. As compared to the surfactin solution, a less pronounced channeling effect was detected for the rhamnolipid or SDS solution. With the presence of foam in the flushing approach, the channeling effect could be inhibited, and one could obtain improved removal efficiency was higher for cadmium ions than for copper ions, which could be explained by the significant adsorption of the cadmium ions in the inter-particle pore regions. Moreover, the cumulative removal efficiency variations with the foam-enhanced solution flushing could be correlated with the dynamic foam capacity of the surfactant solutions.

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1. Introduction

Metal ions in a solution flowing over sand particles may adsorb onto the particle surfaces with various interaction mechanisms [1,2]. The outer-sphere type is the interaction between metal ions and the sand surfaces involving the liquid phase. The inner-sphere type is the interaction between metal ions and functional groups on the surfaces, which does not involve a liquid phase or water molecules between the ions and the surfaces [3–6]. Natural sands may have macropores and mesopores, and the porosity is mostly influenced by particle size, grain shape, and rock type [7,8]. The porosity can be classified into inter-particle porosity and intraparticle porosity. Pores cause not only great surface area, but also high selectivity in adsorption [9]. The interaction between metal ions and sand surfaces will affect the desorption behavior of the metal ions in a remediation process.

Several remediation technologies have been developed for removing heavy metal ions from contaminated soils. A foam-enhanced solution flushing technique has been applied to remediate soils containing metal ion contaminants, and one was able to improve the migration of surfactant solutions with the presence of foam during the solution flushing process [10–12]. The foam could inhibit the channeling effect of solution flow by increasing the resistance of the solution flow and thus by forcing the solution to homogeneously flow throughout the medium. This would enhance the removal efficiency of the soil remediation [10,13,14]. By using a foam-enhanced solution flushing technique, the removal efficiency was increased even in a heterogeneous porous medium [15]. The effectiveness of foam generation is influenced by surfactant concentration [11,16], and this technique is attractive due to the low usage of surfactants [10,11].

Applications of biosurfactants in the field of environmental protection have received much attention due to their biodegradability, low toxicity, effectiveness in enhancing biodegradation, and ability to solubilize hydrophobic compounds [17–20]. Surfactin is a biosurfactant produced by various strains of *Bacillus subtilis* with negatively charged characteristic in rod micelle form [21,22] and excellent foam stability [23]. Rhamnolipid is produced by *Pseudomonas aeruginosa* [24,25] and has excellent quality of foam [26]. Rhamnolipid in water possesses a negatively charged characteristic and may be particularly effective in remediating soils contaminated with metal ions that are less sensitive to ion exchange processes [25].

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The objective of this study is to demonstrate and compare the abilities of two biosurfactants, surfactin and rhamnolipid, to remove strongly adsorbed metal ions from sand surfaces with the foam-enhanced solution flushing technique. The key role of surfactant foaming ability in the flushing approach is then identified. Sands with adsorbed metal ions were prepared by an adsorption process and then were dried to allow the metal ions to interact with the sand surfaces mainly through the inner-sphere interaction. Physical properties of the biosurfactant solutions were characterized, and the removal efficiency of the biosurfactant solutions without or with foam for the adsorbed metal ions from the sand surfaces was evaluated. The removal efficiency obtained by using a popular anionic surfactant, sodium dodecylsulfate (SDS), solution was also determined for the comparison purpose, and the importance of the surfactant foaming ability on the removal efficiency of the foam-enhanced solution flushing approach was discussed.

2. Materials and methods

Surfactin was produced from *Bacillus subtilis* ATCC 21332 [27] with purity about 90%, and rhamnolipid was produced from *P. aeruginosa J4* with purity about 63% [24]. Sodium dodecylsulfate (SDS), a popular anionic surfactant, (purity 99.0%) was purchased from Sigma–Aldrich, Japan. Research-grade copper (II) sulfate pentahydrate (Cu₂SO₄·5H₂O) and cadmium chloride (CdCl₂) purchased from Showa Chemical Co. Ltd., Japan, were chosen as the sources for adsorbed metal ions on sand surfaces. Purified water with a resistivity of 18.2 M Ω cm was obtained from a Milli-Q plus purification system (Millipore, USA) and was used in all experiments.

