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Limestone reservoirs: are they good for CO_2 geological storage?

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A promising method that could drastically reduce the effects of anthropogenic carbon-dioxide emissions is the capture of CO_2 and its storage in geological formations (CCS technology). The processes that can take place in saline aquifers got under the spotlight in the last decades and the most promising options are sandstone reservoirs. However, natural CO_2 trapped in carbonate (limestone) reservoirs are not well studied. The general assumption is that CO_2 aggressively dissolves the limestone (matrix, grains, and cement), which would cause drastic changes in the reservoir properties (e.g., porosity, permeability).
To better understand the processes that CO_2 injection can cause in a carbonate reservoir, a natural CO_2 subsurface occurrence in Ölbő (Hungary) was investigated, where CO_2 has been trapped safely in the limestone on a geological timescale. Core samples of the reservoir from 1700-1900 m depth were studied with various methods like petrography (carbonate facies analysis, nannoplankton determination), scanning electron microscopy, cathodoluminescence microscopy, X-ray diffraction and infrared spectroscopy. Microdrilling of the carbonates was also carried out to determine the C and O isotope composition of different constituents in order to reveal possible dissolution/recrystallization processes which may occur in the CO_2 reservoir.
Two types of cement were found in the samples, a blocky, drusy cement and a syntaxial cement on the echinoderms (early cement). Contrary to the assumption, dissolution features, may be related to the CO_2 inflow, were not observed in the rocks.
The average mineral composition of the samples is the following: 79 m/m% calcite; 6 m/m% dolomite; 3 m/m% ankerite, mica and quartz; 1 m/m% kaolinite, minor feldspar and pyrite. Dawsonite, the indicator mineral of CO_2 flooding in siliciclastic sandstones, was not identified in the samples.
Carbonate components of the rock are Red algae, Foraminifera, Bryozoa, Bivalves, Echinoderms and Brachiopods. Nearly all were originally calcitic. Based on nannoplankton biostratigraphy and literature, the age of the host rock is Upper Badenian (Serrevallian), Middle Miocene.
The stable C and O isotope data of microfossils shows a narrow range, $\delta^{13}C$ is ranging from -1.55‰ to 2.05‰ (average: -0.23‰), $\delta^{18}O$ is between -7.98‰ to -0.25‰ (average: -4.54‰),

expressed on the V-PDB scale. These data do not indicate the effect of magmatic CO_2, which may reside in the Ölbő reservoir (Cseresznyés et al., 2021), in agreement with the petrography. According to our preliminary results, CO_2 inflow did not affect the Ölbő limestone reservoir, i.e., did not imply significant dissolution, neither was involved in cement precipitation. Limestone thus could be an excellent physical trap for CO_2. However, due to limited mineral reactions, our results indicate that limestone reservoirs may not be the best for mineral trapping which is the safest storage mechanism of CO_2 on geological timescale. Further analyses will be carried out with geochemical modeling, to study the water-CO_2-limestone reactions based on the Ölbő CO_2 field.

Reference:
Cseresznyés et al 2021. ChemGeol. https://doi.org/10.1016/j.chemgeo.2021.120536