Effect of CO_2-enriched fluid on three argillite type caprocks

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The sequestration of CO_2 under impervious caprocks as argillite is proposed to reduce the greenhouse effect. CO_2 impact on the containment properties of argillite has to be assessed, as performed by [1] on Upper Toarcian rocks from Tounemire site (France). In the current study, we widened the approach to two other levels from Tournemire (Lower Toarcian and Domerian), displaying distinct carbonate contents (30% and 8%, resp.).

^{36}Cl, HTO, Br^- and D_2O were used as tracers in through-diffusion experiments carried out with and without CO_2-enriched fluids. The through-diffusion technique consists in imposing a concentration gradient of the tracer between the two faces of the sample. For the cells simulating an acidic fluid attack, dissolved CO_2 was injected inside the upstream reservoir. Chemistry evolution was also monitored in the up and downstream reservoirs.

Tracer data analyses indicated a degradation of the containment properties of the samples, increasing from the Paper Shale (unaffected) up to the Toarcian (Fig.1)

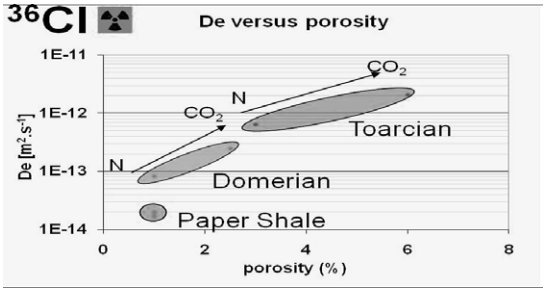

Figure 1: evolution of the diffusive parameters for the 3 levels

Such a difference can be accounted for by the distinct kinetics for achieving the chemical equilibrium regarding carbonates as shown by [Ca2+], [Mg2+] and alkalinity monitoring. This demonstrates the role played by the initial petrophysical properties of the rocks, in addition to their carbonate content.

[1] Berthe *et.al*. (2011) *Energy Procedia* **4**, 5314-5319.

On the use of nucleation barriers in numerical simulation of water-rock interactions

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Numerical simulations of water-rock interactions in diagenetic or CO_2 sequestration systems generally predict mineral alteration processes that are considerably faster than those observed in experiments or diagenetic environments. This has been attributed to several reasons, among which the common disregard for nucleation of precipitating mineral phases in such models.

The effect of the implementation of nucleation barriers was evaluated on a kinetic reaction path model of sandstone diagenesis. Critical levels of supersaturation required for precipitation, based on literature data of formation water compositions, substantially slow down reaction kinetics due to their feedback on the dissolution rates of the minerals supplying the component species of the precipitating phase.

The implementation of nucleation barriers and rates based on classical nucleation theory have a similar effect, but often predict unrealistically high levels of supersaturation. One of the crucial parameters in classical nucleation theory is the mineral-water interface tension. Literature data on mineral-water interface tension is scarce and ambiguous. Different methods for measuring or calculating interface tensions produce very different results.

A methodology, based on capillary rise in powder beds as described by Washburn's equation, was developed for quantifying mineral water interface tensions under conditions typical of diagenetic systems. The interface tensions of several common rock forming minerals were determined. Application of these new data in the classical nucleation theory based kinetic reaction path model results in more realistic levels of supersaturation.