

What controls the mobility of rare earth elements (REE) in critical mineral deposits in acidic vs. alkaline hydrothermal fluids?

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(Per)alkaline complexes and carbonatites evolve through a sequence of magmatic-hydrothermal processes. Most of them are overprinted by auto-metasomatic processes which involves the mobilization, fractionation and/or enrichment of critical elements in hydrothermal fluids, such as the rare earth elements (REE) [1]. However, our current ability to predict the behavior of REE in high temperature fluids and interpret these natural systems using geochemical modeling depends on the availability of the thermodynamic data for the REE minerals and aqueous species. Previous experimental work on REE solubility has focused on acidic aqueous fluids up to ~300 °C, and considered chloride, fluoride and sulfate as important ligands for their transport [2]. However, magmatic-hydrothermal systems that form these critical mineral deposits may cover a wider range of fluid chemistries spanning acidic to alkaline pH as well as temperatures and pressures at which the fluids are supercritical. A few recently published studies have shown that other ligands (e.g., REE carbonates and/or combined fluoride species) could become important in near-neutral to alkaline fluids [3,4], and that REE mobility can also be increased in saline alkaline fluids reacted with fluorite [5]. Here we present new hydrothermal REE hydroxyl/chloride speciation data and REE phosphate/hydroxide minerals [6,7], and calcite and fluorite solubility experiments as a function of pH, salinity and temperature. We use an integrated approach to link a wide array of experimental techniques (solubility, calorimetry, and spectroscopy) with thermodynamic optimizations using GEMSFITS [8], and present the development of a new experimental database for REE and its integration into the MINES thermodynamic database (<https://geoinfo.nmt.edu/mines-tdb>). The latter permits simulating hydrothermal fluid-rock interaction and ore-forming processes in critical mineral deposits to better

understand the behavior of REE during metasomatism.

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[3] Nisbet et al. (2022), *Chemical Geology*, 611, 121122.

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[6] Gysi et al. (2018), *Geochimica et Cosmochimica Acta*, 242, 143-164.

[7] Gysi & Harlov (2021) *Chemical Geology*, 567, 120072.

[8] Miron et al. (2015) *Applied Geochemistry*, 55, 28-45.