High-Precision Isotope Analysis of Uranium and Thorium by TIMS

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The U.S. Geological Survey (USGS) Yucca Mountain Project Branch laboratory in Denver, Colorado, conducts routine high-precision isotope analyses of uranium (U) and thorium (Th) using thermal ionization mass-spectrometry (TIMS). The measurements are conducted by a solid-source mass-spectrometer equipped with a Faraday multi-collector system and an energy filter in front of an active-film-type secondary electron multiplier (SEM). The abundance sensitivity of the instrument (signal at mass 237 over ^{238}U in natural U) with the energy filter is $\sim 15 \times 10^{-9}$ and peak tails are reduced by a factor of ~100 relative to the Faraday cup measurements. Since instrument installation in April 2004, more than 500 rock and water samples have been analyzed in support of isotope-geochemical studies for the U.S. Department of Energy's Yucca Mountain Project.

Isotope ratios of sub-nanogram to microgram U and Th samples are measured on graphite-coated single-filament and double-filament assemblies using zone-refined rhenium filaments. Ion beams less than 5 millivolt (mV) are measured with the SEM, which is corrected for non-linearity on the basis of measurements of National Institute of Standards and Technology (NIST) U-500 and 4321B standards with ion beams ranging from 0.01 to 8 mV. Inter-calibration between the SEM and the Faraday multi-collector is performed for every mass cycle using a ~5 mV beam switched between Faraday cup and SEM ("bridging" technique), because SEM-Faraday inter-calibrations prior to the

measurement failed to produce acceptable results. Either natural (^{235}U) or artificial (^{236}U, ^{229}Th) isotopes were used for the bridging. Separate runs are conducted for minor isotopes using SEM only. These techniques result in high within-run precisions of <0.1 to 0.2 percent for $^{234}U/^{238}U$ and 0.2 to 0.5 percent for $^{230}Th/^{238}U$.

The performance of the instrument is monitored using several U and Th isotope standards. The mean measured $^{234}U/^{238}U$ in NIST SRM 4321B is (52.879±0.004) $\times 10^{-6}$ (95 percent confidence, n=134). The mean fractionation factor in these analyses is 0.09±0.06 percent per mass unit. The $^{236}U/^{238}U$ value of ~1.1×10^{-9} measured in this standard defines an upper limit for the ^{236}U background in the analyses. The mean atomic $^{234}U/^{238}U$ and activity $^{230}Th/^{238}U$ for 29 runs of an internal "secular equilibrium" uranium ore standard is (54.82±0.02) $\times 10^{-6}$ and 1.002±0.002, respectively. The Th isotopic composition in the IRMM-036 standard was obtained after addition of high-purity ^{229}Th tracer and measuring $^{232}Th/^{229}Th$ and $^{230}Th/^{229}Th$ ratios in separate lower- and higher-temperature runs, respectively. The resulting $^{230}Th/^{232}Th$ for 30 runs is 31.09±0.04 $\times 10^{-7}$, in excellent agreement with the certified value of 31.13±0.78 $\times 10^{-7}$.

The USGS laboratory measured four samples of depleted to low-enriched uranium in the recent Regular European Inter-laboratory Measurement Evaluation Program (REIMEP-18). The results of these analyses are within 0.1 to 2 percent of the certified values.