Contribution ID: **47**   Type: **Verbal**

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

Thursday, 15 May 2014 11:30 (15 minutes)

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 93Mo 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 93Zr using inductively coupled plasma mass spectrometry after separation of interfering elements, 93Nb, 92MoH+, etc., and to measure 93Mo using X-ray spectrometry after separation of 93mNb 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 93Zr and 93Mo in the contaminated water. First, distribution coefficients (Kd values) for partitioning of Zr, Nb, and Mo onto the resin from HF, mixture of HF and HNO3, and mixture of HF and HCl solutions have been determined. In the HF media, the Kd 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 Kd 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 HNO3 media, the Kd values of Zr, Nb, and Mo were rapidly decreased in 0.01–1 M HNO3, and they are under 1 when the HNO3 concentration is more than 1 M. This supposes that extracted Zr, Nb, and Mo are recovered by 1 M HNO3 solution. In the 0.5 M HF and various concentrations of HCl media, Kd values of Zr was rapidly decreased with an increase of HCl concentration. Whereas, Kd 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 Kd values of Nb and Mo are more than 100 and Kd 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 Kd 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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Session Classification: Separation Methods, Speciation 4

Track Classification: Separation Methods, Speciation