Separation of Plutonium and Uranium in Spent-Fuel Dissolver Solutions by Extraction Chromatography

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The rapid and accurate measurements of samples derived from spent fuel are a requisite for input accountability analysis and nuclear material control and for on-site verification. One possible means of the timely analysis is to employ on-site assay techniques where samples can be analyzed within the processing facility. One such technique that recently has been developed, isotope dilution gamma-ray spectrometry (IDGS), employs passive gamma spectroscopy to determine the plutonium isotopics and a standard plutonium spike to determine the plutonium concentration in spent-fuel dissolver solutions.

Gamma-ray measurements of plutonium and uranium in highly radioactive dissolver solutions from reprocessing plants require the rapid and efficient separation of fission products. Ion-exchange separation was previously used to satisfactorily purify and recover plutonium for the IDGS measurement. In addition to ion exchange, we have developed a new separation method, extraction chromatography using U/TEVA•Spec resin (for uranium and tetravalent actinides specifically), to rapidly separate fission fragments and recover plutonium and uranium for the high-resolution gamma-ray spectroscopy measurements. U/TEVA•Spec is a novel extraction chromatographic resin composed of diamyl amylphosphonate sorbed on an inert polymeric support (Amberlite XAD-7 or Amberchrom CG-71). The resin is commercially available from EIChroM Industries, Inc. A typical gamma-ray spectrum of spent-fuel dissolver solution after chemical separation with extraction chromatography using U/TEVA•Spec resins is shown in Fig. 1.

Over 30 dissolver solutions with plutonium concentrations varied from 0.62 g Pu/ℓ to 1.58 g Pu/L have been analyzed. The range of plutonium isotopic abundances (wt. %) is 0.34% to 1.24% for ^{238}Pu, 58.24% to 70.84% for ^{239}Pu, 21.21% to 28.57% for ^{240}Pu, 4.00% to 8.9% for ^{241}Pu, and 1.4% to 5.2% for ^{242}Pu. The results of plutonium concentrations and isotopic compositions of dissolver solutions analyzed by IDGS agree very well with those obtained by traditional isotope dilution mass spectrometry (IDMS). Typically a 1-h counting period produces a precision better than 1% for plutonium concentrations and a bias between IDGS and IDMS of less than 0.2%. Precision of 0.5% and 0.2% is typical for ^{240}Pu/^{239}Pu ratio and the ^{239}Pu weight percent, respectively. The agreement between IDGS and IDMS is generally excellent, especially for the ^{240}Pu/^{239}Pu ratio (average IDGS/IDMS ratio is 1.003) and the weight percent of ^{239}Pu (average IDGS/IDMS ratio is 1.000). Success

in these isotopic measurements is essential if an accurate determination of the total plutonium concentration is to be made.

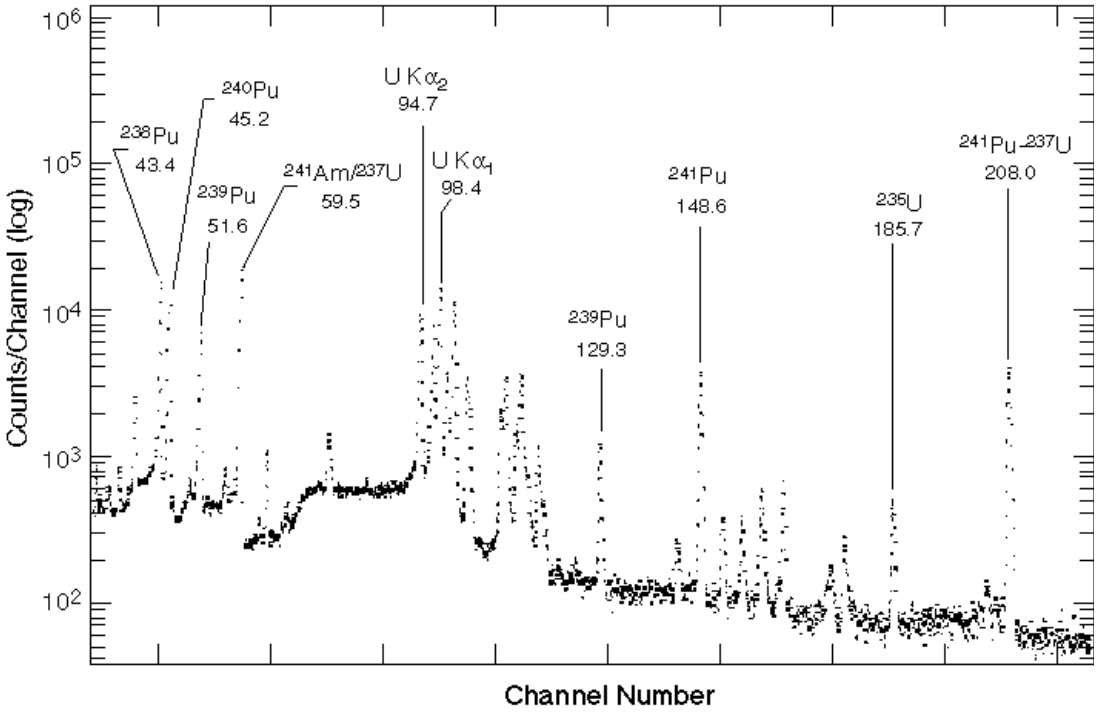

Fig. 1. Gamma-ray spectrum of spent-fuel dissolve solution after chemical separation with extraction chromatography using U/TEVA•Spec resins.

The rapid and accurate IDGS technique with the improved separation method could provide a timely, less expensive, and simpler on-site verification method for the input accountability measurements. This paper will discuss the improvement in fission product separation and measurement methods on input spent-fuel dissolver solutions and will also examine the results of plutonium by using this simplified separation method incorporated into the IDGS technique.

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