Acid Pretreatment as a Strategy for Enhancing CO₂ Mineralization in Basaltic Aquifers

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Basalt carbonation offers a gigaton-scale solution for carbon sequestration due to its high reactivity and global abundance. However, the efficiency of the process is often limited by slow cation release and the formation of pore-clogging secondary phases, which hinder CO₂ mineralization. These barriers to efficient carbon mineralization challenge the scalability of this process at the necessary gigaton-scale level to meet the climate change goals. To address these limitations, this study explores the use of acid pretreatment to accelerate basalt dissolution and enhance the availability of divalent cations for CO2 mineralization.

We conducted a series of far-from-equilibrium dissolution experiments using basaltic glass and crystalline basalt at varying temperatures and pH levels, particularly targeting conditions not previously reported. Dissolution rates were quantified based on the cation release, while scanning electron microscopy and energy dispersive spectroscopy were employed to assess surface alteration and secondary mineral formation. The results showed that acid exposure increased cation release rates by several orders of magnitude while simultaneously reducing silica release, both of which are critical for carbonate precipitation. Reactive transport simulations, using parameters derived from experimental dissolution kinetics, further demonstrated that acid-treated basalt systems retain higher aqueous cation concentrations and promote enhanced carbonate precipitation upon neutralization.

By combining experimental data with geochemical modeling [1], our study demonstrates that exposure to acidic solutions significantly increases dissolution rates, reduces secondary clay formation, and improves mineralization efficiency. The findings suggest that pre-injecting acidic solutions, such as HCl derived from electrochemical CO2 removal processes, can improve the reactivity of basalt formations while neutralizing acid waste streams. Additionally, deep cracks formed throughout the samples after reaction with acid, potentially indicating improved fluid pathways and increased injectivity. Future studies should aim to quantify these effects using geomechanical analyses. These results provide valuable insights into optimizing injection strategies for large-scale carbon sequestration, supporting global

efforts to achieve net-zero emissions.

[1] Q. Zhang, A. N. Awolayo, P. R. Phelps, S. Vadsariya, C. T. Laureijs, M. D. Eisaman, B. M. Tutolo, Enhanced cation release via acid pretreatment for gigaton-scale geologic CO2 sequestration in basalt. *International Journal of Greenhouse Gas Control.* **139**, 104266 (2024).