Geophysical Research Abstracts
Vol. 13, EGU2011-6621-1, 2011
EGU General Assembly 2011

Application of mass spectrometric techniques for the determination of n(U-235)/n(U-238) isotope ratios in depleted to low enriched U material

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Thermal ionization mass spectrometry is a state-of-the-art technique for isotope ratio measurements. As such, in many applications it remains the method of choice in nuclear material sample analysis, in particular when it comes to the determination of U and Pu isotopic fingerprints of IAEA safeguards-related samples. However, the IAEA employs, continuously evaluates and develops complementary analytical techniques in order to obtain a complete picture of a sample's nuclear history and to provide a strong metrological basis for analytical measurements by reference techniques. In the work described in this paper, a multi-collector thermal ionization mass spectrometer (MC-TIMS, Triton, Thermo Scientific) and a multiple collector–inductively coupled plasma–mass spectrometer (MC-ICP-MS, Nu Plasma, Nu Instruments Limited) were utilized for the determination of n(U-235)/n(U-238) isotope ratios in four U-containing solutions that were used for the REIMEP 18 campaign, which was organized by the Institute for Reference Materials and Measurements (IRMM), European Commission Joint Research Centre, Belgium. The REIMEP 18 campaign is an European inter-laboratory measurement evaluation programme, in which the participating laboratories were asked to measure uranium isotope amount ratios; i.e. n(U-234)/n(U-238), n(U-235)/n(U-238) and n(U-236)/n(U-238); in 0.5 M nitric acid solutions containing 2.5 mg of depleted to slightly enriched uranium. These solutions were used for the presented work as they are similar to those available for nuclear material accountancy measurements, in which States' declarations on the quantity and the isotopic composition of nuclear material (mostly U and Pu) are verified.

The aim of this work is to perform a direct comparison and evaluation of both techniques for the determination of n(U-235)/n(U-238) isotope ratios that range from depleted to low-enriched U-235, i.e. n(U-235)/n(U-238) = 0.007 254 2(36), 0.035 470(18), 0.004 379 4(27), and 0.024 233(12). In many applications, MC-TIMS measurement results show superior precision and accuracy. But in comparison to MC-ICP-MS, TIMS often requires more time-consuming sample preparation and larger sample amounts. Two different analytical methods — total evaporation (TE) and modified total evaporation (MTE) — were used for the TIMS measurements. MC-ICP-MS, as well as TIMS measurements (TE and MTE), yielded n(U-235)/n(U-238) isotope ratios that are in agreement with the certified values and respective uncertainties of the REIMEP 18 samples. MC ICP S analyses of the four REIMEP 18 samples yield relative deviations to respective certified values that are smaller than 0.07% (average of five to six replicates per sample). Expanded uncertainties are smaller than 0.3 % (k = 2). TIMS MTE and TE measurements, on the other hand, provide relative deviations that are smaller than 0.02 % (average of 6 replicates per sample) and 0.03 % (average of 6 replicates per sample), respectively. The expanded uncertainties are < 0.08 % (k = 2). Both techniques are therefore appropriate for the measurement of the major U isotope ratio. However, selection of the method has to be adapted according to the requested analytical problem (e.g. analysis time, chemistry and amount of the sample, detection limits, required accuracy and uncertainty).