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Flow-sheet study of MA recovery by extraction chromatography for SmART cycle project

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Abstract

Optimization in a flow-sheet of the extraction chromatography process for minor actinides (MA(III); Am and Cm) recovery from high level liquid waste (HLLW) were carried out through batch-wise adsorption/elution experiments on diluted HLLW and column separation experiments on genuine HLLW. Separation experiments using CMPO/SiO₂-P and HDEHP/SiO₂-P adsorbent columns with an improved flow-sheet successfully achieved more than 70 % recovery yields of MA(III) with decontamination factors of Ln(III) > 10³, and a modified flow-sheet for less contamination with fission products was proposed consequently. These results will contribute to MA(III) recovery operations for SmART Cycle project in Japan Atomic Energy Agency which is planned to demonstrate FR fuel cycle with more than 1g of Am.

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Keywords: Extraction chromatography; minor actinides; CMPO; HDEHP

1. Introduction

Japan Atomic Energy Agency is conducting the SmART (Small Amount of Reuse Fuel Test) Cycle project, which consists of recovering all actinides from MOX irradiated fuel, fabrication of MA (minor-actinides: Np, Am, Cm) -bearing MOX fuel and irradiation experiments, to demonstrate FR fuel cycle as shown in Fig. 1¹. In this project, MA(III) (Am and Cm) are planned to be recovered by the extraction chromatography technology from the

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raffinate generated in U/Pu/Np co-extraction process. In the technology, MA(III) are recovered through extraction/back-extraction reactions inside the packed column with adsorbents. The adsorbent is prepared by impregnating an extractant into the support of porous silica particle coated with styrene-divinylbenzene polymer (referred as SiO₂-P)². The project targets more than 1 g of Am recovery from the 4 Joyo irradiated fuel pins, and MOX fuel containing 5% Am will be fabricated.

MA(III) recovery and denitration of a product solution containing U, Pu, Np, Am and Cm will be carried out at Chemical Processing Facility (CPF) of Japan Atomic Energy Agency. The extraction chromatography experiments on genuine HLLW have been carried out in hot cells of CPF in our previous study³, and a flow-sheet employing CMPO/SiO₂-P adsorbent for MA(III) + Ln(III) recovery and HDEHP/SiO₂-P adsorbent for MA(III)/Ln(III) separation must be promising though it requires some modifications to enhance the MA(III) recovery performance. In those experiments, MA(III) + Ln(III) recovery by the 1st column was successfully achieved, however MA(III) selective stripping from the 2nd column was poorer than expected. Consequently, selection of an appropriate eluent for MA(III) from HDEHP/SiO₂-P was revealed to be a main necessary improvements in the flow-sheet. Based on the previous data, optimization in pH of the current eluent is expected to enhance the selective MA(III) elution performance.

A microwave denitration apparatus is installed inside a glove box in CPF, and high decontamination factors of fission products are required to transport the product solution from hot cell to the glove box in the respect of radioactivity restriction of the glove box. Fission products such as ¹³⁷Cs and ⁹⁰Sr are easily decontaminated satisfactory by the 1st column, however decontamination of Ln(III) is one of the most difficult tasks. Amounts of Ln(III) have to be reduced to at least one-hundredth according to the radioactivity restriction, thus the eluent for the 2nd column should be attentively selected to accomplish the Ln(III) decontamination. At present, target decontamination factors for all fission products are set at 100 tentatively. The required recovery yields of Am for the SmART cycle project is more than 30 % considering losses at reprocessing and denitration processes, therefore targeted recovery yields of MA(III) in the extraction chromatography process are set at 50 %.

In this study, in advance with the actual MA (III) recovery operation, optimization in the flow-sheet of the extraction chromatography process through batch-wise adsorption/elution experiments and MA(III) recovery trials on the genuine HLLW using the modified flow-sheet were carried out to estimate the MA(III) recovery yield for the SmART cycle.

