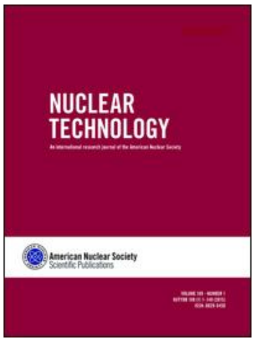

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ENHANCEMENT OF DECONTAMINATION PERFORMANCE OF IMPURITIES FOR URANYL NITRATE HEXAHYDRATE CRYSTALLINE PARTICLES BY CRYSTAL PURIFICATION OPERATION

CHEMICAL REPROCESSING

KEYWORDS: *sweating, melt filtration, crystallization*

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A crystal purification process consisting of sweating and melt filtration was developed to improve decontamination factors (DFs) of fission product impurities from uranyl nitrate hexahydrate (UNH) crystal recovered from a dissolver solution of irradiated fast reactor core fuel. Batch experiments on the sweating and melt filtration processes were carried out at 56 to 80°C. Although the DFs of solid impurities such as Cs and Ba remain the same in the sweating process, those of liquid impurities such as Zr, Nb, Ru, Ce, and Eu were 2.32, 2.40, 2.50, 2.45, and 2.60 at 60°C. On the other hand, the DF of Pu for the UNH crystal slightly increased to 1.25 at 60°C. Because Pu incorporated the UNH crystal in both the solid impurities such as $Cs_2Pu(NO_3)_6$ *and in the liquid impurities, Pu in the liquid fraction was removed by the sweating operation. Decontamination of liquid impurities was effective with sweating time and with a rise in sweating temperature. In the melt filtration process, 0.45- to 5.0-μm–diam filters were used for the separation of the molten UNH crystal. The DF of Ba was approximately ten times as high as the crude crystal with 0.45- to 5.0-μm–diam filters. The particle size of Pu and Cs formed as* $Cs_2Pu(NO_3)_6$ *was quite small. As a proof of this, although the decontamination of Pu and Cs was not effective with a 5.0-μm-diam filter, their DFs rose 2.7 times using a 0.45-μm-diam filter.*

I. INTRODUCTION

Concerning advanced aqueous reprocessing for a fast reactor fuel cycle, U is recovered from the dissolver solution of fast reactor mixed oxide (MOX) fuel as a uranyl nitrate hexahydrate (UNH) crystal by the cooling crystallization method.[1] The dissolver solution of nuclear fuel contains transuranium (TRU) elements and fission products (FPs), which attach to the UNH crystal in the form of liquid or solid. In case of impurity in the form of a liquid, the UNH crystalline particles produced from the dissolver solution of irradiated fast reactor MOX fuel are contaminated by the mother liquor that appears on the surface of or inside the bodies of the crystals. In our previous study,[2] the UNH crystal is washed with HNO_3 solution for its decontamination, and it has been found that the crystal washing was effective for removing liquid impurities on its surface. However, removal of inclusions in the form of liquid within the UNH crystals was not expected by the crystal washing with HNO_3 solution.[3] Removal of inclusion by heating up to as high as the melting point of the crystal has been applied to refine many materials, and this process for the crystal purification is named "sweating."[4] On the other hand, impurity

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TABLE I

Composition of Feed Solution in Crystallization Process

U (g/dm^3)	Pu (g/dm^3)	Ba (g/dm^3)	^{95}Zr (Bq/cm^3)	^{95}Nb (Bq/cm^3)	^{106}Ru (Bq/cm^3)	^{137}Cs (Bq/cm^3)	^{144}Ce (Bq/cm^3)	^{155}Eu (Bq/cm^3)
5.12×10^2	4.60×10^1	7.80×10^{-1}	1.87×10^7	4.16×10^7	3.70×10^8	7.98×10^8	2.82×10^9	1.04×10^8

in the form of a solid was not decontaminated by the crystal washing with HNO_3 solution in the U crystallization process.[3] The melting point of the UNH crystal is 60.2°C and is expected to be low compared to that of solid impurities. If there are differences between the melting points of solid impurities and the UNH crystal, these materials could be separated from each other just by melting the UNH crystal and filtering the solid impurities. The grown crystals are generally purified as a method of increasing purity in the crystallization at an industrial plant. In the U crystallization process, it is proposed that the decontamination factors (DFs) of impurities for the UNH crystal would be improved by the crystal purification process. By removing the FPs, it would bring about reduction in the cost for the recovered U storage and the blanket fuel fabrication due to decreased radiation shielding. For the purpose of further decontamination of TRU elements and FPs, the basic property of crystal purification must be confirmed experimentally.

