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Determination of ²³⁷Np, ⁹³Zr and other long-lived radionuclides in medium- and low-level radioactive waste samples

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The majority of long-lived radionuclides produced in the nuclear fuel cycle can be regarded as “difficult to determine nuclides”due to the low activities and/or the absence of gamma-radiations of medium or high energies in the decay schemes. Most isotopes of actinoides are alpha-emitters, Sr-90 (fission product) and Zr-93 (activation product) emit almost exclusively beta-particles, Nb-93m (activation product) decays by isomer transition or electron capture and emits only X-rays, Nb-94 (activation product) emits low intensity gamma-radiation due to its low activity.
Chemical separation is needed before the nuclear measurement of all the isotopes mentioned above.

A combined radiochemical separation method had been developed that enables the simultaneous determination of Th-230, Th-232, U-234, U-235, U-238, Pu-239-240, Pu-238, Am-241, Cm-242, Cm-244, Sr-89, Sr-90, Nb-93m and Nb-94.
Recently, this method has been extended for determination of Np-237 and Zr-93.
The main steps of the method are addition of tracers and carriers, sample destruction, co-precipitation on iron(II) hydroxide and calcium oxalate, separation by extraction chromatography using supported dipentylpentylphosphonate (UTEVA), supported N,N-octylphenyl-di-i-butylcarbamoylmethyl phosphine oxide (TRU) and supported bis-(t-butylcyclohexano)-crown(18,6)ether (Sr.Resin), separation on anion exchange resin, alpha- and beta-source preparation, alpha spectrometry, liquid scintillation counting (LSC), gamma spectrometry, measurement by inductively coupled plasma mass spectrometry (ICP-MS).

Np-237 and Zr-93 are pre-concentrated by co-precipitation on iron(II) hydroxide and zirconium oxide, and separated by extraction chromatography using UTEVA.
The key parameter of the method is the adjustment of the oxidation state of the actinoides before adding the sample onto the UTEVA column.
This can be done using many redoxi agents (for example potassium bromate, sodium nitrite or ammonium peroxo disulfate).
Highest yields were achieved when ammonium peroxo disulfate was used with silver nitrate as catalyst.
As even traces of isotopes with mass number near 237 or 93 cause considerable interferences during ICP-MS detection, a purification step by extraction chromatography was inserted.
It was determined by model experiments, that even a small amount of fluoride anions inhibits the retention of Zr on UTEVA, but this problem can be eliminated by addition of boric acid.

Analyzing real samples (evaporation concentrates of a nuclear power plant) 66-97% and 31-66% chemical yields were achieved for Np and Zr, respectively.

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