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Solubility and fractionation of light/heavy rare earth elements in hydrothermal fluids up to supercritical conditions

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Hydrothermal aqueous fluids play a key role in the formation of rare earth elements (REE) deposits. The mobility of REE is controlled by the solubility of minerals and the stability of aqueous species. Ligands that complex with REE include Cl^-, F^-, OH^-, CO_3^{2-}, SO_4^{2-}, which depend on pressure, temperature, fluid chemistry and solvent properties. This overview presents thermodynamic optimizations and experimental solubility data for modeling the stability of monazite and xenotime, REE^{3+} aqua ions, and REE aqueous complexes (hydroxyl, chloride, and sulfate species) up to supercritical conditions. Experimental data presented include low-pressure batch-type Teflon-lined reactors (100 to 250°C, *swvp*) and Inconel 625 Parr reactors (350°C and *swvp*, to 450°C and 700 bar), as well as high-pressure in-situ μXRF synchrotron-based measurements in a hydrothermal diamond anvil cell (25 to 550°C, 0.2 to 2 GPa). At low pressure, synthetic single $REEPO_4$ crystals (La, Ce, Nd, Dy, Yb, Y) were equilibrated in hydrothermal solutions from acidic to alkaline across various conditions, including sub- and supercritical conditions, and varying NaCl/HCl/NaOH concentrations. The experimental results indicate that the solubility of monazite and xenotime strongly depends on temperature and pH. REE chloride complexes control the solubility of $REEPO_4$ at acidic conditions, whereas REE hydroxyl complexes, predominantly $REE(OH)_2^+$ and $REE(OH)_3^0$, control solubility at alkaline conditions in supercritical fluids. In acidic fluids, REE concentrations increase significantly, and light/heavy REE fractionation increases with NaCl concentrations due to the stability of REE chloride complexes, leading to higher monazite solubility over xenotime at supercritical conditions. At high pressure and up to 550°C, in-situ μXRF was used to measure the solubility of $NdPO_4$ and $YbPO_4$ in NaCl-HCl and Na_2SO_4-H_2SO_4 solutions down to ~100 ppm. Overall, high pressure lowers REE solubility, and the stability of sulfate complexes can provide an important mechanism for light/heavy REE fractionation. We use an integrated approach to link experimental data with thermodynamic optimizations using GEMSFITS. The updated thermodynamic constants are integrated into the MINES thermodynamic database, which enables simulating hydrothermal ore-forming processes in critical mineral deposits. This work is supported by DOE BES grant DE-SC0026161, and high pressure μXRF experiments through beamtime at the European Synchrotron Radiation Facility.