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Development of High Performance Clarification System for FBR Dissolver Liquor

M.Takeuchi, T.Kitagaki, Y.Noguchi, and T. Washiya

Abstract**—**A high performance clarification system has been discussed for advanced aqueous reprocessing of FBR spent fuel. Dissolver residue gives the cause of troubles on the plant operation of reprocessing. In this study, the new clarification system based on the hybrid of centrifuge and filtration was proposed to get the high separation ability of the component of whole insoluble sludge. The clarification tests of simulated solid species were carried out to evaluate the clarification performance using small-scale test apparatus of centrifuge and filter unit. The density effect of solid species on the collection efficiency was mainly evaluated in the centrifugal clarification test. In the filtration test using ceramic filter with pore size of □0.2μm, on the other hand, permeability and filtration rate were evaluated in addition to the filtration efficiency. As results, it was evaluated that the collection efficiency of solid species on the new clarification system was estimated as nearly 100%. In conclusion, the high clarification performance of dissolver liquor can be achieved by the hybrid of the centrifuge and filtration system.

Keywords**—**Centrifuge, Clarification, FBR dissolver liquor, Filtration

I. INTRODUCTION

ADVANCED aqueous process named New Extraction System for TRU Recovery as "NEXT" has been developed for spent fuel reprocessing of fast breeder reactor (FBR) in Japan Atomic Energy Agency (JAEA). Dissolver liquor includes many kinds of insoluble sludge. It interferes with stable operation of reprocessing plant owing to some troubles such as a blockade in equipments and an entrainment in solvent extraction. Centrifugal clarification is one of the desirable methods for the sludge separation in the dissolver liquor. The technique has been applied for the clarification of dissolver liquor in UP2-800 and UP3 plants at La Hague and it is known that the collection efficiency of insoluble sludge is not enough (about 70%) [1], because it is hard to collect the species with low density in the sludge and small particle size. In case of the FBR dissolver liquor, amount of insoluble species from fission products is significant. In addition, the application of centrifugal contactors is provided for the solvent extraction stage in the NEXT process, so it is desirable to collect the solid species in dissolver liquor under high efficiency for the stable operation. Then, the clarification system with high performance should be required in the advanced aqueous process for FBR reprocessing.

In this study, a new clarification system based on a hybrid of centrifuge and filter unit was proposed to achieve the high separation performance of the insoluble species in this study. The filter system is desirable as a backup of the centrifuge clarification because it can collect the smaller particle size of solid species perfectly. The collection performances of solid species on centrifuge and filtration were evaluated using an experimental scale apparatus and simulated insoluble species. The performance of new clarification system could be demonstrated according to the results of clarification tests.

II. FLOW OF THE "NEXT" PROCESS

The characteristics of the "NEXT" process for FBR reprocessing are a cost merit from a reduction of solvent extraction equipments by introducing a uranium crystallization technique and a recycle of minor actinide such as americium and curium. Some modifications are given in PUREX process from the above viewpoint. The fundamental scheme of NEXT process is shown in Fig.1 [2]-[3]. Firstly, a hexagonal shaped wrapper tube is removed from the FBR fuel assembly by mechanical cutting technique. Cubic boron nitride (CBN) is used as cutting tool for the mechanical cutting and it is demonstrated that the disassembly was successfully carried out using simulated FBR fuel assembly [4]. After the completion of disassembly, a bundle of fuel pins are sheared shortly to dissolve the fuel component in nitric acid solution efficiently. The length of the fuel pins is adjusted to approximately 10mm and the fuel component was more fragmented by the shorter stroke shearing. The fuel in the pins is effectively dissolved according to the fragmentation effect and the heavy metal (HM) concentration in the dissolver liquor is controlled to about 500gHM/L for the following uranium crystallization. More than 70% of uranium in the dissolver liquor is separated as uranyl nitrate hexahydrate (UNH) crystal by cooling crystallization technique [5]. The solvent extraction equipment, which is mainly consisted of centrifugal contactors, can be rationalized by the uranium separation effect. The UNH crystal and mother solution are separated in crystallizer, and uranium, plutonium, and neptunium in mother solution are recovered together in the following solvent extraction process and separated from fission products (FP). Miner actinides (MAs) such as americium and curium are recovered from the raffinite by extraction chromatography method. The amount of

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insoluble species in the NEXT process is significant because of the short stroke shearing and the dissolver liquor with high heavy metal concentration in addition to the high burn-up fuel.

