

A microfluidic study into salt precipitation in saline aquifers induced by continuous CO₂ injection

Introduction

Carbon dioxide sequestration in saline aquifers is one of the most promising methods to mitigate CO₂ emission due to the potential capacity of storage volume. However, salt precipitation (particularly halite) may occur during the injection of CO₂ (both in the gas phase and supercritical phase) due to evaporation. Such precipitation may block pores reducing the porosity and permeability of reservoir formations, which must be taken into account when evaluating saline aquifers for storage (Bacci et al., 2011; Miri & Hellevang, 2016). The field evidence in the Ketzin gas storage reservoir (near Potsdam, German) indicates a one-meter interval of salt accumulation during CO₂ injection, building up extra injection pressure (Baumann et al., 2014).

Core-flooding experiments are normally performed with CO₂ (gas or supercritical phases) and brine under different pressure and temperature conditions. Such conventional methods with CT scans can offer a good assessment of the petrophysical properties of rocks and quantitatively visualise salt precipitation distribution (Chavez Panduro et al., 2020), but observations and insights into salt precipitation at the micro/nano-scale are difficult. Microfluidics can provide extremely high-quality images of single crystals under a microscope. Limited microfluidic experiments were performed to analyse salt precipitation under mimicking the CCS process (Ho & Tsai, 2020; Kim et al., 2013; Nooraiepour et al., 2018). Two types of crystal patterns, bulk crystal, and polycrystalline aggregate, are observed at room temperature (Ho & Tsai, 2020; Kim et al., 2013)

Previous studies suggested that the salinity of the aquifer, the gas flow rate, and capillary pressure play the most influential roles with respect to salt precipitation (Miri & Hellevang, 2016). A lack of study systematically demonstrates how temperature affects salt formation. Increasing temperature causes a noticeable increase in water solubility in the CO₂ phase, resulting in a quicker evaporation rate. In contrast, high temperature will normally lead to a higher solubility of salt, which needs a higher concentration to precipitate. In addition, the temperature will also affect the diffusion rate of substrates and the surface tension of water to gas, leading to complex variations of many parameters.

The motivation of this study is to investigate the temperature effect on salt crystallization and distribution in porous media during dry CO₂ (gas phase) injection into saline aquifers. Potential hypotheses include,

- 1) a higher evaporation rate at elevated temperatures may lead to a faster salt precipitation rate and more small polycrystalline aggregates rather than large single crystals.
- 2) salt precipitation may mainly distribute in the locations, where the residual water stays during gas injection and the front interface between water and the gas.

Valuable insights into the mechanisms of CO₂ injection-induced salt precipitation in saline aquifers may help assess preferable temperatures (shallow or deeper formations) for storage sites containing similar fluid salinity to mitigate the problem of precipitation.

Since this is an ongoing study, we only present some preliminary results on CO₂ injection at room temperature and discuss potential effects and mechanisms. Particularly, we use a microfluidic chip containing real-rock structures, which is more reliable to represent the fluid flow in real rocks.

Sample and experimental methods

- Sample

A microfluidic chip (i.e., Enhanced oil recovery chips from Micronit) mimicking the porous rock structures is used for the experiments, as shown in Figure 1. The chip contains an inlet and an outlet hole for fluid flow, where a rock-structured pattern with a dimension of 10 mm x 20 mm is fabricated

in between. The thickness of the flow pattern is only 0.02 mm. The initial gas permeability of the chip was measured with CO₂ under constant flow conditions, showing a significantly high value of about 2215 mD.

- Fluid

250 g/L NaCl solution was prepared for the experiments. Such salinity is similar to the formation fluid in Ketzin CO₂ storage reservoirs (Na-Cl 220g/L) (Baumann et al., 2014) and in a geothermal reservoir in Groß Schönebeck, Northern Berlin (Na-Ca-Cl type fluids containing 265 g/L) (Milsch et al., 2013), which may represent the real saline fluid in the subsurface. The solution was degassed by vacuum and stored in a Duran glass bottle.

- Experimental setup

The experimental setup is illustrated in Figure 1, where the microfluidic chip is placed in a chip holder (incubator) that can be heated up to 150 °C. The whole assembly is mounted under an inverted microscope. The chip is connected to two syringe pumps, including a fluid pump for the initial saturation and drainage of the chip during gas flow, and a gas pump connected to a CO₂ compressed bottle to ensure sufficient CO₂ volume on the upstream gas flow side. In addition, a vacuum pump is connected inline for the initial saturation of the chip and removing the air in the capillaries before injecting CO₂.

