

Heterogeneity in basalt dissolution kinetics revealed by surface topography measurements under experimental conditions relevant to CO₂ storage.

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Basaltic rocks hold significant potential for CO₂ sequestration, primarily through the binding of CO₂ as carbonate minerals via coupled dissolution-precipitation reactions. The dissolution kinetics of basalt critically influences the efficiency of carbonate mineralization, as the dissolution rate is considered a rate-limiting step in these reactions [1]. The quantification of dissolution rates based on detected changes in fluid composition provides average dissolution rates. However, these bulk methods cannot reveal the heterogeneities in dissolution rates related to the intrinsic properties of the material [2], such as chemical composition, crystallographic properties, and surface structure. Here, we investigate the surface reactivity of a polished micro-crystalline basalt sample through a flow-through experiment conducted under conditions relevant to offshore CO₂ storage. We employed vertical scanning interferometry (VSI) to achieve spatially resolved dissolution rate maps. The composition and orientation of the minerals subjected to dissolution was investigated using Raman spectroscopy, electron microprobe analyses (EMPA), and electron backscatter diffraction (EBSD). This integrated approach enabled us to quantify the dissolution kinetics of various mineral phases, including plagioclase, clinopyroxene, magnetite, and glass, within a single experimental run. The dissolution rate maps show a high sensitivity to compositional variations within individual plagioclase and clinopyroxene phenocrysts, allowing for the measurement of dissolution rates over a wide range of solid solution compositions. For example, for plagioclase, intercrystalline rate variations are reflected in anorthite contents ranging from approximately 20% to 77%. Furthermore, the data indicate local variations in dissolution rates related to crystal orientation and texture. Our findings underscore that subtle variations in chemical composition, a key internal heterogeneity, profoundly influence the dissolution kinetics of basaltic rocks. This deeper quantitative understanding of CO₂ mineralization processes is pivotal for refining computational models, ultimately enhancing

our ability to assess the long-term viability of CO₂ storage in submarine basaltic aquifers.

[1] Snæbjörnsdóttir, S.Ó., Sigfússon, B., Marieni, C., Goldberg, D., Gislason, S.R., Oelkers, E.H. (2020), *Nature Reviews Earth & Environment* **1**, 90–102; [2] Fischer, C., Kurganskaya, I., Schäfer, T., and Lüttge, A. (2014), *Appl. Geochem.* **43**, 132–157.