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Separation and Analaysis of Actinides in Hardly Soluble Materials

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The analysis of actinides in nuclear spent fuel or nuclear debris are important for actinide management, i.e., the decision of how to deal with actinides in these materials. Before almost any analysis, the dissolution of these materials into solution is necessary for prices and accuracy analysis, and the nuclide separations and actinide separation are required in many cases of analyses. In the present report, the pretreatment method for dissolution of these materials, the nuclide separation methods by ion exchange, and the actinide analysis by a triple quadrupole ICP-MS/MS are presented.

The materials with uranium dioxide (UO_2) such as nuclear spent fuels and debris are not easy to be dissolved into acidic solution. We have proposed that these materials are chemically converted by thermochemical reactions. Our proposed conversion consists of the chemical powderization and the chlorization.

The nuclide separation has been proposed by the column techniques based on ion exchange. In this presentation, the Cs removal, Pd recovery/removal, and actinide separations are reported. Cs is desirable to be removed for a reason of its high radioactivity. Pd is advisable to be removed before the separation of other ions because its adsorption behavior on many ion exchanger or adsorbents inhibits the adsorption of other ions. After the removal of these elements, the actinides separations are carried out. The pyridine resin is used for these actinide separations. The actinide separation consists of recovery of U, separation of Pu and Np from trivalent actinides, separation of trivalent actinides from lanthanides, and the mutual separation of trivalent actinides.

The quantitative actinide analysis by a triple quadrupole ICP-MS/MS was also studied. The used nuclides were ²³⁸U, ²³⁷Np, ²⁴⁰Pu, ²⁴¹Am, and ²⁴⁴Cm, respectively. We confirmed the formation percentages of ionic species of each actinides in several kinds of collision reaction cell modes, no gas, hydrogen gas, helium gas, oxygen gas, and BEC (background equivalent to counts) of each actinide ions are confirmed in each modes. And also, we investigated the formation rates of molecular ions in collision reaction cell of oxygen gas mode. We found that the formation rates of actinide ion, oxide ion, and dioxide ion depends on the oxygen flow rate and on actinides; the percentage of actinide monoxide ion with higher atomic number is higher, while percentage of the dioxide ion with lower atomic number is higher. From these results, we concluded that we can discriminate the isobars of these actinides by use of the differences of these actinide molecular ion formation.

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