Experimental study of the co-valorisation of carbon dioxide storage through hydrogen production in ultramafic geological formations

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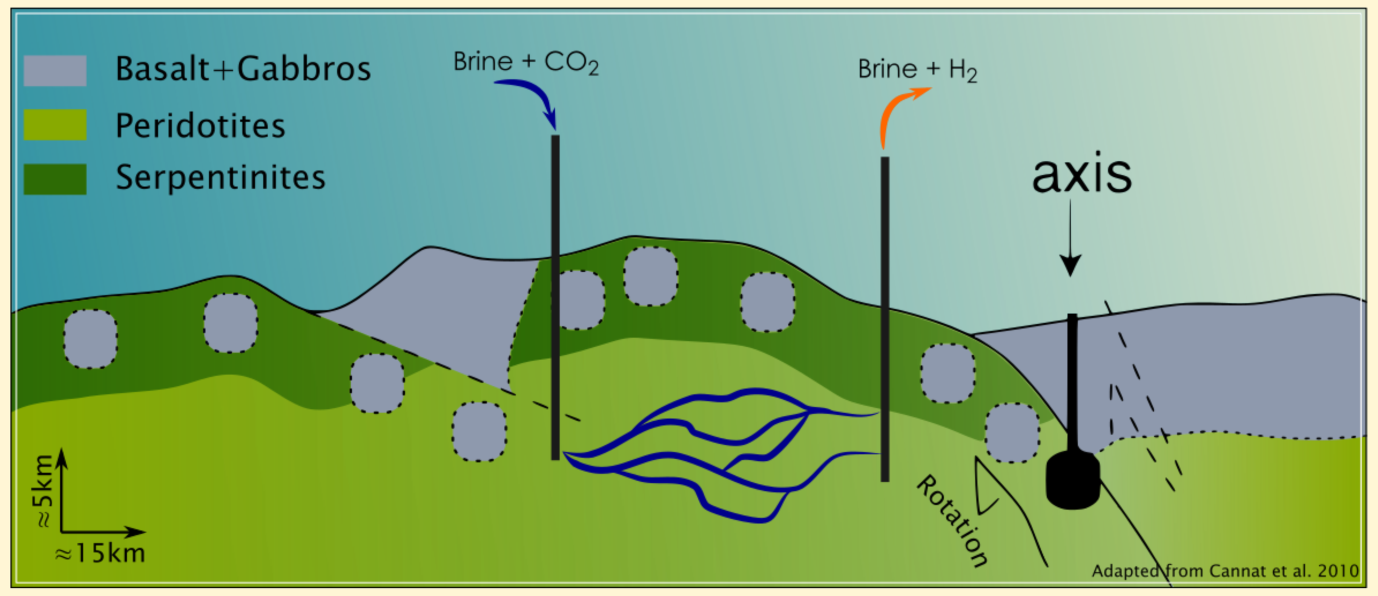
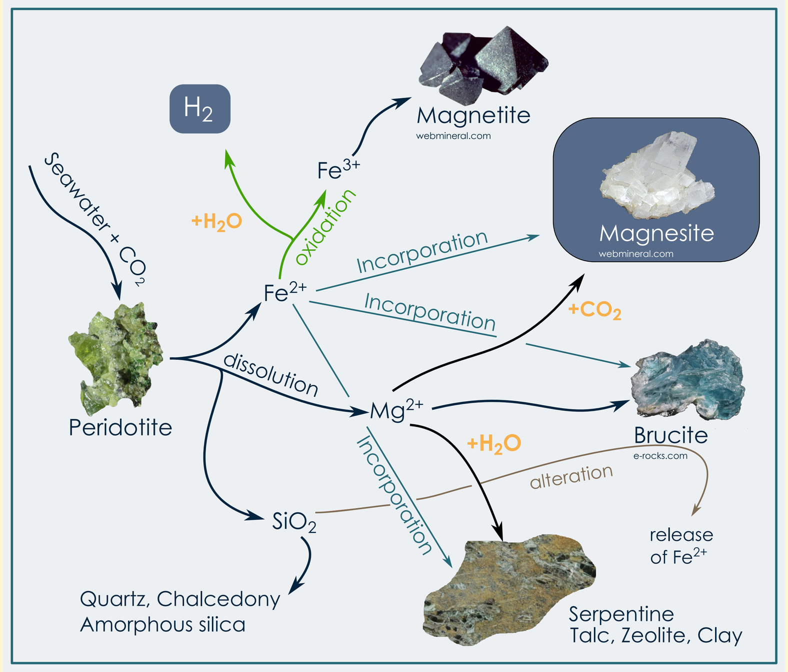
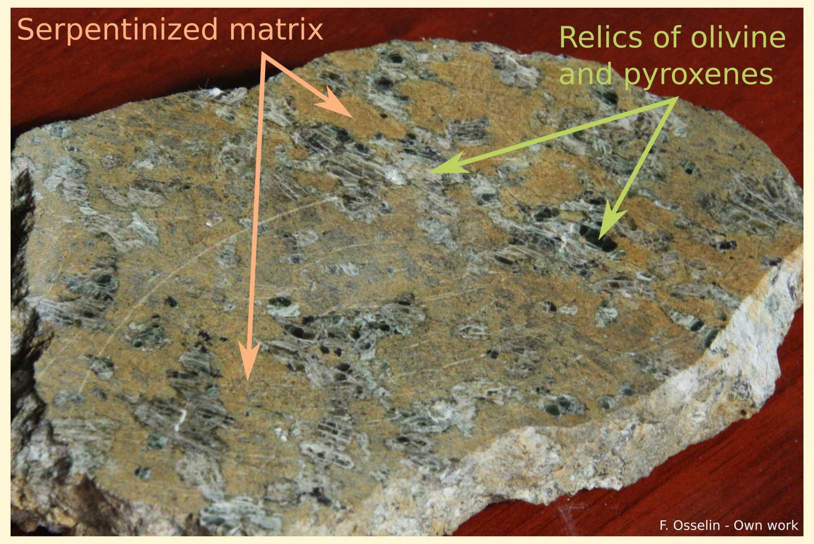
The development of Negative Emission Technologies appears as one of the most appealing solution for the global reduction of anthropic carbon dioxide emissions. In this study, we are leveraging the reactivity of silicate minerals in order to **develop an in situ green hydrogen production coupled with CO_2 mineralization**. The objectives of the project are to (1) optimize the conditions (P, T, protolith, redox) for both processes (2) characterize the hydrochemical coupling during the reactive percolation of the carbonated brine in a partially serpentinized peridotite and (3) integrate the coupling with mechanics and in particular reaction-induced fracturing and crystallization pressure





