Lessons Learned from Batch Experiments for CO_2 Mineralization in Basalts: Implications for Reactive Transport Modelling

Introduction

Long-term CO2 storage in geological formations is critical for climate change mitigation, as it enables the safe and permanent removal of carbon dioxide from the atmosphere. Among the various options available, the main locations for CO2 storage include depleted oil and gas reservoirs, deep saline aquifers, unmineable coal seams, and reactive volcanic rocks such as basalts. CO2 mineralization in mafic/ultramafic rocks like basalts offers potential for permanent storage by forming stable carbonates. This involves CO_2 reaction with silicates to precipitate solid carbonates. Evaluating this potential in basaltic reservoirs is necessary before injection. Evaluation relies on geological characterization, laboratory experiments, and modeling. This work describes the current laboratory methodology for assessing CO2 mineralization (carbonate precipitation) in subsurface basalts. It focuses on the lessons learned, insights, and limitations from conventional laboratory experiments for CO_2 mineralization in basalts. This provides valuable input for Reactive Transport Modelling, improving its accuracy and reliability by grounding simulations in observed geochemical behavior.

Methodology

The conventional methodology involves batch experiments in autoclaves using basalt samples, synthetic brine, and CO_2 under controlled P, T, and time (Cao et al., 2024). The process includes field sampling, preparing samples as plugs and crushed material, and characterizing rock and brine composition before and after experiments. Characterization uses techniques like routine core analysis porosity, permeability, ICP-MS, XRD, and SEM-EDS (Khan et al., 2024). Significant challenges have been identified in the methodology including sample representativeness (outcrop versus wellbore cores), the use of synthetic brines not in equilibrium with the host rock, experimental limitations related to time of the experiment, cost, and limited autoclave capacity, as well as the restricted range of geochemical and physical scenarios typically investigated. This methodology provides a static picture, lacking insight into the process evolution. Addressing these limitations is vital for more reliable and comprehensive CO_2 storage assessments.

Results

The experimental methodology begins with the preparation of basaltic rock samples, typically collected from field outcrops rather than actual wellbore cores. This introduces several limitations, including the potential for meteoric alteration, challenges in obtaining sufficient quantities, and the inability to capture the full lithological diversity of the reservoir. Additionally, field-based classifications often lack the mineralogical resolution needed for detailed analysis. Samples are processed either as crushed material or cylindrical plugs employing sequential analytical phases that gradually modify the pressure-temperature (P-T) conditions throughout the process. For the fluid phase, a synthetic brine (commonly NaCl-based) is used; because this solution is not in equilibrium with the rock, it tends to be highly reactive, and does not realistically simulate in-situ formation water conditions. During the experiment, supercritical CO_2 is introduced into the autoclave, where it interacts with the brine and rock under controlled conditions. Notably, the rock-to-brine ratio in experiments is typically much lower than that in actual reservoir environments, which may influence the observed geochemical dynamics.

Characterization of the samples is conducted both before and after the experimental procedure to assess changes induced by CO_2-brine-rock interactions. This comprehensive analysis includes physical, chemical, and geological properties. Physically, parameters such as porosity, permeability, grain density, and specific surface area are measured to evaluate structural alterations. Chemically, techniques like CHNS/O elemental analysis and Inductively Coupled Plasma Mass Spectrometry (ICP-MS) are

employed to detect changes in elemental composition and trace elements. Geologically, a suite of methods including X-ray Diffraction (XRD), X-ray Fluorescence (XRF), Scanning Electron Microscopy (SEM), SEM coupled with Energy Dispersive Spectroscopy (SEM-EDS), thin section petrography, and cathodoluminescence are used to investigate mineralogical transformations and microstructural alterations. This multi-faceted approach provides a detailed snapshot of the system's evolution, although it remains limited to initial and final states compared with Reactive Transport Modelling (Chen et al., 2024).

