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Mass Balance Evaluation of Pyrochemical Process for Reprocessing used Fuel Salt of Molten Chloride Salt Fast Reactor

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A pyrochemical process is proposed to reprocess used fuel salt taken from a molten chloride salt fast reactor. The proposed process is constituted of several steps using chemical and electrochemical reactions in the chloride salt / liquid Bi system. To investigate the technical feasibility of the proposed pyrochemical process, the mass balances of actinides (U, Pu, Np, Am, Cm) and fission products (La, Ce, Pr, Nd, Gd, Sr and Cs) were evaluated based on their thermodynamic properties (their distribution behaviors between the chloride salt and liquid Bi). Demonstration tests of the pyrochemical steps were performed to investigate the distribution behaviors of U, lanthanides, Sr and Cs in this study. The evaluated mass balances showed that almost all actinides were to be recycled as a new fuel salt and most of lanthanides fission products were to be stabilized in a waste form, which indicates the basic feasibility of the proposed pyrochemical process was confirmed.

KEYWORDS: molten salt fast reactor, pyrochemical reprocessing, chloride salt, actinide, fission product

I. Introduction

Chloride salt has recently been attracting increasing attention worldwide as a fuel salt for a molten chloride salt fast reactor (MCSFR). From the viewpoints of both the effective utilization of nuclear fuel resources and the efficient transmutation of long half-life nuclides, a process for reprocessing the used fuel salt discharged from MCSFR should be developed. Since any feasible reprocessing

processes for the used fuel salt have not been reported yet, we have proposed a new pyrochemical process dedicated to MCSFR.^{1,2)}

Figure 1 shows a flow diagram of the proposed pyrochemical process for the recovery of actinides involving the separation of fission products such as lanthanides, alkaline metals and alkaline earth metals in the used fuel salt. The separation of Zr and noble fission products such as Ru, Rh and Mo is not included in this flow diagram, because these

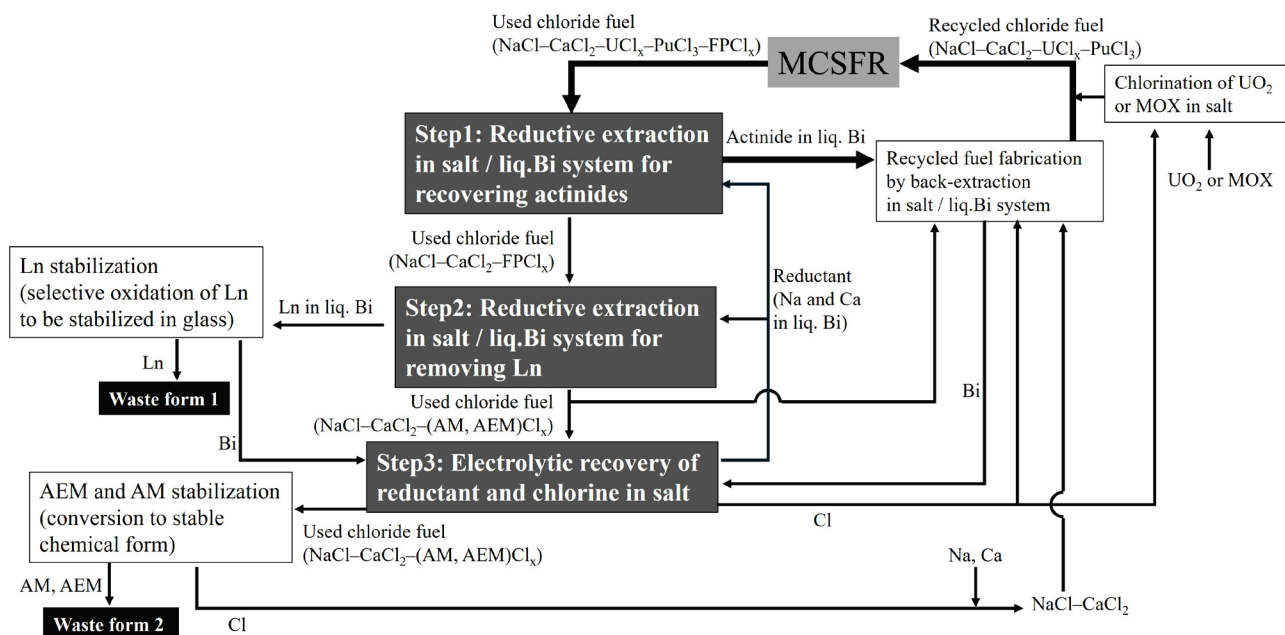


Fig. 1 A flow diagram of the proposed pyrochemical process for reprocessing the used fuel salt from MCSFR.

