A mechanistic interpretation of potential CO₂ leakage through shaly caprocks

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Global warming brought upon by anthropogenic CO₂ emissions into the atmosphere is causing significant impacts on the Earth and represents one of the major concerns of the current century. To be controlled, it is widely accepted that huge amounts of CO₂ at the gigatonne scale have to be captured and injected back into the underground in a process known as Carbon Capture and Storage (CCS). As CO₂ is less dense than the in-situ brine, it tends to flow upward out of the storage reservoir by buoyancy and the injection overpressure. A laterally-extensive and thick non-fractured caprock possessing low permeability and high entry capillary pressure is commonly expected to keep CO₂ within the host reservoir. However, the potential risks of CO₂ leakage through the intact caprock need thorough assessment. This contribution brings together experimental observations and numerical simulations to inspect the sealing capacity of an intact shaly caprock and render an in-depth understanding of the governing flow mechanisms. Reproducing the subsurface conditions of CO₂ intrusion and flow through the caprock, breakthrough experiments are conducted on Opalinus Clay as a representative caprock for CO₂ storage. The adopted approach consists of injecting supercritical CO₂ into the caprock sample lying between two permeable porous disks, all initially saturated with brine. Supplementary experiments are also performed to characterize the pore structure and hydromechanical properties of the specimen. The extracted properties are used to parameterize a two-phase flow model in deformable porous media and simulate the breakthrough experiment carried out on Opalinus Clay to make a mechanistic interpretation of the experimental observations. Simulation results reveal three concomitant CO₂ flow mechanisms into and through the caprock: molecular diffusion, bulk volumetric advection, and transported CO₂ dissolved in the advected brine. It is inferred that the high entry pressure and low effective permeability prevents free phase CO₂ penetration deep into the caprock. The drainage path is followed by the imbibition of brine back into the pores from the downstream until recovering the initial state of being completely saturated with brine. While the contribution of brine advection to CO₂ transport is found to be negligible, we find that CO₂ flow through the caprock is mainly governed by molecular diffusion, whose effects on the potential leakage of CO₂ during geological time scales have to be taken into account.