



The impact of diagenetic fluid-rock reactions on the alteration of central German Rotliegend sandstones - a baseline evaluation on potential CO₂ reservoir rocks

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In Germany Rotliegend deposits form voluminous potential reservoirs for CO₂ storage in the framework of planned CCS-projects. Hence we investigated Rotliegend sandstones from several central German locations (Saxony-Anhalt, Thuringia, Baden-Württemberg) by sedimentological, petrographic, mineralogical, geochemical, and petrophysical means to evaluate former (CO₂-bearing) fluid-rock reactions during diagenetic burial and to predict potential future CO₂-fluid-rock reactions, which are most likely induced by CO₂ injection. We chose pristine red bed facies and bleached sandstones for these studies, because sandstone bleaching is commonly attributed to the alteration of red sandstones by (CO₂-bearing) fluids. Thus these deposits can serve as natural analogues for planned industrial CO₂ injection in clastic sediment sequences.

In general, Rotliegend sandstones deposited in an aqueous environment contain higher amounts of clay minerals and their pristine red to red-brown color is remained. In these rocks the amount of pore filling minerals is commonly high and any features of mineral dissolution are only minor. Thus this kind of sandstones exhibit low to moderate porosities and permeabilities. In contrast aeolian type sandstones show high porosities and permeabilities. This is caused by their primary (depositional) sorting, which enables extensive early diagenetic pore space cementation (by anhydrite and carbonates) and its late stage dissolution during burial diagenesis. This anhydrite- and carbonate dissolution is coupled with Fe-reduction and some Fe-release, leading to enforced sandstone bleaching with an increase in porosity by decreasing Fe whole rock content.

Fe-mobilization is also confirmed by the presence of yellowish-brownish goethite (?) bands in bleached sandstones as well as authigenic chlorite composition. Thereby chlorite laths in bleached rocks contain higher Si-, but lower AlIV- and Fe-contents as maintained in red sandstones, suggesting formation from compositional distinct fluids.

In the red bed sandstones also the whole rocks contents of Al, Na and K and Al/(Na+K) ratio are increased, with ratios up to 1.4, indicating higher illite content in these rocks. Here, negative correlations of these ratios with Ca-whole rock content and porosity are most pronounced. These features suggest that enhanced primary clay (illite) content inhibits early carbonate/anhydrite cementation and preserved lower porosity (and permeability) during diagenetic evolution. This is in contrast to Al/(Na+K) ratios in bleached rocks which scatter at about 1, reflecting the dominance of (alkali) feldspar instead of illite. Grain size and petrographic investigations sustain that in these rocks (illitic/chloritic) clay cutans are commonly less developed and (at least in some rocks) grain sizes < 2 μm (clay fraction) is almost absent. Nevertheless in some of these bleached rocks also some authigenic meshwork illites precipitated in the pore space during diagenetic burial.

Al/(Na+K) ratios are low in Ca-poor, highly porous bleached rocks, suggesting that authigenic illite formation had only minor impact on porosity (and permeability) evolution.

Hence the increase in porosity and permeability from red, fluvial to bleached, aeolian rock suites is accompanied by Ca- and S-, as well as Al-, K- and Fe-depletion reflecting the poverty of carbonate, anhydrite and clay minerals in the altered (bleached) rocks. Thereby in bleached rocks poroperm strongly increases with decreasing Ca-content, whereas in the red facies sandstones poroperm values are almost constant, despite strongly decreasing Ca. Instead, in the red rocks increasing porosity is coupled with decreasing Fe-, Al- and (less pronounced) K-content. These features confirm that in bleached rocks carbonate/anhydrite cement dissolution and in red rocks less pronounced illite/chlorite (cutan) transformation controlled poroperm evolution. Therefore at an initial stage of red bed sandstone bleaching effects on total porosity/permeability due to described reactions will be minor, but they will most pronounced during ongoing alteration/bleaching leading to strongly enhanced rock porosity and permeability – an evolution also most likely taking place during future industrial CO₂ injection.