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ARTICLE

New filter concept for removal of fine particle generated in high level radioactive solution

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Extraction chromatgraphy technology for trivalent minor actinide (MA(III) ; Am(III) and Cm(III)) recovery from the solution generated by an extraction process in reprocessing of spent nuclear fuel has been developed. Since the solution contains many kinds of elements, a fine particle is generated in the solution. The fine particle must be removed before MA recovery operation, because that leads clogging of the extraction chlomatography column. In order to prevent clogging the column, filtration system utilizing porous silica beads packed column has been designed. In this study, a fine particle trapping system was developed and particle removal performance of the system was experimentally evaluated using alumina particles as simulated fine particle. Column experiments revealed that the fine particle with the particle size from 0.12 to 15 μm is cause of clogging of the filtration column. Since simulated fine particles were trapped on filtration experiments, a filtration system using the porous silica beads column is practical.

Keywords: MA recovery; high level liquid waste; prarticle trapping; porous silica beads; filtration column

1. Introduction

Japan atomic energy agency has been developing trivalent minor actinide (MA(III); Am(III) and Cm(III)) recovery from the solution generated by an extraction process in reprocessing of spent nuclear fuel contained high MA [1,2]. An extraction chromatography technology has been applied for the MA recovery. The solution contains many kinds of metallic ions, and they are known to form precipitation [3,4]. Precipitants are the cause of clogging of the extraction chromatography column. Clogging of the column would lead the accumulation of radioactive elements and hydrogen gas. Therefore, these have to be removed from the solution on upper stream of MA recovery operation.

Concentration of nitric acid of the solution will be from 3 to 5 mol/L, and the solution contains MA, fission products (FP) and corrosion products (CP). Some elements in the solution precipitate as a fine particle. Characteristic of the fine particle varies along with the composition of the solution, and the configuration of filtration system depends on the characteristic of the fine particle, therefore it is required to investigate the characteristic.

The sand filtration system is a candidate of a removing system from the fine particle from the solution before MA recovery operation. Because the system is used to treatment of waste water such as natural water and sewage generated by industrial, domestic household, agricultural or commercial activities [5-7]. The system removes the suspended solid such as grid, sand and colloidal materials from a waste water, and this removing materials are same particle size as the fine particle. When the porous silica bead which is used for the extraction chromatography technology is used as the filtrate instead of sand, the waste generated in the filtration system and an extraction chromatography technology will be similar characteristic. Therefore, there isn't needed further a new system and waste stream for removing the fine particle. However, trapping behavior of the fine particle by porous silica bead hasn't been investigated. It is needed to evaluate possibility of trapping the fine particle by the porous silica bead.

In this study, a fine particle trapping system was developed and the fine particle removal performance of the system was experimentally evaluated.

2. Method

2.1. Experiments of filtration system

Bibler et., al. [8] reported the content of fission product and actinide in the fine particle contained in HLLW at the Savannah River site. The tank was made of steel. Main components of the fine particle were Fe and Al, and ^{79}Se, ^{90}Sr, ^{234}U, ^{238}Pu, ^{243}Am and ^{244}Cm were also detected. S. K. Bindal [9] estimated the characteristic of the fine particle by making simulation fine particle. The fine particle generated in pH5.5-6.0 was the mixture of oxide and hydroxide. Density on the simulated fine particle was 1.1 g/cm^3 and particle size of that was 0.005 – 50 μm. E.C. Buck [10] researched the tank in the Hanford site, and revealed that the fine particle was an aggregate of 4 μm fine particles and its composition was Na-Al-Si. Summarized

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Table 1. Summarized characteristic of the fine particle in several high level radioactive solution.

Shape of particle	Aggregate
Particle size	0.005 – 50 μm
Density	1.1 g/cm^3
Main composition	Fe, Al

Table 2. Condition of the bed and simulated fine particles.

		RUN1	RUN2	RUN3	RUN4
Bed	height	15.2 cm	15.4 cm	15.4 cm	14.8 cm
Fine particle	Particle size	0.089 μm	0.12 and 4.5 μm	15 μm	42 μm
	Density	4.0 g/cm^3	3.6 g/cm^3	3.9 g/cm^3	3.8 g/cm^3

characteristic of the fine particle in several high level radioactive solution is in **Table 1**. The fine particle is estimated as a corrosion product from its constituent elements. The corrosion product is originated by tanks and pipes, and it takes in radioactive elements. The characteristic of the fine particle will vary along with characteristic of high level radioactive solution, so the fine particle was simulated on only the particle size. Because there is the big range on assuming the particle size.

