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**ARTICLE**

# Development of high performance clarification system for spent MOX fuel reprocessing

Masayuki Takeuchi [a*], Takeshi Takata [a], Keita Saito [b] and Takahiro Chikazawa [b]

[a] *Japan Atomic Energy Agency, 4-33 Muramatsu, Tokai-mura, Naka-gun, Ibaraki 319-1194, Japan;*
[b] *Mitsubishi Materials Corporation., 1002-14 Mukohyama, Naka-shi, Ibaraki 311-0102, Japan*

Control of insoluble sludge from fuel dissolution process is one of the important issues to secure the safety in reprocessing plant operation. The sludge volume from spent MOX fuel reprocessing are more than that from spent $UO_2$ fuel reprocessing, so it is required to improve the sludge recovery performance of clarification stage. In order to achieve the higher sludge recovery, a new system with centrifugal device and filter unit has been developed for spent MOX fuel reprocessing. Most of sludge are recovered by centrifugal clarifier which is applied to Rokkasho reprocessing plant in Japan, and the remaining sludge is perfectly recovered by the following filter unit which is a back-up for the recovery of sludge with small diameter and low density.

In this study, over-all clarification performance of an integrated system was evaluated on engineering scale as parameters of sludge type, sludge concentration in feed solution and rotation rate of bowl in centrifugal device. As results, total sludge recovery rate of more than 99.5% was achieved in all test conditions and this new clarification system showed excellent clarification performance. The sludge recovery rate of centrifugal device was influenced by test conditions and was 87 ~ 98%. It was confirmed the recovery rate was improved with higher bowl rotation speed and lower viscosity of feed solution. This study showed the integrated technology is one of the promising clarification systems for spent MOX fuel reprocessing to improve the performance of sludge recovery greatly.

***Keywords: reprocessing; clarification; centrifugal clarifier; filter; sludge***

## 1. Introduction

Volume of insoluble sludge from spent MOX fuel is generally more than that from spent $UO_2$ fuel because platinum group elements etc. in fission products are increased owing to increase of plutonium content in spent fuel [1]. Accumulation of sludge causes flow inhibition of process solution, blockage in pipes and equipment, performance degradation of heat transfer devices and formation of third phase in solvent extraction process. Thus, in spent MOX reprocessing, it is more important to improve the sludge recovery performance from dissolver liquor in clarification process. Typical components of sludge in PUREX process include platinum group alloy [2-4] and zirconium molybdenum hydrate (ZMH) [5-7]. However, the formation mechanism of those chemical species is quite different each other. The platinum group alloy is contained in the sludge as undissolved species of spent fuel in hot nitric acid solution, on the other hand, ZMH is a poorly soluble species generated from dissolved species in hot nitric acid solution by chemical reaction. In particular, growth of ZMH crystal is significant as characteristics of insoluble particles. It contributes to adhesion inside heat transfer device including dissolver [6] and plutonium is accompanied in the crystal as Zr-Pu molybdate [8-10]. However, it would be not easy to separate the ZMH efficiently by centrifugal device because it has fine particle and low density. Thus, the behavior of insoluble sludge in spent fuel reprocessing is complicated, so we have developed a simulation tool to understand the material balance and the formation mechanism [11]. In our thermodynamic simulation tool, especially, the precipitation amount of ZMH was accurately simulated with evidence of experimental data in simulated high level liquid waste solution.

High performance clarification system with centrifugal clarifier and filter unit has been developed for spent MOX fuel reprocessing in Japan Atomic Energy Agency (JAEA). The concept of new clarification system is shown in **Figure1**. In the new system, most of sludge are recovered from the dissolver liquor by centrifugal clarifier and the remaining sludge is collected by the following filter unit. The target of sludge recovery rate on this high performance clarification system is 100%. Generally, a centrifugal clarifier has some advantages like treatment of a large amount of sludge and lower maintenance, but it is not easy to recover sludge with small size and low density under centrifugal force. So, the filter unit is required as back-up of the centrifugal clarifier. It has a disadvantage on filter maintenance, but load of maintenance can be reduced