This paper reports on the purification behavior of the UNH crystal recovered from the dissolver solution of irradiated fast reactor core fuel in batch experiments by the sweating and melt filtration processes. These batch experiments were conducted at the Chemical Processing Facility of Nuclear Fuel Cycle Engineering Laboratories, Japan Atomic Energy Agency.

II. EXPERIMENTAL

II.A. Reagents and Feed Solution

HNO_3 from Junsei Chemical Company was used without further purification. Irradiated core fuel of the fast reactor "JOYO" Mk-III with an averaged burnup of 53 GWd/t and a cooling time of 3 yr was used for the crystal purification experiments. The sheared pieces of core fuel comprising 130 g of heavy metal were dissolved with 325 cm^3 of 8 mol/dm^3 HNO_3 solution at 95°C. The valence of Pu in the dissolver solution was changed to Pu(IV) by NO_x bubbling. The concentrations of U and Pu in the feed solution were 512 and 46 g/dm^3, respectively. The HNO_3 concentration in the feed solution was adjusted to 4.0 mol/dm^3. The $CsNO_3$ solution was prepared by dissolving $CsNO_3$[a] powder into 3 mol/dm^3 HNO_3 solution in order to add the dissolver solution so that the concentration of Cs in the feed solution was 3.1 g/dm^3. The heavy metal concentrations in the feed solution were determined based on the design study for advanced aqueous reprocessing.[5] The composition of the feed solution in the crystallization is summarized in Table I.

II.B. Procedure

The crystallizer was made from Pyrex glass, its capacity was ~200 cm^3, and it had a cooling jacket. The crystallizer temperature was controlled by a thermostat. After the dissolution, the dissolver solution was evaporated, was changed to Pu(IV) in the flask, and then was fed to the crystallizer by a pump for the U crystallization experiment. The feed solution was stirred for 15 min after the addition of the $CsNO_3$ solution at 50°C and was cooled to 4°C for 180 min. The crystalline particles were separated from the mother liquor by a centrifuge.[b] Then, the UNH crystal was washed with 8 mol/dm^3 HNO_3 solution at 4°C in order to remove the mother liquor attached to its surface. The separation of crystal from the wash solution was also implemented by the centrifuge.

Crude crystals of ~14 and 8 g were placed in a 21-mm-diam glass vial in the sweating and melt filtration processes, respectively. In the sweating operation, the UNH crystal is purified by heating it up to its melting point, and the liquid impurities flow along defects and grain boundaries. The glass vials were sealed and were immersed in a thermostat bath, which was maintained at 56 to 60°C for a determined period because the melting point of the UNH crystal is 60.2°C. The crystal samples were taken in the upper part of the glass vial for the analysis. In the case of melt filtration, only the UNH crystals melt in the thermostat bath, and then the solid impurities are filtered from the molten UNH crystal. The melting of the UNH crystal was carried out for 120 min at 70 and 80°C. The molten UNH crystal was sampled by a microsyringe with a filter, and the weight of the unmelted crystal was measured. In this study, 0.45-, 0.8-, and 5.0-μm–diam filters were used for removal of the solid impurities.

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[a] Wako Pure Chemical Industries, Ltd.

[b] H-112, KOKUSAN Company, Ltd.

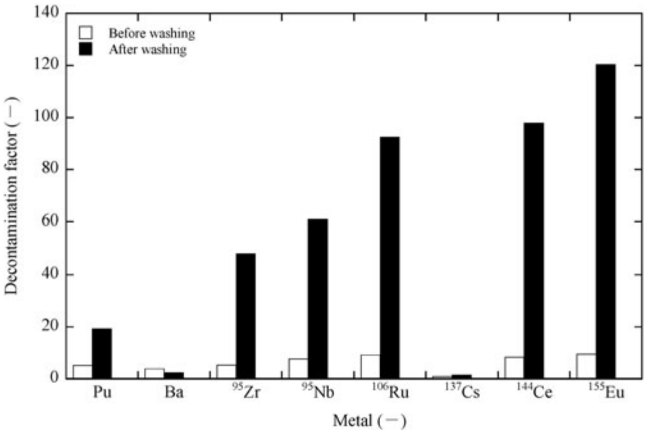

Fig. 1. Influence of crystal washing on DFs of metals for UNH crystal in crystallization process.

