

## HARNESSING PERIDOTITE ALTERATION FOR CARBON CAPTURE AND STORAGE

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Mantle peridotite, composed largely of the mineral olivine with Mg end-member  $Mg_2SiO_4$ , is far from equilibrium with the atmosphere and surface waters. Where peridotite comes to the surface via plate tectonics, faulting and erosion, this creates a huge, accessible reservoir of chemical potential energy, which drives rapid reaction and can support subsurface chemosynthetic ecosystems. Feedbacks in reactive transport lead to two very different outcomes. Hydration, carbonation and oxidation cause large increases in the mass and volume of solid phases, which can fill pore space, destroy permeability, and armor reactive surfaces, in a self-limiting, negative feedback. As a result, we can sample high grade metamorphic rocks on the Earth's surface. Alternatively, heating due to exothermic reactions can accelerate reaction rates. More importantly, volume change due to increasing solid mass and decreasing density converts chemical potential energy to work, and can cause differential stresses that are sufficient to fracture rocks. This reaction-driven cracking process can maintain or enhance permeability and reactive surface area, in a positive feedback system leading to complete equilibration of fluid with altered peridotite.

In some natural geologic settings, complete reaction of peridotite with  $CO_2$ -rich hydrothermal fluid forms "listvenites", rocks in which all Mg and Ca atoms combine with  $CO_2$  to form solid carbonate minerals. For example, olivine  $Mg_2SiO_4$  reacts with dissolved  $CO_2$  in water to form magnesite  $MgCO_3$  + quartz  $SiO_2$ . More commonly, all dissolved carbon is removed from ground water migrating through peridotite-hosted aquifers, by precipitation of Mg carbonates in the subsurface. C-depleted, CaOH-rich, alkaline water (pH 11.5) then returns to the surface, where it draws down additional  $CO_2$  from air to form extensive terraces composed of calcite  $CaCO_3$ .

Various avenues of research focus on emulating these natural processes to design engineered methods for carbon capture and storage that make maximum use of available chemical potential to minimize cost and energy use. One set of ideas focuses on solid storage of carbon which has already been captured via some other technique. Fluids with high  $P(CO_2)$  -  $CO_2$ -saturated water,  $H_2O$ -saturated supercritical  $CO_2$ , "seltzer", perhaps even flue gas - could be injected into a peridotite-hosted aquifer, where they will react rapidly to form solid carbonate minerals. Such a system would be similar to the CarbFix project in Iceland, where "seltzer" is injected into basalt-hosted aquifers. However, reaction-rates and uptake capacities are larger in peridotite-hosted environments. On the other hand, basalt formations are more common, and generally have more ambient, initial permeability than peridotite.

Alternatively, a more faithful emulation of natural systems involves  $CO_2$  removal from air, for example via enhanced reaction of surface water with peridotite. The simplest, "low-hanging fruit" in this context involves various methods to induce enhanced circulation of water through fine-grained mine tailings. Hundreds of millions of tons of  $CO_2$  could be captured and stored in this way, before extant peridotite mine-tailings were totally consumed.

Thermal convection of seawater through subsurface, fractured, peridotite, could also achieve  $CO_2$  removal from air. The simplest, and thus probably lowest cost implementation would involve artesian production of warm, carbon-free, CaOH-rich alkaline waters from existing subsurface aquifers in peridotite. Once at the surface, these waters will draw down  $CO_2$  from air to form solid carbonate minerals. A geologist's level analysis (not an economist's!) suggests that the cost of such a system could be less than \$100 per ton of  $CO_2$  captured and stored, provided that aquifers with permeability  $\sim 10^{-12} m^2$  can be found. Various hybrid technologies can be envisioned, for example geothermal power plants using  $CO_2$ -rich fluids, in which carbon mineralization consumes  $CO_2$  along the fluid circulation path. However, successful implementation of such "hot dry rock" geothermal systems is problematic, with or without  $CO_2$  in the heat transfer fluid.

More generally, if we understood how to induce reaction-driven cracking, there might be other applications for such techniques, including enhanced extraction of oil and gas from tight reservoirs using  $CO_2$  as a reactant to produce a ramified crack network at the grain scale. In addition, it may be important to understand and forestall reaction-driven cracking, in wellbore cement and in natural permeability barriers - such as carbonate-bearing shales - above storage reservoirs for acidic water saturated in supercritical  $CO_2$ .