Salt Precipitation during Geological CO_2 Storage: Effect of Access to Continuous Brine Source

Mohammad Masoudi[1, *], Mohammad Nooraiepour[1] and Helge Hellevang[1]

[1] Department of Geosciences, University of Oslo, P.O. Box 1047 Blindern, 0316 Oslo, Norway
* Correspondence: mohammad.masoudi@geo.uio.no

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[1] Department of Geosciences, University of Oslo, P.O. Box 1047 Blindern, 0316 Oslo, Norway

[*] mohammad.masoudi@geo.uio.no

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ABSTRACT

During CO_2 storage, the near-wellbore environment experiences significant chemical, physical, thermal, and mechanical perturbations. Therefore, it is crucial to characterize the coupled thermo-hydro-mechanical-chemical (THMC) processes in this region through representative experimental tests and numerical simulations to maximize the safety and cost-efficiency of CO_2 storage. One common shortcoming of current experimental methods is failing to facilitate access and replicate the in-situ brine sources within geological repositories, particularly saline aquifers. This failure can significantly alter the dynamics and dimensionality of mineral/crystal growth in porous reservoir rocks due to the availability and continuity of solute. To address this issue, we designed surface mineral precipitation tests and large-scale sandbox experiments to investigate salt precipitation and growth under two different scales and regimes in porous geometries. The laboratory results indicated massive salt accumulation close to the injection port and underlined the effect of solute availability and continuity on intensifying severity of salt accumulation. The observed continued growth highlights the importance of designing representative laboratory experiments in exploring fluid-rock interactions during carbon sequestration in the subsurface geological media.

1. INTRODUCTION

Global warming is causing climate change that could lead to irreversible and devastating consequences for life on the planet. The primary anthropogenic greenhouse gas responsible for global warming is carbon dioxide (CO_2), primarily emitted through fossil fuel combustion at industrial point sources (Pachauri et al., 2014). Carbon Capture and Storage (CCS) technology has emerged as a potential solution to mitigate the effects of climate change by capturing CO_2 from point sources and storing it underground. However, to achieve significant progress, CCS must be implemented on a large scale (giga-tons scale). A detailed assessment of injectivity performance and permeability changes in the near-well region is critical to achieving such substantial injection volumes. The near-well environment, the connection between the CO_2 injection well and storage reservoir, requires maintaining high permeability and injectivity to ensure operational injection rates. The clogged pore space in the near-well region may cause pressure build-up, formation channeling or fracturing, and even enhancement of capillary backflow, leading to increased salt accumulation (Grude et al., 2014; Hansen et al., 2013; Miri and Hellevang, 2016; Ringrose et al., 2009; Torabi et al., 2015). The pore-clogging can occur because of a variety of thermochemical mechanisms (such as mineral deposition, asphaltene/wax precipitation, or hydrate formation) as well as mechanical processes (including particle-grain migration, rock compaction, and shrinkage-swelling (Torsæter and Cerasi, 2018). Among all of them, salt precipitation and growth have been identified to be the most severe issue. These issues threaten the integrity of CO_2 injection wells and flow assurance of CCS projects, making it crucial to maintain high permeability in the near-well region through constant evaluation and monitoring.

Supercritical CO_2 injection into saline aquifers leads to the evaporation of the formation water into the CO_2 stream and the concentration build-up of dissolved salt in the aqueous phase. Excess salt can precipitate out of the aqueous phase when it exceeds its solubility limit, forming salt crystals in the porous or fractured media. This process can affect the properties of both reservoir and top seal sequences during geological CO_2 storage. Several field (Baumann et al., 2014; Grude et al., 2014; Talman et al., 2020), experimental (Berntsen et al., 2019; Kim et al., 2013; Miri et al., 2015; Nooraiepour et al., 2018a,b), and numerical studies (Masoudi, 2021; Masoudi et al., 2021; Parvin et al., 2020; Zeidouni et al., 2009) have provided evidence for salt precipitation during subsurface CO_2 injection.