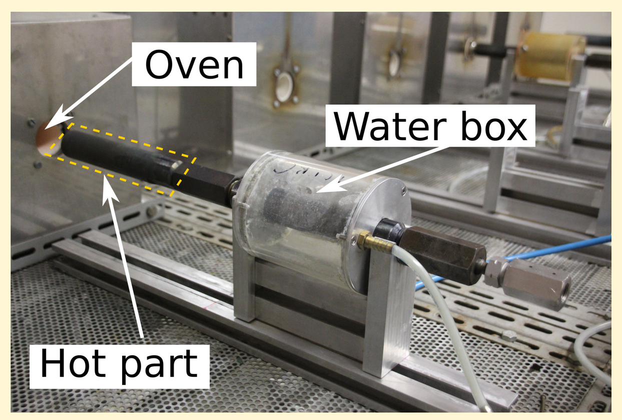


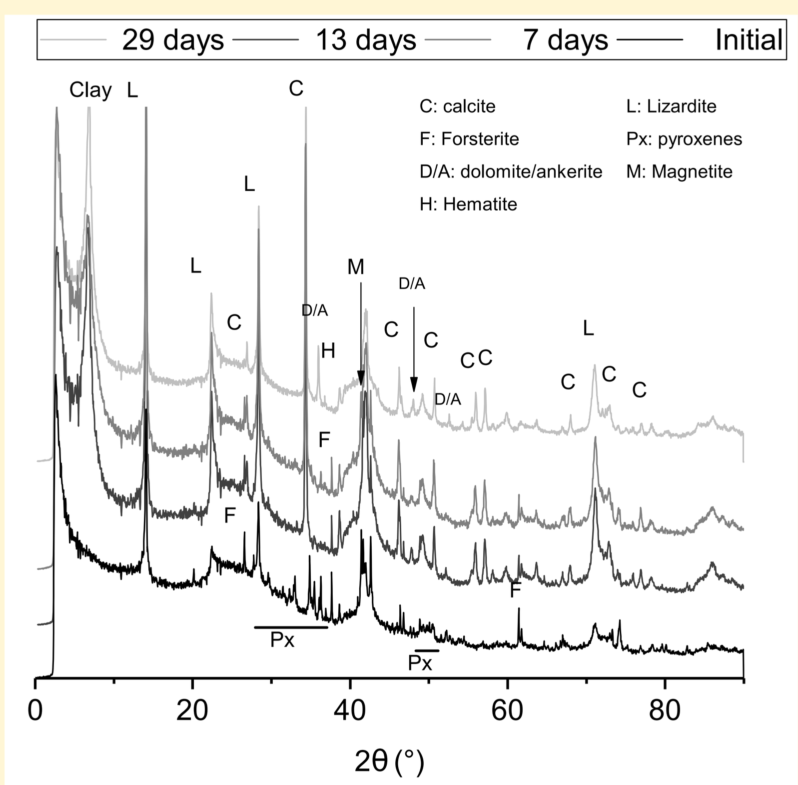
The protolith used for the experiments is a **serpentinite** from the SW Indian Ridge
- Several successive generations of serpentine
- Relics of olivine, ortho and clinopyroxenes
- Traces of magnetite and Cr-spinel

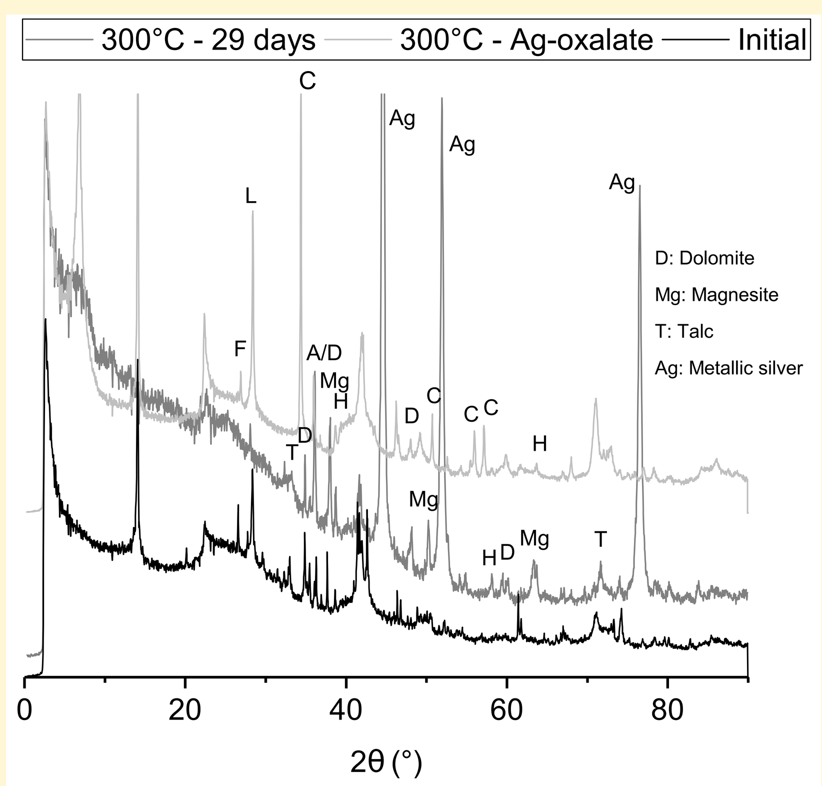
10% porosity. Permeability controlled by fractures (≈100µm)