Table 2 shows the condition of the bed and simulated fine particle. The simulated fine particles were alumina (Kojundo Chemical Lab. Co., Ltd., Japan), and porous silica bead (particle size is 40-75 μm, mean pore size is 50 nm, FUJI SYLISIS CHEMICAL Ltd, Japan) was selected. The porous silica bead was same bead on previous studies about MA recovery operation [2,11,12].

Twenty-six mL of porous silica bead was mixed to distilled water, and this mixture was filled in a column with 15 mm diameter. After packing of the bead, height of the bed was measured. Alumina as simulated fine particle was dispersed to 5.0 mol/L nitric acid with 1.55 g/L. Four experiments (RUN1-4) were conducted on differ particle size of alumina. Each particle size of alumina were showed in Table 2.

Simulated solution was fed to the column with 90 mL/min and effluent was fractionally collected at every 26 mL. Pressure of the column was measured during the experiment. The experiment was finished when the pressure increased to be 0.5 MPa, or 2 L of simulated solution was supplied. Turbidity and acid concentration of each fraction were analyzed. Weight of trapped powder by the bed was estimated by difference of weight of the bed.

2.2. Zeta potential measurement

Zeta potential measurement was carried out for porous silica beads and alumina powder by electrophoretic analysis utilized Zetasizer Nano ZS (Malvern Panalytical) to discuss the mechanism of the fine particle trapping. The fine particle might be trapped by the bed related to electrostatic force. When there is an attractive force between two materials, the fine particle can be trapped efficiency.

3. Results and discussion

3.1. Filtering experiments

On RUN1, the pressure of the column increased up to 0.20 MPa, Turbidity and acid concentration of the effluent are shown in **Figure 1**. Simulated fine particle with diameter smaller than 0.089 μm passed through the column. When the diameter of porous silica beads was smaller than RUN1, the pass of the column gets to narrow, therefore the bed could catch 0.089 μm particles.

Figure 2 shows the pressure of the column on RUN2. The pressure of the column steadily increased up to 0.5 MPa. The bed caught 0.17 g of simulated fine particle. Turbidity of the effluent was under the detection limit. The column could catch the particles with size of 0.12 - 4.5 μm.

The pressure of the column didn't get to 0.5 MPa on RUN3. The bed caught 2.0 g of simulated fine particle. Turbidity of the effluent was under the detection limit. **Figure 3** shows an appearance of the simulated fine particle layer.

The pressure of the column didn't increase on RUN4. Since turbidity of the effluent was under the detection limit, the simulation fine particle was trapped on the column bed. However, a part of the fine particle settling

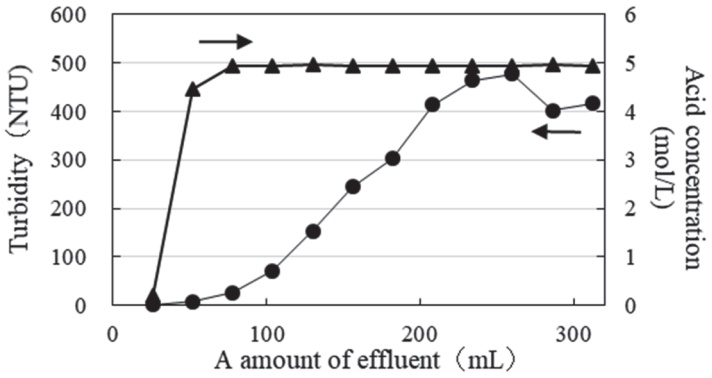

Figure 1. Turbidity and acid concentration of the effluent on RUN1.
NTU (Nephelometric Turbidity Unit): One NTU is turbidity of 1 mg/L of Formazine solution.

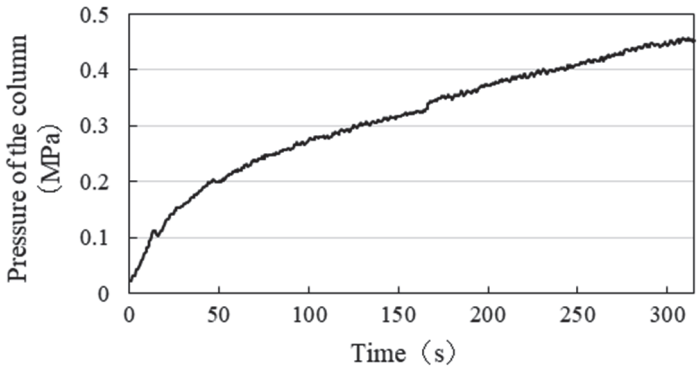

Figure 2. Pressure of the column on RUN2.

