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Redox-controlled dissolution of monazite in fluids and implications for phase stability in the lithosphere

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Abstract

Monazite is an important host of rare earth elements in the lithosphere including redox-sensitive Ce, which may occur as trivalent and tetravalent in terrestrial environments. Here, monazite solubility is explored as a function of oxygen fugacity through a series of dissolution experiments in alkali-rich and H_2O fluids at 925 °C and 1.5 GPa. The oxygen fugacity was controlled with seven different solid-state buffers and ranged from about the iron-wüstite to above the magnetite-hematite equilibrium reactions. The solubility of natural monazite increases monotonically at oxygen fugacities equal to or higher than the fayalite-magnetite-quartz equilibrium. Electron microscopy reveals incongruent dissolution at Ni-NiO and above, where Ce-oxide is observed with monazite as a stable phase. Solubility experiments were also conducted with synthetic crystals ($CePO_4$, $LaPO_4$, and Th+Si-doped monazite). End-member $CePO_4$ exhibits profound changes to the surface of the crystal under oxidized conditions, with erosion of the crystal surface to depths of ~100 μm or greater, coupled with precipitation of Ce-oxide. In contrast, the solubility of $LaPO_4$ shows no sensitivity to the redox state of the experiment. The addition of Th (~3 wt%) and Si (~0.3 wt%) to monazite promotes crystal stability under oxidizing conditions, though small ThO_2-CeO_2 (5–10 μm) crystals are present on the surfaces of these crystals, whose abundance increases at higher oxygen fugacities. In aggregate, these experiments show that the stability and solubility of monazite is affected by oxygen fugacity, and that the redox state of a fluid may be partially responsible for redistribution of rare earth elements and phosphorus in the crust. Lithospheric fluids with oxygen fugacities at or above the fayalite-magnetite-quartz equilibrium may contribute to some of the complex textures, variable chemistry, and age relationships observed in natural monazite.

Keywords: Monazite, solubility, incongruent dissolution, oxygen fugacity