II.C. Analysis

Acid-base titration[c] was used for determining the acidities of the dissolver solution. The Pu valence in the dissolver solution was confirmed as Pu(IV) by spectrometry[d] of the ultraviolet-visible region. The concentrations of U and Pu were measured by colorimetry, and low Pu concentration was determined by alpha-ray spectrometry.[e] The concentrations of FPs were analyzed by gamma-ray spectrometry[f] and inductively coupled plasma atomic emission spectrometry.[g] In the case of the UNH crystal, it was measured in a solution prepared by dissolving it into a 3 mol/dm^3 HNO_3 solution.

III. RESULTS AND DISCUSSION

III.A. Crystallization Process

Figure 1 shows the DFs of metals for UNH crystals before and after washing. The DFs of metals for the UNH crystal in the U crystallization process are defined in Eq. (1):

$$\beta_{c,j} = \frac{\dfrac{C_{F,j}}{C_{F,\mathrm{U}}}}{\dfrac{C_{P,j}}{C_{P,\mathrm{U}}}} \quad , \tag{1}$$

where

$\beta_{c,j}$ = DF of metal j for the UNH crystal in the crystallization process
$C_{F,j}$ = concentration of metal j in the feed solution (g/dm^3)
$C_{F,\mathrm{U}}$ = concentration of U in the feed solution (g/dm^3)
$C_{P,j}$ = concentration of metal j in the crystal (mg/g)
$C_{P,\mathrm{U}}$ = concentration of U in the crystal (mg/g).

The UNH crystal obtained from the dissolver solution was washed with HNO_3 solution in order to remove impurities on its surface. The DFs of liquid impurities such as Zr, Nb, Ru, Ce, and Eu increased to 48.0, 60.9, 92.5, 97.9, and 120 by the crystal washing. Liquid impurities remained in the mother liquor on the surface of the UNH crystal and were efficiently removed after the UNH crystal was washed. Among the FPs, Cs and Ba were difficult to separate from the UNH crystal by the crystal washing because of precipitation as $Cs_2Pu(NO_3)_6$ and $Ba(NO_3)_2$, respectively.[3] The crystal washing with HNO_3 solution was not an effective technique for removing the solid impurities from the UNH crystal. Not only was Pu precipitated with Cs as $Cs_2Pu(NO_3)_6$, but it also remained in the mother liquor. Therefore, the DF of Pu relatively increased by the crystal washing by a factor of 19.1. If all Cs in the UNH crystal after the crystal washing was formed as $Cs_2Pu(NO_3)_6$, the precipitation of Pu as $Cs_2Pu(NO_3)_6$ to Pu in the UNH crystal was 38%, and the other 62% of Pu was contained in the UNH crystal as the liquid impurity.

III.B. Sweating Process

III.B.1. Decontamination of Liquid Impurities for Uranyl Nitrate Hexahydrate Crystal by Sweating

The temperature at the center of the crystal bed was found to change, as shown in Fig. 2. The UNH crystal was initially at 24°C and attained the sweating temperature at ~17 min after the immersion of the glass vial in the thermostat bath. The temperature of the UNH crystal could attain equilibrium, and then these experiments were carried out at a constant temperature. Figure 3 shows the appearances of the UNH crystals during the sweating operation. The sweating time was 60 min at 56 to 60°C. The higher the sweating temperature became, the more the amount of U melt increased. Figure 4 shows the relationship between the sweating temperature and the DFs of the metals after 60 min. The DFs of the metals for the UNH crystal in the purification process are defined in Eq. (2):

$$\beta_{p,j} = \frac{\dfrac{C_{b,j}}{C_{b,\mathrm{U}}}}{\dfrac{C_{a,j}}{C_{a,\mathrm{U}}}} \quad , \tag{2}$$

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[c] COM-2500, Hiranuma Sangyo Company, Ltd.

[d] V570DS, JASCO Corporation.

[e] CU017-450-100: detector and NS920-8MCA: pulse-height analyzer, ORTEC.

[f] GEN10: detector and 92XMCA: pulse-height analyzer, ORTEC.

[g] ICPS-7510, Shimadzu Corporation.