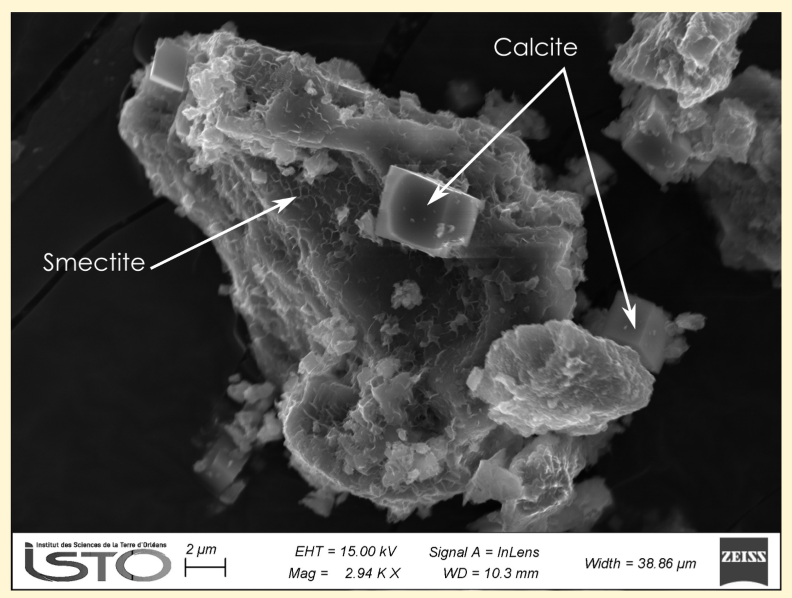
Batch Experiments

Reactions @300°C and 165°C - 500 bar
- 50 mg serpentinite (<50 µm)
- 100 mg Solution 5% $NaHCO_3$ 2% NaCl
- 100 mg silver oxalate for the second series (X_{CO2}=0.5)