Salt precipitation during CO_2 storage is a complex phenomenon because of the interplay of different physio-chemical interactions. Salt can form in different structures depending on the transfer, flow, thermodynamics, and kinetics conditions. Microfluidic experiments showed salt crystals formed in two different structures (Kim et al., 2013; Miri et al., 2015; Nooraiepour et al., 2018a,b): (1) large crystals which form in the bulk of the aqueous phase, and (2) aggregates of micrometer size salt crystals at the CO_2-liquid interface towards the CO_2-rich phase. When a stable salt nucleus is in contact with the sufficient supersaturated solution and has enough time to grow, single, large crystals form (Nooraiepour et al., 2021b). On the other hand, when a salt nucleus forms near the interface in the CO_2 stream (in the brine film along the pore channels), it is not covered with enough supersaturated fluid. As a result, the nucleus consumes all the excess dissolved salt for crystal growth until the solution reaches its solubility limit. The hydrophilic nature of the newly formed crystals results in capillary suction of the fresh brine to the CO_2 stream and connecting residual brine to the drying front. This leads to more evaporation and therefore, new nucleation events and the formation of new salt crystals on the previous small crystals. These processes are occurred repeatedly and lead to the formation of salt aggregates. The aggregates form a porous structure with massive capillarity. This phenomenon is called "salt self-enhancing", and can lead to massive salt accumulation.

Salt precipitation during geological CO_2 storage is a complex phenomenon involving various physio-chemical interactions. Salt formation can occur in different structures depending on factors such as transfer, flow, thermodynamics, and kinetics conditions. According to microfluidic experiments, salt crystals can form in two different structures: (1) large crystals that form in the bulk of the aqueous phase and (2) aggregates of micrometer-sized salt crystals at the CO_2-liquid interface towards the CO_2-rich phase (Kim et al., 2013; Miri et al., 2015; Nooraiepour et al., 2018a, b). Single, large crystals form when a stable nucleus is in contact with a supersaturated solution and has enough time to grow (Nooraiepour et al., 2021b). In contrast, when a salt nucleus forms near the interface in the CO_2 stream, it is not covered with enough supersaturated fluid. As a result, the nucleus consumes all the excess dissolved salt for crystal growth until the solution reaches its solubility limit. In addition, the hydrophilic nature of the newly formed crystals results in capillary suction of the brine saturation in the pore space towards the CO_2 stream and transporting residual brine and available solute to the drying/evaporation front. The continued transport of solute supports more evaporation, new nucleation events, and the formation of new salt crystals on the previous small crystals. These processes are repeated over and over again and contribute to the growth of salt aggregates. This leads to the formation of aggregates of salt crystals, which form a porous structure with massive capillarity. This phenomenon, called "salt self-enhancing", can lead to massive salt accumulation.

Several experimental studies have observed salt self-enhancing growth (He et al., 2019; Ho and Tsai, 2020; Kim et al., 2013; Miri et al., 2015; Nooraiepour et al., 2018a). Additionally, a field study on the Aquistore CO_2 storage site, which is a part of SaskPower's Boundary Dam CCS demonstration project, has revealed the formation of salt crystals inside the injection well. They have stated that salt self-enhancing growth might be the reason behind the water backflow and salt formation inside the injection well (Talman et al., 2020). Despite its significance, most existing numerical models ignore the salt self-enhancing growth phenomenon. To address this issue, Parvin et al. (2020) proposed a modification to the capillary pressure model, enabling the simulation of salt self-enhancing growth at a continuum scale for the first time. In a related study, Masoudi et al. (2021) developed a new numerical approach to simulate the formation of salt aggregates resulting from self-enhancing growth at the pore scale, using a novel probabilistic nucleation model (Fazeli et al., 2020; Nooraiepour et al., 2021a) and specific numerical implementations of nucleation and growth processes at sub-grid resolution.