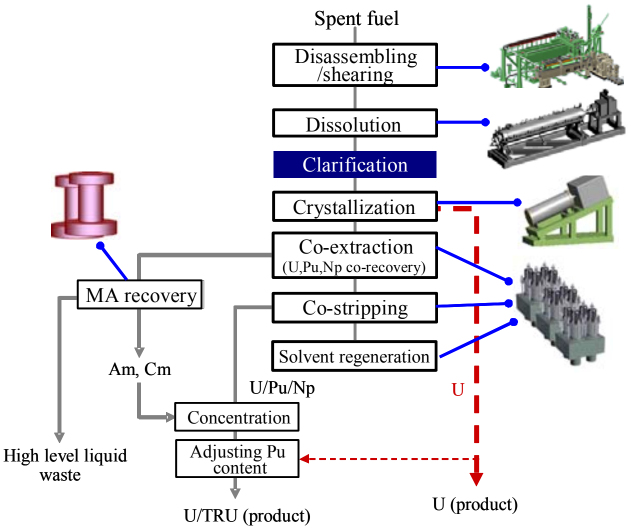

Fig.1 Flow of NEXT process

III. INSOLUBLE SPECIES IN DISSOLVER LIQUOR

Dissolver liquor includes many kinds of insoluble species. It is basically classified to primary insoluble species which do not dissolve in the fuel dissolution process and secondary insoluble species generated according to solution instability after the fuel dissolution. For example, platinum group element from fission products (FPs) and fragments of cladding tube from shearing of fuel pins belong to the primary insoluble species. Some of the platinum group elements such as ruthenium, rhodium and palladium are involved as a multicomponent alloy in the insoluble sludge [6]-[8]. On the other hand, zirconium molybdate hydrate (ZMh) is well known as the secondary insoluble species [9]-[11]. In general, the platinum group element and the cladding tube have relatively high densities, but the ZMh has a low density (approximately 4g/cm^3). Therefore it is generally hard to separate from the liquor by the centrifuge clarification. In order to improve the collection efficiency of the insoluble species on the clarification process, it is quite important to collect the insoluble species with lower density and smaller size of particles efficiently.

Fig.2 shows an example of particle size distribution on insoluble sludge in FBR dissolver liquor [12]. It was prepared by dissolving the spent oxide fuel of experimental fast reactor “JOYO” whose burn-up was 13,800MWd/t. The main particle size of the insoluble sludge is around □1μm and quite small. The collection curve of the particle size distribution for normal distribution is given together in Fig.2. The curve is conservative with regard to the collection of solid species because the bigger particle size was cut down. In this study, the partial collection efficiency every particle size was discussed by considering the correction curve in Fig.2 as a reference of particle size distribution on insoluble sludge from the FBR fuel.

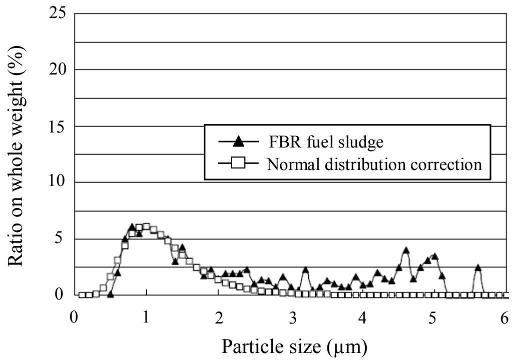

Fig.2 Example of particle size distribution on insoluble sludge in FBR dissolver liquor [12]

IV. CONCEPT OF NEW CLARIFICATION SYSTEM

The advanced aqueous process requires the high performance clarification system of dissolver liquor for FBR fuel. In this study, the clarification efficiency more than 90% at the diameter of 1μm is put as the target of collection performance on the new clarification system. We have proposed the hybrid system of centrifuge and filtration to achieve the target. Fig.3 shows the block flow diagram for the new clarification system. The filtration system is arranged as backup of centrifuges to recover the insoluble species with lower density and smaller particle size.