MCSFR: Molten chloride salt fast reactor, FP: fission products (lanthanides (Ln), alkaline earth metals (AEM) and alkali metals (AM))

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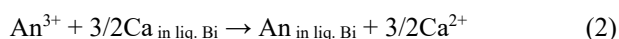
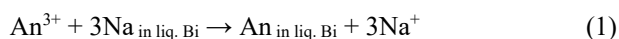
elements are considered to exist as solid precipitates to be separated physically from the fuel salt. Note that the process is designed for not only the used NaCl–CaCl₂ based fuel salt but also the used NaCl based fuel salt from other types of molten salt reactor. The proposed pyrochemical process involves several chemical and electrochemical reactions in the molten chloride salt / liquid Bi system. These steps are constructed similarly to the pyrochemical reprocessing of used metallic fuels, the feasibility of which has been confirmed well.³⁾ The waste products of the proposed pyrochemical process do not contain chlorine. This is a required feature in cases in which the fuel salt is composed of ³⁷Cl-enriched chlorine used to inhibit the formation of long half-life nuclide (³⁶Cl) during irradiation.

To investigate the technical feasibility of the proposed pyrochemical process, the mass balances of actinides and fission products were evaluated. The following were selected as the main elements contained in the used molten chloride fuel salt to be investigated in this study: U, Pu, Np, Am, Cm, La, Ce, Pr, Nd, Gd, Sr and Cs. To evaluate the mass balance in separation among these elements, the thermodynamic properties of representative elements were measured in this study.

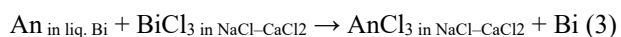
II. Pyrochemical Process to Reprocess used Chloride Fuel Salt^{1,2)}

The main steps of the proposed pyrochemical process are as follows.

Step1 (reductive extraction for recovering actinides): Actinides (An) are recovered from the used fuel salt, where An in the salt are reduced to be extracted in a liquid Bi using Na and Ca as the reductants according to the following chemical reactions.

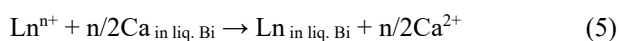


Fission products remain in the salt, realized by controlling the amount of the reductant. The recovered An are then chlorinated through their reaction with BiCl₃ in the NaCl–CaCl₂ melt (back-extraction).



The resulting salt, NaCl–CaCl₂–AnCl₃, is recycled as the new fuel salt.

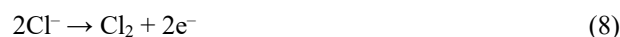
Step2 (reductive extraction for removing lanthanides): Lanthanide fission products (Ln) are removed from the salt, where Ln are chemically extracted in liquid Bi similarly to Step1.



The removed Ln in liquid Bi is selectively oxidized to be contained in stable waste form 1 (for example, borosilicate

glass), leaving most of liquid Bi in the metallic state due to the higher oxygen affinity of Ln than Bi.

Step3 (electrochemical recovery of reductant and chlorine): Na⁺ and Ca²⁺ are formed in the salt after Step1 and 2 (reactions 1, 2, 4 and 5). These cations are electrochemically reduced on a Bi cathode (reactions 6 and 7) in a portion of the salt after Step2, whereas chlorine gas evolves on a carbon anode (reaction 8).



The obtained Na and Ca are used as the reductants in both Step1 and 2. The evolved chlorine is used to prepare BiCl₃ for back-extraction and to chlorinate the feed material of the fuel salt (actinide oxides). It is considered that the fission product iodine gas evolves together with chlorine gas on the anode. The evolved iodine gas must be separated from chlorine gas to be stabilized in a waste form. The separation and stabilization of iodine will be investigated as a future study.