*Corresponding author. E-mail: takeuchi.masayuki@jaea.go.jp

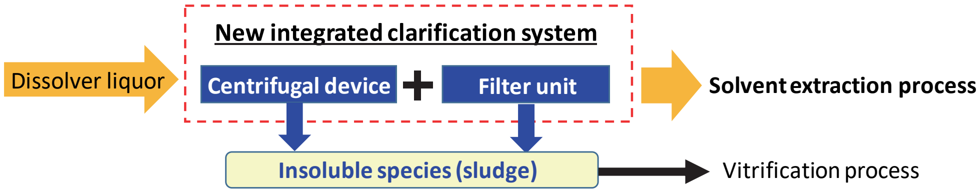


Figure 1. Concept of high performance clarification system.

when the clarification performance of centrifugal clarifier is higher. In previous study, improvement of pulse filter in Tokai reprocessing plant was discussed [12]. It has high sludge recovery ratio more than 96% but corrosion resistance in nitric acid solution should be considered because of metal filter. So, ceramic filter was selected as desirable material with high corrosion resistance in nitric acid solution. In this study, engineering test device of the integrated system was prepared to discuss over-all clarification performance, and engineering clarification tests using some simulants of insoluble sludge were performed to evaluate a prospect for practical use.

## 2. Experimental

### *2.1. Test device*

A clarification test equipment consists of centrifugal device and filter unit including tanks, pumps, pipes. The model of centrifugal test device is AF510SP produced by Tomoe engineering Co. Ltd. in Japan. The throughput is maximum 300 L/h and rotation speed is maximum 6,000 rpm. Appearances of centrifugal device is shown in **Figure.2 (a)**. It is mainly consisted of bowl, driving unit, casing, pipes and pumps. The diameter and height of bowl is 240 mm and 460 mm, respectively. It has 16 bars of baffle plate toward axis direction to grip the collected sludge inside the bowl. Feed solution was put into from bottom of bowl and the sludge was caught at inside wall of bowl by strong centrifugal force. **Figure 2 (b)** shows appearances of rotation unit including bowl in the centrifugal device.

The ceramic filter unit is installed as back-up of the centrifugal device in this system and includes filter, pumps, tanks, compressor. The over view of filter unit device is shown in **Figure3 (a)**. It was manufactured by Noritake Co. Ltd. in Japan. The alumina filter was selected from the viewpoint of corrosion resistance in nitric acid solution and fine pore size. A fine sludge unclarified by centrifugal

a b

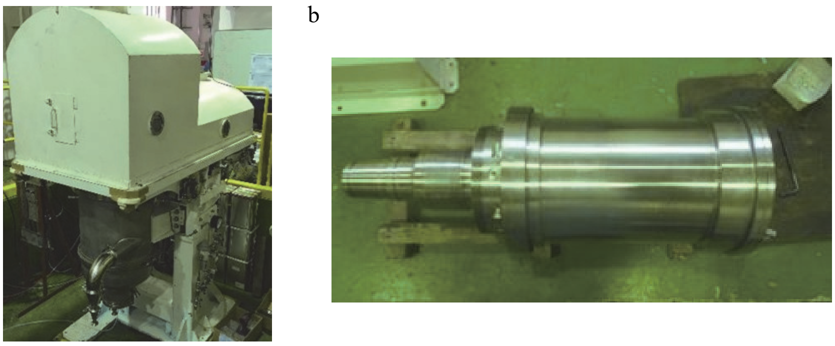

Figure 2. (a) Centrifugal clarifier test device, (b) Bowl and diving unit set-up.

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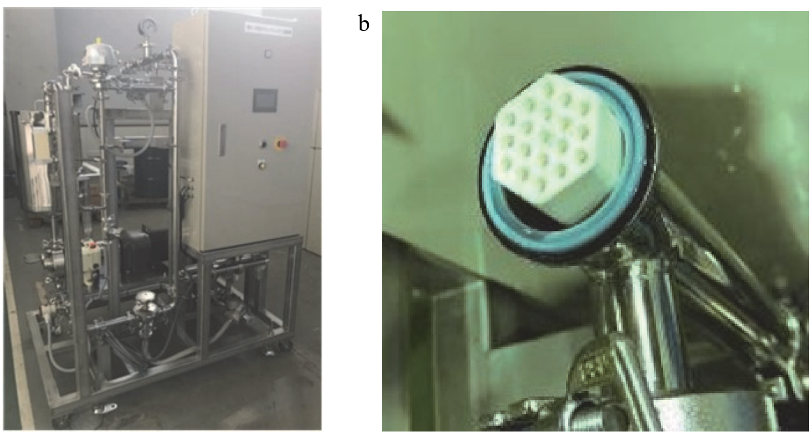

Figure 3. (a) Filter test device, (b) Ceramic filter in unit.

