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Gas generation from intact Opalinus Clay drill cores under elevated temperature conditions

OLIVER HELTEN[1], CHRISTIAN OSTERTAG-HENNING[2] AND THOMAS WEGER[3]

[1]Federal Institute for Geosciences and Natural Resources
[2]Federal Institue for Geosciences and Natural Resources
[3]Federal Institute for Geosciences and Natura Resources

Presenting Author: oliver.helten@bgr.de

Gas generation is one process considered in the safety assessment of features, events and processes in geological repositories of high level, heat-generating nuclear waste. It includes gases being generated from a host rock formation by the elevated temperature conditions[1]. Therefore, data on the molecular composition, quantities, and the temporal evolution of gas generation and limiting processes are needed for modelling the temporal evolution of the chemical environment and the gas pressure in a repository after closure.

Heating experiments (≤200 °C, 3 MPa) were performed in a newly developed experimental setup with intact Opalinus Clay drillcores from the subsurface lab in Mont Terri, Switzerland. Cores with the dimensions 140 × 53 mm (height × diameter; ~0.75 kg) were placed in a glass liner, put in a high-pressure autoclave under helium atmosphere, and were heated for a duration of days up to two weeks. Gas samples were taken in intervals with a gastight syringe and quantified, using a refinery gas analyzer system.

At 200 °C, CO_2 was the main component with up to 141,000 µmol/kg OPA released, followed by butane > propane > methane > ethane (CH_4 <20 µmol/kg OPA). Up to 0.4% molecular hydrogen (H_2) was measured, likely originating from anaerobic iron oxidation and water reduction at the autoclave surface. Trace amounts of hydrogen sulfide (H_2S) were detected. $\delta^{13}C_{CO2}$ indicates that CO_2 originated predominantly from carbonate decomposition/mineral dissolution and exsolution of pore-water dissolved CO_2, with a smaller proportion of CO_2 coming from sedimentary organic matter transformation. Some hydrocarbon gases might have been present initially as residual gases[2], perhaps sorbed to surfaces in the rock or dissolved in the porewater, while methane concentrations over time point to additional generation in the experiments.

Comparison of this experimental setup with experiments carried out on material from the same drillcore with Dickson-type flexible Au-Ti-reaction cells at elevated pressures (20 MPa, 80-200 °C) in water-saturated conditions showed good agreement concerning the amount of gases generated per kilogram rock.

References

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