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Applications of New Synthetic Uranium Reference Materials for Research in Geochemistry

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For many applications in geochemistry research isotope ratio measurements play a significant role. In geochronology isotope abundances of uranium and its daughter products thorium and lead are being used to determine the age and history of various samples of geological interest. For measuring the isotopic compositions of these elements by mass spectrometry, suitable isotope reference materials are needed to validate measurement procedures and to calibrate multi-collector and ion counting detector systems. IRMM is a recognized provider for nuclear isotope reference materials to the nuclear industry and nuclear safeguards authorities, which are also being applied widely for geochemical applications.

Firstly, the double spike IRMM-3636 with a $^{233}U/^{236}U$ ratio of 1:1 was prepared which allows internal mass fractionation correction for high precision $^{235}U/^{238}U$ ratio measurements. The ^{234}U abundance of this double spike material is low enough to allow an accurate and precise correction of $^{234}U/^{238}U$ ratios, even for measurements of close to equilibrium uranium samples. The double spike IRMM-3636 is offered in 3 concentrations: 1mg/g, 0.1mg/g and 0.005mg/g. Secondly, the 236U single spike IRMM-3660 was prepared and is offered in 3 concentrations: 1mg/g, 0.1mg/g and 0.01mg/g. Thirdly, a "Quad"-isotope reference material, IRMM-3101, has been prepared which is characterized by $^{233}U/^{235}U/^{236}U/^{238}U$=1/1/1/1. This material is useful for checking Faraday cup efficiencies and inter-calibration of MIC (multiple ion counting) detectors. The quad-IRM is offered in 3 concentrations: 1mg/g, 0.1mg/g and 0.01mg/g.

As one example for the significant influence of synthetic reference materials for geochemical research, the IRMM-074 series of gravimetrically prepared uranium mixtures for linearity testing of secondary electron multipliers (SEMs) has been applied for the redetermination of the secular equilibrium $^{234}U/^{238}U$ value and the ^{234}U half-life by Cheng et al (2009). Due to the use of IRMM-074, results with smaller uncertainties were obtained, which are shifted by about 0.04% compared to the commonly used values published earlier by Cheng et al. in 2000. This has a significant impact for U isotope measurements in geochemistry.

As another example for a geochemical application, by using the new double spike IRMM-3636, the $^{235}U/^{238}U$ ratios for several commonly used natural U standard materials from NIST/NBL and IRMM, such as e.g. NBS960 (=NBL CRM-112a), NBS950a,b and IRMM-184, have been re-measured at IRMM and other laboratories with improved precision and accuracy. The (preliminary) new result of 137.839(24) for the $^{238}U/^{235}U$ ratio of NBL CRM-112a is deviating by -0.030% from the well-known and widely used old consensus value of 137.88. For this old consensus value no uncertainty has ever been assigned, but it is outside the uncertainty limits of the new measurement result. The new result is based on measurements made at several laboratories worldwide, such as University of Frankfurt (Germany), National Taiwan University, NERC (University of Nottingham, UK), UNM (University of Minnesota, US), Thermo Fisher Scientific, LLNL (Lawrence Livermore National Laboratory, US.DOE), SAL/IAEA and IRMM. The (preliminary) new result of 137.839(24) can therefore be proposed as a new consensus value for the $^{238}U/^{235}U$ ratio of NBL CRM-112a. In contrast to the older consensus value, this new result is traceable to the common SI system of units and has an uncertainty assigned to it.

For the close to natural standard IRMM-184, the re-measured $^{238}U/^{235}U$ ratio of 137.683(23) agrees quite well with the certified value of 137.697(41), the calculated difference is only -0.010(35)% which is insignificant. As a

conclusion, the IRMM-3636 Double Spike has been successfully applied for measurements of important uranium isotopic standards like NBL CRM-112a and IRMM-184, with improved uncertainties at the level of 0.016% and traceability to the SI system.