

Hydrothermal solubility experiments and determination of La hydroxyl complexes at pH of 2 to 5 and temperatures of 150 to 250 °C

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The mobility of rare earth elements (REE) can be assessed in natural systems using thermodynamic modeling [1-2]. Previous experimental studies on REE aqueous speciation have been conducted under acidic conditions for chloride, sulfate, and fluoride complexes [2]. However, the properties of REE hydroxyl species reported by Haas et al. [3] are still used today, despite being theoretical extrapolations from low temperature experimental data [2]. Therefore, our knowledge about the stability and role of REE hydroxyl complexes for REE mobilization in hydrothermal fluids remains unclear.

In this study, we measured the solubility of synthetic La hydroxide in H₂O-HClO₄-bearing aqueous solutions at pH of 2 to 5 between 150 and 250 °C. The experiments were conducted in batch-type Teflon-lined reactors for up to 25 days, after which, the quenched experimental solutions were analyzed for total dissolved La concentrations. Equilibrium speciation, pH, and thermodynamic properties optimizations were calculated using GEMSFITs [4]. A series of kinetic tests indicate that equilibrium between La hydroxide powders and aqueous solutions is approached within 10 to 17 days depending on pH and temperature of the experiments. *A priori*, pH was calculated in the equilibrated solutions using the data by Haas et al. [3]. At 250 °C, the measured logarithm molality of La decreased from -2.4 to -7.2 at pH of 2.9 to 5.0, with a close to linear pH dependence of the measured solubility data. The solubility of La hydroxide predicted from Haas et al. [3] is up to 3 orders of magnitude higher than measured in our experiments, and over predicts the stability of the LaOH²⁺ species. These discrepancies were resolved by optimizing the standard Gibbs energies of aqueous La species (i.e., La³⁺, LaOH²⁺, La(OH)₂⁺ and La(OH)₃⁰). Results from these optimizations indicate that La³⁺ predominates in the studied pH range, and that La hydroxyl complexes are less stable than predicted to date.

[1] Perry and Gysi (2018), *Geofluids*, Article ID 5382480

[2] Migdisov et al. (2016), *Chemical Geology*, 439, 13–42

[3] Haas et al. (1995), *Geochim. Cosmochim. Acta*, 59(21), 4329–4350

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