- Experimental procedures

The experimental procedure can be summarized and listed below,

- Fluid preparation and degassing
- Saturation of the microfluidic chip
- The vacuum of the whole flow-through system
- System pressurization to about 2.5 bar using pure CO₂
- Drainage by injecting CO₂ gas with a constant flow rate of about 1 ml/min from the gas syringe pump to the fluid syringe pump (Figure 1), where the drainage is mainly controlled by fluid withdrawal with the fluid pump.
- Time-lapse imaging, where representative locations are imaged with a time interval of 30 s.

The same microfluidic chip will be reused for all experiments to ensure identical flow patterns. Deionized water will be injected into the chip continuously to fully dissolve and remove the salt precipitation after each experiment.

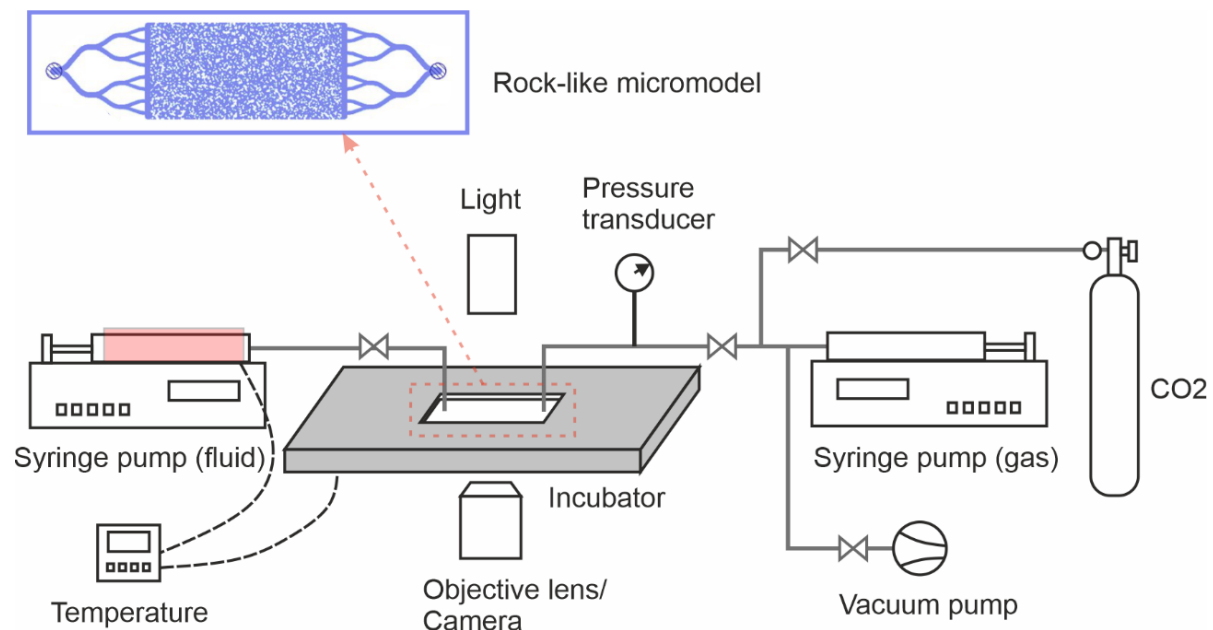


Figure 1 Schematic sketch of the experimental setup (not to scale).

Preliminary results and discussion

Salt precipitation in pores at the final stage demonstrates two crucial salt forms, including large crystals and polycrystalline aggregates (Figure 2), which are similar to the previous findings (Ho & Tsai, 2020; Kim et al., 2013). In addition, the aggregates contain multi-layers and intra-pores.