Alkali metal (AM) and alkaline earth metal (AEM) fission products remain in the salt in the form of their chlorides after Step3. They are converted in another chemical form (for example, phosphate⁴⁾) to be stabilized as waste form 2.

III. Thermodynamic Property Measurements in NaCl–CaCl₂ Melt

For evaluating the mass balances of actinides and fission products, it is necessary to obtain a distribution behavior of actinides and fission products between NaCl–CaCl₂ melt and liquid Bi at Step1, 2 and 3 of the proposed pyrochemical process. Therefore, in this study, demonstration tests of reductive extraction (Step1 and 2) and electrochemical recovery of reductant and chlorine (Step3) were performed to investigate the distribution behavior.

1. Experimental

All experiments were carried out in a glove box in which Ar gas was filled (Fig. 2). The concentration of impurities, O₂ and H₂O, in the glove box was controlled to be below 1 ppm. The eutectic NaCl–CaCl₂ salt (48 mol%NaCl–52 mol%CaCl₂, m.p.777 K, ~107 g, APL Engineered Materials, Inc.) was loaded in an alumina crucible (SSA-S, Nikkato) and melted at 823 K. The salt was used without any pretreatments such as drying. Bi–0.18wt%Na–0.21wt%Ca alloy (and Bi metal) were loaded in a small alumina crucible (9 mm inner diameter, SSA-S) in which a W wire (1 mm diameter, Nilaco Corporation) was inserted to measure the potential of the liquid Bi alloy during the reductive extraction demonstration tests. Bi metal (Rare Metallic Co., Ltd.), Na metal (Rare Metallic Co., Ltd.) and Ca metal (Nilaco Corporation) were used to prepare the Bi–Na–Ca alloy. A liquid Bi electrode was composed of liquid Bi (~2 g) loaded in the small alumina crucible with the W wire lead, which was used for the demonstration tests of electrochemical recovery of reductant and chlorine. For the electrochemical measurements, Ag/Ag⁺

reference electrode and a glassy carbon counter electrode were used. The Ag/Ag⁺ reference electrode consisting of a Ag wire immersed in LiCl–KCl–1.0wt%AgCl (APL Engineered Materials, Inc.) melt, which was loaded in a Pyrex tube with a thin bottom to maintain an electrical contact with the electrolyte.

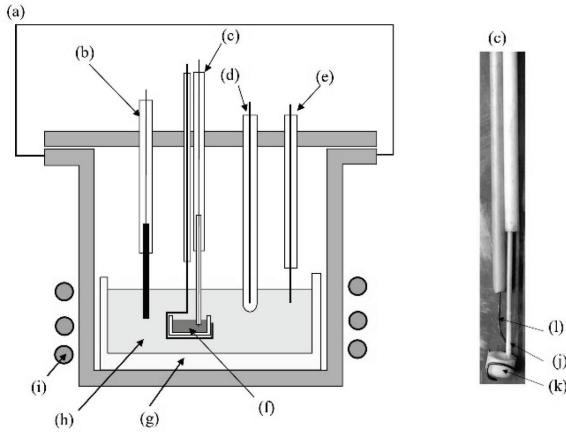


Fig. 2 Experimental apparatus for demonstration tests of reductive extraction and electrochemical recovery of reductant and chlorine. (a) Ar glove box, (b) glassy carbon counter electrode, (c) equipment for Step1 and 2 demonstration holding Bi-Na-Ca (and Bi) alloy or liquid Bi electrode for Step3 demonstration, (d) Ag/Ag⁺ reference electrode, (e) W wire electrode, (f) Bi-Na-Ca alloy and/or Bi, (g) alumina crucible, (h) NaCl–CaCl₂–LnCl₃ (Ln: La, Ce, Pr, Nd, Gd) melt or NaCl–CaCl₂–CsCl–SrCl₂–(CeCl₃) melt at 823 K, (i) electric heater, (j) W wire lead covered by alumina insulator, (k) small alumina crucible, (l) Ta wire to hold the small alumina crucible.