Validation of numerical studies on CO_2 induced salt self-enhancing growth is hampered by the lack of experimental data on the context of permeability reduction caused by the formation of salt aggregates in the CO_2-rich phase. The absence of such experimental data makes it difficult to assess the accuracy of numerical models that aim to simulate salt self-enhancing growth. One of the main reasons for the absence of experimental data is neglecting the access to in-situ brine sources within the actual storage reservoirs, which can dramatically reshape the dynamics and dimensionality of mineral/crystal growth in porous reservoir rocks owing to the availability and continuity of solute. This crucial aspect is often overlooked in experimental works related to salt deposition during CO_2 sequestration, including core flood experiments. In core flood experiments, only one pore volume of brine is available, which is usually pushed out of the porous media relatively quickly. As a result, the effect of several forces and parameters are overlooked, such as gravitational forces that are the main force for the advancement of the CO_2 plume in the reservoir and the capillary forces due to the continuity of the water film.

We conducted a series of experiments at different scales to address the challenges related to the lack of experimental data on brine evaporation and salt precipitation and gain new insights beyond the current state of understanding. These experiments included investigating surface mineral precipitation on reservoir rock and studying salt precipitation resulting from the evaporation of brine in a large sandbox. The experiments were designed to provide new insights into the dynamics of brine evaporation and salt precipitation and growth, and to assess the accuracy of numerical models that aim to simulate salt self-enhancing growth.

2. MATERIALS AND METHODS

To investigate salt precipitation and growth in porous geometries, we designed two experiments: surface mineral precipitation tests and large-scale sandbox experiments. In the surface mineral precipitation test, we partially submerged a homogenous porous reservoir sandstone in brine and monitored it in real-time using a high-resolution Dino-Lite digital microscope and a full-frame Nikon Z 7II mirrorless camera. Scanning electron microscopy (SEM) was later used to analyze the three-dimensional geometry and porous structure of the resulting salt bodies, using backscattered (BSE) and secondary electrons (SE) imaging. We also performed energy-dispersive x-ray spectroscopy (EDS) for chemical analyses and element mapping with a spatial resolution of 1 µm. To investigate possible mechanisms behind large-scale salt precipitation observed in the field, we conducted a sandbox experiment. We filled a Plexiglas box (60×80×2 cm) with quartz-rich beach sand saturated with brine solution. To be specific, 100 g/l (1.71 molar) sodium chloride (NaCl) dissolved in deionized water (DI-water) [Milli-Q water]. High-pressure air was injected through the injection port placed in the middle of the box to mimic the evaporation process in the aquifer. Both experiments were performed under ambient conditions.

3. RESULTS AND DISCUSSION

The results of surface mineral precipitation tests are presented in Figure 1. The experiment started with a dry sandstone core that was then saturated with the aqueous phase, given that the sandstone matrix is a hydrophilic rock (i.e., initially water-wet in Fig. 1a-b). Soon after, salt crystallites appeared on the surface of the rock and near the rock-water interface (Fig. 1c-d). These freshly precipitated crystallites were highly water-wet, forming strong capillary forces that transported the solution through water films (Fig. 1e-h). This led to increased evaporation and further crystal formation. These results demonstrate how reservoir rock in saline aquifers with access to an infinite and continued solute source may form a conductive water film flow across water-wet substrate surfaces towards the evaporation front. However, this continuous and plentiful brine support cannot be precisely reproduced in most flow experiments, which limits their ability to capture the potential issues due to injectivity impairment.