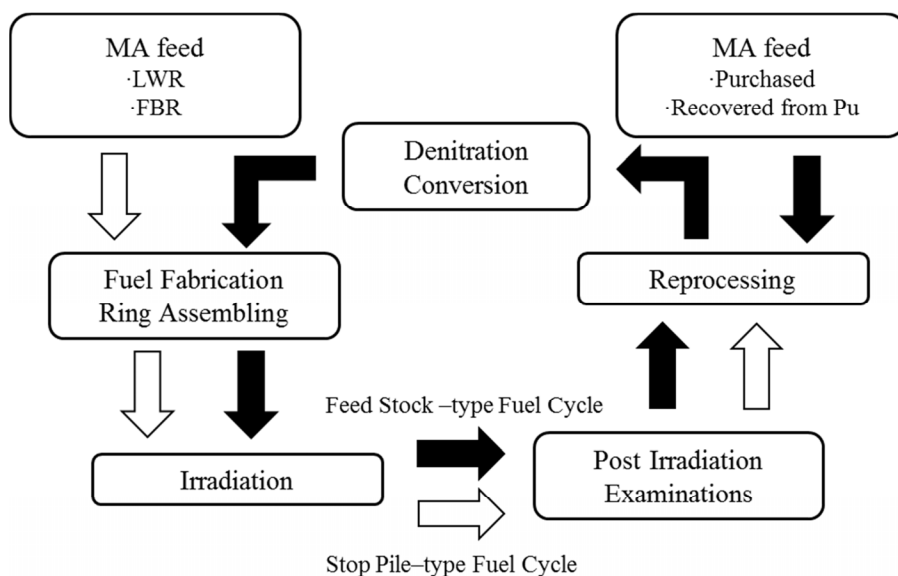


Fig. 1 Concept of the SmART Cycle project

2. Experimental

2.1. Batch-wise adsorption/elution experiments

In order to decide an appropriate pH of the eluent for the HDEHP/SiO₂-P column, Am/Eu separation performance was evaluated through batch-wise adsorption/elution experiments. Genuine HLLW obtained through series of reprocessing experiments on spent JOYO MOX irradiated fuel was diluted to 1000 times with 0.01 M HNO₃ solution. Major composition of the HLLW is shown in Table 1.

50 mL of the diluted HLLW was mixed with 2.5 g of HDEHP/SiO₂-P adsorbent, and then shaken for 3 hours at room temperature. After separation of supernatant solution and the adsorbent, the adsorbent was dried in the atmosphere for 3 days. 4 mL of 50 mM DTPA solutions with various pH were mixed with 0.2 g of the dried adsorbent for elution. After shaking 3 hours at room temperature, supernatant solution was separated from the adsorbent. Gamma radioactivities of ²⁴¹Am, ¹⁵⁵Eu and ¹⁴⁴Ce in the solutions obtained in the adsorption and elution operations were analyzed to evaluate adsorption and elution behavior.

2.2. Column separation experiments on genuine HLLW

CMPO/SiO₂-P or HDEHP/SiO₂-P adsorbents were packed in a glass column with 1cm diameter and 32.5 cm height. The volume of the packed bed; a Bed Volume (BV) was 25.5 cm³. The HLLW used in the above experiments was used as a feed solution for the 1st CMPO/SiO₂-P column. Then, a wash solution (4 M HNO₃) and an eluent (H₂O) were supplied to the column sequentially. The wash solution was supplied to discharge elements which were not adsorbed by the 1st column. Effluent discharged from the column when the eluent was supplied was the interim product of the 1st column, and it was supplied to the 2nd HDEHP/SiO₂-P column without any treatments. A wash solution and an eluent for the 2nd column were 0.01 M HNO₃ and 50 mM DTPA solution, respectively, and pH of the eluent was decided based on the batch-wise experiments.

Temperatures inside the column were controlled to be 323 and 298 K for the 1st and the 2nd column, respectively. Flow velocity of the mobile phase was kept constant at 3.2 mL/min (= 4 cm/min, corresponding to the engineering-scale throughput). Effluents were fractionally collected, and then radioactivity of representative nuclides in them were analyzed to give elution curves. Recovery yields of MA(III) and decontamination factors of fission products were calculated from the chromatograms.

