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Analysis of Th, U, Pu, and Am in radioactive metal waste using extraction chromatography

Lots of metal waste of which surface is contaminated with radionuclides is generated from the decommissioning of nuclear facilities. For the waste management, characterization of radionuclides inventory in the waste is required. In this work, determination method of alpha nuclides, Th, U, Pu, and Am, contained in the metal waste was developed.

Taking into account the half-lives of these alpha nuclides and expected their concentration in the waste, inductively coupled plasma mass spectroscopy (ICP-MS) and alpha-ray spectrometry were selected as analytical tools. The determination using these techniques requires that these alpha nuclides in the metal waste were separated from the large quantities of stable matrix and radioactive interferences nuclides. In our laboratory, we have applied commercially available extraction chromatographic resin to the analysis of alpha nuclides in a dissolved solution of molten products of low-level radioactive waste; UTEVA resin (for uranium and tetravalent actinide resin) for the analysis of Th and U, and TRU resin (for trans uranium resin) for the analysis of Pu and Am. In a similar manner as above, application of these extraction chromatography methods to the metal waste was planned.

In the case of the metal waste, we have to take account for the existence of large amount of Fe, because Fe(III) has significant negative impact on the sorption of trivalent actinides on TRU resin. Hence, the effects of Fe(III) amount and concentration in the sample solution on the uptake of Am, representative of trivalent actinide, on TRU resin were studied in detail with the solution prepared from Fe(NO₃)₃. Fortunately, when Fe(III) is reduced to Fe(II) with ascorbic acid, its effect on the sorption of trivalent actinide is practically negligible. The amount of ascorbic acid required to improve the extraction of Am was also studied.

Based on these study about the effect of Fe(III) on the extraction of alpha nuclides on the extraction resin, the schemes to analyze alpha nuclides in metal waste was optimized. The optimized methods were validated with dissolved solution of stainless steel, SUS304, and adding alpha nuclides. The recoveries of Th and U from dissolved solution of SUS304 were 89±4% and 102±5%, respectively, with optimized scheme using UTEVA resin. The recoveries of Pu and Am were 96±4% and 89±7%, respectively, using TRU resin. Furthermore, Th, U, Pu, and Am in simulated waste prepared by dissolving pipe wastes (the surface ⁶⁰Co concentration was about 70 kBq) sampled from a coolant system of nuclear reactor and adding alpha nuclides were analyzed with the optimized method. The ⁶⁰Co was sufficiently separated from alpha nuclides. The recoveries of Th, U, Pu, and Am were 91±3%, 103±4%, 97±4%, and 91±3%, respectively. These recoveries are comparable to that analyzed dissolved solution of SUS304. These results support the usefulness of the optimized schemes.

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