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Two-phase matrix transport & sorption experiments on low-permeable mudstones and shales within the CO_2SEALS project

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In the CO_2SEALS project (www.co2seals.de/) of the German GEOTECHNOLOGIEN Program, the effect of dissolved CO_2 at subsurface pressures and temperatures on the sealing efficiency of clay-bearing lithotypes has been investigated. The primary objective was to improve the quantitative understanding of CO_2 transport/retention processes within intact (unfractured) fine-grained rocks and the associated CO_2/brine/rock interactions. For a series of generic clay-rich lithotypes, fluid transport and sorption behaviour were determined at elevated pressures and temperatures.

Flow experiments were conducted on pugs (38 mm in diameter, ∼15 mm length) at confining pressures ranging between 15 and 29 MPa and temperatures of 25 to 45°C by different procedures. A selected number of representative samples were investigated, including mature shales/siltstones, a marl/limestone, and immature mudstones. Intrinsic permeability coefficients ranged between 3E-18 and 2.3E-22 m^2.
For the more highly permeable immature mudstones and the limestone, gas breakthrough could be achieved under the experimental conditions. For the limestone sample, capillary breakthrough occurred between 1.3 and 2.9 MPa, with maximal permeabilities of 2.4E-21 m^2. For the immature mudstone no capillary breakthrough of He and CO_2 could be achieved with differential pressures of up to 6 MPa. At 14 MPa breakthrough occurred rapidly with a maximum effective permeability coefficient of 7.2E-22m^2.
The mature shales were shown to be efficient barriers up to a pressure difference of at least 20 MPa for He and 9 MPa for CO_2. No gas breakthrough was observed under these pressure conditions. From these experiments it became evident that CO_2 transport through these caprocks is restricted to diffusion.

High-pressure CO_2 sorption tests were conducted on dried (200°C, >24h) and crushed rock material (500-1000 μm fraction). The measurements were carried out at 45°C and pressures up to 25 MPa.
The maximum CO_2 excess sorption capacities of shale samples ranged from 0.2 to 0.6 mmol/g, while quartz-rich sandstones and calcitic carbonates had lower CO_2 sorption capacities of 0.03 and 0.12 mmol/g respectively. No significant differences in CO_2 sorption behaviour was found for different particle size fractions of the same shales. As expected, sorption capacity decreased with increasing experimental temperature. From the sorption isotherms measured on Opalinus clay at 45°C, 57°C and 70°C an average isosteric heat of sorption of -26 kJ/mol was calculated. This is a realistic value typical for physisorption. A positive correlation was found between CO_2 sorption capacities of shales and specific surface area (BET method), as well as the TOT-clay (smectite + illite and illite-smectite mixed layer clays) content. No statistically significant correlation was found between CO_2 sorption capacity and total organic carbon content (TOC).

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