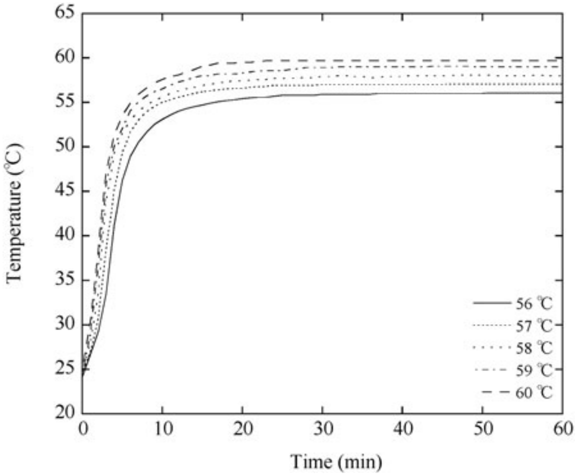

Fig. 2. Temperature change in crystal bed in sweating process.

where

$\beta_{p,j}$ = DF of metal j for the UNH crystal in the crystal purification process

$C_{b,j}$ = concentration of metal j in the crystal before crystal purification (mg/g)

$C_{b,\mathrm{U}}$ = concentration of U in the crystal before crystal purification (mg/g)

$C_{a,j}$ = concentration of metal j in the crystal/melt after crystal purification (mg/g)

$C_{a,\mathrm{U}}$ = concentration of U in the crystal/melt after crystal purification (mg/g).

The DFs of liquid impurities such as Zr, Nb, Ru, Ce, and Eu tended to increase as the sweating temperature increased, but the DFs of solid impurities such as Ba and Cs were constant. On the other hand, since Pu was contained in the UNH crystal not only as a solid impurity but also as a liquid impurity, the DF of Pu increased slightly with the sweating temperature. Figure 5 shows the dependence of the sweating temperature on the yield of the UNH crystal after 60 min. The UNH crystal yield decreased as the sweating temperature was raised. This indicated that high sweating temperature promoted the efficacy of sweating for the UNH crystal. Moreover, a large amount of the melt washed away the mother liquor containing TRU elements and FPs on the surface of the UNH crystal, and the DFs of the liquid impurities for the UNH crystal were increased at high sweating temperature. The relationship between the sweating period and the DFs of the metals at 57°C is shown in Fig. 6. The DFs of the liquid impurities rapidly increased in the initial periods. It can be seen that these DFs are ~2 and became constant after 240 min. The UNH crystals are expected to contain the mother liquor on the surface as the adhered liquid and within the crystal as inclusions. The mother liquor, i.e., liquid impurities in the crystal, were removed by heating up to as high as the melting point of UNH. The DFs of the solid impurities remained the same, and the solid impurities could not be removed by the sweating operation in the batch experiments.

III.B.2. Mechanism of Sweating

A schematic of the sweating mechanism in the UNH crystal purification process is given in Fig. 7. The UNH crystal recovered from a dissolver solution of irradiated fast reactor core fuel contains the mother liquor on its surface as adhered mother liquor and within the UNH crystal as inclusions. The crystal would usually possess some defects attributed to the inclusion during crystal growth and has grain boundaries in the interior of the crystal. These defects continue from the inclusion to the crystal surface. This inclusion in the UNH crystal was mainly liquid impurities such as Zr, Nb, Ru, Ce, and Eu. A heated UNH crystal causes partial melting around the inclusion as liquid impurities and along the defects selectively.[6] The fluid volume in the crude crystal containing the mother liquor increases by heating the UNH crystal. The droplets of the mother liquor and molten UNH crystal within the crystal gradually exude along the way to the surface. The driving force of sweating must be either Ostwald ripening or pressure force arising from the volume increase during the melting.[6] The inclusion is drained away from the inside of the crystal along some defects and grain boundaries in the crystalline particle. In fact, it was reported that an organic crystal had grooves along the directions parallel to the particle after the sweating operation.[7] Although this phenomenon removes the inclusions as liquid impurities in the UNH crystal, the solid impurities are not decontaminated.

III.B.3. Sweating Rate

For the evaluation of crystal purification by the sweating operation, the purification rate coefficient k is generally defined using organic materials, and the equation of the purification rate is obtained with mass balance equations[8]:

$$\frac{dw_s}{dt} = k(w_s^* - w_s) \ , \qquad (3)$$

where

w_s^* = concentration of local equilibrium in the sweating temperature

w_s = concentration of the crude crystal.

