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Effect of sludge behavior on performance of centrifugal contactor

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Abstract

The effects of sludge behavior on the performance of a centrifugal contactor were investigated. Sludge accumulation during the operation of the centrifugal contactor was observed only in the rotor. Based on the sludge accumulation behavior, the effects of rotor sludge accumulation on the performance of phase separation and extraction were investigated using several types of rotors, which simulated different sludge accumulation levels in the separation area. It was confirmed that rotor sludge accumulation would affect the phase separation performance but not the extraction performance. This can be explained by the structure of the centrifugal contactor, wherein the extraction reaction and phase separation mainly proceed in the housing and rotor, respectively.

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Keywords: centrifugal contactor; sludge accumulation; phase separation; extraction performance

1. Introduction

The Japan Atomic Energy Agency (JAEA) has been developing an annular centrifugal contactor for solvent extraction in spent fuel reprocessing, which allows the mixing of aqueous and organic phases in the annular area and their separation from the mixed phase in the rotor. This is made possible by the strong centrifugal force in the rotor. In a reprocessing plant, several kinds of sludge, such as primary sludge (fine, insoluble residue) and secondary sludge (molybdenum zirconium hydrate, crud), are generated after fuel dissolution. Majority of the primary sludge, which is mainly insoluble residue, can be removed in the clarification process; however, secondary sludge, such as molybdenum zirconium hydrate, which is also generated after clarification, is difficult to be removed from the

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process solution due to its small particle size [1-6]. In the extraction process, such sludge might accumulate in the contactor because of the strong centrifugal force and could affect the phase separation and extraction performance. In the present study, we experimentally evaluated the sludge accumulation behavior in the centrifugal contactor and its effect on the phase separation and extraction performance. In addition, the method for cleaning the accumulated sludge in the centrifugal contactor was studied.

2. Experimental Method

2.1. Equipment

The basic structure of the centrifugal contactor is shown in Figure 1. It comprises a motor, driving unit, rotor, and housing. In this study, centrifugal contactors containing rotors with diameters of 55 and 80 mm were used. In our previous studies, the phase separation and extraction performance were investigated using a rotor diameter of 80 mm, which suggested a maximum throughput of 400 L/h and stage efficiencies of nearly 100% [7] [8].

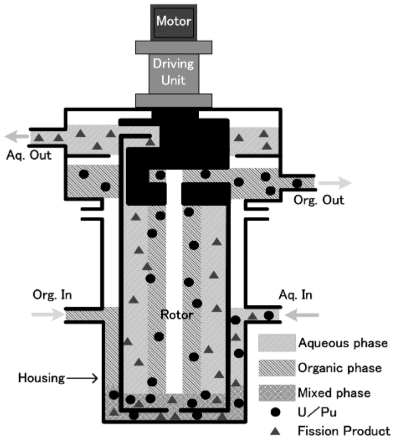


Fig. 1. Schematic of the extraction process in the centrifugal contactor

In the phase separation study, we considered sludge accumulation in several rotors. Schematics of these rotors are shown in Figure 2. Since it was confirmed that sludge accumulation uniformly proceeds in the horizontal direction, sludge accumulation in the rotors was simulated by increasing their wall thicknesses to 10, 30, and 50 vol% of the separation area. In the extraction performance study, the normal rotor and a rotor simulating 30 vol% accumulation were used. The diameter of each rotor was 55 mm.

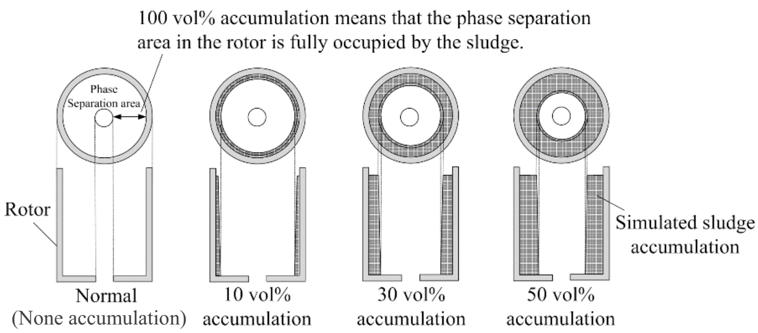


Fig. 2. Rotors simulating sludge accumulation states

2.2. Procedures

At the beginning of the research on the effects of sludge behavior on the performance of the centrifugal contactor, we investigated the sludge accumulation states under the conditions shown in Table 1. These conditions were based on Stokes' law, which can indicate the same critical particle size under the same centrifugal force of 500 G [9].

