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## Mineral Carbonation Sensitivity to Hydrogeologic Heterogeneity of Basaltic Aquifers

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Atmospheric CO<sub>2</sub> concentrations are expected to double over the next three decades, and reducing its emissions alone will not be enough to reduce greenhouse gas emissions and limit anthropogenic warming. The removal of large quantities of CO<sub>2</sub> from the atmosphere or the prevention of newly produced CO<sub>2</sub> from reaching the atmosphere is possible through the combination of carbon mineralization in basalt-hosted hydrogeologic systems with direct air or point source capture technologies. Recent efforts in this direction have shown that rapid mineralization is possible, ensuring the security of underground CO<sub>2</sub> storage. However, predicting how far the injected CO<sub>2</sub> will migrate and how fast it will mineralize as it encounters spatial heterogeneity in basaltic formations presents unique challenges to its full-scale implementation. Previous carbon sequestration research, particularly in sedimentary rocks, has commonly investigated the effects of heterogeneity on CO<sub>2</sub> migration, density-driven convection, dissolution efficiency, formation injectivity, and leakage potential. Nonetheless, the efficiency of CO<sub>2</sub> conversion to carbonate minerals in heterogeneous basaltic aquifers remains underexplored. In this study, we present our recent efforts to constrain mineral surface area in laboratory experiments with the goal of improving predictions of mineralization and evaluating its combined impacts with heterogeneity at the field scale. A combination of gas sorption measurements, Raman spectroscopy, and backscatter electron imaging (500 nm/pixel) with image processing techniques was utilized to quantify mineral-specific surface areas. We developed a continuum-scale reactive transport model based on these measured surface area values and showed that predictions based on mineral-specific quantifications of surface area are in reasonably good agreement with effluent chemistry produced from CO<sub>2</sub> flow-through experiments. While mineral surface area and porosity remain fixed, a spatially varying 3D permeability field, defined by the Dykstra-Parsons coefficients and correlation lengths, introduces heterogeneity to basaltic formations. We systematically generated an ensemble of 100 simulations and computed the mean mineralization efficiency and corresponding correlation of variation in efficiency to analyze the interplay between heterogeneity and the gravity-viscous ratio. Our findings show that variability in permeability yields a greater interfacial area between free-phase CO<sub>2</sub> and formation fluids and thereby facilitates its enhanced transformation to carbonate minerals. In this context, we demonstrate that, despite the uncertainty in heterogeneity and the inability to represent heterogeneity at an adequate scale in most numerical models, variability in permeability can enhance carbon mineralization in basaltic aquifers, which has the potential to significantly reduce the global anthropogenic CO<sub>2</sub> budget and contribute to the achievement of an energy transition future.

### Participation

In-Person

### References

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