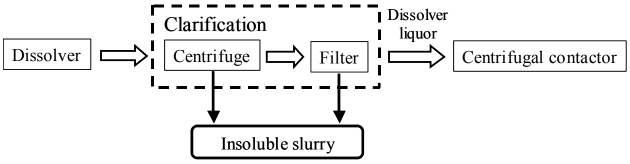

Fig. 3 Equipment structure of new clarification system

The collection performance on centrifuge needs to evaluate the performance of the backup system. In this study, the collection efficiency of solid species on the centrifuge system was theoretically calculated before it is evaluated by the filtration test. The critical particle diameter on the centrifuge is expressed by the stokes law.

$$D_p^{\ 2} = \frac{18\mu_L \ln\left(\frac{r_1}{r_2}\right)}{\omega^2(\rho_p - \rho_L)t} \qquad (1)$$

D_p: critical particle diameter (cm), ω : angle velocity (rad/s),
μ_L: solution viscosity (g/cm·s), t: time for all particle to sediment (s) ,
ρ_p: particle density (g/cm^3), ρ_L: solution density (g/cm^3),
r_1: radius to solution phase (cm), r_2: radius to inside wall (cm)

The partial collection efficiency was calculated according to the equation (1) and the particle size distribution of insoluble sludge in FBR dissolver liquor as shown in Fig.2. Table I shows the main conditions for calculation of collection efficiency. The density effect of solid species on the collection performance was mainly evaluated in this calculation. Fig.4 shows the calculation results of the collection efficiency on the centrifuge. As results, it is obviously found that the density (ρ) effect of the solid species was significant on the collection efficiency. The partial collection efficiency at □1μm was about 93% at ρ=7g/cm^3 and 72% at ρ=4g/cm^3. The total collection efficiency for the whole solid species was about 92% at ρ=7g/cm^3 and 75% at ρ=4g/cm^3, so it is hard to achieve the target of clarification performance using only a centrifuge.

A backup system of centrifuge is required to achieve the target of clarification performance. A filtration technology is desirable as the backup system. It is not necessarily suitable for huge amount of sludge because of the excess of maintenance, but it shows a high performance for smaller particle size of sludge and has some experiences on the nuclear industry [13].

TABLE I
MAIN CONDITIONS FOR CALCULATION OF COLLECTION EFFICIENCY

Bawl diameter	400mm
Rotation	3000rpm
Flow rate	100L/h
Solid species conc.	7g/L
Density of solid species	4g/cm^3, 7g/cm^3
Particle size distribution	Fig.2

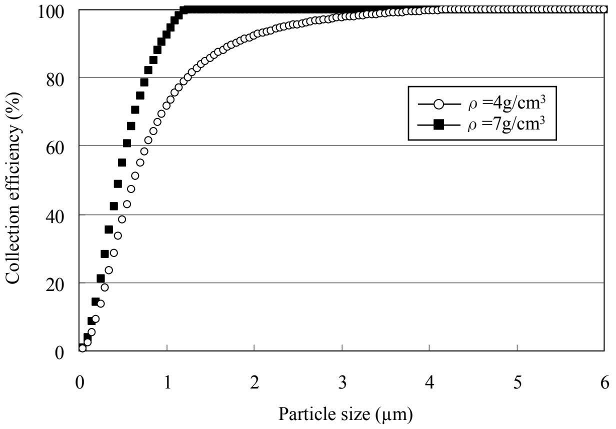

Fig. 4 Partial collection efficiency of solid species with different density

V. EXPERIMENTAL

A. Centrifugal clarification test

The centrifugal clarification test was carried out to demonstrate the estimation of collection efficiency experimentally. Fig.5 shows the schematic diagram of the small-scale centrifugal test apparatus. The centrifuge can operate and separate continuously the solid species from the liquor. The diameter of bawl in the centrifuge was 300mm and the capacity was 4.5L. The rotation condition of centrifuge was 3500rpm to adjust the centrifugal force to about 2000G as well as the calculated condition in Table I. A glycerin solution with solid species was used for the test solution. The glycerin was added to correct the density and viscosity of the test solution for that of actual dissolver liquor. It was assumed that the density of the actual dissolver liquor is 1.85g/cm^3 and the viscosity is 3.71×10^{-3}Pa·s, so the glycerin concentration was adjusted to around 50% in the test solution. Iron oxide and alumina powders were used as the simulated insoluble species to discuss the effect of density of solid species on the collection performance. In case of the iron oxide, the apparent density of solid species was adjusted to 7g/cm^3 by controlling the glycerin concentration. Similarly, the apparent density of alumina in the glycerin solution was adjusted to 4g/cm^3. The particle size distributions of Fe_2O_3 and Al_2O_3 powders are given in Fig.6. The average particle sizes were both approximately □0.9μm and the concentration was adjusted to 7g/L.

