Geological sequestration of carbon dioxide (CO₂), originating from sources like fossil-fueled power plants and contaminated gas production, is presently seen as a large scale disposal option to reduce greenhouse-gas emission (IPCC 2005). Deep saline aquifers and depleted oil and gas fields are potential candidates for CO₂ subsurface deposits. Trapping mechanisms and the degree of trapping are important factors for assessing the storage capacity for sequestration in formation rock and depend to a large degree on the fluid-fluid displacement processes in the subsurface and the mass transfer between the fluid phases.

The process of displacement and mass transfer between CO₂ and brine, which are relevant for the prediction of plume migration and pore-space utilization during and after CO₂ injection in saline aquifers, were studied by conducting unsteady-state core flood experiments in nearly homogeneous Berea sandstone rock. Mutually saturated and unsaturated CO₂ and brine phases were injected in the rock under realistic sequestration conditions. The Berea rock sample had only very low heterogeneity and was sufficiently well described with a homogeneous numerical model. A sensitivity test showed that the degree of heterogeneity in this sample has only very little impact on the resulting parameters. Three drainage experiments were conducted with brine and a model fluid (decane), and with unsaturated and saturated CO₂–brine. In addition, an imbibition experiment was conducted where unsaturated brine was injected into saturated brine and near-residual CO₂.

The unsteady-state drainage experiments were analyzed by “history matching” with a numerical model. For the decane–brine experiments, the relative permeability and capillary pressure obtained were in good agreement with steady-state and centrifuge measurements, respectively, performed on twin samples. The saturated CO₂–brine drainage experiment was conducted with equilibrated phases such that mass transfer between the phases during the experiment is prevented. The relative permeability obtained is clearly different than the one from the decane–brine experiment. The differences are attributed to a different wetting behavior of CO₂–brine which is more intermediate-wet compared to a water-wet situation for decane–brine, an observation which is supported by contact angle data from literature. This finding indicates that with respect to wettability it does matter whether experiments are conducted with CO₂ or with model fluids provided that a model fluid is found that exhibits similar wetting properties. When comparing saturated with unsaturated CO₂–brine drainage experiments, solubility and mass transfer from brine to CO₂ and from CO₂ to brine were observed. The first case, water dissolving in CO₂ (solubility approx. 0.08 vol-%), leads to evaporation at a 2-3 cm wide zone at the inlet, i.e. the length scale $l_{\text{evap}}$ of water transfer at the respective flux is much smaller than the sample length. The decrease of water saturation in the evaporation zone estimated from CT data is larger
but of a comparable order of magnitude to estimations from solubility data. The second effect, CO$_2$ dissolution in brine (solubility approx. 9 vol-% in the CO$_2$ phase), leads to a diminished displacement of brine by CO$_2$ which is most pronounced at the point of breakthrough. The magnitude of the effect is close to predictions from solubility data. This effect influences the saturation profiles from inlet to outlet, meaning that the length scale of the effect $l_{\text{diss}}$ is comparable to or larger than the sample size and also much larger than $l_{\text{evap}}$, $l_{\text{diss}} > l_{\text{evap}}$. These findings may matter in cases where phase equilibration effects are relevant but for numerical modeling, a discretization coarser than $l_{\text{evap}}$ and $l_{\text{diss}}$ is used. Imbibition of unsaturated brine into the rock sample at near-residual (trapped) CO$_2$ and CO$_2$ saturated brine shows first the miscible displacement of saturated by unsaturated brine and then a dissolution of the CO$_2$ phase. This experiment represents the transition from residual trapping to solubility trapping.