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Oxidized fluids fed Earth's earliest hydrothermal systems

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Zircon ($ZrSiO_4$) has shaped our understanding of early Earth and the mineral has helped to establish the conditions of our planet during a time in which life may have emerged. Zircon chemistry has been applied to understand the origins of crust, to provide evidence for weathering at or near Earth's surface, and to infer the subsequent burial and assimilation of this weathered material into igneous systems in ways that would not otherwise be possible [1,2]. Also of importance are high temperature fluid-rock interactions that occurred during the first 700 million years of our planet's history. These fluids represent a critical communication pathway between Earth's interior and the near surface, possibly including hydrothermal pools. We report zircon-fluid partitioning experiments of redox-sensitive Ce and Eu (and other rare earth elements) under oxygen fugacity buffered conditions. The zircon-fluid partition coefficients for Ce and Eu are a function of oxygen fugacity and temperature. When the experimental results are applied to detrital Jack Hills zircons with ages approaching 4 Ga that have subsolidus crystallization temperatures (average = 580 ºC based on the Ti-in-zircon thermometer [3]), the average calculated oxygen fugacity is more oxidized than the fayalite magnetite quartz equilibrium. This is the oldest reported oxygen fugacity for lithosphere fluids, which are more oxidized than the terrestrial mantle during this time. Other geochemical lines of evidence for these zircons ($\delta^{18}O$, $[Cl]_{zrc}$ [4]) provide evidence for communication with near surface aqueous systems – e.g., hydrothermal pools. We also conducted geochemical modeling using the EQ3/6 software package with fluid and rock compositions (and oxygen fugacity/temperature) broadly characterizedÂby the geochemistry of zircon. This modeling constrains the behavior and transport of metals (e.g., Zn, Cu), some of which are hypothesized to be important for the origin of life.

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