Table 1 Major composition of the HLLW

Species	Concentration
H ⁺	3.7 mol/L
²⁴¹ Am	1.04×10 ⁸ Bq/mL
²⁴⁴ Cm	2.11×10 ⁶ Bq/mL
¹⁰⁶ Ru	1.85×10 ⁷ Bq/mL
¹²⁵ Sb	4.50×10 ⁶ Bq/mL
¹³⁷ Cs	3.81×10 ⁸ Bq/mL
¹⁴⁴ Ce	7.21×10 ⁷ Bq/mL
¹⁵⁵ Eu	1.15×10 ⁷ Bq/mL

3. Results and discussion

3.1. Optimization in the flow-sheet by batch-wise adsorption/elution experiments

Adsorbed amount of ²⁴¹Am, ¹⁵⁵Eu and ¹⁴⁴Ce were 6.10×10⁶, 5.02×10⁵ and 1.94×10⁵ Bq/g-adsorbent, respectively. Elution ratios of ²⁴¹Am, ¹⁵⁵Eu and ¹⁴⁴Ce by nitric acid solution containing 50 mM DTPA were shown in Fig. 2, here

the pH was measured after the equilibration. Since elution of ^{144}Ce was not confirmed at $\text{pH} < 2.8$, the elution ratio for ^{144}Ce was calculated from the lower limit value for the analysis i.e. $3.7 \times 10^2 \text{ Bq/cm}^3$.

The elution ratios of the all nuclides increased with increasing in pH, and that of ^{241}Am showed the maximum among them. These results agree with our previous study. At the lower pH region, high decontamination factors for ^{155}Eu and ^{144}Ce are expected although the elution ratio of ^{241}Am might be small.

Figure 3 shows separation factors calculated from ratios of distribution ratios of the 3 nuclides. We selected $\text{pH} = 2.3$ as an appropriate composition to minimize Ln(III) elution and to maximize both MA(III) recovery yield and the separation factor $\text{SF}_{\text{Am/Ln}}$. The amount of the eluent for the 2nd column was decided to be 15 BV tentatively which is about twice as large as the volume employed in the current flow-sheet. The modified flow-sheets for the column separation experiments are shown in Table 2. After supplying eluents into the columns, other wash solutions were supplied to wash out the residual elements.