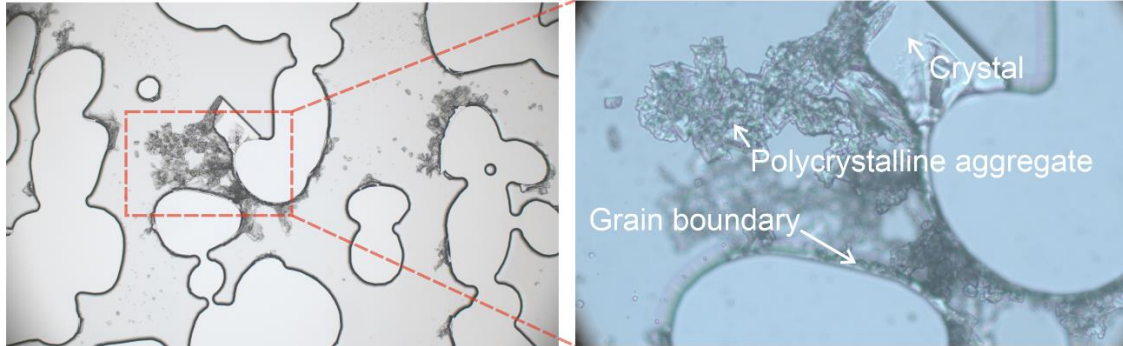


Figure 2 Salt precipitation in pores at the final stage when the chip is completely dry, where large salt crystals and polycrystalline aggregates are founded (the zoomed-in image).