Table 1 Conditions for investigating sludge accumulation states

Rotor diameter size (mm)	Rotor speed (rpm)	Flow rate (L/h)	Critical particle size (μm)
55	4,000	17.5	0.72
80	3,500	44.0	0.72

In this study, aluminum oxide (alumina; mean particle size = approximately 1 μm, density = 3.99 g/cc) was dispersed in water with a dispersing agent (5 wt% concentration of alumina) was used for the feed slurry (the alumina concentration was 15 g/L). Alumina was selected considering its similar density to molybdenum zirconium hydrate (density = 4.0 g/cc), which is generated as a secondary sludge in a reprocessing plant [1-3]. The particle size of alumina was based on that of sludge after the clarification process in a reprocessing plant [4-6]. The feed slurry was supplied to the centrifugal contactor, and the change of alumina concentration after discharging was measured using a turbidity analyzer. The accumulation weight of sludge in the rotor was evaluated using the following equation:

$$A_{(t)} = \int_{t=T_1}^{t=T_n} Q_{(t)} \left(D_{In(t)} - D_{Out(t)} \right) dt \tag{1}$$

$A_{(t)}$: Sludge accumulation weight (g) in the rotor at time t
$Q_{(t)}$: Flow rate (L/h) at time t
$D_{In}(t)$: Sludge concentration (g/L) in the feed slurry at time t
$D_{Out}(t)$: Sludge concentration (g/L) in the discharged slurry at time t

Furthermore, the capture ratio of alumina for each particle size in the rotor was defined using the following equation:

$$P(D_i) = \frac{S(D_i) - D(D_i)}{S(D_i)} \times 100 \tag{2}$$

$P(D_i)$: Capture ratio of sludge with particle size D_i (%)
$S(D_i)$: Concentration distribution of sludge with particle size D_i in the feed solution
$D(D_i)$: Concentration distribution of sludge with particle size D_i in the discharged solution

Based on the study of sludge accumulation states, several rotors were fabricated to simulate sludge accumulation, and the phase separation performance was investigated to understand the influence of sludge accumulation in the rotors.

In the study of phase separation performance, 0.02 mol/L HNO_3 and 30 vol% TBP/n-dodecane were used as the aqueous and organic phases, respectively. Rotor speeds were controlled at 3,500 rpm for both rotor sizes, and the flow rates of both phases were varied as parameters to evaluate the abnormal flow condition, i.e., the inseparable phase state (entrainment). In addition, residence time was measured using the same solution at the same rotor speed. It was calculated from the ratio of the solution's volume in the housing to its flow rate.

In the study of extraction performance, a uranyl nitrate solution (U: 23 g/L, H^+: 3 mol/L) and 30 vol% TBP/n-dodecane were used as the aqueous and organic phases, respectively. Rotor speed was controlled at 3,500 rpm, and both flow rates were controlled at 60 L/h (total flow rate = 120 L/h). Prior to the extraction of uranium, 3 mol/L HNO_3 was supplied to the centrifugal contactor. Subsequently, the organic phase was supplied. After confirming the stable operation of the centrifugal contactor, 3 mol/L HNO_3 was changed replaced by uranyl nitrate solution. Uranium concentrations in the feed and discharged solution were measured to evaluate the stage efficiency of the centrifugal contactor. In this study, the stage efficiency was determined using the following equation (Murphree efficiency):

$$E = \frac{U_{Aq.feed} - U_{Aq.product}}{U_{Aq.feed} - U_{Aq.product\text{-}in\text{-}equilibrium\text{-}\exp.}} \times 100 \quad (3)$$

E: Stage efficiency (%)

$U_{Aq.feed}$: Uranium concentration in the aqueous feed solution (mol/L)

$U_{Aq.product}$: Uranium concentration in the aqueous discharged solution (mol/L)

$U_{Aq.product\ in\ equilibrium\ exp.}$: Uranium concentration in the aqueous discharged solution after equilibrium (mol/L)

2.3. Analysis

The sludge concentration was determined using a turbidity analyzer (2100AN, Hach), and its particle size was measured using a particle size analyzer (Multisizer 3, Beckman Coulter Inc.) applying the Coulter Principle. The uranium concentration was determined via absorption spectroscopy (V660, JASCO Corp.).