2. Demonstration Tests of Reductive Extraction Step1 and 2

The distribution behaviors of lanthanides (La, Ce, Pr, Nd, Gd), alkali metal (Cs) and alkaline earth metal (Sr) between NaCl–CaCl₂ melt and liquid Bi at Step1 and 2 were investigated in terms of the equilibrium separation factor (SF). Separation factor of lanthanide based on Ce is defined as follows,

$$SF_M = (X_{M \text{ in salt}} / X_{M \text{ in Bi}}) / (X_{Ce \text{ in salt}} / X_{Ce \text{ in Bi}}) \quad (9)$$

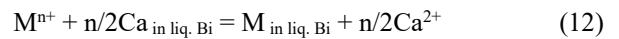
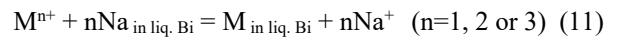
Separation factor of Cs, Sr and Ce based on Na is defined as follows,

$$SF_M = ((X_{M \text{ in salt}})^{1/n} / (X_{M \text{ in Bi}})^{1/n}) / (X_{Na \text{ in salt}} / X_{Na \text{ in Bi}}) \quad (10)$$

(n = 1 in case of M = Cs, n = 2 in case of M = Sr, n = 3 in case of M = Ce)

$X_{M \text{ in salt}}$ and $X_{M \text{ in Bi}}$ represent the concentration of element M in the melt and in liquid Bi, respectively. M represents La, Ce, Pr, Nd, Gd, Cs or Sr. To obtain the separation factor, 2 runs of reductive extraction demonstration tests were performed, one in NaCl–CaCl₂ melt containing LnCl₃ (Ln: La, Ce, Pr, Nd, Gd), while the other in NaCl–CaCl₂ melt containing CsCl, SrCl₂ and CeCl₃ at 823 K each. Since Ce is

one of the primary elements in lanthanide fission products, Ce was chosen as the base to compare SF_M among lanthanides. Since RUN2 was focused on obtaining SF_{Cs} and SF_{Sr}, Na, which was a main element to be extracted in Bi among AM and AEM, was selected as the base. The compositions of the melt and liquid Bi containing the reductant are seen in **Table 1**. The composition of the melt was determined based on the composition of fission products in the used fuel salt. Bi–0.18wt%Na–0.21wt%Ca alloy was used in RUN1 and Bi–Na–Ca alloy prepared by diluting Bi–0.18wt%Na–0.21wt%Ca alloy with Bi metal was used in RUN2. Thus, the amount of Na and Ca in Bi in RUN2 is less than that in RUN1. The liquid Bi containing Na and Ca as the reductant was immersed in the melt until the open circuit potential of the liquid Bi became stable, indicating that the reductive extraction reactions (reaction 11 and 12) could reach the equilibrium.



After reductive extraction demonstration tests RUN1 and RUN2, part of the melt or liquid Bi was dissolved in nitric acid to analyze the concentration of Ln, AM and AEM by ICP-MS (inductively coupled plasma mass spectrometry) and ICP-OES (inductively coupled plasma optical emission spectrometry), and Cs by AAS (atomic absorption spectrometry). Based on the analyzed results, separation factors of Ln, Cs and Sr were evaluated as summarized in **Table 2**, which were used to evaluate the mass balances at Step 1 and 2. Separation factors of lanthanides in LiCl–KCl melt were reported at 773 K and 873 K (SF_{La} = 3.02 (773 K) and 2.80 (873 K), SF_{Pr} = 0.72 (773 K) and 0.66 (873 K), SF_{Gd} = 4.04 (773 K) and 3.43 (873 K))⁵. The separation factors obtained in this study were close to the reported values. This indicated that there was not significant dependence of separation factors of lanthanides on the cations of the melt.

