Isotopic analysis of lead in sub-nanogram quantities by TIMS using ^{202}Pb-^{205}Pb spike: application to geochronology and cosmochronology

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We present a method for isotopic analyses of Pb in sub-nanogram quantities by thermal ionization mass spectrometry with fractionation correction using ^{202}Pb-^{205}Pb spike. The procedure is simple, fast, easy to automate, and requires only one analysis of Pb per sample to acquire both fractionation-corrected Pb isotopic ratios and Pb concentration. The procedure is evaluated by analysis of multiple loads of NIST Pb isotopic standards SRM-981 and SRM-982, and Pb separated from USGS standard rock BCR-1 and chondrite Bruderheim, with the amount of Pb per load between 0.05-3.0*10^{-9} g. The data have been corrected using both the double spike (DS) fractionation correction described here, as well as by conventional external normalization (EN) procedures, thus providing a direct comparison between the correction methods on exactly the same analytical runs. The enhancement in precision and reproducibility achieved by DS normalization varies with the quantity of Pb analysed. For amounts of Pb of less than 0.2 ng, the improvement in precision and reproducibility of ^{207}Pb/^{206}Pb ratios measured on Farady cups (10^{11} Ohm) is small but becomes more substantial with increasing sample size: 3-10 times lower within-run errors, and 2-3 times better reproducibility are obtained for 0.5-3.0 ng loads. Even greater enhancement in precision and reproducibility is achieved for the ^{208}Pb/^{206}Pb ratio, at twice the relative mass difference between isotopes. The advantage of DS for the ^{204}Pb/^{206}Pb ratio is, however, much smaller. This indicates that factors other than fractionation control precision and accuracy of isotopic ratios measured at the ion beam levels below 2-3*10^{-13} A - the intensity typically observed for ^{204}Pb in all samples smaller than 2-3 ng, and for all Pb isotopes in samples smaller than ca. 0.1 ng. The method described here is applicable to high-precision Pb-Pb and U-Pb dating of milligram quantities of meteorites and their components, and single, microgram-size grains of zircon and other U-bearing minerals. The technique described here had been used for dating early solar system materials (Ca-Al rich inclusions and primitive achondrites) containing 0.01-0.3 ng Pb per fraction, and yielded Pb-Pb isochrons with the age error of 0.16 - 0.33 million years, which was previously unattainable for so small samples.