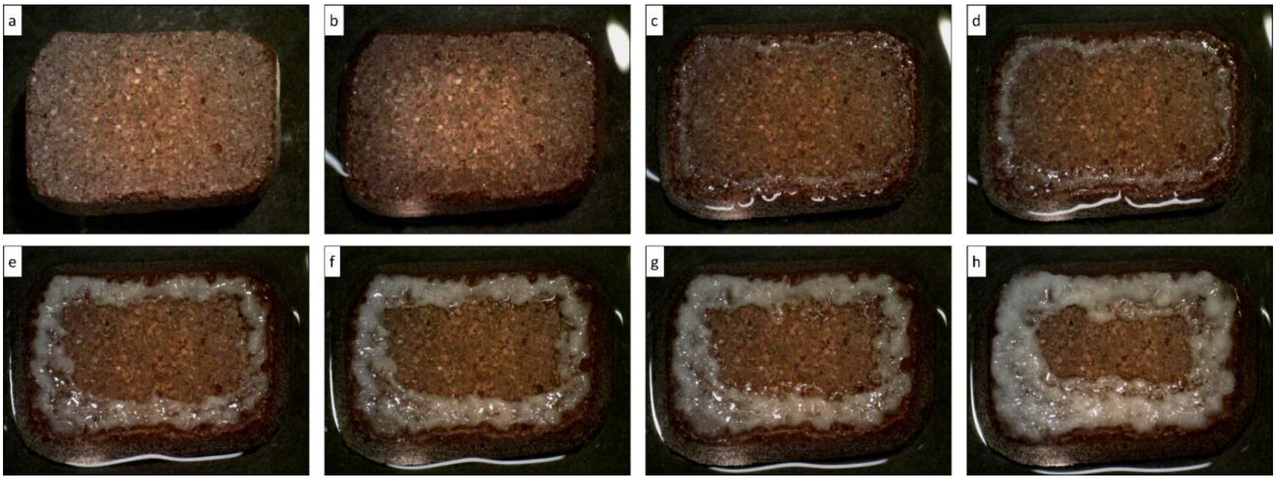

Figure 1: Dynamics of (a-b) saturation and imbibition of brine solution, (c-d) precipitation and (e-h) growth of halite crystal on the surface of a porous reservoir rock. Imbibing brine toward the salt bodies and transporting water films carrying solute toward the evaporation sites are evident in the bottom row.

Figure 1 illustrates how solute transport affects the salt precipitation front. The amount of salt accumulation depends on the evaporation rate, solute accessibility, and the characteristic length scale of capillary-driven backflow of brine and suction towards the evaporation front. Our previous hypothesis suggested that capillary forces arise from salt's surface characteristics and salt bodies' porous structure. Preliminary results of SEM structural identifications indicate the presence of both porous (ranging from micro- to nano-meter) and non-porous (smaller than nano-meter) CO_2-induced salt accumulations within the porous media. By elucidating these porous structures, we can estimate the potential for capillary forces imposed by the growing body of salts within reservoir rocks.

