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Carbon Dioxide Sequestration in Sulfate-Rich Magnesium Hydroxide Slurries

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Mineral carbonation using magnesium-bearing slurries is a durable route for CO_2 sequestration but is limited by the kinetics of CO_2 dissolution and speciation, brucite ($Mg(OH)_2$) dissolution, and magnesium carbonate precipitation. We investigated the effect of three organic additives: ethylenediaminetetraacetic acid (EDTA), gallic acid, and sodium diethyldithiocarbamate (DDC), on the carbonation of a synthetic brucite-containing slurry with ionic strengths exceeding 4 M exposed to 5% CO_2. Time-resolved measurements of pH, dissolved inorganic carbon (DIC), and aqueous magnesium were combined with quantitative x-ray diffraction and geochemical modeling to quantify the rate and extent of carbonation over a period of 5 to 12 h. DDC was found to significantly increase both the rate and extent of carbonation experienced by the slurry in trials where the initial slurry was purged with nitrogen for comparatively short timescales (~1 h). When the durations for equilibration of the initial slurry and carbonation are increased, the differences in the carbonation rates of the slurries became less pronounced. Hydromagnesite is the dominant carbonate that precipitates at early checkpoints; whereas nesquehonite accumulates as carbonation is allowed to progress. Final mass balance results for all trials approached predictions made by geochemical modeling. These results provide insights into the feasibility of mineral carbonation for magnesium solutions resembling concentrated industrial brines made from the tailings of ultramafic rocks, where elevated ionic strength and sulfate activity alter the kinetics of the precipitation of magnesium carbonates, identifies the distinct stages of mineral disassociation and remineralization during carbonation of these slurries, and demonstrates the viability of alkaline, magnesium-rich slurries as a means of recapturing carbon emissions released during mining operations.