Chapter 18

[[Ch. Heading]] Closing Remarks: Future Research Needs for Geological Carbon Storage

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[[H2]] Abstract

Geological carbon storage (GCS) is a rapidly-evolving technology, which can become vital in mitigating the increasing concentrations of atmospheric carbon dioxide (CO_2). While we understand overall phenomena associated with injecting and storing CO_2 in porous subsurface formations, unknowns persist. For successful geological carbon storage, trapping of CO_2 over extended time periods (10^3 to 10^4 years) is required. Existing predictive models examining these time scales include large uncertainties and require improvement. Addressing challenges associated with temporal and spatial scales, characterizing and controlling mechanisms, and kinetics of individual, as well as coupled processes are central to this technology. One of the key future research areas is developing fundamental science to understand process coupling at relevant temporal and spatial scales, and being able to predict, anticipate, and mitigate emergent behavior (e.g. development of preferential flow paths). This chapter summarizes the current state-of-the-art knowledge detailed in the previous chapters and presents future research needs for successful GCS.

Keyword: *Geological carbon storage (GCS), coupled processes*

[[H1]] CO_2 storage in saline formations: scientific discoveries and challenges

Subsurface systems targeted for geological carbon storage (GCS) are heterogeneous and react dynamically to injection of large volumes of CO_2. For GCS to have an impact on atmospheric CO_2 concentration, annual injection of supercritical carbon dioxide ($scCO_2$) on the order of billions of metric tons into deep subsurface reservoirs is required (IPCC, 2014, NETL, 2015). For safe and successful GCS, CO_2 should remain in the subsurface for thousands of years. The injection of such large volumes of CO_2 perturbs the pore pressure, state of stress, chemical, mechanical, thermal, and biological steady-state or equilibrium conditions of subsurface reservoirs. Since $scCO_2$ is immiscible with brine in reservoir, two-phase fluid flow with CO_2 fingering develops and bypasses some of the available pore space. The initially dry $scCO_2$ phase dehydrates the formation and forms a partially wet $scCO_2$ front. Wet $scCO_2$ is chemically reactive, along with the CO_2-acidified formation brines. Following the injection of CO_2, the subsurface starts re-equilibrating towards new equilibrium or steady-state conditions. This re-equilibration proceeds along different time scales for different processes.

Laboratory-scale studies show that carbonation reactions proceed in CO_2-acidified brines, as well as in the wet $scCO_2$ phase, in which the activity of water dictates the resulting products and reaction rates. Carbonation of mineral surfaces exposed to wet $scCO_2$ is as important as reactions in the aqueous phase, with the extent of carbonation dependent on the thickness of the water film (Loring et al., this volume). While sheet silicates are not susceptible to chemical attack by CO_2, expandable clay minerals can undergo shrinkage because of dehydration by neat $scCO_2$, and swelling due to intercalation of water and CO_2 in the interlayer (Loring et al., this volume). These physical changes in swelling clay minerals can impact the bulk mechanical and fracture properties of caprock.

The alteration of brine chemistry and the invading $scCO_2$ phase can alter native microbial communities. Microbial processes have a strong impact on matrix permeability, wettability, solution chemistry, and carbonation reaction rates (Thompson et al., this volume). Field injection tests and laboratory studies of mesocosms show that some microorganisms are resilient and thrive in environments with high CO_2 fugacity.

Field-scale CO_2 injection tests showed that observed changes in reservoir brine chemistry agreed with those predicted based on pre-injection geochemical characterization and modeling. Dissolution of CO_2 into brine takes place rapidly, and the amount of dissolved CO_2 is on the order of laboratory-predicted concentrations (Hovorka and Lu, this volume). In terms of geochemical characterization during field-scale tests, the main difficulty is obtaining representative pore-fluid samples from a multi-phase deep reservoir. Reservoir-scale predictive models for geochemical changes have not been validated at the corresponding field scale so far. For example, it is not possible to quantify the amount of brine within a rock formation that encounters $scCO_2$ at the reservoir scale.

Important lessons for GCS can be learned from experience with CO_2 injections in oil reservoirs for enhanced oil recovery (EOR). The volumetric sweep efficiency observed in CO_2-EOR was low; however, sufficient CO_2 injection rates were achievable without over-pressurizing the reservoirs, and the retention of a significant portion of CO_2 in the subsurface at these EOR sites was observed (Lake et al., this volume).

Understanding multi-phase flow is of paramount importance for successful GCS. Multi-phase flow is complicated due to contrasts in relative permeability, density, viscosity, and capillary entry pressure between different fluids (Jia and McPhearson this volume, Pini and Krevor, this volume, and Bandilla and Celia, this volume). Field tests indicated that years to decades are required for a CO_2 plume to stabilize in the subsurface. The time required for the onset of gravity-driven convection due to density contrast between native and CO_2-acidified brines varies from months to thousands of years (Ennis-King et al., 2005). Depending on the flow conditions, the size of viscous fingers formed by $scCO_2$ can be on the order of millimeters to centimeters, and can only be observed in cm-scale laboratory studies or simulations with sufficiently fine grids. Larger-scale channeling was also observed and was governed by compositional and textural heterogeneities in the target formation. Wettability of reservoir rocks and caprocks may be altered due to interfacial chemical reactions triggered by CO_2, which affects $scCO_2$ plume migration and trapping. Field observations and laboratory core-scale CO_2-flood studies illustrate complexity of the porosity-permeability relationships, which can follow different scaling laws, depending on the location within a reservoir (Pawar and Guthrie, this volume).

