Goldschmidt 2000
September 3rd–8th, 2000
Oxford, UK.

Journal of Conference Abstracts
Volume 5(2), 506

Accurate Measurement of Uranium and Thorium Isotopic Ratios in Low-Uranium Natural Carbonates Using Multi-collector ICP-MS

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Accurate measurement of uranium and thorium isotopic ratios can allow late Quaternary carbonates to be dated using the 238U/234U/230Th decay system. Traditionally, these isotope ratios have been measured by alpha-spectrometry, a slow process involving large sample volumes. Over the last decade, thermal ionisation mass spectrometric methods have enabled more precise measurements on smaller sample volumes, with considerably shorter analysis times. However, this method remains labour-intensive, and is ultimately limited by the very poor thermal ionisation efficiency of thorium. Recently a number of workers have reported isotope measurements in the 238U decay chain using multi-collector ICP-MS, in which thorium ionises considerably more efficiently.

A Micromass Isoprobe MC-ICP-MS instrument at Royal Holloway, University of London typically achieves sensitivity on uranium of 400-500 Volts/ppm, or ca. 2% sample-to-collector efficiency, using a Cetac Aridus desolvating nebuliser. Thorium sensitivity is about half of this figure, allowing precise isotope ratio measurements on greatly reduced sample sizes, relative to TIMS. Internal precisions of 0.1% are routinely attained on 234U/ 238U measurements, from only 5 ng of uranium, equivalent to an acquisition time of a few minutes. Corresponding thorium isotopic ratios can be measured on the same solution, to a precision of 0.2% on 230Th/ 232Th ratios as small as 5ppm.

Accurate measurements of 234U/238U and 230Th/ 232Th require precise determinations of not only the raw ratios, but of mass discrimination, Daly/Faraday collector gain and instrument background. When the cumulative errors from measurements of these factors are propagated through to a final isotope ratio, the internal precisions above typically increase to ca. 0.3% (2 S.E.), for sample and standard measurements of 5 minutes. Repeat measurements of standards indicate that external precision and accuracy of 234U/238U and 230Th/ 232Th ratios calculated in this way are comparable to their fully propagated internal precisions.

The advantages of MC-ICP-MS over TIMS in this field are two-fold. Chemical preparation time is greatly reduced, as samples are run as solutions and do not require the same degree of purification as samples for TIMS measurement. This allows more samples to be analysed for given time and cost constraints, allowing the derivation of more detailed age-depth functions and making part-sample isochron techniques more viable. Secondly, the capability to obtain precise data from considerably reduced sample volumes potentially allows greater sampling resolution, and the dating of samples too small to analyse by TIMS.