671

Quantification and kinetics of H₂ generation during hydrothermal serpentinization experiments

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To quantitatively evaluate the flux of H₂ associated with serpentinization and document the kinetics of the H₂-producing reaction, serpentinization experiments have been performed on fine-grained (<60µm) natural harzburgites powders from the Pindus ophiolite. Experiments are carried out in large-volume Dickson-Seyfried bombs at 250 and 300°C for a total pressure of 500 bar and durations up to 3 months. Either Ti or Au-Ti bags were used as containers, together with artificial seawater (3.2% NaCl). A constant water-rock ratio of ~1.8 was imposed. For each temperature two types of experiments were conducted in parallel. In the first type, the hydrothermal fluid is periodically sampled and its H_2 concentration determined by gas chromatography (GC). In the second type, the H₂ fugacity inside the reaction bag is permanently monitored with a H_2 semi-permeable membrane made of Au₂₀Pd₈₀. Product phases identified after the experiments include serpentine (>80 vol%), magnetite and brucite consistent with a serpentinization reaction.

At 300°C, the GC measurements show that the H_2 concentration increases up to ~50mmol.kg⁻¹ in less than ~10 days. A plateau of ~110 mmol.kg⁻¹ is reached after ~50 days. H_2 concentrations and time evolutions are similar for Ti and AuTi bags. At 250°C, the plateau is lower (~50 mmol.kg⁻¹) and attained after 12 days. An increase of the membrane fH₂ is observed after the 2nd (300°C) and the 3rd (250°C) day. The maximum rate of fH₂ increase occurs during the 6th (300°C) and the 5th (250°C) day. At 300°C, the fH₂ is stabilized at 12.4 bar after 16 days, corresponding to a fO₂ of ~NNO-3.9. At 250°C, using a membrane with a lower free volume, the fH₂ is stabilized at 38 bar after 70 days, corresponding to a local fO₂ of ~NNO-5.5.

Our intrinsic fH_2 measurements are the first performed on a serpentinization reaction and suggest that H_2 generation is surprisingly rapid. The establishment of a quantitative link between the H_2 concentration and the fH_2 is in progress.