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Sequential extraction of U, Np, Pu and Am from sediment samples for AMS studies at the Centro Nacional de Aceleradores (CNA, Spain)

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During the last decades, the scientific community has dedicated efforts to expand the list of radionuclides that can be measured by the ultra-sensitive AMS techniques on either traditional or compact facilities. To date, 239Pu, 240Pu and 236U are consolidated radionuclides in AMS. In the last few years, the most minor actinides isotopes have been put in the limelight: 233U, 241Pu, 242Pu, 244Pu and 241Am [1-3]. To make the most of those AMS advances for environmental studies, it becomes necessary to adapt and/or improve the current sample preparation methods according to the specific requirements of those new and more challenging radionuclides.

The Centro Nacional de Aceleradores (CNA, Sevilla, Spain) has hosted a 1 MV multi-elemental AMS system since 2005. This system was the first prototype of compact AMS designed and manufactured by the Dutch company High Voltage Engineering Europa (HVEE, Amersfoort). The measurement techniques for Pu isotopes (i.e. 239Pu, 240Pu, 241Pu, 242Pu, 244Pu), 233U, 236U and 237Np have been implemented at this facility [4-6] and the possibilities for 241Am and 243Am have been recently explored [7]. They are based on the extraction of monoxide anions (e.g. PuO-) from the ion source, the selection of the 3+ charge state (e.g. Pu3+) after the stripping on He gas at about 650 kV (35% yield), and their final analysis from the total energy signal provided by a gas detector. Complementary, important efforts have been made to develop reliable radiochemical procedures for the sequential extraction of actinides from environmental samples, meeting the requirements of the simple design of the 1 MV CNA.

In this work, we present the latest advances in actinides radiochemistry developed at the CNA for sediment samples. The goal is to set up a sequential method allowing the separation of uranium, plutonium, neptunium, and americium from sediments, so that: i) the extremely low concentrations of 233U, 241Pu, 242Pu,244Pu or 243Am can be analyzed, in addition to those of 239Pu, 240Pu, 236U and 241Am; and ii) 237Np can be reliably normalized to the non-isotopic tracer 242Pu. The proposed procedure is based on a Fe(OH)2 co-precipitation of actinides after the leaching of the samples, followed by a sequential extraction of actinides using TEVA® (Pu+Np), UTEVA® (U) and DGA® (Am) resins. The efficient removal of Th from both Pu and U fractions becomes mandatory, due to the constated survival of the molecules 232Th12C3+ and 232Th1H3+ during the analysis of 244Pu and 233U, respectively. Moreover, for 243Am and 241Am analysis, lanthanides (i.e., Dy isotopes) need to be cleaned from the Am fraction to prevent undesired pile-up events from 2+ molecular fragments, making it necessary to introduce an additional TEVA® resin in the procedure. Results on the chemical yield for the different species and on the performance of the method will be presented and discussed.

References

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Student Submission

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