Table 1 The composition of the melt and liquid Bi containing the reductant used for reductive extraction demonstration tests RUN1 and RUN2

		Weight (g)	
		RUN1	RUN2
Liquid Bi containing reductants	Bi	2.09	2.04
	Na	3.77×10^{-3}	1.35×10^{-3}
	Ca	4.40×10^{-3}	1.58×10^{-3}
Melt	NaCl–CaCl ₂	106	107
	LaCl ₃	0.955	-
	CeCl ₃	3.017	1.691
	PrCl ₃	1.538	-
	NdCl ₃	5.021	-
	GdCl ₃	0.156	-
	SrCl ₂	-	3.165
	CsCl	-	2.501

Table 2 The evaluated separation factor (SF) for RUN1 and RUN2

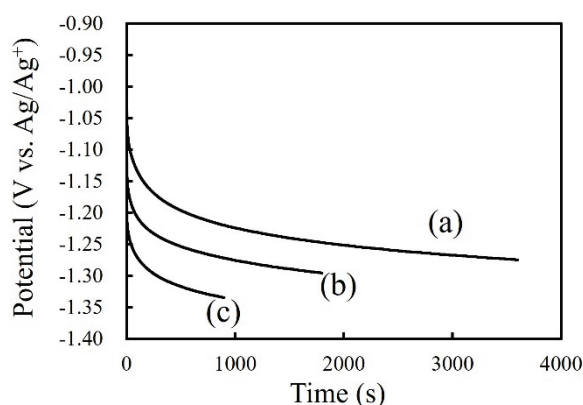
	RUN1*	RUN2**
SF _{La}	3.2	-
SF _{Ce}	1	3.3×10^{-4}
SF _{Pr}	0.84	-
SF _{Nd}	0.86	-
SF _{Gd}	4.1	-
SF _{Sr}	-	3.0×10^{-2}
SF _{Cs}	-	1.8

*Separation factor based on Ce defined by equation 9

**Separation factor based on Na defined by equation 10

3. Demonstration Tests of Electrochemical Recovery of Reductant and Chlorine Step3

For evaluating the mass balances for Step3, the distribution behavior of Cs, Sr, Na and Ca between NaCl–CaCl₂ melt and liquid Bi was investigated by carrying out demonstration tests of electrochemical recovery of reductant and chlorine. To evaluate the dependence of distribution behavior on the applied current, three runs of galvanostatic electrolysis using a liquid Bi electrode at different applied cathodic currents were performed in NaCl–CaCl₂–1.2mol%CsCl–1.6mol%SrCl₂ melt at 823 K which simulated the melt after Step2. **Figure 3** shows the potential changes of the liquid Bi electrode during galvanostatic electrolysis. All electrolyses were continued until the passed electricity reached 18 coulombs. The potential shifted gradually to negative direction with electrolysis time, showing the increase of Na, Ca, Cs and Sr concentrations in liquid Bi by electrolysis.

**Fig. 3** Potential changes of the liquid Bi electrode with reference to Ag/Ag⁺ electrode during galvanostatic electrolysis at (a) –0.005 A (RUN3), (b) –0.01 A (RUN4) and (c) –0.02 A (RUN5) in NaCl–CaCl₂–1.2mol%CsCl–1.6mol%SrCl₂ melt at 823 K

Part of the melt or liquid Bi after electrolysis was dissolved in nitric acid to be supplied to ICP-MS or AAS to analyze the concentration of Na, Ca, Cs and Sr in both the melt and the liquid Bi. By using the analysis results, the distribution ratio between the melt and the liquid Bi as defined by Eq. (13) was evaluated.

$$(\text{distribution ratio}) = (X_{\text{M in salt}} / X_{\text{M in Bi}}) / (X_{\text{Na in salt}} / X_{\text{Na in Bi}}) \quad (13)$$

Table 3 lists the obtained distribution ratio. The distribution ratio of Cs was found to be much larger than 1, indicating that the tendency of the preferential recovery of Na than Cs. In addition, the distribution ratio of Cs decreased with increasing applied cathodic current. This might be explained by that $X_{\text{Ce in Bi}}$ tends to increase at lower Bi electrode potential (at higher applied cathodic current as shown in Fig. 3) because Cs would have lower deposition potential on Bi than Na. The distribution ratio of Sr was similar to that of Ca and did not show a significant dependence on the applied cathodic current.

