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Hydrothermal REE partitioning experiments between fluorite and acidic to alkaline aqueous fluids from 100 to 250 °C

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Fluorite veins are commonly found in critical mineral deposits overprinted by hydrothermal processes [1]. Fluorite can incorporate significant REE through substitution with Ca^{2+} and potentially be used as a vector of fluid-rock interaction. However, the mechanisms controlling fluorite-fluid REE partitioning behaviors have not yet been determined extensively; experimental REE partitioning data are currently limited to 60 °C [2]. This study aims to evaluate the effect of fluorite dissolution on REE mobility, the predominant REE aqueous complexes in equilibrium with fluorite, and the role of secondary mineral precipitation on REE solubility.

Batch-type fluorite solubility experiments were conducted between 100 and 250 °C by equilibrating natural fluorite crystals with acidic to alkaline (pH of 2–10) aqueous solutions of variable salinities (0–15 wt. % NaCl) and REE concentrations. Major elements and REE concentrations of the reacted fluid were used to calculate fluorite solubility, aqueous REE speciation, and secondary REE mineral saturation using the GEMS code package [3].

Etch pit dissolution of the reacted fluorite, coupled with fluid chemistry, indicate that fluorite solubility is favored at acidic pH and with the addition of NaCl, even at alkaline pH. Secondary REE fluorides were only observed to form in the REE-doped experiments. In acidic fluids, REE fluoride complexes were observed to be dominant, whereas the REE hydroxyl complexes controlled the REE solubility in alkaline fluids. The REE chloride complexes only controlled REE solubility when the fluid was both saline and acidic. The REE concentrations of the fluids were dominantly controlled by the dissolution behavior of fluorite. While previous modeling work indicates that the REE are transported via chloride complexation in acidic aqueous fluid [4], new insights gained from our experiments indicate that the REE can also be soluble in: 1) acidic fluids dominated by REE fluoride complexes and 2) in alkaline saline fluids. Both conditions promote the transport of REE in fluids potentially equilibrated with fluorite veins.

[1] Gysi et al. (2016), *Econ. Geol.* 111, 1241-1276; [2] van Hinsberg et al. (2010), *Geology* 38, 847-850; [3] Kulik et al. (2013). *Comput. Geosci.* 17, 1-24; [4] Migdisov and Williams-Jones (2014), *Mineralium Deposita* 49, 987-997.