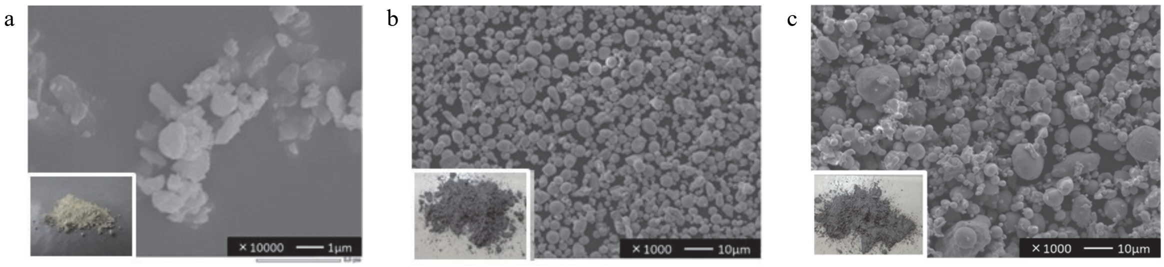


Figure 4. Micro observation results of simulated sludge (a) ZMH, (b) SUS with smaller particle size, (c) SUS with larger particle size.

Table 1. Clarification test conditions.

| Run No. | Simulated sludge | Sludge conc. in feed solution (g/L) | | Solvent | Operation conditions | | |
|---|---|---|---|---|---|---|---|
| | | SUS | ZMH | | Rotation speed (rpm) | Flow rate (L/h) | Test time (min) |
| 1 | ZMH | 0 | 5 | Water | 3000 | | |
| 2 | SUS*+ZMH | 5 | 5 | 44wt% Gly.*** | 3000 | | |
| 3 | SUS*+ZMH | 5 | 5 | 44wt% Gly.*** | 1500 | 100 | 30 |
| 4 | SUS*+ZMH | 7.5 | 7.5 | 44wt% Gly.*** | 3000 | | |
| 5 | SUS**+ZMH | 5 | 5 | 44wt% Gly.*** | 3000 | | |

*SUS powder with average grain size of 8 μm, ** SUS powder with average grain size of 4 μm, ***glycerin solution

device must be separated by the filter unit. Appearance of ceramic filter in the unit is shown in **Figure 3 (b)**. The feed solution was gone through some holes as observed in the Figure3 (b) and filtrated in fine layer with pore size of 0.2 μm by shutting outlet side. Differential pressure is increased with filtration time of sludge and finally back-washing of the ceramic filter is required to recover the filtration performance. So, trend of differential pressure is more important to discuss the maintenance performance and was also researched in this study.

### *2.2. Test conditions*

Five test conditions were selected for the engineering clarification test as parameters of viscosity of feed solution, type and concentration of sludge, rotation speed of bowl in centrifugal device to discuss each effect on the performance of sludge recovery. The powders of stainless steel with average grain size of 4 μm and 8 μm and zirconium molybdenum hydrate with average grain size of 1 μm were prepared as simulant of insoluble sludge in this study. The ZMH is representative sludge component in reprocessing and the stainless steel powder was selected for the simulant of sludge with high density such as shearing chips and platinum group. The ZMH particles were synthesized by KOJUNDO CHEMICAL LABORATORY Co..Ltd. and the stainless steel particles were prepared by EPSON ATOMIX Corporation. **Figure 4** shows appearances of sludge samples for this study. The agglomeration of ZMH particles can be observed from the result. The simulant of sludge was put into water or glycerin solution for preparation of feed solution. The glycerin concentration was controlled as 44 wt% to simulate the assumed viscosity (3.71E-03 MPa·s) of dissolver liquor. Flow rate of feed solution and clarification test time was 100 L/h and 30 mins in all conditions. The flow rate and rotation speed of rotor were consistently stable during test time. These test conditions are summarized in **Table 1**.