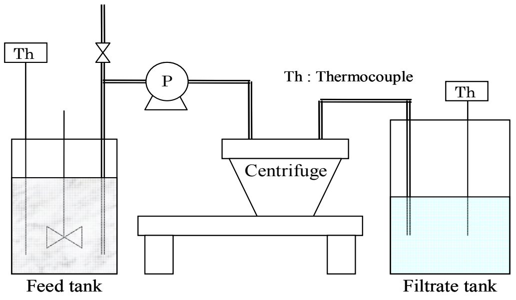

Fig. 5 Structure of small-scale centrifuge apparatus

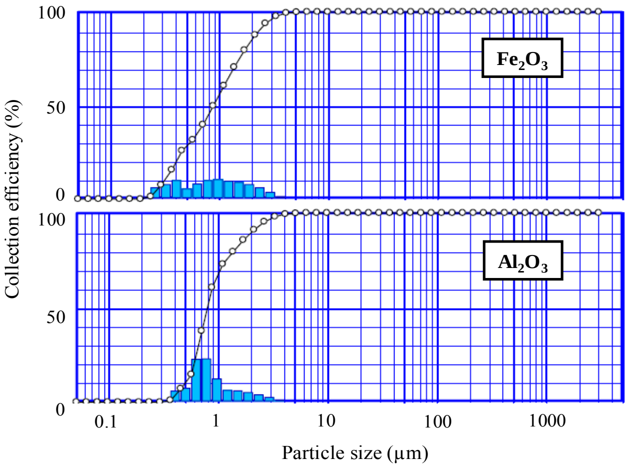

Fig. 6 Particle size distribution of solid species in test solution

The procedure of the centrifugal clarification test is as follows. The test solution was prepared in feed tank. The centrifuge ran and the rotation was controlled at 3500rpm. Next, the test solution was fed at flow rate of 30L/h to control the residence time of solution in the bawl and the solid species were forcibly separated from the test solution under strong centrifugal force. The solid species in solutions after the clarification was collected by filter with pore size of □0.4μm, and the weight was measured to evaluate the concentration of solid species in the filtrate and collection efficiency. The collection efficiency was calculated according to the following equation (2). In addition, the particle size distributions of the collected solid species were measured by laser diffraction type analyzer. The main conditions of centrifugal clarification test are given in Table II.

$$\mathrm{CE} = \left(1 - \frac{C}{C_0} \right) \times 100 \qquad (2)$$

CE : Collection efficiency (%)
C_0 : Concentration of solid species in solution before clarification (g/L)
C : Concentration of solid species in solution after clarification (g/L)

B. Filtration test

The filtration tests were carried out in consideration of the back-up of the centrifugal clarification. The schematic of test apparatus is given in Fig.6. It is mainly consisted of a compressor, a filter element with tube type, and a pressure gauge. The whole filter element is made of alumina. The cross section structure of the ceramic filter is shown in Fig.7. The inner diameter was 7mm and the length was 250mm. The surface of filter film was arranged inside the tube and the surface area was approximately $0.0055\mathrm{m}^2$. The test solution flows through the holes and is extruded from the inside wall through the filter by closing the head of holes as shown in the Fig.7. The ceramic filter is consisted of the surface layer, intermediate layer, and base material made of α-alumina. The filtration performance of solid species is basically controlled by the surface (filter) layer and the pore size is □0.2μm. In a previous study, it was reported that the filter element has a good backwashing performance [13]. The alumina powder was used as simulated solid species as well as the centrifugal clarification test. The performances such as filtration efficiency, permeability, and specific cake resistance were evaluated and the basic filtration mechanism was discussed from the test results.