Table 3 The distribution ratio of Ca, Sr and Cs at different applied current

	RUN3	RUN4	RUN5
Current (A)	–0.005	–0.01	–0.02
Ca	1.5	1.6	1.5
Sr	1.4	1.4	1.3
Cs	19.9	18.9	14.4

The distribution ratio at applied cathodic current of –0.02 A was used to evaluate the mass balance.

4. Distribution Behavior of Actinides

Concerning the distribution behaviors of actinides (U, Pu, Np, Am and Cm), their separation factors were evaluated as follows. Since one of the authors had reported the formal standard redox potential of U (E^0_{U}) to be –2.325 V (vs. Cl₂/Cl[–]) in NaCl–CaCl₂ melt at 823 K,⁶⁾ separation factor of U based on Ce (SF_{U} defined by equation 9) was evaluated to be 1.2×10^{-3} according to the following equation,

$$\ln(\text{SF}_{\text{U}}) = 3F/RT (E^0_{\text{Ce}} - E^0_{\text{U}}) + \ln(\gamma_{\text{U in Bi}}/\gamma_{\text{Ce in Bi}}) \quad (14)$$

R, T and F mean the gas constant, temperature in Kelvin and the Faraday constant, respectively. $\gamma_{\text{U in Bi}}$ and $\gamma_{\text{Ce in Bi}}$ are activity coefficient of U and Ce in liquid Bi, respectively, which are summarized in a review paper.⁷⁾ E^0_{Ce} represents the formal standard redox potential of Ce in the melt, which was measured in this study. After Ce metal was deposited on W wire electrode by galvanostatic electrolysis at –91 mA cm^{–2} for 10 s in NaCl–CaCl₂ melt containing different concentration of CeCl₃ at 823 K, the applied current was changed to 0 mA to measure the stable potential of the Ce metal which was considered to be the redox potential of Ce³⁺/Ce (E_{Ce}) in the melt. E_{Ce} are plotted against the concentration of CeCl₃ in the melt as seen in **Fig. 4**. According to Nernst equation (equation 15), E^0_{Ce} was evaluated to be –2.934 V (vs. Cl₂/Cl[–]) from the value at the intercept of the regression line with the theoretical gradient, 0.0236 (= RT/3F), which was presented as a dotted line in Fig. 4.

$$E_{\text{Ce}} = E^0_{\text{Ce}} + RT/3F \times \ln X_{\text{Ce in salt}} \quad (15)$$

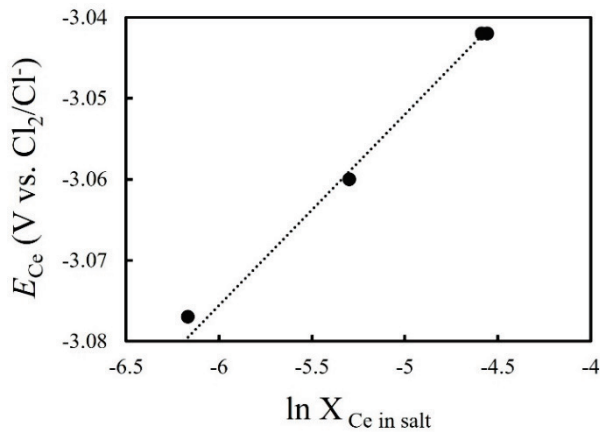


Fig. 4 Redox potential of Ce^{3+}/Ce (E_{Ce}) plotted against the concentration of CeCl_3 in NaCl-CaCl_2 melt at 823 K

Since few thermodynamic properties of Pu, Np, Am and Cm were reported in NaCl-CaCl_2 melt, reported values of their separation factors in LiCl-KCl melt and liquid Bi system^{8,9)} were used for evaluating mass balance in the present study.

IV. Mass Balance Evaluation of the Proposed Pyrochemical Process

The mass balances of the selected actinides and fission products for the proposed pyrochemical process were evaluated in a case of reprocessing the used fuel salt taken from MCSFR of 700 MWth. Throughput of the process was assumed to be around 122 liters of the used fuel salt in 30 days. The equilibrium composition of the used fuel salt to be reprocessed in 30 days is summarized in **Table 4**.

