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Mineral Alteration and Changes in Transport Properties in Caprocks due to CO_2 Treatment – an experimental Study

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Interaction of CO_2 with pelitic caprocks (shales, marls) may lead to alterations of mineralogy and transport properties. In order to investigate these processes and their potential effects on seal integrity, fluid flow experiments were conducted with selected caprock samples under subsurface P/T conditions. The experimental program comprised permeability, gas breakthrough and diffusion tests on cylindrical plugs of 10 - 20 mm thickness and 28.5 mm or 38mm diameter, respectively. The samples were characterized by petrophysical methods and mineralogical analyses in their original state and after the flow experiments.

Before each gas breakthrough and diffusion experiment with CO_2, single phase flow tests with water were performed to determine permeability coefficients and ensure complete water-saturation of sample plugs. Permeability tests were repeated after each CO_2 experiment to detect changes in the fluid transport properties.

Capillary gas breakthrough tests were performed according to the procedure described by Hildenbrand et al. (2002). To test for reproducibility and petrophysical changes due to the interaction of the samples with CO_2, repetitive runs were carried out on the same sample. Gas breakthrough experiments with Helium and CO_2 under the same conditions were conducted to compare the transport properties of inert and reactive

gases.

Diffusion experiments with CO_2 were carried out using the method of Schloemer and Krooss (2004). These tests revealed unexpectedly high CO_2 storage capacities for certain shales and marls. Significant increases in (water) permeability coefficients were observed after CO_2 diffusion experiments.

Changes in petrophysical properties were also observed after repetitive CO_2 gas breakthrough tests. These comprised an increase in (water) permeability as well as increases in effective gas permeability in subsequent gas-breakthrough tests. On the other hand the capillary entry pressures of the samples decreased during the tests, indicating a lower capillary sealing efficiency as a result of repetitive exposition to CO_2.

Mass balance calculations for a clay-rich sample indicated significant CO_2 loss from the gas phase during the first breakthrough tests. This loss exceeded the amount of CO_2 that could be dissolved in the pore water and was attributed to sorption and mineral reactions. No further CO_2 loss was observed in subsequent tests with the same sample. A calcite rich sample did not show similar effects, probably due to the absence of clay minerals and organic matter.

Unexpectedly high CO_2 storage capacities of shales and marls were also evidenced by manometric sorption experiments on powdered samples. In order to clarify whether these are due to sorption or chemical reactions, XRD analyses, BET tests, and Hg porosimetry measurements are presently being performed on the original and CO_2-exposed samples.

References:

Hildenbrand et al., 2002. Gas breakthrough experiments on fine-grained sedimentary rocks. Geofluids (2002) 2, 3-23

Schloemer S. and Krooss B., 2004. Molecular transport of methane, ethane and nitrogen and the influence of diffusion on the chemical and isotopic composition of natural gas accumulations. Geofluids 4(1), 81-108.