In situ U-Pb zircon dating using LA-MC-ICPMS and a multi-ion counting system

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LA-MC-ICPMS allows the rapid and precise *in situ* determination of isotope ratios in minerals. The aim of this work was to date zircons using the U-Pb isotope system.

In order to achieve this, it was necessary to obtain $^{207}Pb/^{206}Pb$ and $^{206}Pb/^{238}U$ ratios with a precision of around 2% and 5% respectively (95% confidence limit). This rapid method allows a relatively large number of individual data to be collected, which enables statistics on ages suitable for sound geochronological investigation. The currently available 213 nm UV laser, which uses ablation in He, provides a more controlled ablation and smaller particles than lasers of longer wavelength. Because the laser ablation process produces rather erratic transient signals, it is necessary to collect the data over a short period of time and in static mode. In addition, a soft ablation at high spatial resolution (20 µm) produces a limited amount of ionised particles. For these reasons, multi-collection is required. Furthermore, for young zircons containing a low amount of Pb (e.g. 10 ppm), Faraday detectors are insufficient and a multi-ion counting system is thus required. The dispersion of the variable multi-collector array is specifically designed to simultaneously measure ^{204}Pb, ^{206}Pb, ^{207}Pb, ^{208}Pb masses on four ion counters, while ^{238}U ions can be collected either with a Faraday cup or an attached ion counter. In addition, at low mass levels, another ion counter is dedicated to simultaneous ^{202}Hg measurement.

We corrected for ^{204}Hg originated from gas and mass discrimination of the actual mass spectrometer. Then, after common-Pb correction, the resulting radiogenic $^{207}Pb^*/^{206}Pb^*$ ratio is quite close to the expected value for the zircon standard. This means that the actual laser system does not significantly fractionate Pb/Pb ratios. In contrast, the precision on U/Pb determination is constrained by the contrasting volatile properties of U and Pb and any subsequent ablation-related fractionation. An analytical procedure is proposed that provides reliable ages and results on reference zircons already dated using ID-TIMS or SIMS.

Characterization of Nd, Te and U isotope ratios in UO_2 using SIMS

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Isotopes provided the initial evidence for the 2 Ga naturally occurring fission reactors discovered at Oklo and Okelobondo in Gabon, Africa and have been subsequently applied to characterize reactor operation conditions and the migration of actinide and fission product elements. However, the application of SIMS has been largely limited due to analytical challenges. Uraninite (UO_2), which forms the core of these natural reactor zones represents an extremely complex matrix, containing measurable concentrations of most elements between $70<A<160$. Furthermore, the isotopic ratios for a given element have been measurably altered from the range in typical terrestrial rocks. Therefore, nearly all isotopic measurements of reactor zone material have been based on chemical separation and analysis using TIMS or ICP-MS. This has limited the spatial resolution as well as the number of elements and samples analyzed. Note that these same complicating analytical factors also provide the evidence for our understanding of reactor operation.

Ratio	Reactor Zone Range
$^{235}U/^{238}U$	0.00543-0.00722
$^{143}Nd/^{144}Nd$	0.859-0.978
$^{145}Nd/^{146}Nd$	0.591-0.658
$^{146}Nd/^{144}Nd$	0.516-0.589
$^{125}Te/^{128}Te$	0.107-0.244
$^{130}Te/^{128}Te$	3.965-5.387

Here we present the use of SIMS to measure isotopic ratios of Nd, Te and U isotopes (Table). Elements and isotopes were selected based on instrumental considerations (e.g., interferences and relative abundances) and for their sensitivity to reactor operation conditions. To minimize compound ion interferences, extreme energy filtering (100 - 200V) was used with O^- (for U and Nd) and Cs^+ (for Te) primary beams. Instrumental mass fractionation was corrected by measuring reactor zone uraninite previously characterized by TIMS or ICP-MS. Analysis of samples distributed across the Okelobondo reactor zone provided insights into the heterogeneity of isotopic ratios at various length scales (µm to m). In conjunction with computer simulations of reactor operation, these isotopic ratios were used to constrain the reactor operating conditions.