Table 4 The amount of the elements in the used fuel salt from MCSFR of 700 MWth to be reprocessed in 30 days.

	Amount in the used fuel salt from MCSFR to be reprocessed in 30 days (mol)
U	6.21×10^2
Pu	1.28×10^2
Np	1.26
Am	6.20
Cm	2.06
La	5.85
Ce	1.84×10
Pr	9.14
Nd	3.21×10
Gd	1.42
Sr	1.72
Cs	1.30
Na	8.85×10^2
Ca	8.85×10^2

To evaluate the mass balance, the recovery and decontamination ratios at Step1, 2 and 3 defined by the following equations were evaluated as listed in **Table 5**.

Recovery ratio (RR) = (Amount recovered in liquid Bi at the Step) / (Initial amount in the used fuel salt at the Step) (16)

Decontamination ratio (DR) = (Initial amount in the used fuel salt at the Step) / (Amount contaminated in liquid Bi at the Step) (17)

Table 5 Evaluated recovery ratio (RR) and decontamination ratio (DR) for selected actinides and fission products at Step1, 2 and 3 of the proposed pyrochemical process

	Step1		Step2		Step3
	RR	DR	RR	DR	RR
U	0.999	-	1	-	-
Pu	0.999 (input)	-	1	-	-
Np	0.999	-	1	-	-
Am	0.997	-	1	-	-
Cm	0.957	-	1	-	-
La	-	2.8×10^2	0.969	-	1
Ce	-	77	0.999 (input)	-	1
Pr	-	62	0.999	-	1
Nd	-	61	0.999	-	1
Gd	-	3.6×10^2	0.937	-	1
Sr	-	1.7×10^3	-	3.9×10^2	0.970
Cs	-	2.4×10^3	-	1.2×10^3	0.229

RR of actinides and RR and DR of lanthanides were evaluated, assuming that a batchwise countercurrent extraction with three stages¹⁰⁾ would be applied for Step1 and 2. DR of alkali metal and alkaline earth metal were the ones at the single stage reductive extraction. RR and DR were uniquely determined from the separation factors mentioned in section III under the condition of 99.9% recovery of Pu for Step1 and 99.9% recovery of Ce for Step2. The detail of the above calculation was reported by Sakamura.¹⁰⁾ RR at Step3 was determined using the distribution ratio at RUN5 seen in Table 3, when the electrolysis was continued until the amount of recovered reductant corresponded to the required amount for Step1 and 2. Here, to reduce the volume of the waste form 2, the electrolysis was assumed to be performed in a half amount of the salt after Step2 and the rest half of the salt was used at back-extraction.

The evaluated mass balance is summarized in **Table 6**. It was noticed that almost all actinides were recovered from the used fuel salt to be recycled as a new fuel salt, while most of lanthanide fission products were stabilized in a waste form 1. 1.5 % of Sr and 39 % of Cs were stabilized in a waste form 2. The rests of Sr and Cs were used as the reductant at Step1 and 2 or contained in the NaCl-CaCl_2 base salt which was used as the melt at back-extraction step. This means that the concentrations of Sr and Cs in the fuel salt would change to reach certain values in equilibrium after repeating the proposed pyrochemical process. The equilibrium concentrations will be investigated as a future study. The

amounts of the produced waste form 1 and 2 were evaluated to be 97 kg/month and 6.9 kg/month, respectively, assuming that the concentration of lanthanides in the glass waste form (waste form 1) would be 10 wt% and the phosphate glass containing 26 wt% of alkali metal and alkaline earth metal would be selected as the waste form 2.