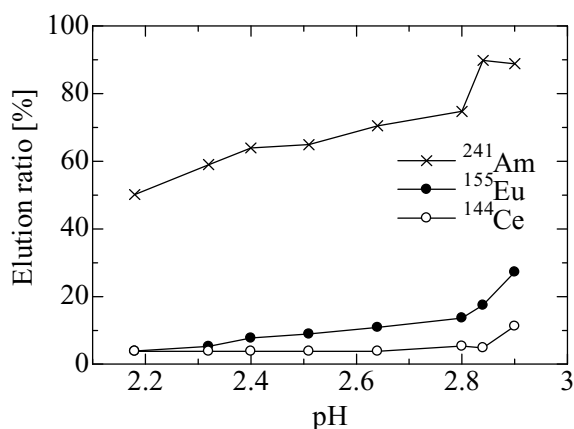


Fig. 2 Elution ratio from HDEHP/SiO₂-P adsorbent

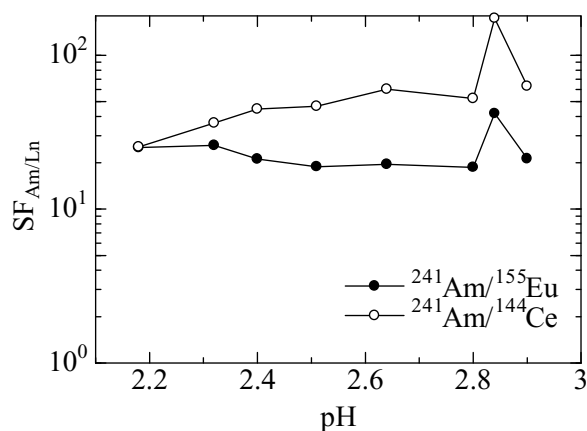


Fig. 3 Separation factors of ^{241}Am for ^{155}Eu and ^{144}Ce

Table 2 Experimental conditions of the column separation experiments

	CMPO/SiO ₂ -P column		HDEHP/SiO ₂ -P column	
	Flow velocity: 4cm/min		Flow velocity: 4cm/min	
	Temperature of the column: 323 K		Temperature of the column: 298 K	
	Solution	Amount	Solution	Amount
Conditioning	4 M HNO ₃	2 BV	0.01 M HNO ₃	2 BV
Feed	Raffinate from simplified PUREX	2 BV	Intermediate product of the CMPO/SiO ₂ -P column	10 BV
Wash solution 1	4 M HNO ₃	6 BV	0.01 M HNO ₃	2 BV
Eluent	H ₂ O	10 BV	50 mM DTPA (pH 2.3)	15 BV
Wash solution 2	50 mM DTPA (pH 3)	5 BV	1 M HNO ₃	5 BV

3.2. Demonstration of MA(III) recovery by column separation experiments on genuine HLLW

3.2.1. MA(III) + Ln(III) recovery by CMPO/SiO₂-P column

Elution curve obtained by CMPO/SiO₂-P column is shown in Fig. 4, where C_0 and C are the concentrations in the feed solution and the effluent, respectively. Radioactivity of ^{241}Am was expressed as the sum with ^{238}Pu due to the analysis. Nevertheless, amount of ^{238}Pu must be quite small since radioactivity of $^{239}\text{Pu} + ^{240}\text{Pu}$ in the feed solution was negligible and isotope abundance ratio of ^{238}Pu in the spent fuel was less than 1 %.

^{137}Cs was eluted from the column with the feed solution and the wash solution, and it was not detected in fractions of the eluent and the wash solution 2. Cs and some fission products such as Sr are not extracted by CMPO⁴, thus they can be discharged properly from the column in the early stage of the process. ^{125}Sb was found in the fractions of wash solution and the eluent. ^{106}Ru showed similar behavior with ^{125}Sb , however some part was discharged with the last wash solution. ^{125}Sb seems to be eluted only by the acid, whereas the DTPA solution is also effective to discharge the adsorbed ^{106}Ru . The complicated elution property of ^{106}Ru must be explained by various chemical forms of Ru in nitric acid solution⁵. ^{125}Sb and ^{106}Ru are inevitably mixed in the interim product if all the H₂O fractions are used as the product, however they are possible to be excluded reasonably from the product by selecting an appropriate area for the product. Elution of MA(III) and Ln(III) by the H₂O eluent and washing out of the residual part of them were proceeded as expected. In this study, all fractions of H₂O were employed as the interim product and supplied to the next HDEHP/SiO₂-P column.

Table 3 Material balance, decontamination factor and recovery yields obtained by column experiments

Nuclides	CMPO/SiO ₂ -P column			HDEHP/SiO ₂ -P column			2 steps operation	
	Material balance [%]	DF	Recovery yields [%]	Material balance [%]	DF	Recovery yields [%]	DF	Recovery yields [%]
^{106}Ru	53.1	1.11×10^1	17.0	139	3.59×10^1	2.00	3.98×10^2	53.1
^{125}Sb	38.5	1.88×10^1	13.8	85.1	3.67×10^0	32.0	6.90×10^1	32.8
^{137}Cs	105	6.36×10^2	0.15	102	1.60×10^1	6.17	1.02×10^4	100
^{144}Ce	144	7.17×10^{-1}	96.8	85.1	$> 5.87 \times 10^3$	$< 2.00 \times 10^{-2}$	$> 4.21 \times 10^3$	85.1
^{155}Eu	111	9.11×10^{-1}	98.7	89.9	$> 3.17 \times 10^4$	$< 3.50 \times 10^{-3}$	$> 2.89 \times 10^4$	89.9
$^{238}\text{Pu}+^{241}\text{Am}$	108	—	92.7	136	—	79.4	—	73.6
^{244}Cm	109	—	98.0	103	—	74.3	—	72.8

Table 3 shows material balances, decontamination factors and recovery yields into the interim and MA(III) products calculated from the elution curves. The material balance was the ratio of sum of the eluted amount to the supplied amount. The decontamination factors were calculated by

$$DF = C_{M,F} V_F / (\sum C_{M,i} V_i),$$

where $C_{M,i}$ and $C_{M,F}$ are the concentration of M in i th fraction of the products and feed solution, respectively, and V_i and V_F are volumes of i th fraction and feed solution, respectively. Recovery yield was the ratio of the collected amount into the product to the total of the discharged amount.

