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Lanthanide fractionation in calcite records fluid evolution in basalt-hosted and high-pCO_2 systems

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Basalt carbonation, or the engineered conversion of CO_2 to carbonate minerals via reaction with basalt, represents a promising potential climate change solution. Yet reliable verification of the efficacy of this process remains challenging. Here, we explore the potential of rare earth element and yttrium (YREE) fingerprinting of calcite as a potential verification methodology. YREE in calcite is widely used to reconstruct fluid sources and hydrothermal input in basalt-hosted systems, and is therefore a potential low-cost, high-fidelity internal discriminator between previously existing and neo-formed, anthropogenic calcite. However, because YREE contents of calcite could reflect intrinsic mineral–fluid partitioning during precipitation or prior, open-system fluid evolution, the potential of this technique remains unproven. Here, we investigate the relative roles of these partitioning processes via in situ LA–ICP–MS analyses of calcites from IODP Sites 1027C, 1301B, and U1362A in the Cascadia Basin and measurements of fluids and solids from inorganic calcite precipitation experiments conducted under simulated basalt carbonation conditions. Basalt-hosted calcites exhibit heavy-REE enrichment (Nd/Yb = 0.12–0.95), variable positive Eu anomalies (Eu/Eu* up to >10), muted Ce anomalies (Ce/Ce* = 0.82–1.14 relative to East Pacific seawater Ce/Ce* ≈ 0.06), positive Y anomalies (Y/Y* = 1.08–1.97), and systematic variation with depth and porosity type, consistent with progressive seawater–basalt interaction and hydrothermal modification in an open crustal system. The experimentally precipitated calcites cluster tightly in $\log_{10}$(La/Yb)–$\log_{10}$(Yb/Nd) space and overlap their parent fluids (Nd/Yb ≈ 0.08–0.11; Eu/Eu* ≈ 5; Ce/Ce* ≈ 0.2–0.3; Y/Y* ≈ 0.9–1.1). Despite a ~30–60% decrease in dissolved YREE concentrations during precipitation, PAAS-normalized pattern shapes and anomaly indices remain nearly invariant ($T_0 \approx T_F$), demonstrating that closed-system precipitation introduces minimal additional lanthanide fractionation and distinguishes closed-system inorganic precipitation from hydrothermally modified basalt-hosted calcite. Together, the magnitude and geometry of these multi-proxy differences (including enhanced HREE enrichment relative to LREE in natural samples) show that YREE fractionation in calcite primarily records prior open-system fluid–rock interaction rather than precipitation chemistry alone. YREE systematics provide a quantitative fingerprint of system openness and fluid evolution in altered oceanic crust and basalt-hosted CO_2 storage environments.