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Potential of caprocks as CO_2 storage reservoirs

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Shale lithotypes of various compositions occur abundantly in sedimentary basins and act as natural seals for petroleum and natural gas reservoirs over extended geologic periods. Based on to their mechanical, petrophysical and chemical/mineralogical properties, shales are becoming increasingly of interest in the context of long-term isolation of anthropogenic (e.g. radioactive) waste and subsurface storage of fluids. The efficiency and long-term integrity of seal formations (caprocks) is also one of the central issues for CO_2 storage in saline aquifers, depleted oil and gas reservoirs and coals. Due to its chemical reactivity and physico-chemical properties, CO_2 is expected to differ substantially from other natural gas components in terms of transport behaviour and interaction with the mineral/water system.

An experimental procedure has been developed to measure molecular diffusion of CO_2 in water-saturated shales. This non-steady state method provides information on the effective diffusion coefficients and the CO_2 storage capacity of the shales. Effective diffusion coefficients for CO_2 were found to range between 10^{-9} and 10^{-11} m^2/s. Storage capacities were found to vary significantly but can be as high as 0.14 mmol CO_2/g sample. This storage capacity was found to be significantly higher than CO_2 solubility in water present in the pore system.

Therefore, volumetric sorption experiments with CO_2 at pressures up to 20 MPa have been performed on dry and moist shale samples to test for additional storage options of CO_2 in shales. The results of these experiments revealed unexpectedly high storage potentials of the same order of magnitude as those from the diffusion measurements. The CO_2 storage capacities are not only related to organic carbon content. In further experimental studies it was shown that the sorptive CO_2 storage potential of clay min-

erals (montmorillonite, kaolinite, etc.) is significantly high.

These findings may provide a new view on the issue of caprock integrity. In addition to their sealing properties, natural shale sequences could represent a significant sink for carbon dioxide deposited in the subsurface by fixing and immobilising it and hence reduce the risk of leakage to the surface.