Material balances of all nuclides except for ^{106}Ru and ^{125}Sb exceeded 100 %. Sum of errors in the analyses and volume of the collected liquid must be one of the reasons of the too large material balances, and all supplied amount of them are assumed to be discharged properly from the column. Half of ^{106}Ru and 60 % of ^{125}Sb supplied might stay inside the column even after the operation. They could be washed by nitric acid as mentioned above, thus conditioning process between the operations is expected to discharge those nuclides. Additional experiments are required in advance with repeated operations.

Decontamination of ^{137}Cs was predictably succeeded by the 1st column, and the decontamination factors 10-20 were achieved even for ^{106}Ru and ^{125}Sb . As discussed above, those nuclides can be more decontaminated by optimization in the range the interim product. Ln(III) have to be significantly decontaminated by the following HDEHP/SiO₂-P column to achieve the targeted decontamination factors.

A part of MA(III) and Ln(III) were recovered in fractions of the wash solution 2, then the recovery yields of them were less than 100 %. Though fractions of the DTPA are necessary to be involved in the interim product in order to increase the recovery yields of MA(III) up to about 100 %, those fraction were excluded from the product to suppress the influence of DTPA in the feed solution on the HDEHP/SiO₂-P column performance. The amount of MA(III) washed by the DTPA solution was a few % of the supplied amount, those MA(III) are desirable to be included in the product depending on the performance of the 2nd column. Optimization in the structure of the adsorbent may contribute to enhance the elution property as seen in the case of TODGA/SiO₂-P adsorbent⁶. The

similar tunings in structure of CMPO/SiO₂-P adsorbent are expected to lead the higher recovery yields of MA(III) by H₂O eluent.

3.2.2. MA(III)/Ln(III) separation by HDEHP/SiO₂-P column

Elution curve obtained by HDEHP /SiO₂-P column is shown in Fig. 5. Concentrations of ¹⁰⁶Ru and ¹²⁵Sb in the feed solution were lower than the detectable limits, thus those values were calculated from elution behavior of the 1st column. ¹⁰⁶Ru was discharged from the column with the feed solution and the wash solution, and was not mixed into the product solution. Ln(III) were found only in the fractions of the wash solution 2, and the selective stripping of MA(III) from HDEHP/SiO₂-P by the DTPA solution with pH = 2.3 was successfully achieved. Whereas ¹²⁵Sb was also detected in the product solution as well as MA(III), further decontamination of ¹²⁵Sb might be required. Not all MA(III) were recovered in the fractions of DTPA solution, and the residuals were eluted with Ln(III) by 1 M HNO₃ solution. This study put priority on the decontamination of Ln(III) rather than on recovery yields of MA(III), then the final product is set at fractions of the DTPA solution.

Material balances, decontamination factors and recovery yields into the final product calculated from the elution curves are shown in Table 3, where decontamination factors and recovery yields of ¹⁴⁴Ce, ¹⁵⁵Eu were calculated from the analytical limit 3.7×10^1 Bq/cm³ as they were not detected in the product solution. Material balances of MA(III), ¹⁰⁶Ru and ¹³⁷Cs were more than 100 %, thus all of supplied amount of them are considered to be discharged from the column. More volume of the nitric acid solution might be required to wash out all Ln(III) charged in the column. Errors in analysis and in the assumption of C₀ might cause the bad material balance for ¹²⁵Sb.