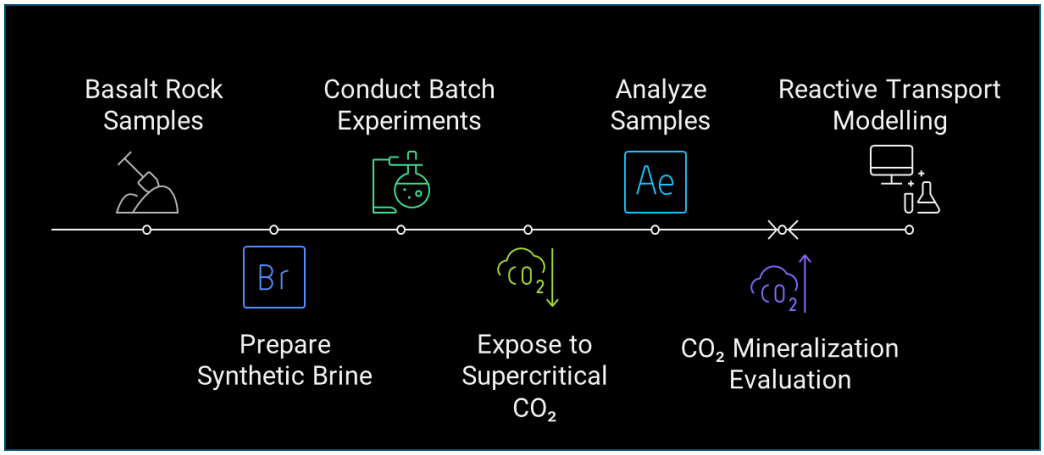

Figure 1 *Cartoon illustrating the laboratory methodology steps, compared to Reactive Transport Modelling used to assess CO_2 mineralization (carbonate precipitation) in basalts.*

Conclusions

Evaluating the potential for CO_2 mineralization is a critical step in assessing the viability of geological storage. The batch autoclave method has become a standard experimental approach for simulating subsurface conditions and assessing carbonate precipitation. This technique relies on detailed pre- and post-experiment characterization to understand the geochemical interactions involved. However, several limitations affect the reliability and representativeness of the results. The use of outcrop samples, which often differ significantly from actual reservoir rocks in composition and alteration state, limits the applicability of findings. Similarly, the synthetic brine used in experiments is not in equilibrium with the rock and may follow reaction pathways that differ from those in natural settings due to its high reactivity and pH. The experimental setup also suffers from scale and duration constraints: the rock-to-brine ratio is lower than in situ conditions, and the relatively short experiment times, typically from weeks to months, may not capture long-term processes. Additionally, the limited number of autoclaves and high costs restrict the number of scenarios that can be tested in parallel. As a result, the method provides only a static snapshot of the system, lacking insight into the kinetics and temporal evolution of reactions. A key observation is that increasing CO_2 concentrations beyond a certain threshold tends to enhance mineral dissolution rather than promote carbonate precipitation. Addressing these limitations through more representative sampling, improved brine formulations, extended experiment durations, and real-time monitoring would significantly improve the accuracy of field-scale assessments. Nevertheless, the insights and constraints derived from these experiments are invaluable for Reactive Transport Modelling, offering essential data to calibrate and validate simulations, and ultimately enhancing their predictive power in real-world applications.

References

Cao, X., Li, Q., Xu, L., Tan, Y., 2024. A review of in situ carbon mineralization in basalt. Journal of Rock Mechanics and Geotechnical Engineering, 16, 1467-1485. https://doi.org/10.1016/j.jrmge.2023.11.010

Chen, Y., Clennell, B., Zhang, J., Tang, M., Ahmed, S., 2024. Reactive transport modelling of in-situ CO2 mineralization in basalt formations. Capillarity, 13 (2), 37-46.

Khan, M.N., Siddiqui, S., Thakur, G.C., 2024. Recent Advances in Geochemical and Mineralogical Studies on CO2–Brine–Rock Interaction for CO2 Sequestration: Laboratory and Simulation Studies. Energies, 17, 3346. https://doi.org/10.3390/en17133346