



Experimental Studies on Seal Efficiency for CO₂ Storage

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One of the key factors in subsurface storage of CO₂ is the assessment of the efficiency and long-term integrity of top seals which, in many instances, consist of argillaceous rocks. Main parameters determining the seal quality are the capillary gas sealing efficiency and gas permeability after capillary breakthrough. Failure of an initially water-saturated capillary seal (“membrane seal”) will occur once the gas pressure exceeds a critical value (capillary entry pressure, breakthrough pressure) and the gas phase forms an interconnected pathway along the pore system of the rock. The effective gas permeability after breakthrough will depend on the gas saturation and, correspondingly, on the gas pressure gradient. Capillary breakthrough tests with methane, nitrogen and carbon dioxide have been performed to provide fundamental information on the gas sealing efficiency of various mudrocks. By monitoring the spontaneous re-imbibition process the effective gas permeability was determined as a function of the gas pressure gradient. The residual pressure difference at the end of the re-imbibition process was interpreted as the (minimum) capillary breakthrough pressure.

Single phase permeability measurements with water on pelitic rock samples typically yield permeability coefficients in the microdarcy down to the sub-nanodarcy range ($k=10^{-18}$ - 10^{-22} m²). Effective gas permeability coefficients after gas breakthrough are significantly lower than the single phase coefficients with values as low as 10^{-24} m². Capillary breakthrough pressures for all three gases were found to correlate to the maximum effective permeabilities. CO₂ generally showed lower capillary breakthrough pressures than the other two gases.

Based on the experimental results a dynamic model for gas leakage through shale

caprocks was designed taking explicitly into account the variation of effective gas permeability as a function of pressure.

Diffusion experiments with CO₂ performed on water-saturated shales, siltstones at elevated pressures (up to 6 MPa) have yielded effective diffusion coefficients in the range from 0.2 – 2·10⁻¹⁰ m²/s and revealed unexpectedly high CO₂ storage capacities for individual shale samples. Diffusive loss of CO₂ from underground storage sites through shale caprocks is not considered a relevant leakage mechanism. However, the effects of this chemically reactive gas on the mineralogy and, in consequence, on the rock-mechanical properties will be controlled by diffusion rates.