Decontamination factors of Ln(III) were far larger than the targeted value 100. Decontamination factors of ¹⁰⁶Ru and ¹³⁷Cs were more than 10. They seem to be large enough considering that large part of them were already removed by the 1st column. Not only ¹²⁵Sb but also ¹⁰⁶Ru are possible to be decontaminated further not in HDEHP/SiO₂-P column but in CMPO/SiO₂-P column by changing first 3 BV of the interim product where elution of MA(III) was not confirmed into waste solution.

Recovery yields of MA(III) were 74-79%, and the residuals were washed out with Ln(III) by 1 M nitric acid solution. Additional volume of the DTPA solution will give larger recovery yields of MA(III). The long tailings of the MA(III) elution curves at fractions of the product might be improved by some modifications in the structure of the adsorbent since the optimizations in TODGA/SiO₂-P adsorbent succeeded in reduction of the tailing⁶. Reduction in the volume of the eluent and increase in the MA(III) recovery ratio might be possible simultaneously by the improvements in the adsorbent.

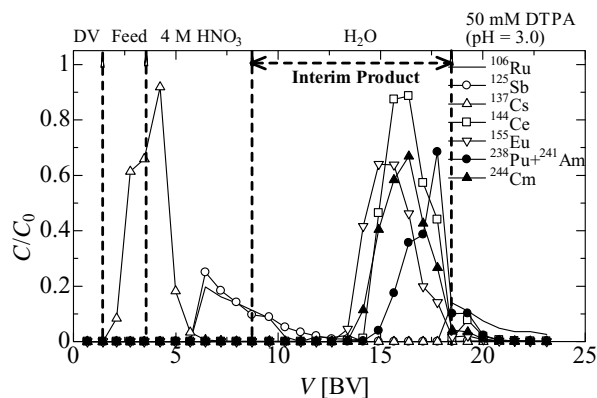
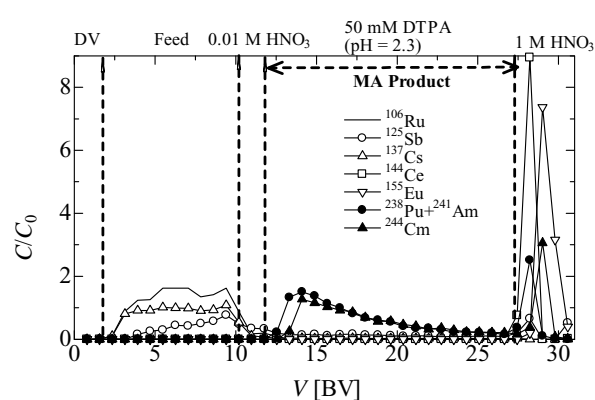
3.2.3. Performance of the 2steps column separation process

Decontamination factors and recovery yields obtained by the 2 steps column operations are shown in Table 3. Decontamination factors were simply multiplying the two results. Recovery yields of the fission products were calculated by multiplying results from two columns and the values exceeding 100 % were set at 100 %. Recovery yields of MA(III) were obtained by multiplying recovery yields evaluated for two columns. Thus, recovery yields of the fission products and MA(III) in right column of Table 3 mean ratios of discharged from the two columns and of recovered in the final product solution, respectively.

About a half of ¹⁰⁶Ru supplied must be stayed in the CMPO/SiO₂-P column, and ¹²⁵Sb remained both in the 1st and 2nd columns. Those nuclides should be washed out by supplying nitric acid in the conditioning process after the operation as discussed above. Decontamination factors of ¹⁰⁶Ru and ¹²⁵Sb are easily improved by reduction in the volume of the interim product from the 1st column. Reduction in the first 3 BV of the interim product leads to the decontamination factors of ¹⁰⁶Ru and ¹²⁵Sb in the 1st column to be 1.32×10^1 and 1.25×10^2 , respectively, and the results implies that the decontamination factors by 2steps operations exceed targeted 100. For ¹³⁷Cs, the targeted recovery yield and decontamination factor were achieved and further improvements in the flow-sheet is not necessary. Decontamination factors of ¹⁴⁴Ce and ¹⁵⁵Eu were far beyond the targeted value, and recovery yields of them should be improved by supplying more volume of the last wash solution for the 2nd column.

