New experimental insights on hydrogen/hydrocarbon generation and simultaneous carbonation of olivine $(Mg,Fe)_2SiO_4$

KANCHANA KULARATNE[12], OLIVIER SISSMANN[1], ISABELLE MARTINEZ[2], MICHEL CHARDIN[1], SONIA NOIREZ[1], HELENE VERMESSE[1], ISABELLE BOUBOUNE[1], JULIEN LABAUME[1], JOEL LOPES DE AZEVEDO[1], FANNY LUTZ[1], PASCAL HAYRAULT[1], ERIC KOHLER[1], LAURE CORDIER[2], JULIE CARLUT[2] AND FRANÇOIS GUYOT[3]

[1]IFP Energies Nouvelles, 92852 Rueil-Malmaison, France
[2]Institut de Physique du Globe de Paris, 75005 Paris, France
[3]IMPMC, MNHN, UPMC, 75005 Paris, France
(*correspondence:kanchana.kularatne@ifpen.fr)

Olivine is considered one of the best candidates for mineral carbonation under 200°C, because of its high Fe/Mg content and rapid dissolution kinetics. In addition, its alteration at higher temperatures (300°C) has also shown potential for hydrogen generation, as Fe^{2+} released during the serpentinization process may reduce water to produce H_2. Though those processes have different optimum temperatures, they may occur simultaneously at intermediate ones.

Hydrothermal batch experiments were thus performed on an olivine-bearing mine waste (New Caledonian mine slags) at P_{CO2} = 15 - 30 MPa and at 200°C-300°C, in order to determine the optimal environmental conditions that could lead to the generation of hydrogen and reduced carbon molecules in a CO_2-rich environment (through Fischer-Tropsch Type reactions), while producing mineral carbonation at the same time. The fluid was sampled regularly and the reaction path followed was thermodynamically modelled thanks to cation composition analysis. In addition, organic species (aldehydes and acids) dissolved in the fluid phase were characterized, in order to identify intermediate products formed during the FTT reactions. Finally, mineral and textural analyses at the nm-scale were performed on secondary surface layers (through FIB-TEM observations), to characterize the phases controlling the dissolution kinetics.

Batch experiment performed at 15 MPa and 200 °C yielded ~20 wt.% of ferroan-magnesite and ~2.0 mmol/g of hydrogen after a 4-weeks reaction on a ~50 μm powder, but no detectable reduced C-componds. At 30 MPa and 250 °C - 300 °C, though analyses are still underway, the study also shows evidences of the presence of fomaldehyde and acetaldehyde in the fluid phase. Furthermore both hydrogen and methane were detected in the gaseous phase, suggesting reduction of inorganic carbon species into light hydrocarbons.