Goldschmidt 2026 Abstract
https://doi.org/10.46427/gold2026.36981

Carbon Fixation using Seawater for Sustainable Large-Scale Carbon Capture and Storage—Calcium Stable isotopes to Quantify Mineralisation

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Subsurface carbon mineralization is considered a safe and permanent method of carbon capture and storage; a process where dissolved CO_2 is injected into the subsurface, reacting with mafic or ultramafic rocks (e.g., basalt) to release cations (e.g., Ca^{2+}, Mg^{2+}), ultimately precipitating carbonate minerals. Existing methods of mineral carbonation are water intensive, thus sustainably implementing mineral carbonation carbon storage on the Gigatonne scale will require alternate fluids for CO_2 dissolution depending on the location, and seawater is an attractive replacement[1]. Carbon mineralization efficiency in seawater has been demonstrated in experiments[2].

Stable isotope ratios of calcium ($d^{44}Ca$) can be used to compare the efficiency of mineral carbonation in seawater to that in freshwater, however, carbon mineralization processes must reliably be distinguished from mixing between sources (e.g., seawater and basalt) of aqueous calcium. Furthermore, accurate isotopic fractionation factors (α) for carbonate mineral precipitation at relevant conditions are required. Here, we measure $d^{44}Ca$ in carbon mineralization experiments in seawater at varying temperatures and CO_2 partial pressures to understand how different conditions impact the calculated α for carbonate precipitation. We use radiogenic strontium ($^{87}Sr/^{86}Sr$) as a proxy for basalt dissolution to disentangle mixing and mineralization processes. Estimates of carbon mineralization using $d^{44}Ca$ are compared to alternative, direct methods (acidification and non-dispersive infrared (IR) CO_2 gas analyzer, furnace and solid state IR, mass balance calculations) to assess the accuracy of the approach. Findings of this work will be used in conjunction with post-injection measurements of $d^{44}Ca$ to evaluate the efficiency of mineral carbonation in seawater. This study highlights key variables to consider when using $d^{44}Ca$ to investigate subsurface processes and lays a foundation for the first field-scale investigation of carbon mineralization in seawater.

[1] Marieni, Chiara, et al. "Mineralization potential of water-dissolved CO_2 and H_2S injected into basalts as function of temperature: Freshwater versus Seawater." *International Journal of Greenhouse Gas Control* 109 (2021): 103357.

[2] Voigt, Martin, et al. "An experimental study of basalt–seawater–CO_2 interaction at 130° C." *Geochimica et Cosmochimica Acta* 308 (2021): 21-41.