

Reactive transport experiments of coupled carbonation and serpentinization in a natural serpentinite. Implication for hydrogen production and carbon geological storage

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Carbonation of ultramafic rocks is currently a hot topic in the energy landscape as the engineered sequestration of mineral CO₂ in these formations could help reduce atmospheric emissions and cope with climate change. Ultramafic minerals react with dissolved CO₂ to precipitate magnesium and calcium carbonates, sequestering CO₂ in a safe manner. In the meantime, oxidation of Fe(II) leads to the reduction of water to H₂. This reaction, which is usually associated with serpentinization is spontaneously occurring in natural settings (mostly at mid-oceanic ridges) and is responsible for an important production of natural hydrogen; and could potentially be leveraged along the carbonation of these rocks to sequester CO₂ while producing green hydrogen.

In this study, we present four reactive percolation experiments proceeded on a natural serpentinite dredged from the ultraslow South-West Indian Oceanic Ridge. Two serpentinite cores (length 3-4 cm and dia. 5.6 mm) were subjected for 10 days to the continuous injection of a NaHCO₃-rich brine at 160°C and 280°C, while two other cores were subjected to the injection of a CO₂-free brine for comparison. Petrographic and petrophysical results as well as outlet fluid compositions were compared to PHREEQC batch simulations, allowing the reconstruction of the mineralogical evolution of the cores.

The most striking observation is the fast and dramatic decrease of the permeability for carbonation experiments from the precipitation of carbonates. In contrast, precipitation of serpentine was found to be limited and less impacting as permeability for the CO₂-free experiments showed a much slower permeability decay. Despite the permeability drop, carbonation was found to be quite successful with a 31% yield at 280°C, while hydrogen production presented a yield of 2%. Experiments without CO₂ presented a higher H₂ production.

The modeling highlight the fundamental role of spatio-temporal length-scales, controlled by the competition between advection and chemical kinetics. In particular, experimental results show that the behavior is strongly dependent on the initial pore size distribution. The presented semi-quantitative modeling is based on the concept of penetration length, a powerful and adaptable tool for the analysis of multi-mineral water-rock interactions, which can easily be extended to any natural or engineered system.