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Local structure analysis of fission products and rare earth elements adsorbed on the adsorbent impregnated with HONTA extractant

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1 Introduction

Development has been carried out for the realization of a recovery system for trivalent minor actinides (MA: Am and Cm) by separation processes such as solvent extraction and extraction chromatography for high-level radioactive liquid waste. At present, a process using hexaoctylnitrilotriacetoamide (HONTA) extractant in MA recovery process has been considered. For HONTA impregnated adsorbents, the problem was that some fission products such as molybdenum, zirconium, paradium from upstream may disturb the adsorption of MA and rare earth elements. It is necessary to establish the optimum conditions when using HONTA extractant for MA and rare earth elements effectively, the nitric acid concentration dependence of the complex structure of variety of elements (Nd, Sm, Eu, Mo, Zr, Pd) in the adsorbent impregnated with HONTA extractant was investigated systematically by EXAFS.

2 Experiment

An adsorbent was prepared by impregnating porous silica particles coated with a styrene-divinylbenzene copolymer (referred to as SiO_2-P) with 5 wt% of HONTA extractant. As an adsorption treatment, it was shaken for 3 hours at a ratio of 1:10 of an adsorbent and a metal solution containing combination of 10 mM of one of rare earths (Nd, Sm and Eu and 10 mM of one of fission products (Mo, Zr and Pd), using nitric acid concentration as a parameter. After the shaking, solid-liquid separation by centrifugation, 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 BL27B, in transmission mode, using Mo, Zr K absorption edges and Nd, S, Eu L_3 absorption edges.

3 Results and Discussion

By the previous research, radial structural functions are almost constant even though several rare earths co-existing system. Thus optimum condition of elution is well understood by the results obtained for mono-element system.

Figure 1(a) shows the radial structural functions of rare earths and 1(b) shows the RSFs of zirconium. By fitting analysis using typical EXAFS equation, local structure around rare earths are not well influenced by co-existing molybdenum or zirconium, while local structure around molybdenum and zirconium are affected by co-existing rare earths, especially, coordination number decreases by co-existing rare earths. If elution using the same nitrate concentration are applied to once co-adsorbed sample, rare earths are eluted and the RSFs are very similar to mono-element adsorbed ones. Therefore, molybdenum and zirconium are not affected to the mechanism of adsorption/elution of rare earths, thus it is necessary to find cleaning process before extraction chromatography process for separation of MA and rare earths.

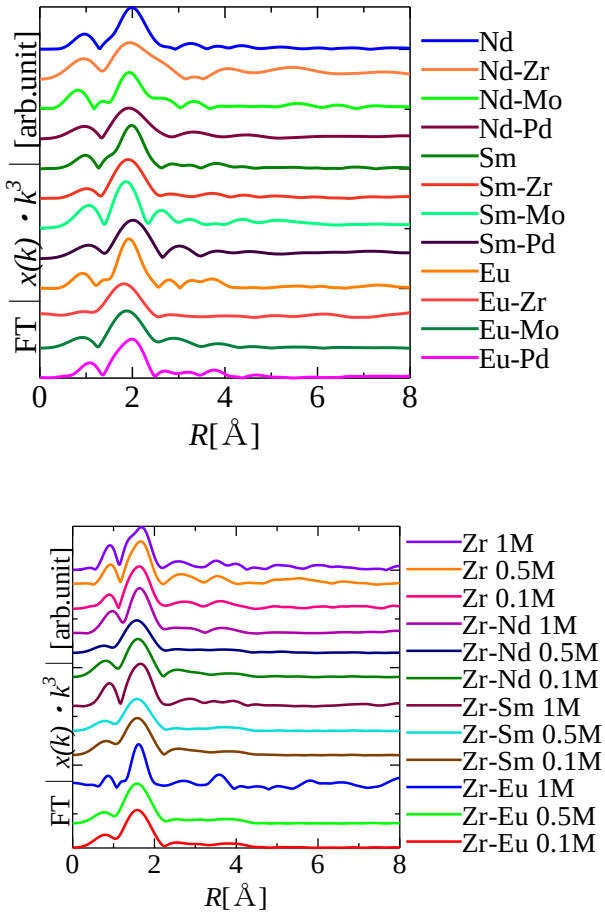

Fig. 1: RSFs of (a) rare earths and (b) zirconium.

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