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Experimental study of pure mineral phases/supercritical CO_2 reactivity - Kinetics of portlandite carbonation measurement

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Carbon dioxide sequestration in deep aquifers and depleted oil-fields is one of the technical solutions proposed to reduce greenhouse gas release in the atmosphere. The gas containment relies on several trapping mechanisms and a low permeability cap-rock to prevent CO_2 from leaking upwards. During the several phases of the sequestration, the host-rock, the cap-rock and the borehole seals will be in contact with a supercritical CO_2 ($scCO_2$) phase, containing various amounts of dissolved water. However, the reactivity of the rock minerals in $scCO_2$ fluids is thoroughly unknown.

Preliminary tests have demonstrated the effective reactivity of several pure mineral phases, including in water-free experiments [Regnault, 2005]. In particular, the results have shown that the portlandite, chosen as an analogue of the cement phases, was entirely recrystallized into calcite in a short amount of time.

We emphasized this mineral because of the importance of the borehole seals behaviour during the sequestration. In fact, the strong reactivity of portlandite suggests that reactions with cement structures around the borehole may occur. Thus, the purpose of the study is to measure the kinetic rate of portlandite carbonation in initially dry $scCO_2$.

The experimental setup consisted in batch reactions between pure portlandite and an initial anhydrous $scCO_2$ fluid at 160 bar and 120 to 200°C. The experiments were carried out using crushed pure synthetic portlandite. The fluid was maintained at constant temperature and pressure using a thermal press. The kinetic rate of the above reaction has been approached using three different ways. First, the CO_2 consumption rate as a function of time has been recorded throughout the experiment by following the position of the compressor piston. Secondly, the amount of formed calcite by reacted portlandite has been measured, after different reacting times, by X-Ray Diffraction analysis of the solid fraction. Finally, the vapour phase has been sampled with the microsampler ROLSI^{TM} and analysed using a gas chromatograph to follow the amount of water released in the course of the reaction.

The experiments carried out in this study showed an effective reactivity of portlandite in an anhydrous $scCO_2$ fluid with a total transformation into calcite. Furthermore, the kinetics of this reaction is very fast: the carbonation is complete within less than 10 hours at 200°C.

The data gathered by this study can be used in chemical models (and reactive transport models) to simulate portlandite alteration. As an analogue to more complex cement phases, portlandite can help start modelling the alteration of borehole seals due to $scCO_2$ injection. Complex feedbacks can then be addressed: increase/decrease in porosity, shortcuts creation, and more generally modification of the cement structure.

References

Regnault, O., V. Lagneau, H. Catalette, H. Schneider, 2005: Experimental study of pure mineral phases/supercritical CO_2 reactivity, implications for geological CO_2 sequestration. C. R. Geosciences, 337, 1331-1339.