Changes of the crystal composition with time can be evaluated by Eq. (3) if the crude crystal is heated in a higher temperature atmosphere than its crystallization temperature. The final composition that the purity approaches is not the equilibrium composition but some

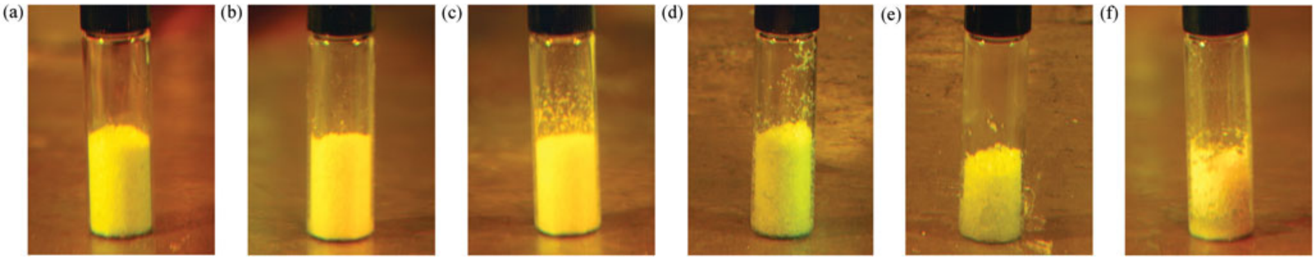

Fig. 3. Appearances of the UNH crystals in sweating process: (a) 25°C, (b) 56°C, (c) 57°C, (d) 58°C, (e) 59°C, and (f) 60°C.

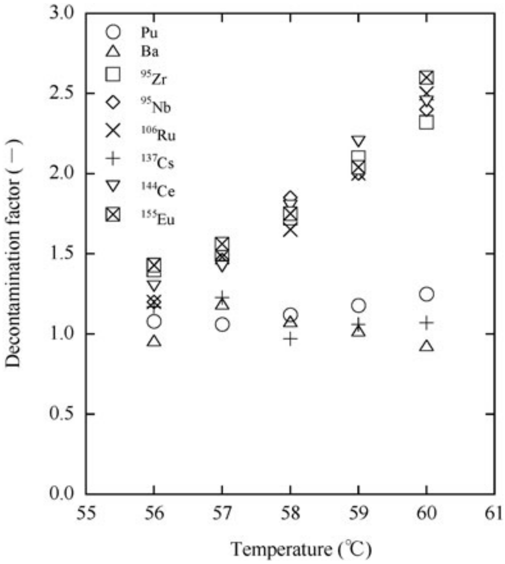

Fig. 4. Influence of sweating temperature on DFs of metals for UNH crystal after 60 min in sweating process.

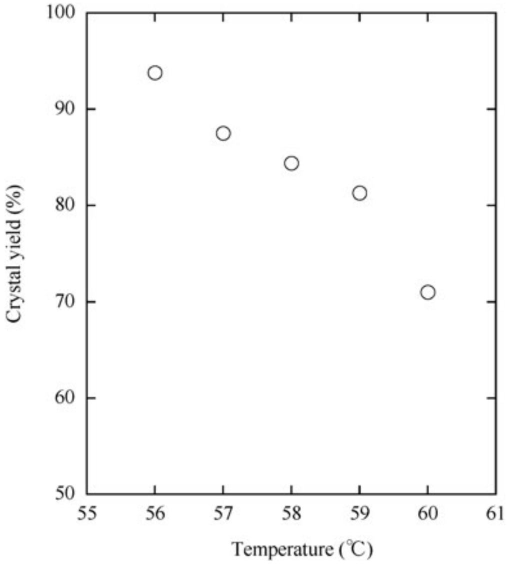

Fig. 5. Relationship between sweating temperature and UNH crystal yield after 60 min in sweating process.

constant value. Therefore, if this value is written as $w_{s\infty}$, then Eq. (4) can be rewritten as

$$\frac{dw_s}{dt} = k_p(w_{s\infty} - w_s) \ . \tag{4}$$

The degree of refining for the reprocessing product is usually expressed as the DFs of the impurities. The final DFs of the metals for the UNH crystals approach constant value $\beta_{p,j\infty}$, and the modified purification rate equation is written as follows:

$$\frac{d\beta_{p,j}}{dt} = k_d(\beta_{p,j\infty} - \beta_{p,j}) \ . \tag{5}$$

With the boundary condition $\beta_{p,j} = \beta_{p,j0}$ at $t = 0$, this gives the changes in the DF $\beta_{p,j}$ for the isothermal sweating process as

$$\beta_{p,j} = \beta_{p,j\infty} - (\beta_{p,j\infty} - \beta_{p,j0})\exp(-k_d t) \ . \tag{6}$$

The purification rate coefficient k_d of the UNH crystal for the metals is summarized in Table II. The purification rate coefficient of the UNH crystal for Pu is smaller than that of the UNH crystal for other liquid impurities. This is the reason why a part of Pu precipitates as $Cs_2Pu(NO_3)_6$ in the course of the crystallization and is incorporated in the UNH crystal during the sweating process.

