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Combined radiochemical separation and multicollector ICP-MS approach to determine 135Cs and 135Cs/137Cs isotopic ratio

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The radiological characterization process conducted during the lifecycle of a nuclear facility is paramount in all operative phases of decommissioning, waste management, and site environmental monitoring to enhance safety of workers and public and mitigate the environmental footprint of nuclear technology. Most of radiocesium contamination can be easily detected like 134Cs and 137Cs, and continuously monitored because of the short/medium-term risk of radiation exposure. Besides, the nuclide vector includes hard-to-measure radionuclides like 135Cs (t1/2 above 10E6 y), one of the major radionuclides responsible for the long-term environmental impact of a waste repository because of its high mobility. The recent development of advanced non-radiometric methods coupled with selective radiochemical separations enables the detection of the low abundant pure beta emitter 135Cs by the evaluation of 135Cs/137Cs isotopic ratio that could identify different contamination sources and monitor their dispersion downstream of plant operations. The low abundance of 135Cs needs a high recovery yield from the matrix and an effective separation from potential interfering elements before accurately assessing 135Cs/137Cs isotopic ratio. The removal of polyatomic (95,97Mo) and especially of isobaric (135,137Ba) interferences is the most challenging issue. Notably, the procedure of Cs recovery based on the selective ammonium molybdophosphate (AMP) ion-exchanger and its subsequent complete dissolution introduces a large amount of Mo and Ba contaminants in the eluted Cs solution. This approach requires a complicated purification step. So far, several strategies have been attempted by using combined anion and cation chromatography systems following the Cs pre-concentration.
In this work, a chromatographic system based on the ammonium molybdophosphate polyacrylonitrile (AMP-PAN) resin is being developed and combined with a Multi-Collector ICP-MS (Plasma 3 –Nu Instruments). Firstly, the conditioning, loading, and stripping conditions of AMP-PAN resin have been verified with a surrogate waste containing 200 ppm Cs and 80 ppm Co, Sr, Ni in 1 M HNO3. The effluent concentrations measured at different time intervals showed full breakthrough of Co, Sr and Ni and high retention of Cs (> 90%). Thereafter, the use of a 5 M NH4NO3 stripping solution has led to high Cs recovery (~ 90%) and a negligible amount of Mo. The optimization of selectivity towards interfering elements, especially Ba, is being conducted with further experiments on a multi-element feed solution of 1 –4 M HNO3 containing 20 ppm Cs and 2 ppm Ba, Mg, Al, K, Ca, Mn, Fe, Co, Ni, Zn, Sr, Pb, Bi. To this purpose, several purification strategies are being investigated. Removal of contaminants has been attempted by preliminary co-precipitation (i.e. calcium phosphate), extraction chromatography (i.e. Sr-resin), and washing (i.e. 5 M HNO3) from the AMP-PAN resin to improve Cs selectivity and decontamination factors. To evaluate the proposed method, Cs recovery yield and decontamination factors of interfering elements have been assessed by Q-ICP-MS, while Ba pattern has been detected by magnet scanning of the MC-ICP-MS.
The optimized radiochemical method is being tested and validated with some real samples and reference materials (IAEA 385, 446), where the concentration of interfering elements could be orders of magnitude higher. In these cases, 134, 137Cs have been also monitored by gamma spectrometry. Moreover, laboratory intercomparison could be fostered for the calculation of 135Cs/137Cs isotopic ratio.

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