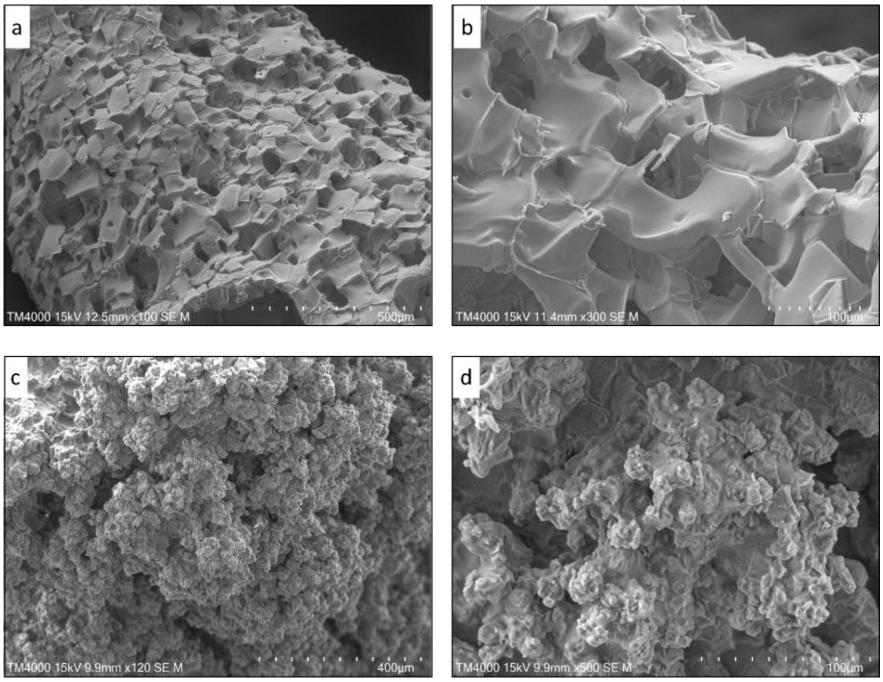

Figure 2: Scanning electron microscopy of porous and non-porous CO_2-induced salt precipitation within porous media. Halite crystal accumulation on primary and secondary substrates where accumulations constructed (Top) porous and (Bottom) non-porous growth structures. Subplots in b and d show enlarged sections for better representation.

To investigate the interplay between gravitational, capillary, and viscous forces in a more extensive system, we conducted the sandbox experiment for 50 days. Gas injection into the sandbox displaced water due to a pressure gradient. The capillary forces act to resist the infiltration of the gas into the porous media. However, the viscous forces overcome the capillary forces and enable the gas (non-wetting phase) to displace the water (the wetting phase) and spread throughout the pore spaces. As the gas infiltrated the porous media, gravitational forces caused gas bubbles to rise upward in the sandbox. Although no displacement front was observed, the gas phase exited the box as discontinuous gas bubbles. Figure 3 illustrates salt accumulation in the sandbox over time, with most accumulation occurring near the injection port, indicating a significant issue of salt accumulation that could impede the injectivity of CO_2 injection wells.

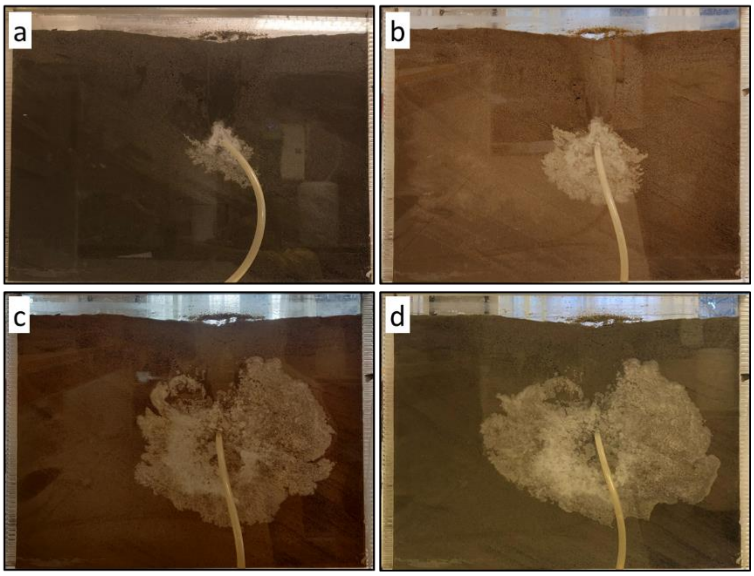

Figure 3: Salt precipitation magnitude and distribution in the sandbox apparatus after (a) 19, (b) 25, (c) 34, and (d) 48 days. The initial salt accumulation appeared next to the injection port, and then it started to grow and spread.

Water saturation is indicated by sand tone/color changes, as demonstrated in Figures 3-5. The dried areas in the sandbox varied, revealing the water displacement in the box. The area near the injection port remained moist throughout the experiment. Salt accumulation appeared next to the injection port after 9 days (Fig. 4a), but the area remained moist. Salt formation increases capillary forces, resulting in a backflow that transports brine to the evaporation front, intensifying salt precipitation. Crystal growth near the injection port advanced significantly afterward as shown in Figures 4 b-c. The area close to the newly formed salt crystals was always moist. The biggest and thickest salt accumulations increased in size by attracting more brine.