3. Results and Discussion

3.1. Sludge accumulation behavior

The tendencies of the sludge accumulation weight in the rotor with operation time are shown in Figure 3. Sludge concentration in the discharged solution reached that of the feed solution at a time of approximately 300 min for both rotor sizes. The maximum accumulation weights were approximately 400 and 1,500 g in the 55- and 80-mm rotors, respectively.

Figure 4 shows the change in the capture ratio of each rotor with particle size at an operation time of 5 min. Both rotors showed a similar tendency. The supplied sludge was captured in the rotor at the same ratio for every critical particle size regardless of the rotor size.

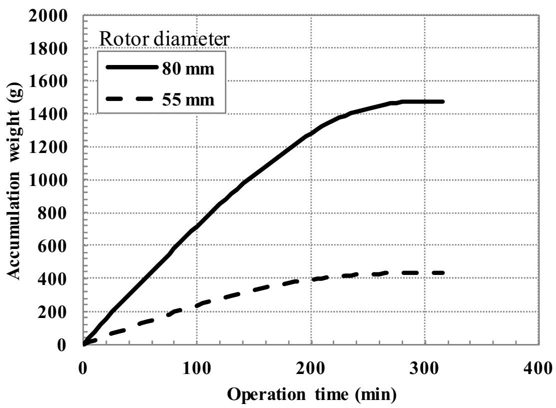


Fig. 3. Change in sludge accumulation weight with operation time

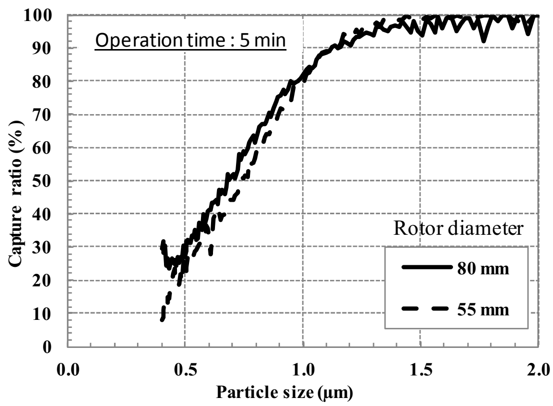


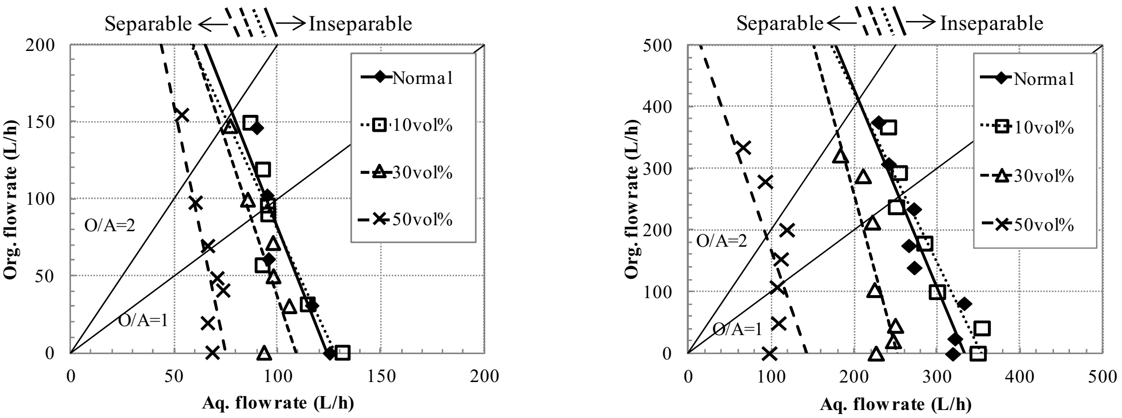
Fig. 4. Change in capture ratio with particle size

During the operation with sludge inclusion condition, sludge accumulation was solely observed in the rotors and there was no accumulation in the housings. This result can be explained by the strong centrifugal force in the rotor, which causes the sludge accumulation. On the other hand, no sludge accumulated in the housing because liquid and sludge sufficiently mix in the housing, preventing sludge accumulation.