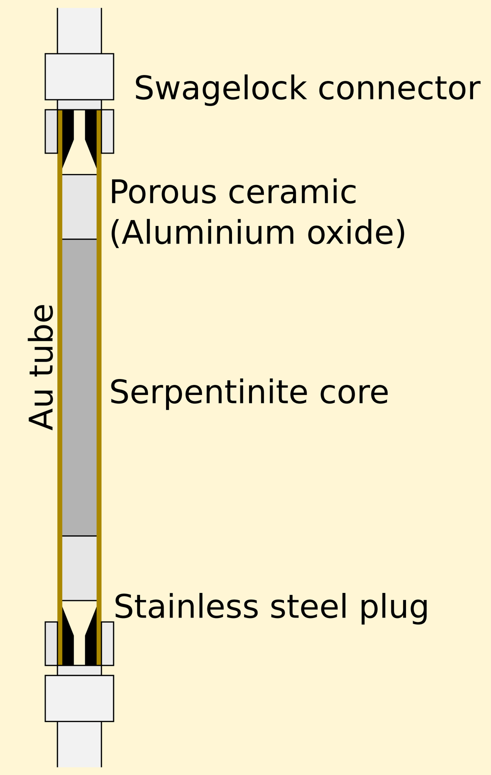


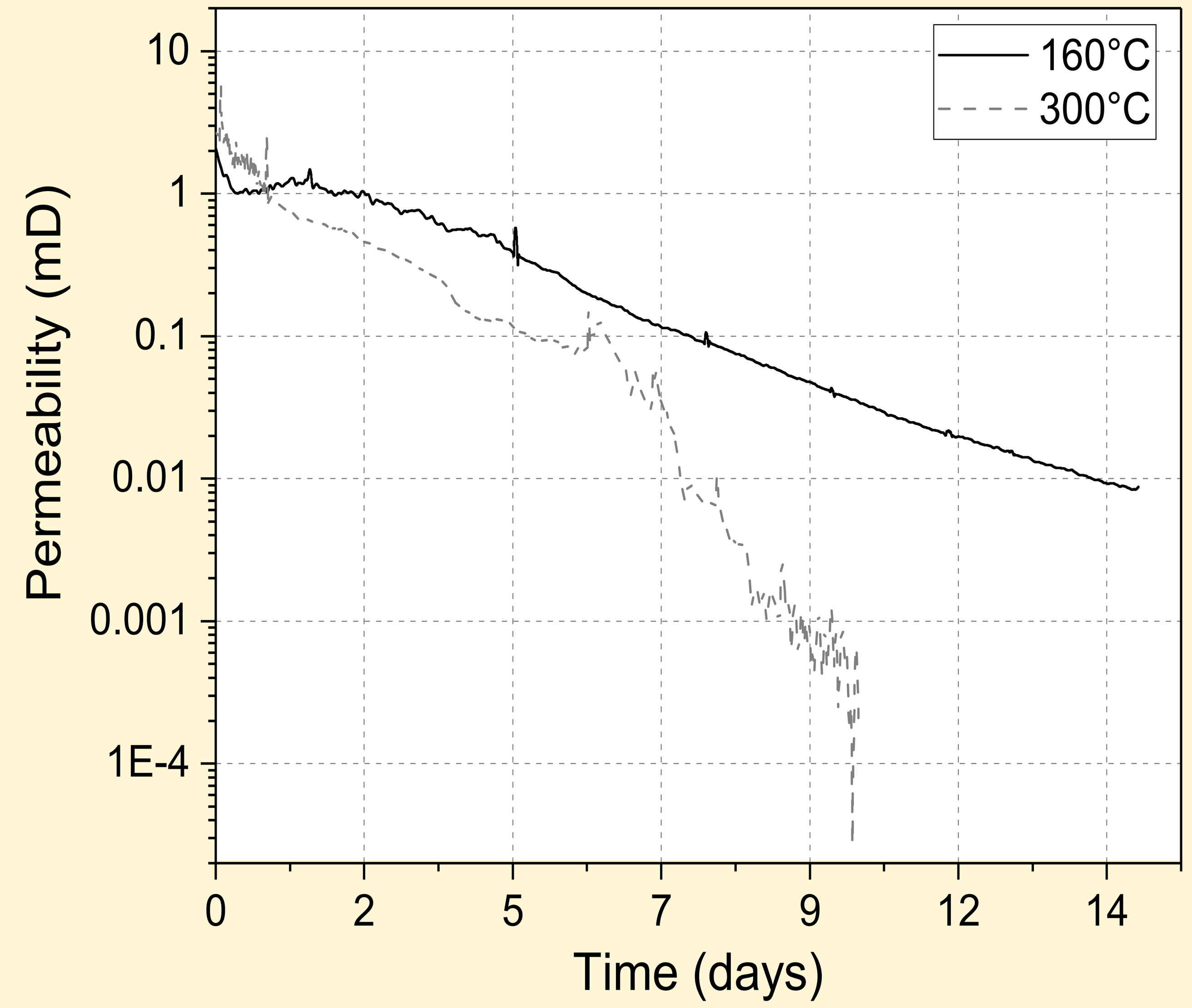




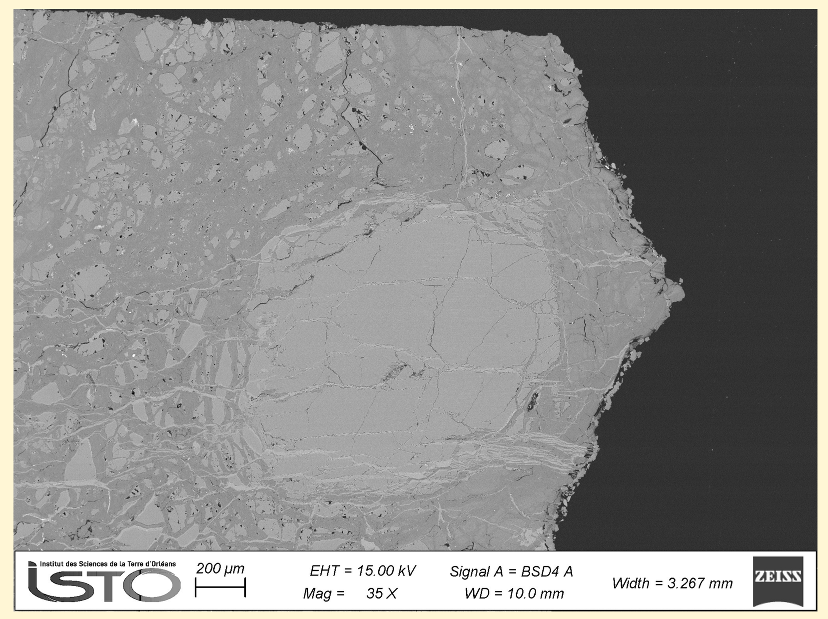
In each case, **carbon dioxide is the limiting reactant**
- Disappeareance of pyroxenes and the majority of forsterie
- Formation of carbonates (magnesite, calcite and solid solutions)
- Only a little magnetite but presence of hematite (oxidation)
- Reaction of serpentine in the case of oxalate
- Reduction of Ag^+ to Ag(s)
- No reactivity with CO_2 in the absence of water

