Geophysical Research Abstracts
Vol. 14, EGU2012-9309, 2012
EGU General Assembly 2012

CO_2 Geological Storage in Olivine Rich Basaltic Aquifers: New Insights From Flow-Through Experiments

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Injection of CO_2-rich fluids into (ultra-)mafic aquifers is one of the methods envisaged for mitigation of increasing atmospheric CO_2 concentrations. These rocks, rich in Mg, Fe and Ca, have a high potential to trap CO_2 as carbonates minerals. However, the role of reaction-transport processes has not been investigated to predict the capacity and sustainability for CO_2 storage in these highly reactive systems.

We performed three percolation-reaction experiments on the ICARE-2 bench (Luquot et al., 2012) at 180°C and total pressure of 12 MPa. $NaHCO_3$ rich water (0.5 mol/L) mixed with CO_2 (Pco2 = 10 MPa) were injected at an initial injection rate of 1 mL/h for exp. 1 and 2, and 0.1 mL/h for exp. 3 into sintered olivine analogues of accumulation zones in basaltic flows. The sintered samples comprise mainly olivine Fo87 (Mg1.75Fe0.24Ni0.01SiO4) with minor basalt and spinel (Mg0.57Cr1.19Ti0.03Mn0.01Fe0.66Al0.54O4). The initial porosity and permeability of samples ranges from 4 to 7 % and 30x10-18 to 400x10-18 m2 respectively.

All experiments resulted in a strong permeability decrease (down to 10-18 m2) after 90 hours for exp. 1 and 2 and 55 hours for exp 3. The enrichment in several major and trace elements observed in outlet fluids indicates that dissolution of olivine (Si, Co), basalt (Al, Zr) and spinel (Cr, V) occurred, while the depleted Ca and Mg composition of outlet fluids indicates precipitation of Ca-Mg rich phases. Analyses of reacted samples indicate that all basalt grains were dissolved and the spatial distribution of etch pits on forsterite surfaces suggest the anisotropic dissolution of olivine. Ankerite and calcite is observed in pores of samples after exp. 1 and 2, and well-developed magnesite ($Mg0.88Fe0.11Ca0.01CO_3$) and dolomite replacing dissolved olivine is found in exp.3 sample. The (001) planes of magnesite and dolomite are parallel to the (100) plane of forsterite. Downstream, no carbonates were found but the growth of relatively large (up to 5 μm) Mg-Fe rich phyllosilicates plates perpendicular to (at the expense of ?) olivine surfaces was observed. The precipitation of these serpentine type minerals may explain the strong decrease in permeability during experiments.

Carbonation was relatively efficient in our experiments: between 0.003g (exp.3) and 0.015 g (exp.1 & 2) of CO_2 per gram of sample was trapped as carbonates. When these analyses are upscaled to the size of an injection site, they correspond to an average yield of 3.6 to 10.6 Mtons of CO_2/km3/day.

Our results indicate a strong control of flow rates on carbonation, and also on hydration reactions. This implies not only variations of the CO_2 storage capacity of the basaltic aquifer with distance to the injection well, but also that controlling the injection rate would allow enhancing the efficiency of in situ carbonation.

Luquot L., Andreani M., Gouze P., Camps P. (2012) CO_2 percolation experiment through chlorite/zeolite-rich sandstone (Pretty-Hill Formation – Otway-Basin–Australia), Chemical Geology 294-295, 75–88, doi:10.1016/j.chemgeo.2011.11.018.