III.C. Melt Filtration Process

III.C.1. Decontamination of Solid Impurities for Uranyl Nitrate Hexahydrate Crystal by Melt Filtration

The temperature change at the center of the crystal bed is shown in Fig. 8. The temperature of the crystal would be enough to attain equilibrium after 17 min. Figure 9 shows the appearances of the UNH crystals before the melt filtration operation and after 120 min. The lemon yellow UNH crystal at 25°C changed to

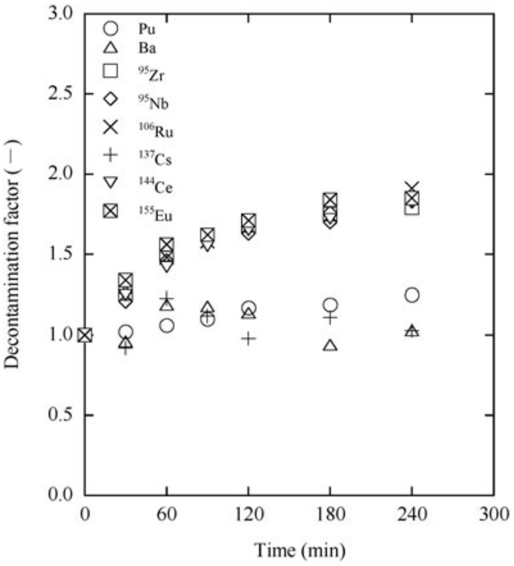

Fig. 6. Influence of sweating time on DFs of metals for UNH crystal at 57°C in sweating process.

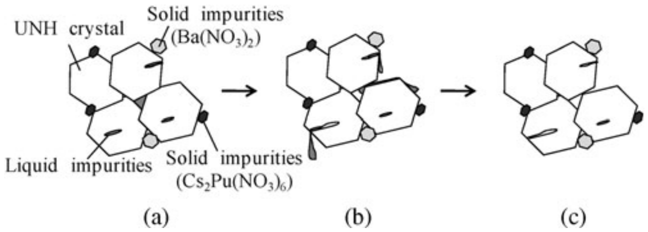

Fig. 7. Schematic of sweating mechanism in UNH crystal purification process: (a) before sweating, (b) partial crystal melting during sweating, and (c) after sweating.

dark green melt at 70 and 80°C after 120 min. At 70 and 80°C in the melt filtration operation, almost all of the UNH crystal was molten, but 23 and 21 wt% of the precipitates of the crude crystal did not melt, respectively. The behavior of the metals was not observed in the difference between 70 and 80°C in the melt filtration process. The DFs of the metals for the UNH crystal in the melt filtration process at 80°C is shown in Fig. 10. In Fig. 10, the DFs of the metals were calculated using Eq. (2). Regardless of the filter mesh size for filtration, the DF of Ba for the UNH crystal was ~10, and Ba was thoroughly removed from the U melt by the melt filtration operation. On the other hand, Pu and Cs show similar behavior, which depends on the mesh size of the filter for separation of the molten UNH crystal. The DFs of Pu and Cs increase approximately 2.7 times from before melt filtration with a filter of 0.45-μm diameter. The molten UNH crystal could not be separated from Pu and Cs using a filter of 5.0-μm diameter. The particle size of the Pu and Cs compound is not measured in these experiments, but the compound is assumed to be <5.0 μm in size and to pass through the 5.0-μm-diam filter. The particle size of $Ba(NO_3)_2$ is ~50 μm and is assumed to be larger than that of $Cs_2Pu(NO_3)_6$. Therefore, Ba tends to be purified in comparison to Pu and Cs in this method.