The sands were cleansed with purified water and were adsorbed by copper and cadmium ions through mixing the cleansed sands with the metal ion solutions. 100-g sand was mixed with 100 mL of 50-ppm metal ion solution, and the mixture was shaken for 24-h with 150 rotations per minute. Sands with adsorbed metal ions were dried by introducing N₂ gas to remove the aqueous phase with the metal ions adsorbing through the inner-sphere interaction [5,6]. After the sands were completely dried, the metal ion concentration in the solution was analyzed by using an atomic absorption spectrometer (model SensAA Dual, GBC Scientific Equipment Pty Ltd., Australia) and the adsorption density of the metal ions on the sand surfaces was determined.

For a foam-enhanced surfactant solution flushing operation, a glass column with a length of 5 cm and an outside diameter of 3.5 cm was used as the foam-generator with inlets for surfactant solution and N_2 gas, respectively [12]. The continuous flow of surfactant solution into the foam generator was controlled by using a peristaltic pump, and the dynamic foam capacity of a surfactant solution was assessed when the volume of foam in the foam generator was constant. The dynamic foam capacity was determined by dividing the constant volume of foam (mL) by the flow rate of N_2 gas (mL/min). A glass column with a length of 7.5 cm and an outside diameter of 1.5 cm was used in the sand-packed column experiments to simulate the soil remediation condition. Sand particles with an average diameter of 320 μ m were used as the porous medium in the column.

All experiments were performed at room temperature. Surfactin solution was prepared in a 10^{-3} M phosphate buffer with a pH value of 8.0 [23,28,29]. Rhamnolipid was dissolved in pure water with a pH of 5.6 to prepare the aqueous solution [30]. The SDS solution was also prepared with pure water of pH = 5.6. Flow rates were fixed at 2 mL/min for surfactant solution and at 20 mL/min for N₂ gas. Biosurfactant solutions with a concentration of 5× critical micelle concentration (cmc) and SDS solution with a concentration of 2.5× cmc were used to flush the metal

ion-adsorbed sand-packed column. The effluent from the packed column during the flushing operation was collected every 4 porevolumes (PVs, one PV is about 2.2 mL), and the metal ion concentration in the effluent was analyzed by atomic absorption spectrometry.

Images of the cleansed sand surface were obtained by using a scanning electron microscope (SEM) (JOEL JSM-6700F, Japan). The surface tension-lowering abilities of surfactin and rhamnolipid in aqueous phase were evaluated with a Wilhelmy plate tensiometer (CBVP-A3. Kyowa Interface Science Co. Ltd., Japan). The zeta potentials of surfactin and rhamnolipid micelles or aggregates were measured by using a zeta potential analyzer (model 3000HS, Malvern Instrument, UK).

3. Results and discussion

3.1. Sands with adsorbed metal ions

The sand surface morphology was observed by SEM and a typical image is shown in Fig. 1. The porous characteristic of the sand surfaces with the presence of intra-particle regions was demonstrated in the SEM image and was expected to affect the metal ion adsorption density.

When a 100-mL aqueous solution containing 50-ppm metal ion was mixed with 100-g sand, the metal ions would adsorb onto the sand surfaces and the adsorption density was increased with time until the adsorption equilibrium was reached [2]. The metal ions in the solutions may interact with the sand surfaces with outer-sphere and inner-sphere types [3]. After drying the sand particles with N₂ gas, adsorption densities of cadmium and copper ions on the sand surfaces were found to be 5.85 and 13.45 mg/kg, respectively (Fig. 2). With the N₂ drying treatment, the adsorbed metal ions with the outer-sphere interaction type on inter-particle sand surfaces would be removed, and the adsorbed metal ions with the inner-sphere interaction type were expected to remain on the sand surfaces.

3.2. Physical properties of surfactant solutions

The abilities of two biosurfactants, surfactin and rhamnolipid, to lower surface tension of aqueous phase are demonstrated in Fig. 3. Based on the surface tension data, the critical micelle concentrations of surfactin and rhamnolipid were estimated to be about 20 mg/L and 40 mg/L, respectively. The surface tensions of surfactin and rhamnolipid aqueous solutions at corresponding critical micelle concentrations were 31 mN/m and 35 mN/m,



Fig. 1. SEM image of the sand surface.

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