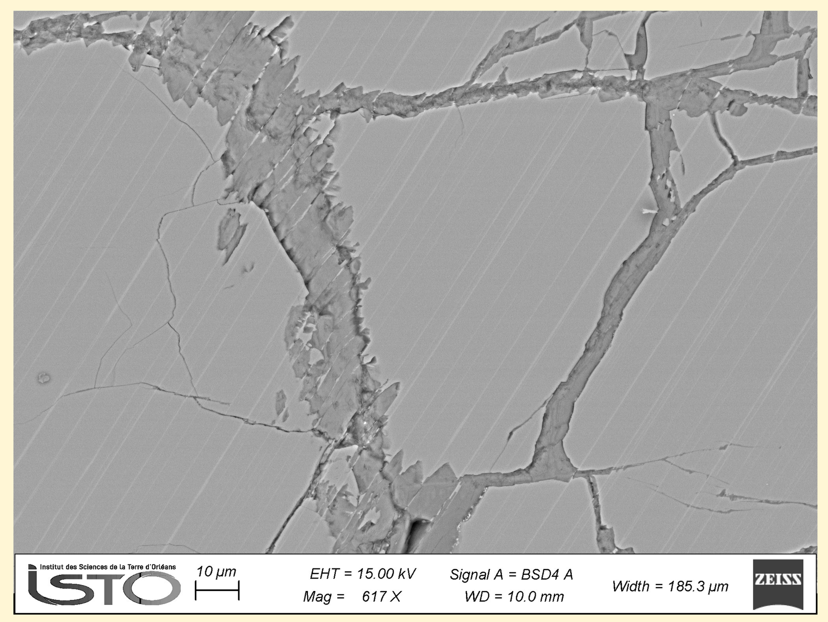
Reactive percolation experiments

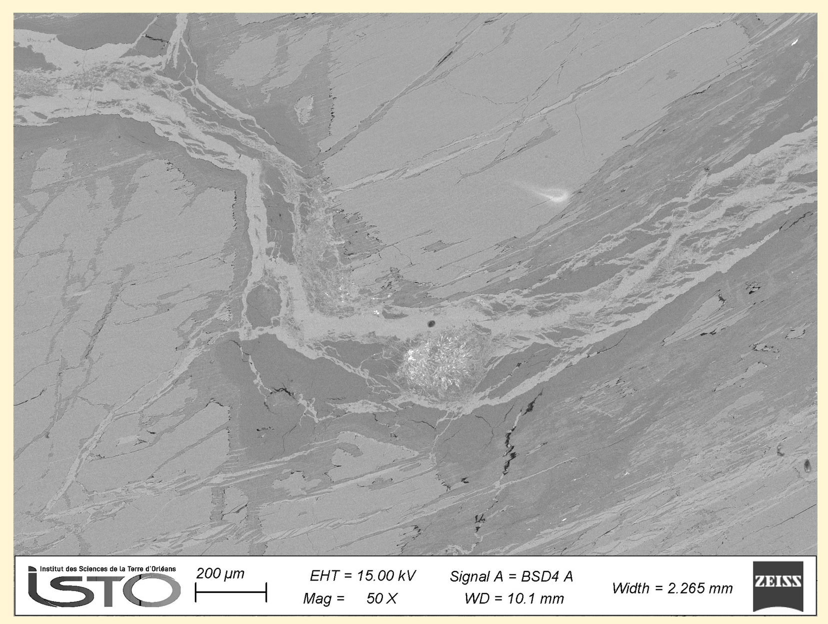




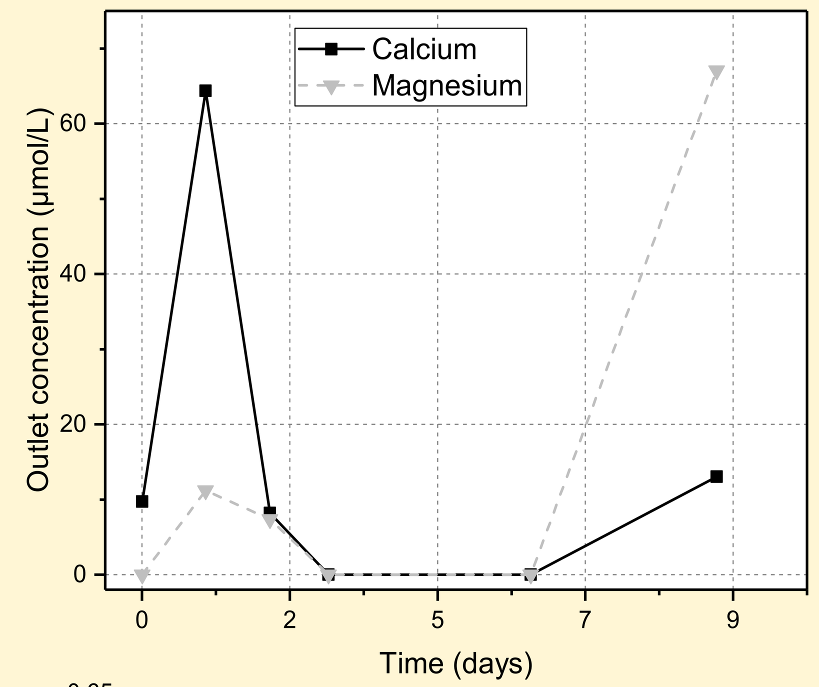
Reactive percolation apparatus for the study of hydrothermal reactions
- Drilled cores ø5.6mm - few cm long
- Up to 500 bars and 300°C
- Inlet solution identical to batch reactions


