Impact of CO_2-induced geochemical reactions on the mechanical integrity of carbonate rocks

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Abstract

Climate change due to human use of fossil fuels leads to carbon dioxide (CO_2) emissions. Carbon Capture and Storage (CCS) has a key role to play in curbing CO_2 emissions from thermal-power generation and industrial processes alongside renewables, energy efficiency, nuclear and other mitigation options. However, CO_2 storage can be regarded as a key challenge for a timely CCS deployment to substantially reduce carbon emissions. Up to the present, a number of industrial scale and pilot CO_2 storage projects have been operating worldwide. The idea behind subsurface storage is to inject CO_2 into deep (> 800m) suitable geological formations; CO_2 enters the host rock pore space initially occupied by one or more fluids. Its fate can then follow several possible paths, depending on specific circumstances: for example, CO_2 can migrate upwards as a separate phase, due to buoyancy and becomes physically trapped; alternatively, it can dissolve in the *in situ* fluids, leading to changes in pH and redox states, along with changed density. These changes could lead to fluid motion, or to *in situ* preccipitations of solid carbon-bearing minerals, along complex process pathways.

The scope of this work is to gain an initial understanding of how brine-saturated carbonate rock systems behave after CO_2 injection, which could have applications in places where carbonate rocks form either the host or the cap rock of the storage site (depending on its porosity and permeability). We intend to investigate the impact of CO_2-induced geochemical reactions on the mechanical integrity of oolitic limestone samples, which have been subjected to reservoir conditions at the laboratory scale. To achieve this target, we perform high pressure – high temperature tests on brine-saturated samples aiming at identifying changes in fluid chemistry and limestone mineralogy upon the completion of each test. Moreover, by preparing pre- and post-treatment thin sections from regions close to the sample edges we aim at identifying possible deformation micro-processes and mineral changes that could occur due to the thermo-chemo-mechanical loading of the samples.

SEM imaging on the study material before the experiments has shown that this oolitic limestone has a relatively low porosity. Calcite cement grew into the pore space from the ooids' margin, as a fine-grained radially orientated cement phase, followed by a coarser pore occluding cement. However, calcite is not everywhere well attached to the ooids. Moreover, micro-porosity and pressure solution have been identified within some of the ooids and at their edges, respectively.

Grain-scale investigations based on thin section observations (SEM imaging) on a brine-saturated limestone sample, exposed to 230 bar CO_2 pressure and 37.5^o C (supercritical conditions) for 2 weeks, have shown no obvious fractures due to the thermo-chemo-mechanical loading of the sample. Micro-porosity in a few ooids together with occasional big voids in cement between ooids have been identified locally in the post-deformation/treatment sample, however similar observations were made in the pre-treatment sample. Moreover, a few sporadic fractures along ooids have been identified in both pre- and post-deformation samples. All micro-scale observations were made in regions 1-3 mm from the sample edges, indicating no actual grain-scale deformation. On the other hand, the increase in concentration of calcium ions in the post-treatment brine has been linked to dissolution of the limestone, which has possibly taken place along the surface of the sample. Thus, it can be argued that under the current test conditions (230 bar, 37.5^o C, 2 weeks exposure) CO_2 does not much damage this repository. Further research on different conditions (particularly longer exposure and more acid solutions that can be linked with faster dissolution of calcite) is currently underway.

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