Weight of collected sludge from centrifugal clarification and filter unit were separately measured after washing and drying to evaluate the recovery ratio. It was calculated according to the Eq. (1). Sludge concentration and grain size distribution in the feed solution and the clarified solutions from centrifugal device and filter unit were measured to understand the behaviour of sludge during the clarification test. In addition, the sludge in clarified solution was also observed by microscope after drying and chemical composition was discussed by Energy dispersive X-ray spectroscopy (EDS) analysis.

$$\textit{Sludge recovery rate (wt\%)} = (W / W_0) \times 100 \qquad (1)$$

*W : Recovered sludge weight by clarification (g),*
*$W_0$ : Sludge weight in feed solution (g)*

## 3. Results and discussions

### *3.1. Recovery rate of sludge on the new integrated system*

Sludge recovery rates of sludge on the integrated clarification system are listed in **Table 2**. The flow rate and rotation speed of rotor were consistently stable during clarification test time. As results, more than 99.5% of sludge in feed solution was recovered by the new system in all conditions and showed excellent clarification performance. The performance of sludge recovery on the

Table 2. Sludge recovery rates in centrifugal and filter treatments

| Run No. | Sludge recovery rate (%) | |
|---|---|---|
| | Centrifugal clarifier (C.C.) | C.C.+ Filter unit |
| 1 | 98.2 | 100 |
| 2 | 96.2 | More than 99.6 |
| 3 | 86.8 | 100 |
| 4 | 96.7 | More than 99.5 |
| 5 | 95.9 | More than 99.5 |

centrifugal clarifier was influenced by variation of the test parameters and it showed the range from 87% to 98%. **Figure 5** shows SEM-EDX result of sludge in clarified solution of Run No.3. It shows only ZMH component in collected sludge from clarified solution after centrifugal treatment was detected. The result means most of SUS sludge with larger density was collected by centrifugal treatment. In addition, nearly all of the remaining sludge in the clarified solution was recovered by the following filter unit. Here, the recovery weight of particle with less than 0.2 μm was estimated from the particle distribution result of clarified solution and conservatively considered as the recovery loss. Actually, most of particles less than pore diameter would be also collected according to the principle of cake filtration. As an example, appearances of feed and clarified solutions after centrifugal and filter treatments in Run No.3 are shown in **Figure 6**.

In addition to sludge in clarified solutions, the remaining sludge in centrifugal device and filter unit was also recovered to understand the over-all behavior of sludge distribution. Total weight of recovered sludge was about 97% of that in feed solution. The uncomfirmed sludge would be still remained in the loop of test device such as pump and tube however washing operation in test equipment was thoroughly carried out. We consider the unconfirmed sludge as mixture of ZMH and SUS powder and it stayed in loop of test device including inside filter. It is recognized the effect of unconfirmed sludge on reprocessing performance is very low because the weight of the unconfirmed one is a few percent of all sludge.

### *3.2. Effect of operation condition of centrifugal device on performance of sludge recovery*

Performance of sludge recovery on centrifugal device depends on the operation conditions. Especially, from the results of clarification test, it was demonstrated the recovery performance was improved with increase of bowl rotation speed. This trend can be understood by the following Eq. (2) derived from Stokes law.

$$v = r\,\omega^2\,(\rho_p - \rho_t)\,D^2 / 18\,\eta \qquad (2)$$

$v$ : *Sedimentation velocity (m/s)*, $r$ : *Radius of bowl (m)*, $\omega$ : *Angular velocity (rad/s)*,
$\rho_p$ : *Particle density (kg/m³)*, $\rho_t$ : *Solution density (kg/m³)*,
D : *Particle diameter (m)*, $\eta$ : *Viscosity (kg·sec/m²)*

The sedimentation velocity ($v$) toward inside the centrifugal device is directly relation to sludge recovery