From these results, it is estimated that sludge accumulation will mainly affect the phase separation performance.

3.2. Phase separation performance

Figure 5 shows the flow rate region causing entrainment with rotor diameters of (a) 55 mm and (b) 80 mm based on the hydraulic test. These rotors were simulated for several levels of sludge accumulation. The aqueous and organic phases can be separated under the lower flow rate condition for each linear approximation (Figures 5a and 5b). The same tendency was observed for the flow rate causing entrainment with the normal rotor and with the rotor simulating a 10 vol% accumulation. However, the flow rate causing entrainment decreased the sludge accumulation by more than 10 vol%.



(a) 55-mm rotor diameter (b) 80-mm rotor diameter

Fig. 5. Changes in flow rates causing entrainment with sludge accumulation

In the PUREX process, the U and Pu extraction stages must handle the greatest volume of sludge. Since the O/A ratio, which is the flow rate ratio of the organic phase to the aqueous phase, is controlled at approximately 1 or 2 in the extraction stages of the PUREX process, the flow rates causing entrainment at O/A = 1 and 2 were measured. Figure 6 shows the decreasing ratio of the inseparable phase condition, which can be defined as the percentage of the flow rate causing entrainment for each rotor simulating sludge accumulation to that of the normal rotor. The decreasing ratio of the inseparable phase condition can be ignored for sludge accumulations of up to 10 vol%. However, the ratio decreases for sludge accumulations of more than 10 vol%, which is more significant for larger rotor sizes. This is because the rotor diameter would be made smaller by the sludge accumulation, and the effects of the sludge accumulation would increase with rotor size at a given sludge accumulation ratio.

The flow rates causing entrainment were also calculated using a theoretical approach, which could be determined by the relation between the residence time of solutions (ΔT_m) and separation time (ΔT_s) in the rotor (Figure 7). In the calculation, it is defined that the organic and aqueous phases can be separated when ΔT_m determined from the flow rate (Q) and phase separation length (L) is smaller than ΔT_s, which can be determined by the centrifugal force (a). Figure 8 shows the change in flow rates causing entrainment, which showed a similar tendency both experimentally and theoretically. As shown in Figure 8, it can be concluded that the flow rate causing entrainment can be estimated from the decrease in rotor diameter, considering that the change in sludge accumulation is more significant for larger rotor sizes.

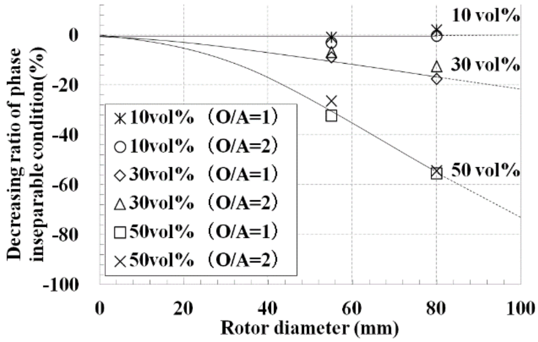


Fig. 6. Influence of sludge accumulation on phase separation performance

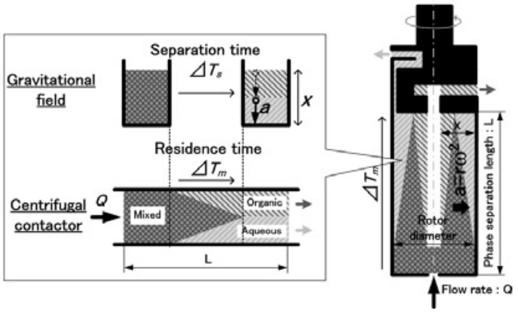


Fig. 7. Image of phase separation calculation.

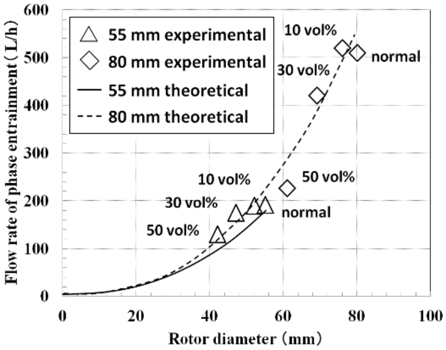


Fig. 8. Change in entrainment flow rate

From these results, the acceptable sludge accumulation ratio for phase separation depends on the rotor size and flow rate at the same rotor speed.

