Goldschmidt 2023 Abstract
https://doi.org/10.7185/gold2023.18464

High temperature monazite-(Nd) solubility experiments to predict Nd complexation at variable pH and salinity in supercritical fluids

DEBARATI BANERJEE[1], LAURA E WATERS[1], NICOLE C HURTIG[2], ALEXANDER P GYSI[1,3], DANIEL HARLOV[4], PROF. CHEN ZHU, M SC PHD[5] AND ARTACHES MIGDISSOV, PHD[6]

[1]New Mexico Institute of Mining and Technology

[2]New Mexico Institute of Mining and Technology, Department of Earth & Environmental Sciences

[3]New Mexico Institute of Mining and Technology, New Mexico Bureau of Geology and Mineral Resources

[4]GeoforschungsZentrum

[5]Indiana University

[6]Los Alamos National Laboratory

Presenting Author: debarati.banerjee@student.nmt.edu

Rare earth elements (REE) mobility in hydrothermal fluids depends on solubility of REE minerals and formation of aqueous complexes, which vary based on fluid composition, pH and temperature. Experimentally-derived thermodynamic properties for monazite solubilities were determined between 100 and 250 °C [1,2]. Here, we determined the solubility of synthetic monazite ($NdPO_4$) crystals at 500-700 °C and 1.7 kbar in aqueous hydrochloric acid solutions of known starting pH ~2 at 25 °C, salinities (0-0.5 mol/kg NaCl), and equilibration times of 96-144 h. Experimental solutions were spiked with ^{145}Nd (0.252-0.265 ppm Nd; 91.18% ^{145}Nd) using the isotope-doping method presented by [3]. Analysis using MC-ICP-MS show total dissolved $NdPO_4$ ranging from 3185.48 to 28.07 ppm comparable to the study by [3] and about 5-6 orders of magnitude higher than at subcritical conditions [1]. For a given temperature, the pH of the saline fluid is higher than for a non-saline fluid. At 500 °C, the most saline fluid has the highest $NdPO_4$ solubility (589.76 ppm) while the non-saline fluid has the lowest Nd solubility (58.82 ppm). At 700°C, the trend reverses, with the non-saline fluid having highest Nd solubility (1867.25 ppm) in comparison to the saline fluid (1045.21 ppm).

The GEMS code package [4] and the MINES thermodynamic database (https://geoinfo.nmt.edu/mines-tdb/) were used to calculate speciation and predict monazite solubility in our experiments. Figure 1 shows that our current models fail to reproduce the experimental data in supercritical fluids >400 °C and necessitate revision of the thermodynamic properties of aqueous Nd hydroxyl and chloride species currently extrapolated from data collected at low temperature. Although GEMS predicts the patterns of Nd solubility as a function of pH, it underestimates total Nd solubility by more than one order of magnitude (14.849 to 4.335 ppm $NdPO_4$), suggesting that thermodynamic properties associated with Nd-speciation reactions must be optimized to understand dissociation mechanisms of $NdPO_4$-monazite at high temperature and pressure.

[1] Van Hoozen et al. (2020), Geochim. Cosmochim. Acta 280, 302-316.

[2] Gysi et al. (2018), Geochim. Cosmochim. Acta 242, 143–164.

[3] Pourtier et al. (2010), Geochim. Cosmochim. Acta 74, 1872-1891.

[4] Kulik et al. (2013), *Comput. Geosci. 17*, 1-24.

Figure 1:Total dissolved NdPO4(ppm) vs Temperature