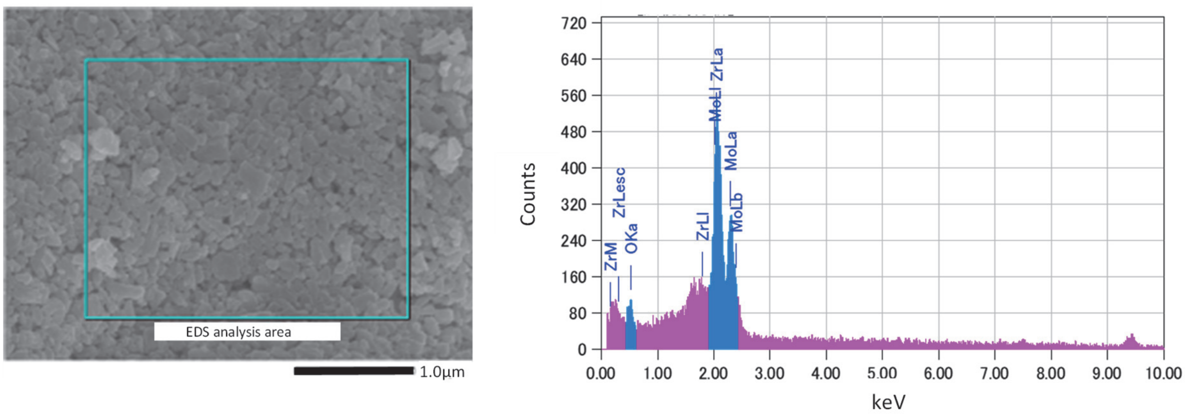


Figure 5. SEM-EDS analysis of recovered sludge from clarified solution after centrifugal treatment (Run No.3).

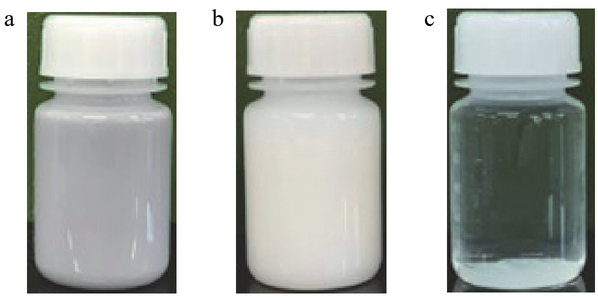


Figure 6. Appearances of (a) feed solution, (b) clarified solution after C.C., (c) clarified solution after filter treatment in Run No.3.

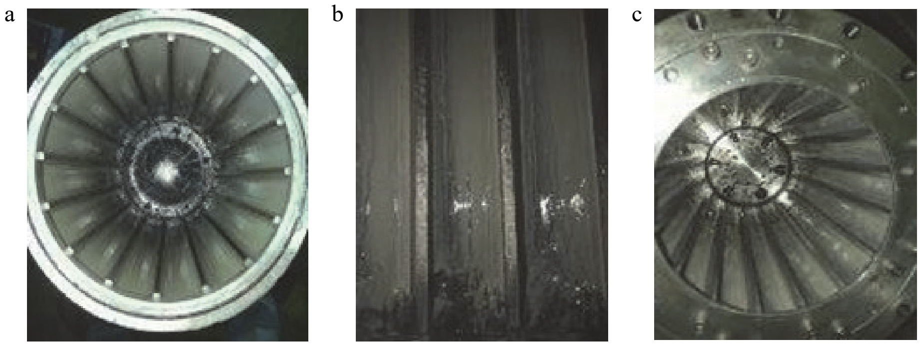


Figure 7. Status of recovered sludge of (a) bottom, (b) side, (c) top in bowl (Run No.3).

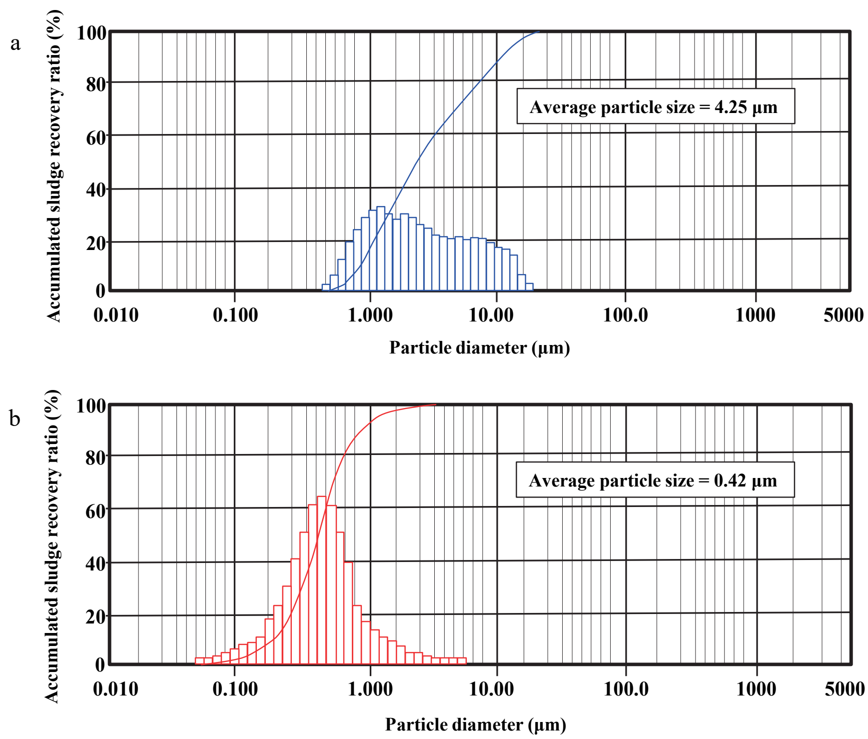


Figure 8. Particle distributions of sludge in (a) feed solution and (b) clarified solution after centrifugal treatment (Run No.3)