TABLE II
CENTRIFUGAL CLARIFICATION TEST CONDITIONS

Bawl size	□ 300mm
Rotation	3500rpm
Flow rate	30L/h
Solid species	Al_2O_3, Fe_2O_3
Conc. of solid species	7g/L
Size of solid species	□ 0.9μm (Ave.)
Temperature	Room
Test time	5hr

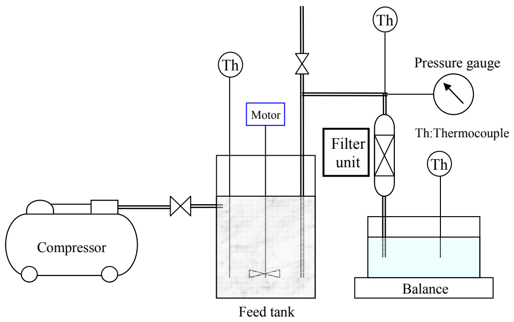

Fig. 6 Structure of filtration test apparatus

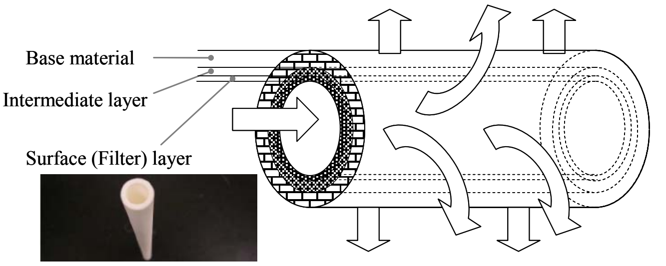

Fig.7 Cross-section of tube type ceramic filter

The solid species in the test solution was sufficiently dispersed by supersonic method and consistently mixed in feed tank. The inside of feed tank was pressurized at 0.12MPa by air and the pressure was kept during the filtration test. The test solution was fed to the filter unit and the total volume of the filtrate was measured with the filtration time. After the filtration, the filtrate was collected for analysis of solid species and the weight of the solid species on the filter was also measured. The filtration efficiency was calculated from the concentrations of solid species in the feed solution and the filtrate. The concentration of solid species in the feed tank was adjusted to 0.7g/L on the assumption that the collection efficiency of the solid species on the centrifugal clarification is 90%. The main conditions of filtration test are given in Table III.Firstly, the specific cake resistance (α), which is an index of flow resistance on cake layer, of the solid species was discussed by a preliminary filtration test. It was carried out using a filter with pore size of 0.1μm as a parameter of the pressure in order not to go through the filter. The filtrate volume and the weight of filter cake were measured under constant pressure condition, and the relationship between the filtration time per unit volume of filtrate (dt/dV) and the filtrate volume (V) was discussed to understand the resistance coefficient (K) from the slope. It was calculated from equation (3) according to Ruth's equation which is generally applied for analysis cake filtration. As a result, the average specific cake resistance (α) of the alumina particle was calculated as 1.4×10^{-12} m/kg from equation (4). The resistance did not change in the range from 0.05 to 0.25MPa.

TABLE III
FILTRATION TEST CONDITIONS

Filter element	Alumina
Pore size	□ 0.2μm (filter layer)
Solid species	Alumina
Solid species conc.	0.7g/L
Size of solid species	□ 0.87μm (Ave.)
Temperature	Room
Pressure	0.12MPa
Test time	5hr

$$\frac{dt}{dV} = \frac{2}{K}(V+V_0) \qquad (3)$$

t: filtration time (s), V: filtrate volume,
V_0: resistance constant, K: resistance coefficient (m^6/s)

$$K = \frac{2\Delta pA^2}{\alpha \ \kappa \ \mu_L} \qquad (4)$$

Δp: pressure for filtration (Pa), κ : cake weight per unit volume of filtrate(kg/m^3)
A: surface area for filtration (m^2), α : average specific cake resistance (m/kg)
μ_L: viscosity of solution (Pa·s)

VI. RESULTS AND DISCUSSION

A. Centrifugal clarification test

The collection performance of solid species with different density was evaluated using a centrifuge apparatus. Fig.8 shows the transition of collection efficiency on the whole particle size with the clarification time. The density effect of the solid species on the centrifugal collection performance was significant as well as the calculation result in Fig.4, and the collection efficiencies of the whole solid species were 93-94% for Fe_2O_3 and 75-77% for Al_2O_3. They showed good coincidence with the calculation data and did not effect on the clarification time.