3.3. Extraction performance

Figure 9 shows the stage efficiencies in the extraction performance tests using the normal rotor and the rotor simulating 30 vol% sludge accumulation. The stage efficiencies showed no change against operation time and same values regardless of the sludge accumulation.

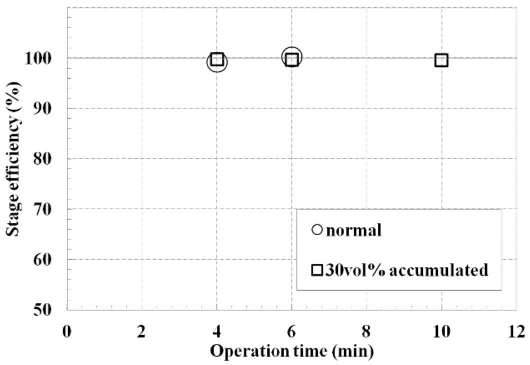


Fig. 9. Influence of sludge accumulation on extraction performance

From these results, sludge accumulation in the centrifugal contactor influenced the phase separation performance but not the extraction performance. This tendency is expected to depend on the centrifugal contactor structure wherein the extraction reaction and phase separation mainly proceed in the housing and rotor, respectively. The aqueous and organic phases, which contain the sludge, are uniformly mixed in the housing. Therefore, the effect of sludge on the mass transfer between phases is not very significant. In addition, as shown in Figure 10, the residence time of each phase in the housing is unaffected by the sludge accumulation in the rotor; in this extraction condition (total flow rate = 120 L/h), both rotors have a similar residence time of approximately 4 s. Therefore, sludge accumulation in the rotor does not affect the extraction performance under the separable phase condition.

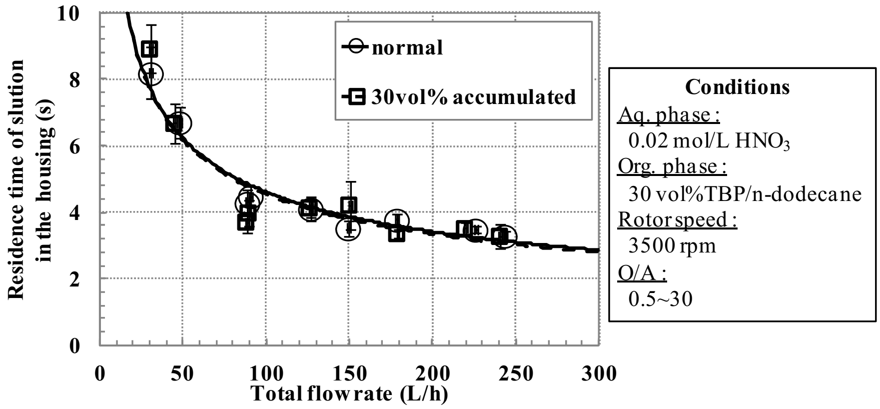


Fig. 10. Influence of sludge accumulation on residence time in the housing

To avoid the effects of rotor sludge accumulations of more than 10 vol% on the phase separation, we have attempted to develop a cleaning method for the sludge accumulated in the rotor. It was confirmed that supplying a washing solution (in this study: water) to the centrifugal contactor and maintaining the rotor rotation at a low speed for a few hours could be an effective cleaning method. The improvement and optimization of cleaning methods, such as the application of a spray nozzle in the rotor, are now in progress.

4. Conclusions

In this study, we investigated the effect of sludge behavior on the performance of centrifugal contactors. The sludge solely accumulated in the separation area of the rotor in the centrifugal contactor. Several types of rotors, which simulated different sludge accumulation levels in the separation area, were fabricated, and the effects of sludge accumulation on the phase separation and extraction performance were evaluated. The sludge accumulation of more than 10 vol% affected the phase separation performance; however, no significant effect on the extraction performance was observed. This tendency is expected to depend on the structure of the centrifugal contactor, wherein the extraction reaction and phase separation mainly proceed in the housing and rotor, respectively.

To prevent excess sludge accumulation during long operation times of the centrifugal contactor, it will be necessary to develop an appropriate cleaning method for the sludge accumulated in the rotor. Several effective cleaning methods have been tested and will be improved and optimized in the future.

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