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Mobilization and concentration of Nd in fluorobastnaesite-bearing hydrothermal systems

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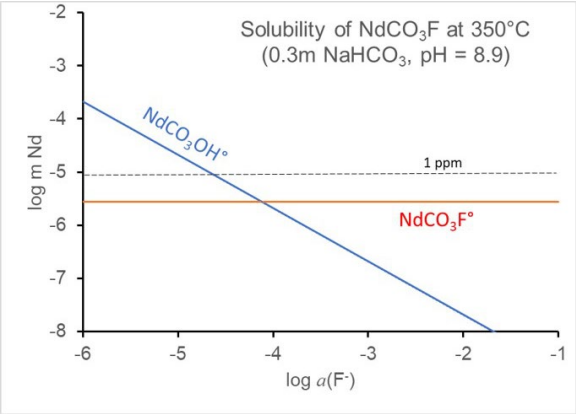

Fluorobastnaesite is one of the most common REE ore minerals and a major industrial source of Nd, a critical element in high demand. It typically forms LREE solid solutions dominated by Ce (bastnäsite-(Ce)) or La (bastnäsite-(La)), with a generalized formula (Ce, La, LREE)CO3F. Bastnaesite ores are commonly linked to alkaline, carbonate-rich hydrothermal fluids. However, the conditions under which Nd concentrates in bastnaesite cannot be quantitatively evaluated because thermodynamic data are lacking for Nd-bearing bastnaesite solids and for the aqueous Nd species that control Nd mobility in carbonate-bearing solutions.

This study is a first step toward closing this gap by determining thermodynamic properties of the Nd-bastnaesite end member (NdCO3F) and investigating its speciation and solubility in aqueous fluids. NdCO3F was synthesized by a hydrothermal sol–gel method in autoclaves at 165–200 °C and held for three months to ensure full recrystallization; products were verified by XRD and Raman spectroscopy. The enthalpy of formation was measured by high-temperature oxide melt drop-solution calorimetry, repeatedly dissolving the sample in sodium molybdate at 800 °C and applying an appropriate thermochemical cycle. Heat capacity (Cp) was measured from 1.8–300 K using a Quantum Design PPMS and from 50–450 °C using a Labsys Evo STA 1600 with a 3D Cp rod. These measurements enabled full thermodynamic characterization of NdCO3F, including enthalpy of formation, Cp(T), and entropy (from integration of low-temperature Cp).

Solubility experiments were then conducted at 100–550 °C in NaHCO3–NaF solutions to constrain Nd aqueous speciation in equilibrium with NdCO3F. Across the full temperature range, the dominant aqueous species was NdCO3F° (congruent dissolution). The results indicate that NdCO3F(s) buffers dissolved Nd to concentrations generally below 1 ppm, supporting the conclusion that fluoride in natural systems acts primarily as a binding rather than mobilizing ligand. Higher Nd concentrations occur only at fluoride activities below ~10^{-4} m, where competition between NdCO3OH° and NdCO3F° becomes significant (figure 1)