Ion microprobe U-series dating and cathodoluminescence of secondary opal at Yucca Mountain, Nevada

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Understanding past unsaturated-zone (UZ) hydrology in the proposed high-level radioactive waste repository at Yucca Mountain, Nevada, is important for understanding responses to future climate change. Secondary opal and calcite on floors of natural cavities record water flow through fractured tuff over the past 12.8-m.y. Because of the very slow mineral growth rates (μm/k.y.), sampling and dating methods must use the finest possible spatial resolution. Opal with tens to hundreds of μg/g U and negligible Th was analyzed by sensitive high-resolution ion microprobe (SHRIMP) using a 30-μm-diameter primary O^- beam. Precision for $^{230}Th/^{238}U$ and $^{234}U/^{238}U$ data depends on U content and age, and ranges from 5 to 25% (2σ) due to small $^{230}Th^{16}O$ and ^{234}U ion-beam intensities. Despite these analytical errors, $^{230}Th/U$ and model $^{234}U/^{238}U$ ages more accurately represent timing of deposition than ages obtained from milligram-sized samples by high-precision thermal ionization mass spectrometry.

U-series ages for profiles across individual opal hemispheres increase from 20 to 50 ka for outermost layers to >1,200 ka for inner layers. Depth-age relations yield growth rates from 0.2 to 3 μm/k.y. for different samples. Although growth rates change abruptly in some profiles, these changes are not cyclic as expected for climate-induced variations in fluid flux. Instead, growth rates are remarkably uniform for the outer 200 to 400 μm of individual hemispheres, which spans the last several climate cycles. Depth-age relations for opal intergrown in the outer parts of calcite-rich coatings indicate similar μm/k.y. growth rates for U-poor calcite.

Oscillatory zoning in opal is present in cathodoluminescence (CL) images. CL intensity is positively correlated (R^2=0.95) with 100-fold differences in U concentration between lightest and darkest zones. Patterns of CL cyclicity and intensity correspond to ~100-k.y. climate cycles in some profiles; however, evidence for correlations in CL patterns between different samples is not obvious. Causes for cyclic U variation within single profiles may be related to variable U concentration in infiltration or differences in the degree of subsurface evaporative concentration at different times.

These CL data, along with the age framework and growth rates provided by opal SHRIMP dating, help elucidate the hydrologic response to climate change at Yucca Mountain.

In situ U-series dating by laser-ablation MC-ICPMS

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The capabilities and potential applications of *in-situ* dating of Quaternary materials using laser ablation-MC-ICPMS are explored. $^{234}U/^{238}U$ and $^{230}Th/^{234}U$ can be measured with precision sufficient for dating at a spatial resolution of better than 100 μm in samples that contain as a little as 1 ppm uranium. Moreover, U and Th concentrations and U-series isotope ratios can be continuously profiled to determine changes in age that occur with sample growth (e.g. in speleothems). These capabilities further permit the dating of bones, teeth and possibly molluscs, which are subject to post-mortem open-system behaviour of U-series isotopes (cf. Pike et al., 2002). They can also be employed to elucidate processes of U-series migration during weathering and diagenesis. A drawback of laser ablation-MC-ICPMS is that it cannot in general provide U-series age estimates with precision equivalent to conventional TIMS or solution MC-ICPMS methods. This reflects, in part, the inability to reproduce with very high precision the elemental Th/U ratio due to fractionation processes that occur at the site of ablation and within the Ar-ICP. However, sample preparation is straightforward, the amount of sample consumed negligible, and the technique may be used to rapidly characterise and/or screen and select samples from which more precise and accurate dates can be obtained using conventional methods. Given further instrumental developments and the establishment of suitable matrix-matched standards for carbonates and other materials, laser ablation-MC-ICPMS will play an increasingly important role in Quaternary dating and research.

Reference

Pike, A.W.G., Hedges, R.E.M., and Van Calsteren, P., 2002. *Geochim Cosmochim Acta* 66, 4273-4286.