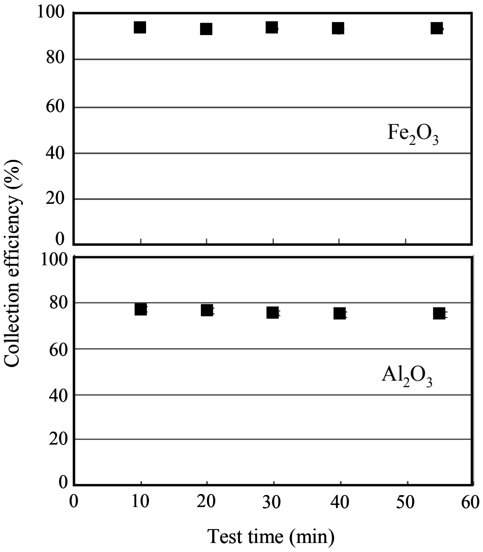

Fig. 8 Transition of collection efficiency with the clarification time

Fig.9 shows the particle size distributions of the solid species in the filtrate. The higher particle size of the solid species was easy to collect, consequently the average diameters of solid species in the filtrate were 0.45µm for Fe_2O_3 and 0.73µm for Al_2O_3 and smaller than that of the feed solution. Fig.10 shows the partial collection efficiencies of every particle size on Fe_2O_3 and Al_2O_3. They are good agreement with the calculated data as shown in Fig.4. The partial collection efficiency was more than 99.9% for Fe_2O_3 by centrifuge in the range of more than □1.5µm. On the other hand, it was more than 99.9% for Al_2O_3 in the range of more than □3µm but dropped for the smaller size. Thus, in case of including the solid species with low density, it is hard to achieve the target of clarification performance by centrifugal clarification.

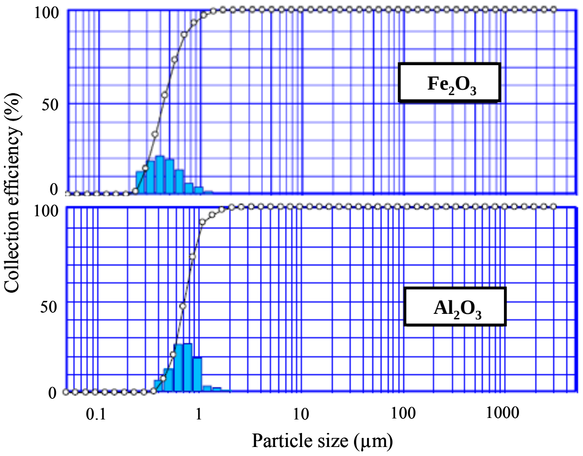

Fig.9 Particle size distributions of the solid species in filtrate

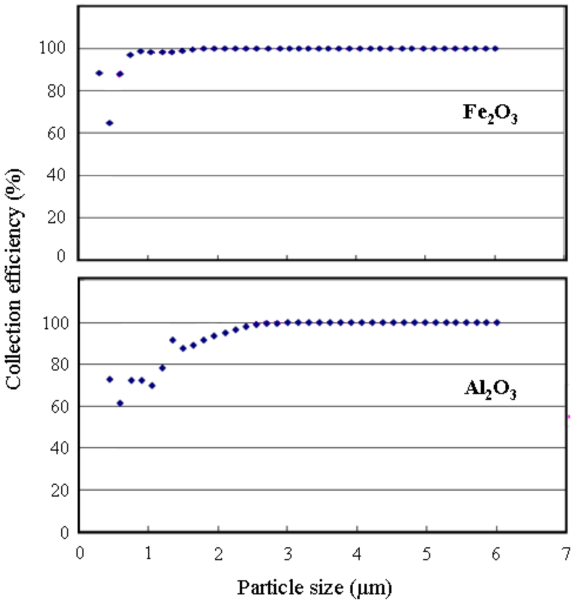

Fig.10 Partial collection efficiency of every particle size

B. Filtration test

The filtration test was performed to discuss the applicability for a backup of centrifugal clarification. Fig.11 shows a permeability of the ceramic filter. The permeate flux increased linearly with pressure and the ceramic filter has good permeability within 0.2MPa. From the results of filtration test under constant pressure condition, the concentration of solid species in the filtrate was less than 0.005g/L, which is the detection limit of analysis, and the filtration efficiency was more than 99%. Fig.12 shows the transition of total filtrate volume with filtration time. The calculation curve, which was evaluated from the average specific cake resistance according to the equation (5), is given together on the figure. The test result of total filtrate volume was higher than that of the calculated. It shows that the solid species makes the cake layer on the filter and the filtration is due to mechanism of cake layer.