rate. Therefore, based on the Stokes law, it is improved by rotation speed of rotor ($\omega$) and particle density ($\rho_p$), but the sludge concentration and particle size of SUS powder did not give severe impact as shown in this study.

**Figure 7** shows distribution of recovered sludge in bowl after the clarification test on Run No.3. It was almost uniformly distributed between baffle plates except the bottom part of bowl because a distance to separate solid component from liquid one needs and the sludge was not recovered at the bottom of bowl. From this sludge distribution in bowl, the effect of sludge accumulation on the stability of driving unit at high rotation speed is quite low. The distribution of the recovered sludge on other test conditions is the same as that on Run No.3. **Figure 8** shows particle size distribution on feed solution and clarified solution after centrifugal treatment in Run No.3. Average particle size in the clarified solution became smaller than that of feed solution, in addition, SUS powder which has higher density than that of ZMH was perfectly recovered by centrifugal device regardless of particle size and test condition. Accordingly, based on the Eq. (2) from Stokes law, it is recognized the average particle size in the clarified solution is much smaller than that of feed solution by collecting most of stainless steel particles by centrifugal force.

### *3.3. Differential pressure in filter unit*

In this integrated clarification system, back-washing operation of the filter unit is essential to maintain a filter performance for a long time however it is desirable to suppress the load of filter maintenance by improving sludge recovery performance on centrifugal clarifier. So, trend of differential pressure with filtration time is important database as standard for the judgement of back-washing operation. **Figure 9** shows transition of differential

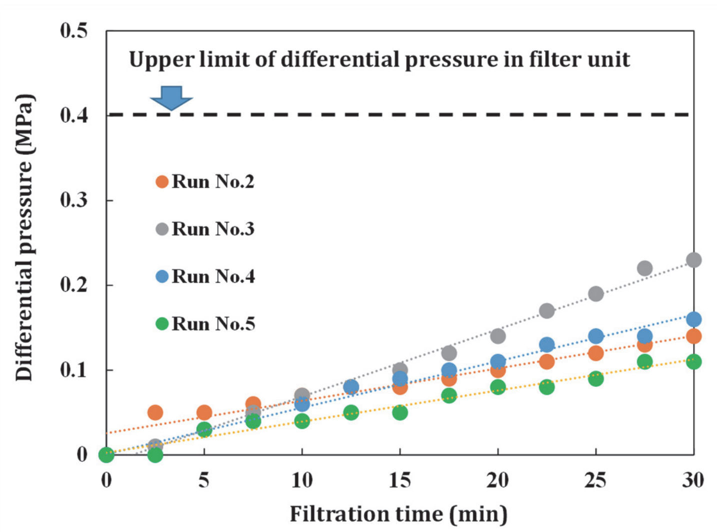


Figure 9. Transition of differential pressure in filter unit during filtration operation.

pressure in filter unit during the filtration test. Here, the trend on Run No.1 was deleted because the sludge recovery rate was quite high and there was slight effect on the variation of differential pressure. As shown in Figure 9, the differential pressure in filter unit was monotonically increased with filtration time and peculiar variation was not observed. So, it is possible to predict timing of back-washing roughly from the trend of differential pressure with filtration time. The increasing rate of differential pressure in Run No.3 was higher than that of other conditions because the sludge recovery rate was lowest on centrifugal device. The back-washing operation was not required through all filtration tests in this study owing to less than 0.4 MPa decided as standard of the differential pressure for back-washing.

## 4. Conclusions

Sludge recovery performance of new clarification system with centrifugal clarifier and filter unit was evaluated on engineering scale. As results, the total sludge recovery rate was more than 99.5% in all conditions and it showed desirably technological prospect as high performance clarification system. In future plan, many types of engineering tests for practical use such as optimization of device structure, clarification condition and back-washing performance of filter system will be carried out to discuss the accuracy and reliability of the integrated clarification system.

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