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Determination of 93Zr, 237Np and 135Cs in radioactive waste by inductively coupled plasma mass spectrometry

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The safety and the behavior of radioactive wastes on a 1000000 year time scale are basically dependent on the radionuclide composition. During such a long time period leakage from the waste packages and the depository into the environment cannot be excluded and radionuclide migrations have to be considered. Therefore it is important to know the exact composition of the various waste forms including the knowledge of the extremely long-lived radionuclides even if they occur at micro or trace quantities. 93Zr, 237Np and 135Cs belong to these exotic nuclides that have half-lives of 1.53·106 y, 2.14·106 y, 2.3·106 y, respectively and that are formed in nuclear fission and activation in nuclear reactors. These nuclides can be detected only after pre-concentration and removal of the sample matrix. They do not emit easy to measure gamma radiation and generally nuclear methods do not offer sufficient sensitivity to measure µBq - mBq activities. These nuclides are preferentially detected by high sensitivity mass spectrometric techniques.
Radiochemical methods have been developed for the separation of Zr, Np and Cs based on extraction chromatography using UTEVA resin*for Zr and Np and AMP resin* for Cs separation. Radioactive tracers were used for chemical recovery determination. Radionuclides were detected by inductively coupled plasma mass spectrometry (ICP-MS). Since isotopic standards are not available for 93Zr and 135Cs determination sensitivity factors were determined by non-isotopic tracers. Three different techniques were used for method validation, i) determination of an isotopic standard solution (237Np), ii) isotope dilution mass spectrometry using enriched 96Zr stable isotope standard solution (93Zr), and iii) measurement by an independent analytical technique, i.e. NAA (135Cs).
Radiochemical procedures, measurement and calibration techniques, as well as results of analysis of 93Zr, 237Np and 135Cs in evaporation concentrates originating from a nuclear power plant will be presented.

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