Table 6 Evaluated mass balance of selected actinides and fission products in the proposed pyrochemical process. Values of Pu, Np, Am and Cm were evaluated based on their reported thermodynamic properties in LiCl–KCl / liquid Bi system.^{8,9)}

	Recovered at Step1 to be recycled as new fuel salt (mol)	Removed at Step2 to be stabilized in waste form 1 (mol)	Stabilized in waste form 2 (mol)
U	6.20×10^2	6.21×10^{-1}	-
Pu	1.28×10^2	1.28×10^{-1}	-
Np	1.26	1.26×10^{-3}	-
Am	6.18	1.86×10^{-2}	-
Cm	1.97	8.86×10^{-2}	-
La	2.1×10^{-2}	5.7	-
Ce	2.4×10^{-1}	1.8×10	-
Pr	1.5×10^{-1}	9.0	-
Nd	5.5×10^{-1}	3.2×10	-
Gd	4.0×10^{-3}	1.3	-
Sr	1.0×10^{-3}	4.3×10^{-3}	2.6×10^{-2}
Cs	5.5×10^{-4}	1.1×10^{-3}	5.1×10^{-1}

The electrochemical conditions will be optimized to increase the distribution ratio of Sr and Cs at Step3 (Eq. (13)) as a future study. For example, an inert cathode will be used to recover the reductant (Na and Ca) leaving most of Cs and Sr in the melt at Step3, which could lead to the increase of the ratio of Sr and Cs in the waste form 2.

V. Conclusion

The authors have promoted research on the pyrochemical reprocessing of the used fuel salt from the molten chloride salt fast reactor. The proposed pyrochemical process uses chemical and electrochemical reactions in reaction media of the molten chloride salt and liquid Bi. Based on the thermodynamic properties obtained in this study as well as those of reported values, mass balances of actinides (U, Pu, Np, Am, Cm) and fission products (La, Ce, Pr, Nd, Gd, Sr,

Cs) in the proposed pyrochemical process were evaluated for the molten chloride salt fast reactor with 700 MWth. Almost all actinides in the used fuel salt were evaluated to be recycled as the new fuel salt, while most of lanthanide fission products were to be stabilized in a waste form (97 kg/month). 1.5% of Sr and 39% of Cs were to be contained in a stable waste form (6.9 kg/month). The obtained results indicates the basic technical feasibility of the proposed pyrochemical process.

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References

- 1) M. Yamawaki, H. Mochizuki, T. Koyama, Y. Arita, K. Mitachi, Y. Shimazu, M. Kinoshita, R. Yoshioka, *J. Atom. Energ. Soc. JPN (ATOMOS)*, **63**, 791 (2021) [in Japanese].
- 2) T. Murakami, T. Koyama, K. Uozumi, M. Yamawaki, “Reprocessing of spent fuel salt from molten chloride salt fast reactor based on the developed technology of pyrochemical reprocessing of spent metallic fuel,” International Workshop on the Chemistry of Fuel Cycles for Molten Salt Reactor Technologies (organized by IAEA and OECD/NEA), Vienna, Austria (hybrid event), 2–6 Oct. 2023.
- 3) T. Koyama, M. Iizuka, “Chap. 12 Pyrochemical Reprocessing Technology development,” R. Taylor & K. Nash edited., “Reprocessing and Recycling of Spent Nuclear Fuels for Advanced Nuclear Fuel Cycles,” Woodhead Publishing 2015.
- 4) B. J. Riley, J. A. Peterson, J. D. Vienna, W. L. Ebert, S. M. Frank, *J. Nucl. Mater.*, **529**, 151949 (2020).
- 5) M. Kurata, Y. Sakamura, T. Matsui, *J. Alloy. Compds.*, **234**, 83 (1996).
- 6) T. Murakami, Y. Sakamura, K. Takamiya, *KURNS Progress report 2022*, CO9-2.
- 7) J. Zhang, E.A. Lahti, E. Wu, *Prog. Nucl. Energy*, **81**, 67 (2015).
- 8) M. Kurata, Y. Sakamura, T. Hijikata, K. Kinoshita, *J. Nucl. Mater.*, **227**, 110 (1995).
- 9) H. Moriyama, H. Yamana, S. Nishikawa, S. Shibata, N. Wakayama, Y. Miyashita, K. Moritani, T. Mitsugashira, *J. Alloy. Compds.*, **271-273**, 587 (1998).
- 10) Y. Sakamura, *Nucl. Technol.*, **210**, 147 (2024).