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Quantifying the replacement of metamict zircon by hydrothermal zircon using isotope doping at mid-crust P-T conditions

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The replacement of metamict zircon by hydrothermal zircon involves structural and compositional changes, making it an excellent system for studying the timing of hydrothermal events. Hydrothermal zircon is often used as geochronometer for post-magmatic alkaline silicic magmatic intrusions, fluoride mineralization, and orogenic gold deposits. However, the answer to how hydrothermal zircons were formed remains elusive. One plausible explanation is that compared to crystalline zircon, radiation-damaged zircon (metamict zircon) has a higher Gibbs free energy and aqueous solubility, enhancing the dissolution of metamict zircon in hydrothermal fluids, which provides Zr4+ to precipitate hydrothermal zircon. The characteristic porosity in hydrothermal zircon appears to support this hypothesis. Thermodynamic modeling using the SUPCRTBL (Zimmer et al., 2016) database showed that hydrolysis of NaF can enhance the solubility of zircon (Ayers and Zhu, 2025), and previous studies have shown enhanced replacement in fluids containing NaF. To test the hypothesis, we performed cold-seal pressure vessel experiments at 600-800° C and 0.2 GPa using zircon with varying degrees of crystallinity. The experimental fluids contained isotopically doped SiO_2 ± NaF and significantly depleted in ^{18}O. The run products showed partial replacement of the metamict zircons in fluids containing NaF but not in fluids without NaF. SIMS Si-O isotope analysis revealed distinct isotopic compositions between the hydrothermal zircon and the metamict zircon. The extent of replacement calculated from isotopic dilution was 30% after 14 days in 1 molal NaF at 800° C. The theoretical and experimental findings will help improve our understanding of the genesis and preservation of hydrothermal zircons and their U-Pb ages.

Zimmer K., Zhang Y. L., Lu P., Chen Y. Y., Zhang G. R., Dalkilic M. and Zhu C. (2016) SUPCRTBL: A revised and extended thermodynamic dataset and software package of SUPCRT92. Computers & Geosciences 90, 97-111.

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