Figures 3b and 4d are taken at the same temporal stage from the front and back of the sandbox, respectively, showing the impact of capillary forces on brine saturation, with darker sands in Figure 4d near the precipitated salt crystals. Images of the front and back of the sandbox were captured to visualize the extent of salt accumulation at the end of the test, as shown in Figure 5. Salt accumulation in such large quantities results from the system's access to a large amount of brine, which is not observed, documented, or reported in the literature on core flooding laboratory studies. Figure 5b shows that water exists near the injection port, indicating the presence of a continuous water film and strong capillary forces causing the capillary backflow of water towards the deposited salt crystals.

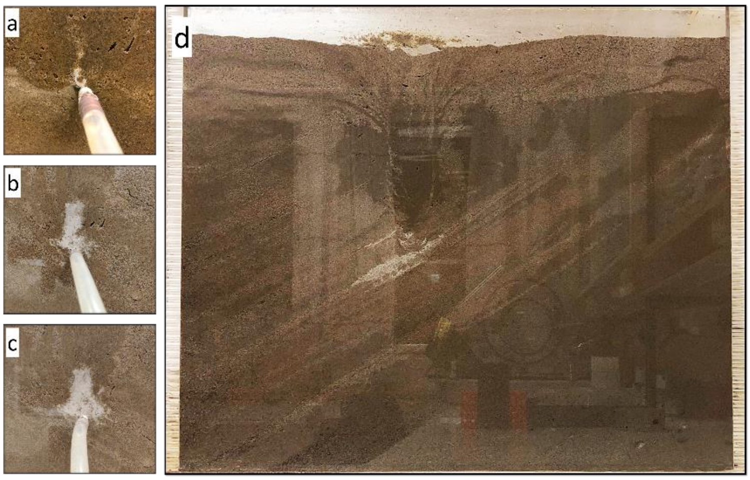

Figure 4: (a) Onset of salt precipitation next to the injection port after 9 days. (b-c) Advancement of salt formation close to the injection port after 12 and 13 days. (d) Back view of the sandbox after 25 days. The changes in sand color show the movement of water in the sandbox.

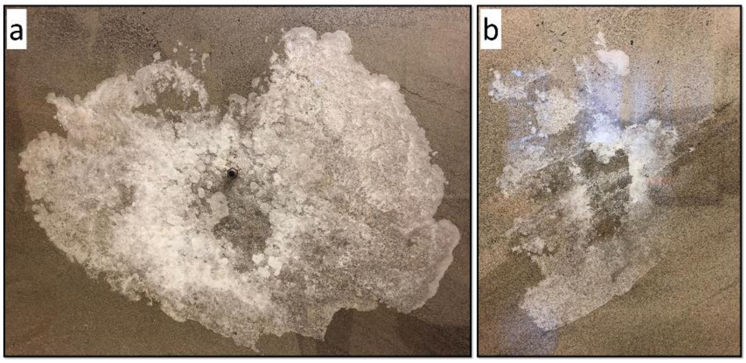

Figure 5: The final stage of the experiment after 50 days. (a) The front side: massive salt accumulation. (b) The backside: the capillary forces suck the water toward the strongly water-wet salt crystals.

4. CONCLUSION

This paper addresses the issue of salt precipitation during subsurface CO_2 storage, which can significantly affect the injectivity and safety of CCS operations. We conducted experiments at various scales to investigate the interplay of solute availability and continuity on salt precipitation and growth. Our results demonstrate the importance of access to infinite solutes in saline aquifers for delineating the manganite, distribution and structure of salt accumulations. Additionally, our findings highlight the complexity of the processes involved in salt precipitation induced by CO_2 injection, some of which are not yet fully characterized.

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