

How fast can CO₂ be mineralized in basalt? Results from hydrothermal experiments on mixtures of plagioclase and clinopyroxene

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Carbon capture and storage (CCS) could accommodate up to 10¹⁰ tons of CO₂ per year if globally deployed. R&D on subsurface storage has been focused on physical trapping in sedimentary formations like sandstone, but there is increasing interest in basalt, which is common and has an abundance of alkaline earth elements (Ca, Mg, Fe). Field studies using small amounts of CO₂ suggest that CO₂ can be mineralized in basalt within years. However, other work suggests slower rates due to alkalinity build-up when injection rates are scaled up (Tutolo *et al.*, *ES&T*, 2022). The extreme sensitivity of silicate dissolution rates to alkalinity (Daval *et al.*, *GCA*, 2010) and decrease with time (Zhang and DePaolo, *ACR*, 2017) need further study. We measured dissolution rates of mixtures of plagioclase (PLAG) and clinopyroxene (CPX), the two major cation-bearing minerals in basalt, under CO₂ storage conditions. We use differing proportions of the two minerals (80:20 and 20:80) in stirred solutions containing seawater Na, Cl, and Mg under an atmosphere of N₂ or CO₂ at 100 bar and 80°C for a period of 275 days. Solutions are sampled periodically to monitor chemistry. Ca concentration provides bulk dissolution rate, and Sr/Ca gives relative proportions of CPX and PLAG dissolving. Total dissolution is 0.4 to 1.1%; only 1.5x faster with CO₂. CPX dissolution rates start at values close to the published rate constant for augite (but 10x slower than diopside; Palandri and Kharaka, *USGS OFR 1068*, 2004). Within 6 months, rates decrease by more than 100x, roughly like 1/*t*, where *t* is experiment duration. PLAG behaves similarly when in larger proportion, but dissolves *faster* than expected when it is the minor mineral. Total cation release rates also decrease as 1/*t* and slow by 100x to 1000x after 9 months. The pronounced 1/*t* slowing is a general feature of dissolution rate studies related to CO₂ mineralization; it may be attributable to non-linear kinetics and mineral surface evolution. This slowing, not generally accounted for in reactive transport models (although see Aradottir *et al.*, *IJGGC*, 2012), implies that other trapping mechanisms must keep CO₂ sequestered until mineralization is complete.