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Development of mutual separation method of Zr, Nb, and Mo with extraction chromatography using TEVA resin

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The reactors at Fukushima Daiichi Nuclear Power Station were damaged severely because of the accident. Cooling water is leaking from the reactor vessels and groundwater is flooding into the basement of reactor and turbine buildings. Consequently, contaminated water is accumulating. The accumulated water processing facility was installed to decontaminate the radionuclides and to desalinate. As a result, secondary waste such as spent zeolite and sludge are generated. In order to evaluate the radioactivity inventory in the waste, water was sampled from inflow and outflow of the apparatus. Zirconium-93 and ^{93}Mo are candidates of the important nuclides because of their toxicity, half-lives, and relatively large effect on the safety assessment. It is considered reasonable to measure ^{93}Zr using inductively coupled plasma mass spectrometry after separation of interfering elements, ^{93}Nb , $^{92}\text{MoH}^+$, etc., and to measure ^{93}Mo using X-ray spectrometry after separation of ^{93}mNb emitting same energy X-ray.

In the present study, mutual separation of Zr, Nb, and Mo using extraction chromatographic resin impregnated quaternary amine-based liquid anion exchanger, TEVA resin, was investigated for the analysis of ^{93}Zr and ^{93}Mo in the contaminated water. First, distribution coefficients (K_d values) for partitioning of Zr, Nb, and Mo onto the resin from HF, mixture of HF and HNO_3 , and mixture of HF and HCl solutions have been determined. In the HF media, the K_d values of Zr, Nb and Mo were gradually decreased with an increase of HF concentration in 0.01–4 M, ca. 3000 to 13 for Zr, ca. 2700 to 220 for Nb, and ca.1600 to 17 for Mo, and approximately constant in 4–8 M, ca. 220 for Nb, ca. 10 for Zr and Mo. The K_d values suppose that these metal ions are extracted on the TEVA resin from diluted HF solution (0.01–2 M HF), whereas Zr and Mo are eluted by >4 M HF but Nb is not eluted by HF solution. In the 0.1 M HF and various concentrations of HNO_3 media, the K_d values of Zr, Nb, and Mo were rapidly decreased in 0.01–1 M HNO_3 , and they are under 1 when the HNO_3 concentration is more than 1 M. This supposes that extracted Zr, Nb, and Mo are recovered by 1 M HNO_3 solution. In the 0.5 M HF and various concentrations of HCl media, K_d values of Zr was rapidly decreased with an increase of HCl concentration. Whereas, K_d values of Mo and Nb are once decreased with an increase of HCl concentration in 0.01–1 M and then increased with an increase of HCl concentration in 1–7 M. The K_d values of Nb and Mo are more than 100 and K_d value of Zr is less than 1 at the 0.5 M HF–7 M HCl. This supposes that Zr is separated from Nb and Mo.

Based on the obtained K_d values, chromatographic separation scheme was designed and mutual separation of Zr, Nb, and Mo was successfully carried out. In the future, the influence of interfering nuclides will be studied to apply the separation method to the contaminated water.

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