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# Development and validation of isotopic analysis methods by thermal ionization mass spectrometry for small amounts of americium

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One of the missions of the French Alternative Energies and Atomic Energy Commission (CEA), and the Laboratory of Analytical Nuclear Isotopic and Elemental development (LANIE), is to characterise the isotopic and elemental composition of fission products and actinides in irradiated fuels and experimental samples. These analyses are of great importance to extend the fields of qualification of neutron codes and for studies related to the reprocessing and storage of nuclear waste, requiring consequently the lowest possible uncertainties. The isotopic ratios determination of americium, a minor actinide, is particularly challenging by multi-collection Thermal Ionization Mass Spectrometer (TIMS) and Inductively Coupled Plasma Mass Spectrometer (MC-ICP-MS) as the Am abundance in irradiated samples is in the µg range.

Measurement methods on the TIMS Triton Plus for low quantities of Am were developed with an in-house material, an Am solution purified from an $UO_2$ pellet doped with Am, and with the newly released reference material IRMM-0243.

The implementation of the historical (so-called “sequential”) method for 500 ng showed an excellent accuracy, repeatability and reproducibility of a few per thousand for the ratio $^{243}Am/^{241}Am$ and the ratio $^{242m}Am/^{241}Am$, smaller by three orders of magnitude. An alternative data acquisition method, the Modified Total Evaporation (MTE) based on a particular heating program, was also tested for Am at 250 and 500 ng. This method has so far only been applied for U isotopes, with about 4 µg loaded on the filament to measure precisely minor U isotopes. This approach has both the advantages of a Total Evaporation (TE) for the major $^{241}Am$ and $^{243}Am$ isotopes by integrating the entire signal received and those of a “sequential” method for the minor $^{242m}Am$ isotope by applying half-mass corrections and multiple signal refocussing. It is then possible to get rid of the initial mass deposited and thus facilitating the Am isotopic characterisation for the operator. The first MTE measurements displayed the same level of analytical performance and accuracy than the “sequential” method.

The inter-comparison between three mass spectrometers, the former TIMS VG Sector 54 now decommissioned, the TIMS Triton Plus and the MC-ICP-MS Neptune Plus validated these results.