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Data Evaluation Strategies for U Isotope Ratio Measurements of Single Particles by Laser Ablation – Multi Collector – Inductively Coupled Plasma Mass Spectrometry

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The knowledge of the U isotopic composition of single particles is highly relevant for international safeguards and nuclear forensics as it helps to detect hidden and undeclared nuclear processes and activities. International Atomic Energy Agency (IAEA) inspectors are collecting U containing particles during inspections of nuclear facilities either in the facility itself (e.g. nuclear reactor, enrichment facility) or in the nearby environment with sizes typically in the low micrometer range. Such particles are emitted during all kinds of operations where nuclear material is involved. Thus, the U isotopic composition of single particles does reflect past and on-going nuclear processes. The advantage of the direct analysis of individual particles is that signatures of unknown isotopic compositions can be detected even if the particles are embedded in a dust matrix or in a matrix with known or natural U isotopic composition. Therefore the U isotopic signature of individual particles is an excellent tool for identifying the origin and the history of nuclear material.

The aim of this work is to fully validate a method for U isotope ratio measurements of single, μm-sized particles analyzed with laser ablation – multi collector – inductively coupled plasma mass spectrometry (LA-MC-ICPMS). A 'Nu Plasma HR' (Nu Instruments) MC-ICPMS was coupled to a 'NWR-193' (Electro Scientific Industries, Inc.) ns-laser ablation system for the simultaneous determination of ^{234}U/^{238}U, ^{235}U/^{238}U and ^{236}U/^{238}U isotope ratios. The data evaluation strategy for isotope ratio measurements is regarded as a crucial step for the determination of the U isotopic compositions of single particles. Emerging questions are mainly related to correction procedures (e.g. mass bias correction, dead time correction, correction of ^{238}U^{+} peaktailing, UH^{+}/U hydride rate correction,...), uncertainty calculations and the assessment of main contributors. In addition, strategies have to be found for dealing with large amounts of data in a most efficient and operator-independent way. The evaluation strategy for the U isotope ratios of glass particles doped with U of certified isotopic compositions will be discussed as well as the strategy applied for the data reduction of single U particles with multiple isotopic compositions deposited on graphite planchets. The latter originated from the NUSIMEP-7 (Nuclear Signatures Interlaboratory Measurement Evaluation Programme) campaign, which was organized by the Institute for Reference Materials and Measurements (IRMM), European Commission Joint Research Centre, in 2011.