III.C.2. Mechanism of Melt Filtration

A schematic of the melt filtration mechanism in the UNH crystal purification process is given in Fig. 11. The melting point of a UNH crystal is 60.2°C. When the melting points of the solid impurities are higher than that of UNH, the molten UNH crystal may be separated from the solid impurities with a filter. Judging by the result of thermal analysis,[9] $Cs_2Pu(NO_3)_6$ and $Ba(NO_3)_2$ are assumed to exist in the solid state at the melting point of a UNH crystal. With these properties, the UNH crystal is expected to melt at a slightly higher temperature than its melting point, to be separable from solid impurities by filtration. For the remaining liquid state of the molten UNH crystal, the liquid impurities can be difficult to separate from the U melt by melt filtration.

III.D. Decontamination Performance of Impurities in Crystallization and Crystal Purification Processes

Table III shows the DFs of the metals for the UNH crystal in the crystallization and crystal purification processes in the batch experiments. The DF of Eu showed a maximum of 334 in the whole process. The crystal purification for organic materials by sweating is generally carried out with countercurrent equipment. With a view to confirm the performance of the equipment on UNH purification, bench-scale experiments of the crystal purifier, Kureha Crystal Purifier[10] (KCP), were carried out with a UNH crystal containing Sr and Eu as the liquid impurities or containing Type 304L stainless steel powder as solid impurities.[3] The DFs of the liquid impurities and the solid impurities for the UNH crystal were evaluated as ~50 and 100, respectively, in this purifier. Unlike the batch experiments, these results indicated the KCP worked not only for liquid impurities but also for solid impurities. The KCP also has a heating unit for melting a portion of the UNH crystal, and a crude crystal was washed with molten UNH by countercurrent operation. This would bring about high DFs of liquid impurities in the KCP compared with the batch experiments. Although the solid impurities were not removed from the UNH crystal by sweating in the batch experiments, the DFs of the solid impurities improved in the KCP. The probable reason for it being highly effective for solid impurities despite the results of batch experiments lies in the characteristic of the KCP. The UNH crystal and solid impurities were transferred by a double-screw conveyor, so two kinds of particles that had different densities were anticipated to be separated from each other by gravity

TABLE II

Purification Rate Coefficient k_d of UNH Crystal for Metals at 57°C in Sweating Process

Pu (s^{-1})	Ba (s^{-1})	^{95}Zr (s^{-1})	^{95}Nb (s^{-1})	^{106}Ru (s^{-1})	^{137}Cs (s^{-1})	^{144}Ce (s^{-1})	^{155}Eu (s^{-1})
1.02×10^{-4}	—	2.93×10^{-4}	1.96×10^{-4}	2.05×10^{-4}	—	1.99×10^{-4}	2.97×10^{-4}

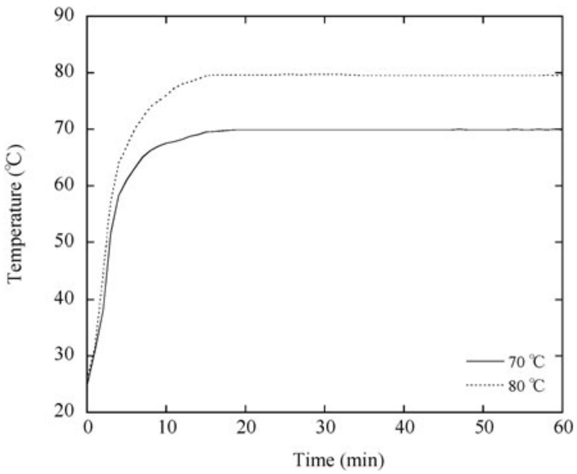

Fig. 8. Temperature change in crystal bed in melt filtration process.

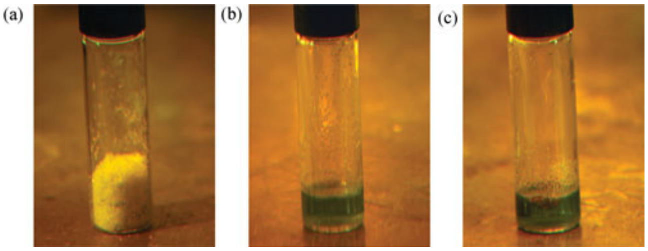

Fig. 9. Appearances of the UNH crystals in melt filtration process: (a) 25°C, (b) 70°C, and (c) 80°C.

and mixing. The DFs of Cs and Ba as solid impurities were not so high in the batch experiments from crystallization to the crystal purification process. However, Cs and Ba might be removed by the countercurrent crystal purifier KCP because of its equipment properties. Meanwhile, the precipitation behavior of Cs depends on the condition of the feed solution in the U crystallization process.[9] Since low concentrations of HNO_3 and Cs avoid the generation of $Cs_2Pu(NO_3)_6$ and Cs exists as the liquid impurity in the U crystallization, further decontamination of Pu as well as Cs is expected by a factor of 50 in the KCP.

