XAFS analysis on manganese in molten salt precipitation method

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1 Introduction

The spent salt bath generated in the research and development relating to the pyro reprocessing method of spent fuel has a risk of residual unrecovered nuclear fuel material and corrosion of equipment and piping due to chloride ions. Therefore, appropriate processing is required. In research on the process development of salt bath, analogous simulants are used as substitutes for nuclear fuel materials. However, the cerium used in the previous research has a lower valence than the uranium in the actual bath salt, and is less likely to form an oxyanion. Therefore, the nuclear fuel material cannot be fully simulated. Based on the above discussion, we focus on manganese alternatively, which exists up to 7 valences, and compare it with cerium by conducting the same experiment procedure as in the previous research [1].

2 Experiment

A mixture of LiCl-KCl eutectic composition salt or NaCl-2CsCl salt, manganese(II) chloride and lithium oxide was placed in a quartz tube in a glove box. Manganese (II) chloride was added at 10wt% to the bath salt, and lithium oxide was added at a stoichiometric amount of 150% or 200% to the amount of manganese. The prepared samples were melted at 700°C for LiCl-KCl bath and 800°C for NaCl-2CsCl bath for about 4.5 hours using an electric furnace in a glove box. After heating, the sample was solidified together with the quartz tube, and salt bath and sediment were separately collected. In this study, only a batch test was conducted in which a precipitant was added in advance before molten.

The collected sample was pulverized and mixed with boron nitride. The mixture was divided into 80 mg and 120 mg and subjected to XAFS measurement by the transmission method at Photon Factory BL-27B of KEK. The EXAFS radial structure function and XANES spectra were analyzed by the measured data, and the coordination structure and valence of the precipitates were investigated.

3 Results and Discussion

Figure 1 shows the EXAFS radial structure functions of the precipitate, and Figure 2 shows the XANES spectra. The EXAFS radial structure functions of the sediments have very close feature of that of manganese(II) oxide than to manganese(II) chloride. From this fact, it can be seen that the added manganese(II) chloride was precipitated as an oxide due to the reaction with lithium oxide. Also, the XANES spectrum confirmed that the precipitate was similar to the manganese oxide. This indicates that the precipitate is divalent. Even though manganese(II) chloride is melted after adding lithium oxide, it precipitates as a divalent oxide with the same valence.



Figure 1 EXAFS radial structure functions of precipitates generated by 1.5 and 2 times larger molar amount of Li_2O to chemical stoichiometric $MnCl_2$



Figure 2 XANES spectrum of the manganese in precipitates generated by 1.5 and 2 times larger molar amount of Li₂O to chemical stoichiometric MnCl₂

Acknowledgement

This research is part of the results of joint research with Tokyo City University and Japan Atomic Energy Agency.

References

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