

CONVECTIVE CO₂ DISSOLUTION IN AQUIFERS

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Summary. The storage of carbon dioxide (CO₂) in geological formations has been proposed as a technological means to reduce anthropogenic emissions of this greenhouse gas. The buoyancy of supercritical CO₂ relative to the ambient brine, may lead to leakage along imperfections in the geological seal, which is of considerable concern for the security of long-term storage. Dissolution of CO₂ into the brine, which results in stable stratification, increases security of storage over time and has recently been identified as a major sink for CO₂ in natural accumulations. The rate of CO₂ dissolution is determined by convection in the brine driven by the increase of brine density with CO₂ saturation. Here we present a new analogue fluid system that reproduces the nonlinear density behaviour of CO₂ and brine. We show that the convective flux is proportional to the Rayleigh number to the 4/5 power through a combination of laboratory experiments and high-resolution numerical simulations, in contrast with a classical linear relationship. The identification of this relationship allows us to extrapolate from the laboratory scale to geophysical scales. A scaling argument that incorporates the effect of the large-scale flow on mixing at the CO₂-brine interface confirms this nonlinear relationship for the convective flux and provides a physical picture of high Rayleigh number convection in a porous medium. The resultant model makes quantitative predictions of the CO₂ dissolution rates in natural and anthropogenic CO₂ accumulations. For example, at the Sleipner field we estimate a dissolution rate of roughly 10% of the annual

injected mass. This suggests that convective dissolution can significantly enhance storage security. Convective mixing provides the key mechanism controlling carbon mobility in the deep terrestrial carbon cycle and therefore sheds light on geochemical observations that require large amounts of CO₂ to dissolve into the brine.