

Perspective

Recent research advances in enhanced CO₂ mineralization and geologic CO₂ storage

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Abstract:

Enhanced CO₂ mineralization and geologic CO₂ storage have received increasing attention as two prominent approaches in combating climate change and fostering sustainable development of human society. This paper aims to explore three emerging areas of research within the realm of enhanced CO₂ mineralization and geologic CO₂ storage, including enhanced rock weathering, numerical modeling and validation of CO₂ storage accounting for the interplay of various trapping mechanisms, and the examination of how reservoir heterogeneity influences the migration of CO₂-brine multiphase flow. Discussions highlight the effectiveness of the spectrum induced polarization for monitoring changes in petrophysical and geochemical properties of rocks during enhanced rock weathering. Additionally, the multi-scale heterogeneity of geological formations needs to be carefully characterized, due to the fact that it plays a vital role in CO₂ migration. Further research is required to achieve accurate and reliable simulations of convective mixing for field-scale applications.

1. Introduction

CO₂ is widely recognized as the primary contributor to global warming (Soeder et al., 2021). The implementation of enhanced CO₂ mineralization and geologic CO₂ storage strategies is expected to play an important role in mitigating CO₂ emissions (Zhang et al., 2023). Within this dynamic field, a multitude of new research directions have emerged, including investigation of CO₂ mineralization through enhanced rock weathering, numerical modeling of CO₂ trapping mechanisms in geologic CO₂ storage, and exploration of the effects of reservoir heterogeneity on the migration of CO₂-brine multiphase flow. In subsequent sections, interesting research works related to the aforementioned research directions are discussed in detail.

2. Enhanced rock weathering for permanent CO₂ sequestration

Climate change presents a pervasive global challenge that necessitates the development and implementation of diverse strategies for carbon dioxide removal. One such strategy, known as enhanced rock weathering (ERW), represents a group of methods that accelerates naturally occurring rock weathering, enabling permanent trap of CO₂ (Kelemen et al., 2019; Beerling et al., 2020; Snæbjörnsdóttir et al., 2020). ERW can be implemented through natural and surficial deployments by spreading rock powders on diverse land types, as well as through ex-situ and in-situ engineered mineral sequestration. Silicate and carbonate rock powders on farmland and forests can facilitate the conversion of gaseous CO₂ into aqueous bicarbonate or carbonate ions (HCO₃⁻,

CO_3^{2-}). These ions can either be mineralized on land or be transported through rivers to the ocean, where they can persist as solutes for an extensive duration ($>10,000$ years) and finally precipitate as solid carbonate minerals (e.g., calcite, magnesite, dolomite, etc.) in the subsea. The ex-situ process, typically employed on olivine or alkaline industry wastes, is usually performed in high temperature and high pressure reactors. The in-situ process, which involves injecting CO_2 into the subsurface pore spaces of rocks like basalt and peridotite, has been effectively demonstrated in projects like Iceland's Carbfix Project (Snæbjörnsdóttir et al., 2020). Besides removal of CO_2 , ERW offers additional benefits such as enhanced agricultural productivity, soil remediation, and ocean alkalization, thereby enabling sustainable socio-economic advancement (Köhler et al., 2010; Beerling et al., 2020).

During the calcite precipitation, the microstructure of weathering rocks is altered, leading to highly heterogeneous and dynamic fluid-rock interactions, which significantly impacts the enhanced weathering process. Effective and robust monitoring of the ERW processes not only help quantify the amount of removed carbon achieved through enhanced weathering reactions, but it is imperative to mitigate risks such as induced seismicity and groundwater contamination. The mineralization processes within soil, water, and plant biomass are typically monitored through the analysis of collected samples, yielding accurate yet spatially limited information. Geophysical monitoring and imaging techniques have the potential of providing high spatiotemporal resolution information of the evolution of petrophysical properties during calcite precipitation. Within the geophysical methods, complex conductivity or spectral induced polarization (SIP) method has shown great sensitivity to pore fluid and mineral interactions (e.g., Binley and Kemna, 2020; Zhang et al., 2012). During the application of SIP, the polarization of charge carriers like calcite is associated with the polarization of the electrical double layer (EDL) surrounding the mineral particles under an external alternating current supply (Jougnot et al., 2010; Revil et al., 2014). Studies (Wu et al., 2010; Izumoto et al., 2020; Saneiyani et al., 2021) have shown the potential of SIP in characterizing the distribution of the induced calcite precipitation at laboratory scales.