$$\Delta p = \alpha \kappa \mu_L u^2 t + K \mu_L u \qquad (5)$$

Δp: pressure for filtration (Pa), κ : cake weight per unit volume of filtrate(kg/m^3)
α : average specific cake resistance (m/kg), μL: viscosity of solution (Pa·s)
u : filtrate volume per unit surface area for filtration (m^3/m^2/s)
t : filtration time (s), K: resistance coefficient (m^6/s)

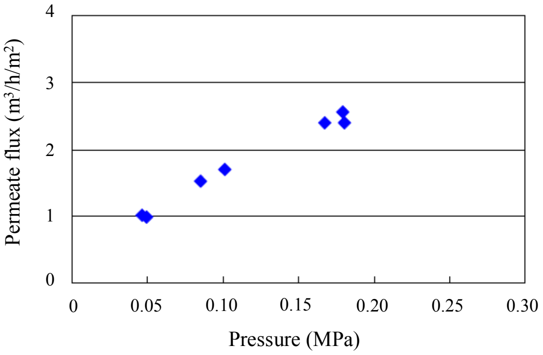

Fig. 11 Permeability of ceramic filter

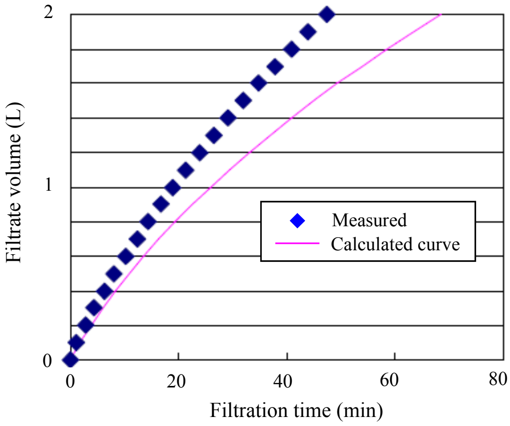

Fig. 12 Transition of filtrate volume with filtration time

Fig.13 shows the relationship between the filtration time and the filtration time per unit volume of filtrate. The resistance coefficient (K) was calculated from the slope as 3.4×10^{-9} m^6/s. according to the equation (3). Moreover, the specific cake resistance (α) was calculated as 8.1 ×10^{11} m/kg according to equation (4). It is nearly same as the calculated value, consequently the filtration is appropriately based on the mechanism of the cake filtration and there is no clogging up in the filter pores within this test conditions. From the results of centrifugal clarification and filtration tests, it is estimated that the collection performance of the solid species, which has particle size as shown in Fig.2, on the new clarification system is more than 99.9% even if the solid species have a low density such as ZMh.

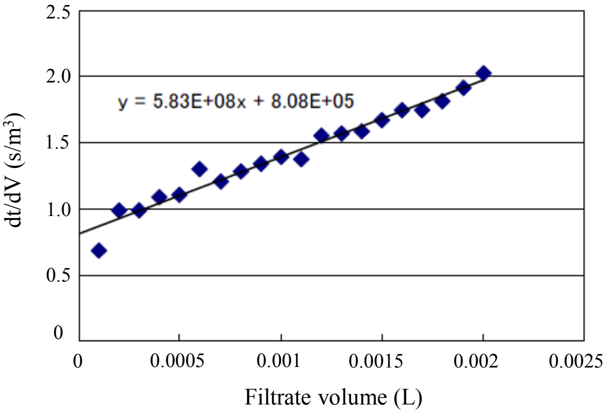

Fig. 13 Relationship between filtrate volume and dt/dV

VII. CONCLUSION

A hybrid process of centrifuge and filtration has been experimentally studied in JAEA as a high performance clarification system for advanced aqueous reprocessing of FBR spent fuel. The collection performance of the whole solid species including lower density and smaller particle based on the new clarification system is more than 99.9% and satisfied the target of collection performance for the advanced aqueous reprocessing. As a next step, we will promote to gain many engineering data such as collection performance and maintenability using a scale-up clarification system to make the new clarification system more practicable.

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