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Development of Kinetic Interface Sensitive Tracers (KIS-Tracer) for Supercritical Carbon Dioxide Injections into Deep Saline Aquifers

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The storage of captured CO_2 into geological formations is recently one of the most promising technologies to mitigate anthropogenic greenhouse gas emissions into the earth's atmosphere. Deep saline aquifers are considered as the most potential sequestration sites of CO_2 due to their huge storage capacities of several thousand Gt.

Ongoing research deals mainly with the investigation of relevant physico-chemical processes, the fate of CO_2 and the risk assessment during and after supercritical CO_2 ($scCO_2$) injections. The occurring processes at the interface between injected $scCO_2$ and formation brine play a major role to evaluate the fate and behavior of $scCO_2$ in the reservoir. This is because the interface represents a reactive zone where numerous physico-chemical processes like dissolution of $scCO_2$ in water as well as dissolution and precipitation of minerals take place. In most cases it is desired to maximize the interface size to increase the storage efficiency. Therefore, knowledge on interface size and dynamics would allow the observation of plume spreading and the detection of mixing or fingering effects. In order to gain this information innovative tracers are necessary which are able to quantify the temporal and spatial development of $scCO_2$/water interfaces. As a result, it may be possible to assess the storage efficiency and to optimize subsequent injections. Up to now, such time-dependent tracers for reservoir studies are not available and limited to equilibrium tracers (known as partitioning and interfacial tracers, respectively). Therefore, novel reactive tracers (KIS-Tracers) are developed to overcome this gap. The idea is to find suitable molecules which allow the implementation of a defined chemical reaction at the interface. Due to the known kinetic constants the change of interface size can be characterized over time.

The new tracer is injected together with the supercritical CO_2 ($scCO_2$) into a deep saline aquifer. Afterwards, the tracer adsorbs at the interface and undergoes hydrolysis in contact with water. As a consequence, two water soluble reaction products are formed and can be measured in the water phase over time. Here, the reaction kinetics is the rate-limiting step for the phase transfer and strongly dependents on reservoir properties, such as temperature and pH. Such tracer molecules must have the following properties: i) low polarity (high log K_{OW}) to ensure high $scCO_2$ solubility and to minimize distribution into the water phase; (ii) at least one highly water soluble reaction product, which does not do partitioning back into the $scCO_2$ phase; (iii) low detection limit.

On the basis of naphthalenesulfonic acid, an established geothermal tracer, different molecules with the desired properties were synthesized and tested in the laboratory. For studying the occurring processes at the interface under atmospheric pressure conditions the $scCO_2$ was replaced with a non-polar organic solvent. The experiments were conducted in a static batch system with constant interfacial area as well as in a dynamic system with changing interface size. In parallel, a macroscopic model which couples mass transfer and reaction kinetics is developed to interpret the data. In conclusion, experiments indicate that the integration of hydrolysis kinetics is possible and even one of the reaction products may be used as additional partitioning tracer, i.e. for measuring the residual saturation.