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Structural analysis on complexes of rare earths formed in the adsorbent impregnated with NTA amides extractant for recovery of minor actinides

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1 Introduction

Trivalent minor actinides (MA(III): Am and Cm) in high-level radioactive liquid waste (HLLW) have long half-lives and decay heat, thus those elements should be recovered in order to reduce radiotoxic impact to global environment [1]. Separation of MA (III) from trivalent lanthanide (Ln (III)) is a main issue due to their chemical similarity. In this research, we aim to construct a hybrid type process that complements the shortcomings while maximizing the features of both the solvent extraction method and the extraction chromatography method. One of promising flow-sheets consists of two steps of MA(III)+Ln(III) co-recovery and MA(III)/Ln(III) separation, the latter adopts extraction chromatography in order to achieve higher separation performance in a compact system, NTA amide with uniform diameter and large pore diameter that aims to achieve rapid adsorption and elution while suppressing pressure loss, which is a problem of the same technology. By applying an adsorbent, the operating conditions will be eased and the amount of waste liquid generated will be suppressed. In this study, influence of side chain structures of the extractants on local structure of rare earths complexes and adsorption/desorption behavior were investigated to gather valuable information for optimum of process design.

2 Experiment

An adsorbent was prepared by impregnating porous silica particles coated with a styrene-divinylbenzene copolymer (referred to as SiO_2-P) with 33 wt% of HONTA and H2EHNTA extractant, respectively. As an adsorption test, it was shaken for 3 hours at a ratio of 1:10 of an adsorbent and a solution containing Eu or Nd, using nitric acid concentration as a parameter. After the shaking, solid-liquid separation, the supernatant was subjected to ICP-OES analysis, and the dried adsorbent was subjected to EXAFS analysis. EXAFS measurement was carried out at High Energy Accelerator Research Organization Photon Factory BL-27B, in transmission mode, using NdL_3 and EuL_3 absorption edge.

3 Results and Discussion

Fig. 1 shows the adsorption rates of Eu^{3+} and Nd^{3+} on NTA amides adsorbents. It can be seen that adsorption rates decreases in both cases as the nitric acid concentration increases. In addition, HONTA showed a large difference in the adsorption rate of Eu used as a surrogate of Am and the adsorption rate of Nd that is RE in comparison with HEHNTA when the nitric acid concentration is low. Thus, HONTA is expected to have better separation performance than HEHNTA. In addition, EXAFS revealed that HONTA had a clear difference in the number of nearest oxygen coordination numbers around Eu^{3+} and Nd^{3+} compared to HEHNTA for each element, suggesting that this tendency affects the adsorption rates. This work is supported by the Innovative Nuclear Research and Development Program from the Ministry of Education, Culture, Sports, Science and Technology of Japan.

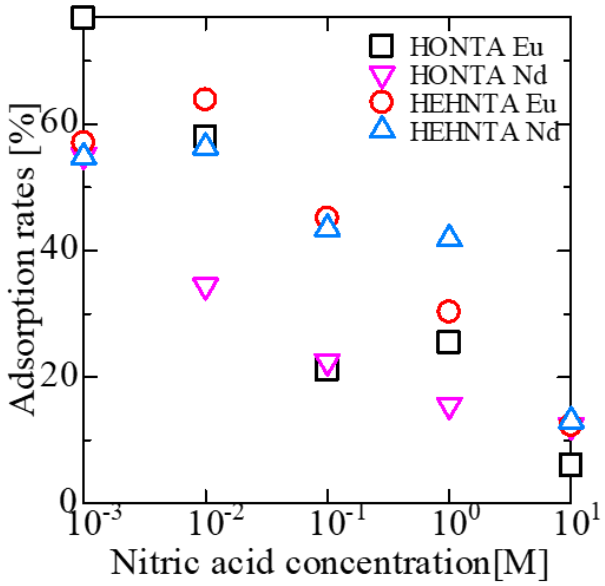

Fig. 1 Adsorption rates of Eu^{3+} and Nd^{3+}

References

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