The majority of ERW research centers around silicate rocks. However, enhancing the weathering of carbonate rocks may also have a substantial contribution to carbon dioxide removal, given much higher dissolution rates of carbonates than silicates. Over half of the atmospheric CO_2 absorbed by rock weathering each year is attributed to carbonate rocks (Gaillardet et al., 1999). In a recent carbonate rock ERW study, laboratory columns were utilized to emulate natural carbonate weathering processes, while geophysical responses were correlated with simulated geochemical reactions and solution geochemistry. Such geophysical investigation of ERW processes elucidates and distinguishes different stages of calcite formation from fine grains to larger aggregates. The combination of geophysical monitoring, geochemical analysis, and numerical modeling not only provides deeper insight into calcite precipitation, but also contributes to the broader understanding on ERW.

3. Numerical simulation accounting for the interplay of trapping mechanisms

The understanding of CO_2 trapping mechanisms within geological formations has been well-established, encompassing structural, residual, dissolution, and mineral trapping (Duan and Sun, 2003; Juanes et al., 2006; Szulczewski et al., 2012; Zhang et al., 2019). These trapping mechanisms operate across varying time scales. Structural and dissolution trapping come into effect immediately after CO_2 injection. Dissolution trapping continues to work in the post-injection stage due to the establishment of gravity-induced convective mixing. Mineral trapping is achieved through geochemical reactions, with the rate of these reactions determined by the composition of the host rock. To comprehensively evaluate the interactions among these trapping mechanisms, the construction of efficient and predictive numerical models is imperative.

Incorporation of the aforementioned trapping mechanisms into the numerical model necessitates the integration of hydraulic, thermal, and chemical processes, which pose considerable challenges to large-scale numerical simulations (Gasda et al., 2011; Wang et al., 2022). To improve the computational efficiency, the equations governing different processes can be decoupled and solved in a sequential manner via non-iterative operator splitting method (Postma et al., 2022). Moreover, in a fully coupled system, evaluation of the physical properties and the derivatives regarding the unknowns can be expedited using a parameterized space (Lyu et al., 2023). In addition to the physics-based numerical simulations, the rise of machine learning has sparked enthusiasm for surrogate models. These models can predict the spatiotemporal evolution of the state variables without the need to run numerical simulations (Wen et al., 2021; Yan et al., 2022; Zhao et al., 2023). However, it is worth noting that the developed models have been typically trained based on simplified physical models (e.g., immiscible two-phase system). Also, it is very important to gather data of physical observations that are open to the public for numerical model validation purpose. At the current stage, ensuring the accuracy of numerical models remains challenging due to the limited availability of comprehensive physical observations. An effective approach to address this challenge is to conduct laboratory-scale CO_2 injection benchmark studies with laboratory-designed heterogeneous permeable medium to evaluate the performance of numerical models (Flemisch et al., 2023). Research groups can utilize their respective custom-developed or open-source numerical simulators and validate their numerical simulation results based on the comprehensive characterization of the heterogeneous permeable medium employed in the benchmark studies.

The key take-away messages can be summarized as follows: (i) The efficacy of the classic continuum model, formulated upon Darcy's law with a multiphase extension, is validated in accurately explaining pertinent physical observations. The simulation results effectively capture the characteristics of both structural trapping and residual trapping, which are influenced by the migration of supercritical CO_2 . (ii) The dissolution trapping, facilitated by convective mixing driven by gravity, is found to enhance the dissolution process of

CO₂. However, predictions concerning the dynamics of dissolved CO₂ exhibit notable disparities among the participating research groups. This variability can be attributed to several physical and modeling parameters, including CO₂ solubility limits, phase densities, and grid resolutions. Consequently, further investigations are warranted to refine the simulation of convective mixing, particularly when applied to scenarios of a larger scale. (iii) Most benchmark laboratory studies are conducted in a controlled system characterized by well-defined heterogeneity. In contrast, real-world subsurface formations are typically characterized through seismic surveys with significantly coarser resolutions. Given that spatial heterogeneity notably influences plume migration (Jackson and Krevor, 2020), field-scale simulations may experience considerable uncertainties in reservoir properties stemming from the relatively coarse geological characterization.