Geochemical modeling studies indicate that re-equilibration of mineral systems and carbonation reactions may proceed for hundreds to thousands of years, which is mainly due to slow dissolution rates of most minerals encountered in CO_2 storage reservoirs and caprocks (Dai et al., this volume). Carbonate minerals, however, have relatively fast dissolution kinetics, compared to silicates, and carbonate dissolution and re-precipitation have been quantified in laboratory studies and linked to geomechanical alteration of reservoirs and caprocks (Vilarassa et al., this volume, and Ilgen et al., this volume). Geochemical modeling coupled to laboratory experiments have also highlighted an additional risk of CO_2 leakage, namely, if the $scCO_2$ plume reaches shallow groundwater, it may mobilize trace metals and compromise water quality (Dai et al., this volume).

Field, laboratory, and numerical experiments relevant to GCS have recognized that individual processes cannot be viewed independently. Strong coupling has been identified between hydrological, mechanical, thermal, and chemical processes in subsurface reservoirs and caprocks (Zhang and Wu, this volume, and Kim et al., this volume). Core-scale laboratory and field tests indicate that mechanical response to CO_2 injection is dictated by pore pressure changes, thermal, and chemical processes (Vilarassa et al., Rutqvist et al., this volume, Ilgen et al., this volume). The injected $scCO_2$ is typically cooler compared to the formation temperature, and a cooled region is usually observed near an injection well, resulting in an increased density and viscosity of CO_2 in this region (Ivanova et al., this volume). This rock cooling can also create thermal stress near the wellbore, which may push the system closer to shear and/or tensile failure conditions.

Fluid flow in fractured formations is another example of observed coupled phenomena. Recent scientific advances have identified a critical link between fluid flow through fractures, fracture deformation and seismic response (Pyrak-Nolte, this volume). Fracture stiffness can be used to describe both deformation and flow in fractures at multiple length scales.

Induced seismicity is a concern for large-scale CO_2 storage sites. Limited data exists on microseismicity in critically-stressed basements underlying CO_2 injection formations, and it is anticipated that with CO_2 storage reaching the required volumetric scales, induced seismicity may become an important process to consider (Vilarassa et al., this volume, and Rutqvist et al., this volume).

[[H1]] Future research needs for geological carbon storage

Based on the properties of saline storage systems, current challenges for safe and economical storage include, but not limited to: (i) sustaining large CO_2 injection rate and volume, capable of keeping up with the rates of anthropogenic CO_2 emissions; (ii) efficient use of available pore space to avoid a large subsurface footprint of CO_2 storage sites, and (iii) avoiding leakage of CO_2, ground deformation, and seismic events. To address these challenges, some major fundamental research needs are outlined below.

[[H2]] Fundamental science needs

- One of the fundamental science challenges is establishing porosity-permeability relationships with appropriate scaling laws, applicable to relevant time scales. Heterogeneity ranging in scale from individual mineral grains, to cm-, m-, and km-scales, multi-phase flow, and flow through matrix and fractures makes this a complex task.
- More data is needed for flow characterization in mixed-wet systems, and for assessing the hysteretic behavior of capillary pressure and relative permeability, and their effects on CO_2 mixing with brine. This data can be used to develop transport parameters describing mixing and migration of supercritical and dissolved CO_2 at storage sites.
- The need to understand leakage through fractures in heterogeneous shale caprocks necessitates extending the flow-stiffness relationship to non-elastic rocks and mixed mineralogy fracture-matrix systems.
- Development of new constitutive models for rock formations is necessary; for instance, caprocks may display visco-elastic or plastic behavior due to contributions from swelling clay minerals.
- Additional data is needed for identifying the microbial feedback to long-term carbon storage. Microbial populations that persist in environments with high CO_2 fugacity could

modify permeability of reservoir and caprock, mineral trapping capacity, and secondary minerals from carbonation reactions.

- More data needs to be acquired to predict the effect of CO_2 injection on crystalline basements, faulted and damaged rocks, and the potential for induced seismicity.
- Thermo-mechanical effects due to cooling-induced contraction near wellbores and its effect on caprock integrity are not fully understood and require further research.

[[H2]] Needs for improving existing tools

- Cross-well seismic surveys to map subsurface CO_2 flow have measurement resolution of 1-2 m, with the imaging area limited to the space between the two monitoring wells. Both 2D and 3D seismic surveys have spatial resolutions exceeding 10 m, while allowing CO_2 plume monitoring at the reservoir-scale. For predicting the CO_2 flow path, both effects of local heterogeneity and viscous fingering must be accounted for. The scale of heterogeneity ranges from mm, to cm, to m, to km. The scale of seismic surveys (meters) is typically much larger than the scale of viscous fingering of CO_2 (millimeters to centimeters) and local heterogeneity, and therefore both viscous fingering and flow deviation due to local heterogeneity cannot be directly probed in the field. Future research needs to improve measurement resolution to minimize uncertainties when measuring subsurface migration of CO_2.
- Further advancement in imaging stress and chemical constituents in brine and $scCO_2$ plume are necessary to understand stress state and distribution of CO_2 and brine in the subsurface.
- To decrease the uncertainties of assessing geochemical reactions in the subsurface, and to quantify the amount of CO_2 dissolved in formation brine, improved sampling approaches are necessary. Also, the development of sensing techniques that would allow quantifying the "contact area" between brine and $scCO_2$ during (i) the injection period, (ii) CO_2 plume development due to gravitational and capillary forces, and (iii) long-term convective flow would be beneficial.
- Advancement in existing mathematical, numerical, and statistical frameworks to extend geological parameters from core and boreholes to locations far from the boreholes are essential for safe GCS.

- Advances are needed in computational tools to simultaneously and seamlessly aid interpretation and integration of data collected across all length-scales.

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