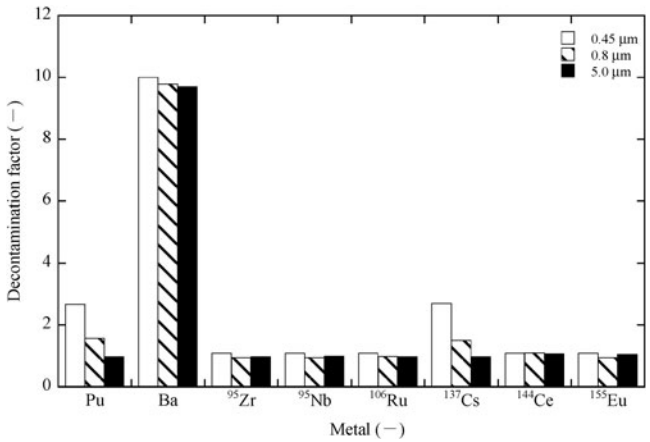

Fig. 10. Influence of filter mesh size on DFs of metals for UNH crystal at 80°C in melt filtration process.

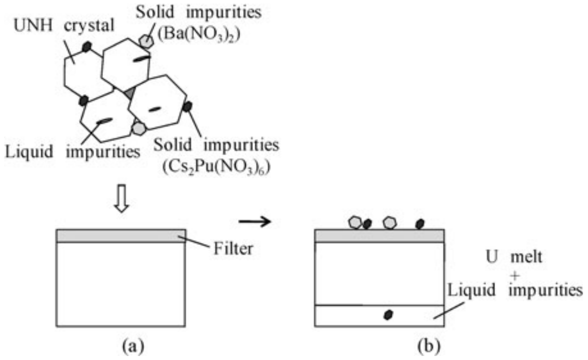

Fig. 11. Schematic of melt filtration mechanism in UNH crystal purification process: (a) before melt filtration and (b) after melt filtration.

IV. CONCLUSIONS

New technology for the crystal purification process has been proposed in order to perform further decontamination of TRU elements and FPs for the UNH crystal. Two crystal purification methods, sweating and melt filtration, were carried out for UNH crystals recovered from the dissolver solution of irradiated fast reactor core fuel.

TABLE III

DFs of Metals for UNH Crystal in Crystallization and Crystal Purification Processes in Batch Experiments

Process	Pu	Ba	^{95}Zr	^{95}Nb	^{106}Ru	^{137}Cs	^{144}Ce	^{155}Eu
Crystallization (after washing)	19.1	2.31	48.0	60.9	92.5	1.50	97.9	120
Sweating (60°C)	1.25	0.93	2.32	2.40	2.50	1.07	2.45	2.60
Melt filtration (0.45 μm)	2.67	11.5	1.09	1.01	1.00	2.70	1.04	1.07
Final product	63.7	24.7	121	148	231	4.33	249	334

The sweating of the UNH crystal was performed at 56 to 60°C in the batch experiments. Although solid impurities such as Cs and Ba remained in the UNH crystals, the DFs of liquid impurities such as Zr, Nb, Ru, Ce, and Eu had a tendency to increase in a high sweating temperature condition. The DF of Eu achieved a maximum of 2.60 in the batch experiments by the sweating operation. Plutonium was incorporated into the UNH crystal in the liquid impurities as well as in the solid impurities, $Cs_2Pu(NO_3)_6$, and was slightly removed from the UNH crystal having a DF = 1.25 at 60°C for 60 min. In the melt filtration process, the UNH crystal could melt at 70 and 80°C, and the molten UNH crystal was relatively separated from the solid impurities with some filtration. The DF of Ba for the U melt was ~10 with 0.45-, 0.8-, and 5.0-μm-diam filters. The DFs of Pu and Cs increased only 2.7 times with a filter of 0.45-μm diameter because the particle size of $Cs_2Pu(NO_3)_6$ is comparatively smaller than that of $Ba(NO_3)_6$. However, the DFs of Pu and Cs will increase to ~250 in the batch experiment by sweating and melt filtration when $Cs_2Pu(NO_3)_6$ does not precipitate in the course of U crystallization.

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