Goldschmidt 2026 Abstract
https://doi.org/10.46427/gold2026.36694

Experimental Study on Perovskite Dissolution for Improving REE Recovery from Kimberlite (Diamond Mine) Tailings

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Perovskite ($CaTiO_3$) is the principal host mineral for rare earth elements (REE) in most kimberlites. However, perovskite is unstable under certain kimberlite post-emplacement conditions. At the Snap Lake kimberlite (Canada), hydrothermal alteration of perovskite and its replacement by anatase (TiO_2) has resulted in the formation of secondary monazite [1]. The development of monazite is economically significant due to its high REE concentrations and favourable extraction characteristics. Understanding the factors controlling perovskite alteration is therefore critical for assessing REE recovery from kimberlite tailings. Although thermodynamic calculations suggest perovskite is unstable in hydrothermal environments, its breakdown kinetics are slow and unstudied under kimberlite conditions. The effects of temperature, pressure, and CO_2 content are poorly understood due to limited experimental data, mostly from studies on SYNROC, a radioactive waste storage material. This study aims to fill that gap. Experiments were conducted over temperature range from 25 to 450 °C and pressures from 50 to 500 bar in H_2O fluids and H_2O–CO_2 fluids with X_{CO_2} (=CO_2/CO_2+H_2O, mol%) = 0.1, 0.25, and 0.5. Polished synthetic $CaTiO_3$ pellets reacted with the fluids inside titanium reactors heated in a box furnace. Oxalic acid and hydrogen peroxide was used to generate the CO_2–H_2O fluid phase. After the runs, the surface of the pellets was characterized using scanning electron microscopy (SEM) to examine textural changes, X-ray photoelectron spectroscopy (XPS) to assess leaching of Ca^{2+} from perovskite, and experimental fluids were analysed by inductively coupled plasma emission spectroscopy (ICP-OES) to determine dissolved Ca^{2+} concentrations. The Ca:Ti ratio of untreated pellets was close to 1, consistent with stoichiometric perovskite, whereas reacted samples showed Ca:Ti ratios <1, confirming preferential Ca^{2+} removal. Dissolution rate (Ti:Ca ratio/duration*1000) was enhanced in CO_2-bearing fluids, with 48 to 5979 min^{-1} compared to 15 to 1405 min^{-1} in H_2O-only experiments, and dissolved Ca concentrations reaching up to 2317 ppm in CO_2-rich fluids compared to pure H_2O which has up to 127 ppm. These findings demonstrate that fluid composition, particularly CO_2 activity, plays a key role in perovskite instability and has important implications for REE redistribution during post-emplacement alteration and recovery in kimberlite tailings.

[1] Mohammed et al., 2025