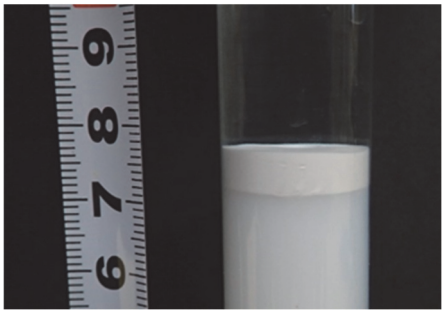

Figure 3. Appearance of simulated fine particle layer on RUN3.

down on a pass by their tare weight was observed, so the > 42 μm fine particle will be remained in a tank and/or settle down in a pass. These results were suggested > 42 μm fine particle doesn't get to cause of the clogging of the filtration column, but investigation of counterplans about sinking fine particle is needed.

3.2. Trapping mechanism

Figure 4 shows the zeta potential of alumina particles and porous silica beads. The zeta potential of alumina particles depended on particle sizes in pH 7-8. The difference of zeta potentials by particle sizes in pH 2 is smaller than in pH 7-8. The zeta potential of alumina particles in 5.0 mol/L nitric acid must be around +60 mV estimated by the dependence of zeta potential on pH. The zeta potential of porous silica beads seems around +1 mV on 5.0 mol/L nitric acid. The zeta potentials of both alumina particle and porous silica bead are considered to be positive values, therefore alumina particles should be trapped physically by the porous silica bead and electrostatic force between particles can be neglected. Consequently, the particle size of the fine particle and the size of flow channel formed in the packed bed dominate the filtering mechanism of this system.

Column experiments revealed that the fine particle with the particle size from 0.12 to 15 μm is cause of clogging of the filtration column filled porous silica beads with the diameter of 50 μm. Zeta potential was suggested the fine particles were trapped with physically. When the fine particles trapped with only physically, the pass of the beads cannot over 0.12 μm. So, these results suggested that the pass of 50 μm beads is under 0.12 μm. When the diameter of porous silica bead is changed, the size of the pass can be controlled, and then the size of passed fine

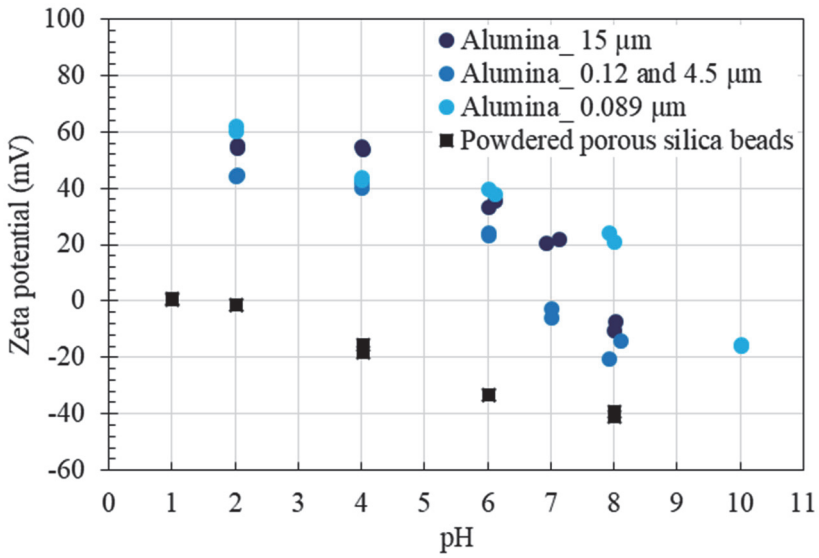

Figure 4. Zeta potential of alumina particles and porous silica beads.

particles can be controlled. Since the diameter of porous silica bead is same diameter as adsorbents of extraction chromatography system, under 0.12 μm fine particles can pass the extraction chromatography system. So, this particle does not lead clogging of the extraction chromatography column. Instead of considering clogging, the MA recovery operation which under 0.12 μm fine particles passes has to be considered.

4. Conclusion

Trivalent minor actinides recovery system has been developing, and it needed a fine particle trapping system from high level radioactive solution for preventive hazardous events. Simulated solution (5.0 mol/L HNO_3, alumina particle) were fed to the column filled porous silica bead which particle size and pore size are same as previous study about MA recovery operation. Column experiments revealed that 0.12 to 15 μm fine particles are cause of clogging of the filtration column. Zeta potential of porous silica bead and alumina were revealed alumina particles is trapped physically by the porous silica bead. Since the > 0.12μm fine particle does not get to cause of clogging of the filtration column, the particle of this size does not get to cause of clogging of the extraction chromatography column, because both column are used same diameter bead. The MA recovery operation which approves passing > 0.12 μm fine particle has to be considered.

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