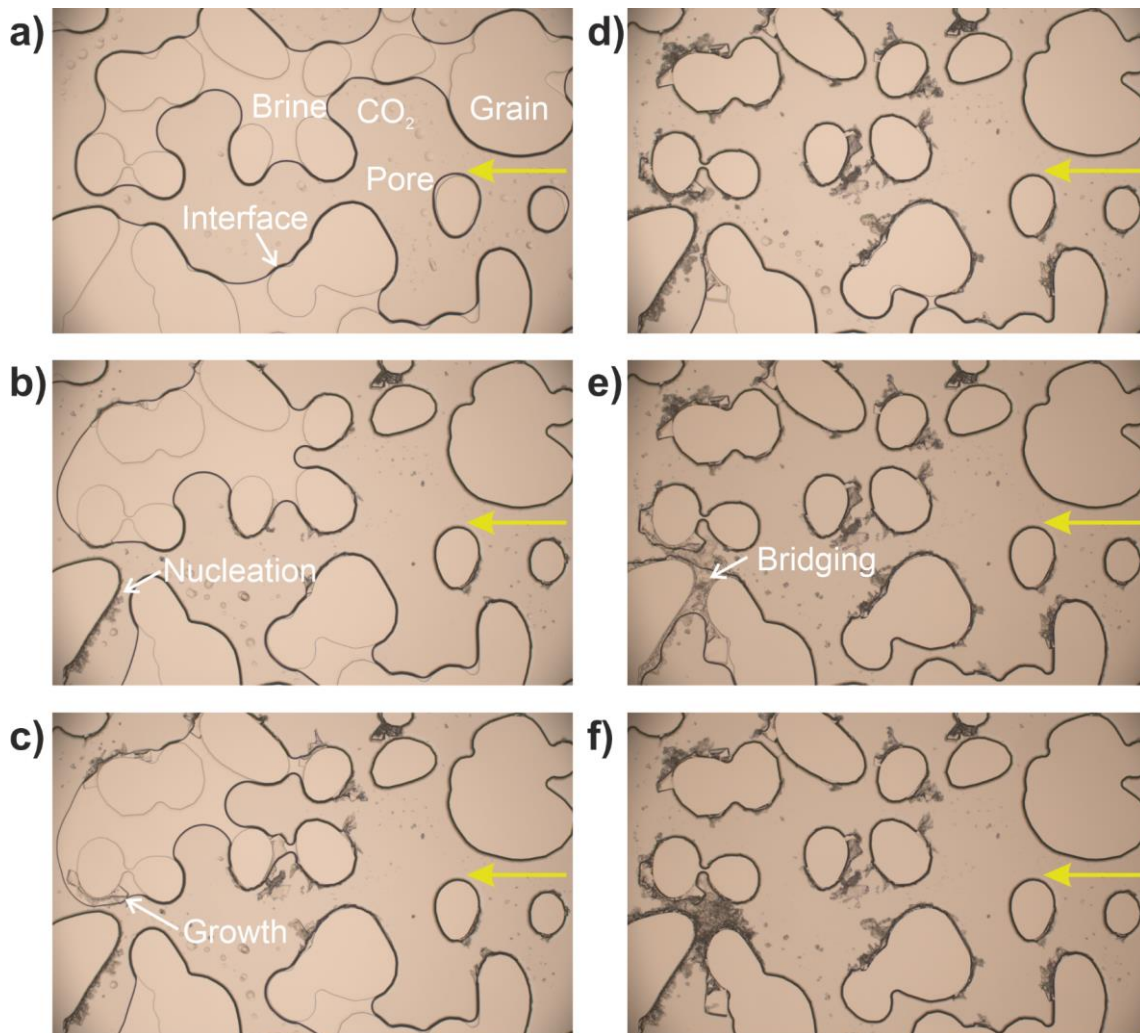


Figure 3 Sequential images of salt precipitation in pores during continuous CO_2 injection into the chip, where the yellow arrow indicates the gas flow direction. a) initial water-gas distribution and interface, b) small crystalline initiation, c) large crystal growth within the water phase, d) late state of salt precipitation, e) water film migration bridging the adjacent grains, f) final stage of salt precipitation containing large crystals and polycrystalline aggregates.

Furthermore, based on the sequential images in Figure 3, we found that large crystals grow within the fluid (Fig. 3c and d), but the front of the polycrystalline aggregates can grow (expand) beyond the fluid interface. Particularly, water can migrate into the polycrystalline aggregates and intensify the density of the small crystals (Fig. 3e and f). The driving force of water diffusion might be the concentration gradient or the difference in evaporation rate at different sites. Additionally, salt precipitation mainly distributes at the pore throats, which may have a significant effect on permeability, which is under current investigation.

Conclusions

This study demonstrates salt precipitation in “real-rock pores” during CO₂ injection into saline-saturated porous media. Large crystals and polycrystalline aggregates are found, where the former occurs within the fluid phase and the latter can grow beyond the water-gas interface. The kinetics of salt precipitation needs to be studied further to gain a better understanding of the potential effects on reservoir performance. Our ongoing study will investigate permeability and porosity changes during the injection, and how temperature may potentially affect the process. Elucidation of such questions will help to better manage geological CO₂ sequestration and minimize risks of formation impairment.

Acknowledgements

The authors wish to express their gratitude to Martin von Dollen (KIT-AGW) and Kevin Altinger (KIT-AGW) for their technical support and contribution to establishing the microfluidic setup.

Reference

- Bacci, G., Korre, A., & Durucan, S. [2011] Experimental investigation into salt precipitation during CO₂ injection in saline aquifers. *Energy Procedia*, **4**, 4450-4456.
- Baumann, G., Henningsen, J., & De Lucia, M. [2014] Monitoring of saturation changes and salt precipitation during CO₂ injection using pulsed neutron-gamma logging at the Ketzin pilot site. *International Journal of Greenhouse Gas Control*, **28**, 134-146.
- Chavez Panduro, E. A., Cordonnier, B., Gawel, K., Børve, I., Iyer, J., Carroll, S. A., Michels, L., Rogowska, M., McBeck, J. A., & Sørensen, H. O. [2020] Real time 3D observations of Portland Cement Carbonation at CO₂ storage conditions. *Environmental Science & Technology*, **54**(13), 8323-8332.
- Ho, T.-H. M., & Tsai, P. A. [2020] Microfluidic salt precipitation: implications for geological CO₂ storage. *Lab on a Chip*, **20**(20), 3806-3814.
- Kim, M., Sell, A., & Sinton, D. [2013] Aquifer-on-a-Chip: understanding pore-scale salt precipitation dynamics during CO₂ sequestration. *Lab on a Chip*, **13**(13), 2508-2518.
- Milsch, H., Giese, R., Poser, M., Kranz, S., Feldbusch, E., & Regenspurg, S. [2013] Technical paper: FluMo—a mobile fluid-chemical monitoring unit for geothermal plants. *Environmental Earth Sciences*, **70**(8), 3459-3463.
- Miri, R., & Hellevang, H. [2016] Salt precipitation during CO₂ storage—A review. *International Journal of Greenhouse Gas Control*, **51**, 136-147.
- Nooraiepour, M., Fazeli, H., Miri, R., & Hellevang, H. [2018] Effect of CO₂ phase states and flow rate on salt precipitation in shale caprocks—a microfluidic study. *Environmental Science & Technology*, **52**(10), 6050-6060.