



Preliminary investigation on the chemical response of cementitious grouts used for borehole sealing in geologically stored CO₂

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The successful geological storage of CO₂ in underground reservoirs aims to immobilize the injected CO₂ stream in the form of secondary minerals through reaction with primary minerals or pore fluids in the host rock formations. Injection wells and other boreholes within the reservoir represent a major potential pathway for CO₂ to leak back to the surface. Therefore, the stability of well seals is a critical factor for the risk assessment of existing and the design of new CO₂ injection wells. Cement-based grouts emplaced within the steel borehole liner, and between the liner and the rock formation, must seal the well against leakage, both during the CO₂ injection stage and for a significant time after well abandonment, to allow for the CO₂ to be immobilized through rock-water interaction in the reservoir. The injected super-critical CO₂ (scCO₂) experiences temperatures up to 180°C and pressures at depths greater than 800m, and when dissolved in rock formation waters create chemically reactive species that could impact the stability of cement seals. In an attempt to evaluate the impact of scCO₂-saturated fluids in class G oilfield grouts, batch experiments at 80bar and 60°C/ 120°C were carried for pure cement and cement-steel cylindrical samples immersed in a realistic formation porewater composition. Destructive and healing features were observed by means of backscattered scanning electron microscopy (BSE) and energy-dispersive X-ray microanalysis (EDS) elemental mapping; both phenomena were evident in Ca leaching from, and deposition on, the surface of the samples, respectively. Structural cement components like Si appear to have retained their original particle-like shape in the regions affected by the CO₂ in the 60°C experiments, but their preservation at 120°C is vaguer. The liberation of Ca²⁺ from the hydrated cement particles (indicated by local decrease of the Ca/Si ratio), and the reactions with the incoming carbonate/bicarbonate anions seem to evolve progressively in distinct zones formed parallel to the outer surface that are evident in BSE images. The proposed mechanism of pH induced release of Ca²⁺ from C-S-H and Ca(OH)₂ particles due to CO₂ fluids, and re-precipitation as carbonates seem to be controlled by the fluid to solids ratio and as a result from the confinement of the exposed surface.