More than 70 % of the MA(III) recovery yields satisfy the requirements from the SmART cycle project. Nevertheless, it seems to be poor as MA(III) recovery performance for reducing heat, radiotoxicity and volume of radioactive waste, and the residual 30 % of MA(III) are desirable to be recovered. Since DTPA does not form complex with MA(III) or Ln(III) at acidic environment⁷, MA(III) and Ln(III) washed out from the 1st column by the

wash solution 2 can be separated from DTPA by the 1st column if they supplied to the column again after the acidity is appropriately adjusted. Then a part of MA(III) can be recovered by the 2nd column. MA(III) washed out from the 2nd column with Ln(III) by nitric acid can be separated from Ln(III) by supplying into the HDEHP/SiO₂-P column again after adjustment in acidity. Eventually, a certain ratio of MA(III) discharged with Ln(III) from the 1st and 2nd columns by the last wash solutions are possible to be recovered if the effluent are added to the feed solution of the 1st or 2nd column and processed again. In addition to that, further studies of optimization in adsorbents are also expected to achieve more efficient MA(III) recovery.

Fig. 4 Elution curves obtained by CMPO/SiO₂-P columnFig. 5 Elution curves obtained by HDEHP/SiO₂-P column

The final flow-sheet modified according to the above discussion for the SmART cycle project is shown in Table 4. A column used in this study could process 50 cm³ of the raffinate in the 1st column. Volume of the raffinate produced from reprocessing of 4 irradiated Joyo fuel pins was about 3 dm³. We have designed and fabricated larger scale column with 3 cm diameter to get higher throughput which is the maximum acceptable size to be handled by master-slave manipulators equipped in CPF. 12 times separation operations (6 times each for the 1st and 2nd columns) with the large columns are able to process all the raffinate to recover 2.6 g of Am. Performance of CMPO/SiO₂-P and HDEHP/SiO₂-P adsorbents during the repeated operation are experimentally guaranteed by 10 times repeated operations with an engineering scale devise⁸ and by their durability against radiations^{9,10}. The actual MA(III) recovery operation will be finished in 2016.

Table 4 The final flow-sheet proposed for the SmART cycle

	CMPO/SiO ₂ -P column		HDEHP/SiO ₂ -P column	
	Flow velocity: 4cm/min		Flow velocity: 4cm/min	
	Temperature of the column: 323 K		Temperature of the column: 298 K	
	Solution	Amount	Solution	Amount
Conditioning	4 M HNO ₃	5 BV	0.01 M HNO ₃	2 BV
Feed	Raffinate from simplified PUREX	2 BV	Interim product of the CMPO/SiO ₂ -P column	7 BV
Wash solution 1	4 M HNO ₃	6 BV	0.01 M HNO ₃	2 BV
Eluent	H ₂ O	10 BV	50 mM DTPA (pH 2.3)	15 BV
		(Last 7 BV is the interim product)		(All Effluent is the final product)
Wash solution 2	50 mM DTPA (pH 3)	5 BV	1 M HNO ₃	7 BV
		(First 3 BV is mixed with the feed solution of the 1 st column)		(First 4 BV is mixed with the feed solution of the 2 nd column)

4. Conclusion

Optimization in a flow-sheet of the extraction chromatography for MA(III) recovery by CMPO/SiO₂-P and HDEHP/SiO₂-P adsorbent was carried out, and MA(III) recovery performance was demonstrated on genuine HLLW using the improved flow-sheet. Recovery yields of MA(III) and decontamination factors of Ln(III) evaluated by the experiments are more than 70 % and 10³, respectively, and they satisfied the requirements from the SmART cycle project of JAEA. In order to decontaminate fission products further in the project, a modified flow-sheet was proposed. The actual MA(III) recovery operation will be carried out based on the flow-sheet in 2016. As one of subjects of further study for the SmART cycle is the microwave denitration process on the solution containing actinides and DTPA, and fundamental studies of the process are currently underway.

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