Chemical state of Ce obtained by distillation tests to alkali chloride baths

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1 Introduction

Since the waste salt contaminated with uranium generated from pyro-reprocessing test can easily capture moisture from the air and corrode equipment and piping further materials, treatment technology for decontamination and waste stabilization should be developed. The aim of this study is to develop a simple separation method for decontamination of bath salts. A two-step process has been considered. In the first step, the oxygen donor is added to a melt of the bath salt, and then uranium is separated from the salt as precipitates. In the next step, melt bath components are evaporated by a vacuum distillation. Melt baths were LiCl-KCl eutectic or NaCl-2CsCl salts. Lithium oxide and cerium chloride were used as a precipitant, and uranium surrogate for the tests, respectively.

On precipitation process, 90% of cerium was recovered from the bath salts when 1.5-2 times more lithium oxide added relative to cerium. Results of structural analysis to the precipitates showed oxychloride (CeOCl) was mainly formed substance [1].

In this study, a distillation equipment has been constructed. The best distillation condition has been searched using the precipitates co-existing melts and a local structure analysis of cerium in the residual of distillation test.

2 Experiment

3.75 g of bath salt, 5 wt% to the bath salt of cerium chloride, and 150% or 200% of stoichiometrically lithium oxide were put into a quartz cell, and melted in an electric furnace in the argon circulation glove box (GB) in the controlled area of the Atomic Energy Research Lab., Tokyo City University. The temperature was 700 $^{\circ}$ C for LiCl-KCl eutectic and 800 $^{\circ}$ C for NaCl-2CsCl salt.

After cooling and solidification, the salt was crushed and transferred to a vacuum distillation unit. The furnace was heated to 800°C and the pressure reduction was maintained for about 2-6 hours. Two types of distillation units were available, horizontal (H) and vertical(V). The cooling system of horizontal type was air, and that of vertical type was water.

After the distillation test, the collected salts were subjected to ICP-OES and Atomic absorbance photometry analysis, and the residues were subjected to XAFS analysis. XAFS measurement was carried out at Energy Accelerator Research Organization Photon Factory BL-27B, in transmission mode and fluorescence mode, using $Ce-L_3$ X-ray absorption edge.

3 Results and Discussion

The collected ratio of salt on the distillation tests is shown in Table 1. The longer the distillation time, the higher the recovery ratio. The vertical unit with a watercooling system achieved a salt recovery ratio of 98%.

Table 1: Collected ratio of the bath salt

	Time (h)	Collected ratio (%)
Li-K 150% (H)	4	89.8
Li-K 200% (H)	2	34.0
Na-Cs 200% (H)	6	58.8
Na-Cs 200% (V)	6	97.5

As shown in Fig. 1, the structure of the residue resembles to that of cerium oxide on the Li-K bath, while the structure of the residue in the Na-Cs bath shows a mixture of oxychloride and cerium oxide. Cerium oxide was generated by evaporation of the baths with excess of lithium oxide.

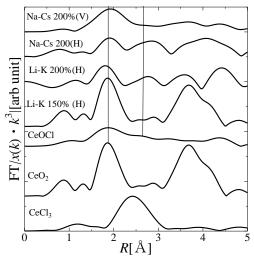


Fig. 1: EXAFS structure function of cerium after distillation tests (Ce-L₃ X-ray absorption edge)

<u>References</u>

- [1] J. Ibe *et al.*, Photon Factory Activity Report, Vol 37 (2020).
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