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Uranium isotopic analysis in micrometer-size particles using multi-collector Secondary Ion Mass Spectrometry (SIMS)

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Secondary Ion Mass Spectrometry (SIMS) technology has been applied for a decade to the analysis of swipe samples collected by IAEA inspectors in and around nuclear facilities, in order to assess the absence of undeclared nuclear activities. The main analytical task is to analyze the uranium isotopic abundances of individual micrometer-size particles present on the swipe. For this purpose, the IAEA implemented in 2010 a new CAMECA IMS 1280 instrument as a replacement for its CAMECA IMS 4f. The IMS 1280 is the only large geometry SIMS commercially available with a design allowing the simultaneous measurement of as many as 5 species in the uranium mass range (e.g., U-234, U-235, U-236, U-238, and U-238H, the latter being used to correct the U-235 hydride contribution to the U-236 signal) using 5 electron multiplier detectors (EMs).

There are two difficulties inherent to particle analysis using SIMS: i) it is not possible to increase the sputtered volume (which is limited by the particle size) in order to improve the counting statistics of a respective measurement. As a result, useful ion yields (referring to the resultant of ionization yield, ion transmission and detection factors) need to be improved if higher precision analysis are desired; ii) the sputtering of sub-micrometric particles to a nearly complete consumption results in transient signals having a large dynamic range (typically from 105 cps to as low as a few cps). To overcome this challenge, the use of multi-collection proved to be highly beneficial for isotopic analyses in particles compared to mono-collection (i.e. a single collector), since it considerably increases useful ion yields and negates measurement uncertainty terms inherent to sequential measurement of transient signals.

However, multi-collection is likely to introduce additional measurement uncertainty terms in isotopic measurements compared to mono-collection. These have been carefully investigated and an evaluation of the capability of the IMS 1280 multi-collector system to measure U-234 and U-236 at ppm-level abundances will be presented in this paper.

First, we demonstrate the performance of the miniaturized EMs fitted to the multi-collector system and compare to common mono-collector EM systems, paying special attention to detector gain linearity and drift. Secondly, we quantify the contribution of cross-talk effects between EMs to the measurement uncertainty (i.e. the noise recorded on a detector when one of the adjacent detectors is exposed to an intense signal). We observed that the cross-talk contribution is a result of the limited abundance sensitivity, hence peak-tailing, that can be measured on the mono-collector EM detector. The contribution of the U-238 peak tail to the U-236 peak is estimated as being 1 ppm. Finally, a detailed monitoring of the crucial EM parameters over several months revealed different ageing rates between EMs dedicated to major and minor isotopes. An adequate gain inter-calibration strategy has been established to correct for this effect.