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U redox state tracked in mineralized hydrothermal carbonate: implications for U-Pb geochronology

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In-situ U-Pb carbonate geochronology is a well-established technique that directly constrains the timing and rates of important geological processes including fluid flow, diagenesis, and tectonic events. However, the fundamental controls on U behaviour in carbonate minerals remain unclear, limiting geological interpretations. Knowledge gaps include the controls on U incorporation, the highly heterogeneous distribution of U at a crystal scale, and the incorporation of U with respect to other (redox-sensitive) elements. The application of synchrotron X-ray microspectroscopy (µXAS) is ideal for investigating these topics, as it can map chemical changes and measure the valance state of key elements at the micron-scale.

Here we combine µXAS and in-situ laser ablation U-Pb carbonate geochronology to temporally track U distribution and redox state in a porphyry-epithermal system. Multiple generations of carbonate minerals record fluid conditions and processes which control the solubility and deposition of metals, including U. Results show that temporally distinct generations of carbonate record both oxidized UO_2^{2+} and reduced U^{4+} species within a single sample section. Mapping of individual carbonate crystals reveals that UO_2^{2+} and U^{4+} also occur within individual growth bands at a sub-millimetre scale, and in rare samples, appear to coexist. µXAS data from the sample suite demonstrate that local fluid conditions in the case-study mineralized system changed from more oxidized to more reduced over a period of ca. 16 Ma.

The preservation of two U oxidation states during discrete precipitation events requires U retentivity within older domains, indicating that the U-Pb carbonate geochronometer is robust under hydrothermal conditions (e.g., ~200–350ºC) and through rapid local redox state changes. Furthermore, crystal zones with abundant fluid/vapour inclusions linked to boiling processes coincide with higher levels of U in the carbonate and favourable U/Pb. Our results suggest redox changes and boiling conditions may be critical for both the deposition of ore minerals, as well as increased U uptake in carbonate minerals. Targeting carbonate domains with these features may therefore increase success for U-Pb geochronology. U-Pb carbonate dating combined with µXAS can track the temporal evolution of processes critical for metal deposition in long-lived and multistage hydrothermal-magmatic ore deposit settings.