Performance of Silica-gel as a Geochemical Barrier under CO$_2$ Storage Conditions

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Abstract

The risk of CO$_2$ leakage must be considered during the planning, injection and monitoring stages of a Carbon Capture and Storage (CCS) operation in order to meet regulatory standards. Various strategies can be used to reduce the risk of a CO$_2$ leak including forming hydraulic barriers, permanent cement- or reactive-barriers, which include the use of chemical reagents that react upon contact with CO$_2$ to form a geochemical barrier. Solutions of concentrated sodium silicate, an industrial by-product, have been proposed as a promising reagent for producing geochemical barriers under CO$_2$ storage conditions. Interaction of CO$_2$ with this reagent leads to the rapid formation of an amorphous silica gel that can form a barrier to reduce permeability in a reservoir [1-2]. To investigate the formation of the silica gel barrier under CO$_2$ storage conditions, we combine laboratory work and numerical modeling. Here we present the results of reagent optimization from flow through column experiments at ambient conditions using an unconsolidated quartz sand. Multi-phase core flooding experiments were performed at reservoir conditions to assess the associated reduction in the permeability of a typical sandstone reservoir rock (Berea Sandstone) and the stability of the gel and barrier formed over time. Finally, reactive transport modeling of the experiments was conducted using TOUGHREACT, with modifications made to the thermodynamic database to account for the properties of the amorphous silica gel, which forms under CO$_2$ storage conditions.

An optimal composition for the sodium silicate reagent was identified as a pH 10.8 solution of 7.15 wt % SiO$_2$ with a 3.21:1 Na$_2$O:SiO$_2$ ratio. This reagent composition has a density of 1.1 g cm$^{-3}$ and viscosity of 0.774 centipoise at 60°C, physical properties similar to a typical low salinity groundwater, therefore was found to be suitable for injection in flow through columns for forming silica-gel barriers in situ. A pH 4 buffer solution was used as a proxy for CO$_2$ saturated waters and reaction with the sodium silicate reagent led to reduced permeability in the flow through columns due to the formation of the silica gel barrier. Micro-CT was used to image the barriers in situ showing the near complete occlusion of porosity in a column flushed with a pH adjusted sodium silicate reagent, which set as a gel within 10 minutes of injection. Implementing the reagent in this manner led to permeability being completed impeded in the column (change from c. 5 D to < 0.05 mD).

Core-flooding experiments were conducted at 60°C and with a back-pressure of 100 bar to simulate CO$_2$ storage conditions. Berea Sandstone cores had an initial permeability of c. 110 mD and porosity of c. 17.5%. The 10 cm long, 3.8 cm diameter, core samples were initially saturated with the sodium silicate reagent, simulating a scenario where the reagent is used to mitigate the risk of CO$_2$ leakage by injecting the reagent into a reservoir overlying the cap-rock of a CO$_2$ storage reservoir. Changes to the permeability of the 3 sandstone cores were monitored during the injection
and incubation of CO₂-saturated water and supercritical CO₂ in separate cores. Figure 1 summarises the results, showing that permeability in the core is reduced by up to three orders of magnitude after barrier formation (0.11 mD) and increases by an order of magnitude after aging for around a month.

![Figure 1. Permeability of three Berea Sandstone cores (Core 1: triangles, Core 2: circles; Core 3: squares) pre-saturated with barrier forming reagent following the injection of A) CO₂-saturated water and B) supercritical CO₂.](image)

Numerical models were setup with the same geometry and conditions of the laboratory experiments, including testing continuous versus intermittent injections. In order to account for the large molar volume of silica gel the thermodynamic properties for amorphous silica (SiO₂(am)) in TOUGHREACT’s database were changed in order to optimize the volume of SiO₂(am) which precipitates. The precipitation of SiO₂(am) is associated with a gradual change in pH at the reaction front in the model and a mineral saturation index for SiO₂(am) greater than zero. A subsequent reduction in porosity is observed as SiO₂(am) precipitates. Figure 2A illustrates the simulation of the flow through column experiments for 3 days with 3 curing periods. The porosity change is greatest at the inlet with subsequent reduction in porosity further into the columns after the defined curing periods. Significantly, this indicates that a curing period is needed to enhance barrier formation at the reaction front in the model. The results simulating core-flooding experiments depict a 50% reduction in porosity near the inlet of the core (Figure 2B) after 20 minutes of reaction, which corresponds well with the permeability decrease seen during the injection phase of core-flooding experiments (Figure 1). The rate of SiO₂(am) precipitation is being further optimized in the model to observe the effect on changes to the porosity in columns and core.

![Figure 2. TOUREACT modelling results of injection of CO₂-enriched water into a silicate saturated column, with sequential curing periods under A) ambient conditions; B) reservoir conditions (60°C and 100 bar).](image)
The laboratory work together with the modeling indicate that using sodium silicate reagents to form a silica gel barrier is a viable strategy for mitigating the risk of a CO$_2$ leakage. Reduction in the porosity (up to 50%) and permeability (up to 3 orders of magnitude) of unconsolidated sand packed columns and a sandstone reservoir rock (Berea Sandstone) were observed during barrier formation. Modeling and experiments under field-scale conditions are needed to further validate the use of the silica gel to form a geochemical barrier under CO$_2$ storage conditions.