Investigation of the Chalk Dissolution Kinetics in Brines Loaded with Carbon Dioxide

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CO₂ injection for carbon capture and storage (CCS) and enhanced oil recovery (EOR) projects is largely performed in high permeability sandstone formations or carbonate reservoirs worldwide. The oil and gas production in Denmark is mainly from high-porosity and low-permeability chalk reservoirs. For CO₂ storage in the offshore depleted fields in Denmark, it is therefore critical to evaluate the feasibility of storing CO2 in chalk reservoirs. The higher reactivity of chalk with CO2 as compared to other types of rock can weaken the storage site safety with fault reactivations and impact the caprock and well integrity. Knowledge of the reactivity between calcite minerals and CO₂-saturated water/brine thus represents a major challenge to assess the suitability of chalk formations for CCS and EOR. In order to develop accurate reactive transport and thermodynamic models in reservoir simulators to describe the interactions between the brine phase with dissolved CO2 and the available mineral phases, experimental data for the kinetics of dissolution/precipitation of chalk exposed to CO₂-saturated brines at the in-situ conditions are required. One possibility is to quantify the proton activity induced by these chemical reactions through the pH in brine. However, measuring the pH at high pressures, temperatures and salinity is challenging due to the fast reactive behavior of chalk. In this work, a high pressure and high temperature glass-based pH probe combined with an internal standard Ag/AgCl reference electrode has been employed to measure the pH of CO₂-saturated water/brine in the presence of calcite/chalk samples at temperatures ranging from 20 to 80 °C and at pressures from 0.1 to 9 MPa. The dissolution rates of calcite and chalk were then determined for different solid-liquid ratios. The outcome of our study revealed that the high specific surface area of chalk results in a higher dissolution rate than for calcite and that the presence of CO₂ further accelerates the chalk reactivity. In parallel, real-time monitoring of the mineral dissolution rate was performed by using a pH-sensitive fluorescent molecule and an optical probe. The response of quartz crystal resonators with mineral samples glued on their surface when immersed in CO₂-saturated brines was also studied.