4. Effects of reservoir heterogeneity on CO₂-brine flow properties

Deep saline aquifers have been regarded as ideal candidates for gigaton-scale CO₂ storage due to their wide geographical spread. Investigation on controlled CO₂ storage in deep saline aquifers is a highly interdisciplinary challenge at the intersection of geology, geochemistry, petrophysics, and engineering. Given the prevalence of reservoir heterogeneity such as the presence of cross-bedding layers, it becomes imperative to examine the influence of reservoir heterogeneity on the multiphase flow properties of CO₂-brine systems, thus paving the way for high-accuracy field-scale simulations.

Given the diverse nature of CO₂ injectivity, flow patterns, trapping mechanisms, migration, and storage capacity across different rock types, it is essential to classify reservoir rocks for accurate geological modeling and reservoir simulation in the context of geological carbon storage (Padhi et al., 2014; Zheng et al., 2021). Rock typing entails classifying specific units of reservoir rocks by considering lithofacies, petrophysical properties of the rock and its pores, interactions between rock and fluid, and/or historical production and injection data (Rebelle et al., 2014). Presently, rock-typing studies often depend on combining two or more clustering methods (Farshi et al., 2019). Unlike oil and gas reservoirs, which have extensive data collected during exploration and development, there is limited data for deep saline aquifers in the context of geological carbon storage. This scarcity underscores the need for reservoir characterization and flow unit classification specifically tailored to saline aquifers.

Following reservoir characterization, studying the impact of rock heterogeneity (i.e., variations in physical and petrophysical properties of rock) on fluid distribution at the sub-core scale enhances our fundamental knowledge of rock relative permeability. It also plays a crucial role in advancing the development of multiphase flow simulation techniques at the field scale. Hence, comprehending how sub-core heterogeneity affects CO₂ flow properties holds paramount importance in the realm of CO₂ geological storage. Currently, the majority of existing studies have concentrated solely on exploring the impact of a singular type of sub-core heterogeneity on CO₂

flow properties, and further research is needed to explore the impact of two or more types of heterogeneity (i.e., the combination of pore distribution and mineral distribution heterogeneities) on CO₂ flow. Also, it is challenging to correlate sub-core heterogeneity with reservoir-scale heterogeneity. Sub-core scale heterogeneities often do not fully represent the entire range of reservoir characteristics, even within a single target layer, due to a large gap between sub-core scale and reservoir-scale (Perrin et al., 2010). Consequently, it is reasonable to anticipate variability in CO₂ sweep efficiency and distribution across different locations within deep saline aquifers during CO₂ injection.

Additionally, when injecting CO₂ into saline aquifers containing reactive minerals, varied interactions between CO₂ and reactive minerals are anticipated within the host rocks. To elaborate, a diverse pore size distribution often indicates widespread heterogeneous dissolution and precipitation within a core sample (Wang et al., 2020). Mineral dissolution tends to happen predominantly in channels or fractures, where fluid flow encounters less resistance (Mohamed et al., 2018). Furthermore, studies have confirmed that mineral dissolution and precipitation within a core sample depend on the specific position during fluid flow-through tests (Huq et al., 2015; Kou et al., 2023). As a result, changes in pore architecture often lead to time-dependent responses in CO₂-brine multiphase flow. After identifying sub-core scale and core-scale properties related to multiphase flow in saline aquifers, these findings are recommended to be integrated into field-scale studies to assess the forthcoming CO₂ injectivity and trapping mechanisms.

5. Summary

As the global energy transition gains momentum and the urgency to effectively remove CO₂ becomes unprecedented, forthcoming research in enhanced CO₂ mineralization and geological CO₂ storage will be paramount in establishing a sufficiently large carbon sink for CO₂, thereby achieving net-zero CO₂ emissions and addressing the challenges of climate change. Future research in this field will focus on improving the efficiency and reliability of CO₂ mineralization, as well as the predictive capability of reservoir simulators for field-scale CO₂ storage operations. Developing better monitoring techniques and ensuring the long-term integrity of storage sites will be key areas of investigation as well. Additionally, efforts will be made to explore new storage options, such as basalts and subsea storage, to increase the total storage capacity.

Currently, research endeavors pertaining to CO₂ mineralization are witnessing a surge, with particular emphasis on swiftly converting CO₂ into stable minerals. Improving the efficiency and reducing the cost of the CO₂ conversion processes will be crucial to make them economically viable, and enhanced rock weathering is a hot research spot in this field. In addition, studies on the numerical modeling that incorporates the interaction between different trapping mechanisms and the impact of multi-scale reservoir heterogeneity are crucial to ensure the efficiency and safety of geologic CO₂ storage.

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Conflict of interest

The authors declare no competing interest.

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