Carbonate veins running through a fractured pyroxene

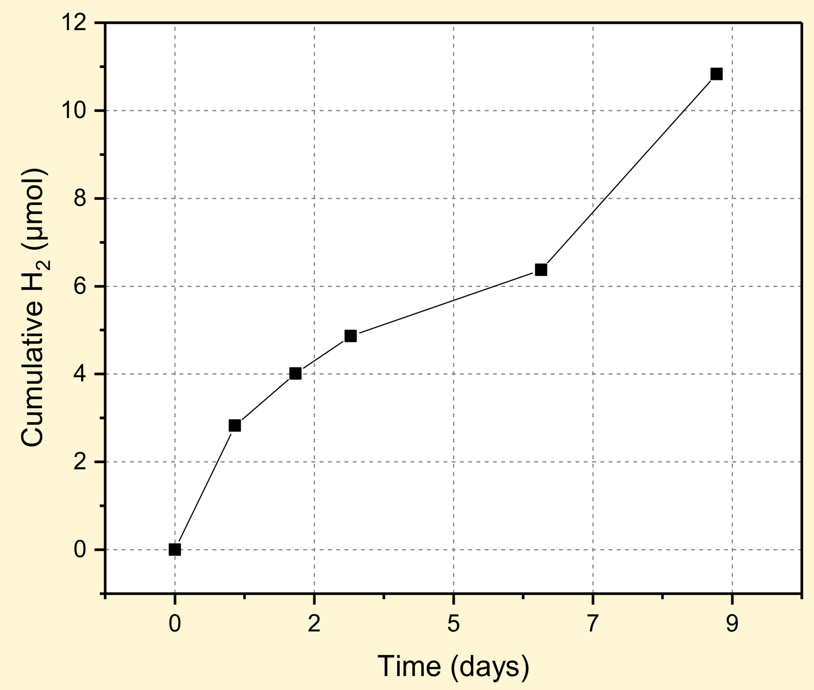

Fractures induced by the growth of the secondary minerals in the etch pits created by pyroxene dissolution

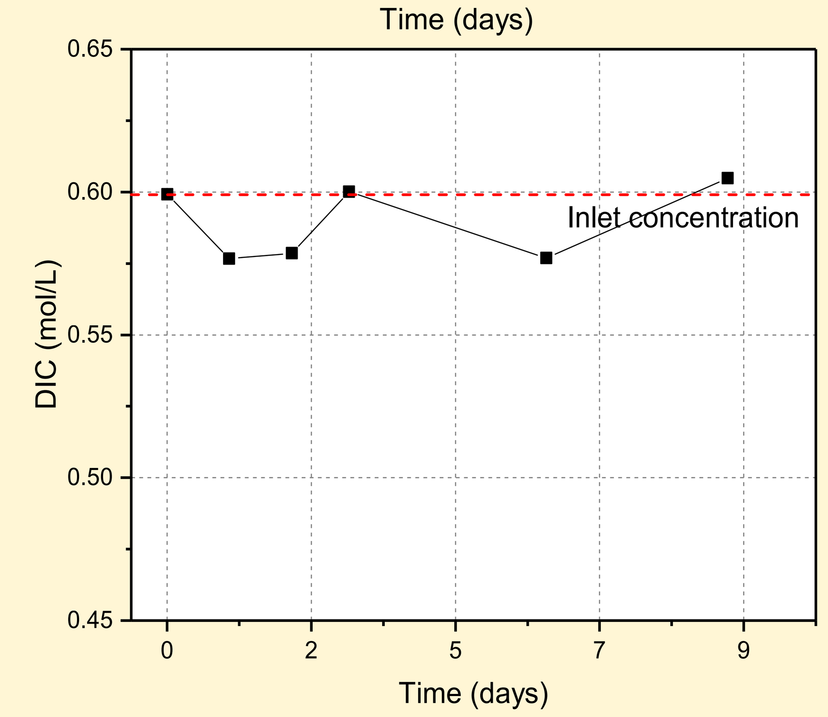

Carbonate vein running through the sample

Experiment @160°C
- Injection during 15 days
- Pressure drop from 2 to 30 bars
- Flow rate ≈ 0.01 - 0.02 ml/min
- 2500 pore volumes injected



Experiment @300°C
- Injection during 5 days
- Pressure drop from 2 to 80 bars
- Flow rate ≈ 0.01 - 0.02 ml/min
- 1600 pore volumes injected





In 9 days, we obtained 1.6% of the H2 yield and 13% of the carbonation yield

Conclusion

Experiments show clearly **the potential for carbonation and hydrogen production** of a peridotite, even partially serpentinized. However, the clogging of the core limits dramatically the yield. Controlled Reaction-Induced Fracturing can potentially enable a sustained injection.
- Continue characterizing the chemistry of the system with batch experiments
- Pursue reactive percolation experiments at different temperatures and with varying fluid chemistries
- Develop a coupled modeling of the THMC behavior, in particular including mechanical feedbacks