

Large-scale thermodynamic analysis of mineral carbonation under elevated CO₂ fugacity

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Mineral carbonation is a compelling strategy for the long-term sequestration of anthropogenic CO₂. Many mineral carbonation efforts have focused on reacting CO₂ with minerals containing divalent cations, such as Ca²⁺, Mg²⁺, Fe²⁺, etc. However, under elevated CO₂ fugacity, a variety of other oxide minerals can become thermodynamically-activated to transform into carbonates. Here, we conduct a large-scale analysis of mineral carbonation thermodynamics, surveyed over all known oxide minerals in the Inorganic Crystal Structure Database (ICSD). First, we benchmark the fugacity of CO₂; in the gaseous state as a function of T - p CO₂, in the aqueous state as a function of p H and dissolved [C] concentration, and in the supercritical state as a function of T - P . By combining these calculated CO₂ chemical potentials with DFT-calculated thermochemical data from the Materials Project database, we construct a large map that visualizes the carbonation thermodynamics of all known crystalline oxide minerals. Our findings quantify the propensity of different minerals to transform into carbonates, thereby establishing a thermodynamic baseline for the design of strategic mineral carbonation solutions.