

EXPERIMENTAL AND VISUAL ANALYSIS OF NATURAL CONVECTION DUE TO CO₂ DISSOLUTION UNDER RESERVOIR CONDITIONS

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ABSTRACT

Dissolution of CO₂ into brine forms a mixture that is denser than the ambient brine. The density gradient causes gravitational instability and triggers convective fingers, which is a favorable process for CO₂ sequestration in saline aquifers. A series of experiments of density-driven natural convection due to CO₂ dissolution were conducted in a transparent vertical Hele-Shaw cell under different conditions. A spectrophotometric method for measuring the concentration of dissolved CO₂ using an acid-base color indicator. The visualization of CO₂ dissolving into brine process demonstrated that the onset time of fingers occurrence was too short to be captured. Our results showed that higher pressure and temperature can promote the fingers' growth; Brine salinity increase inhibits fingers' growth, thus decreasing the concentration of the dissolved CO₂.

Keywords: CO₂ sequestration, visualization, natural convection, Hele-Shaw, density driven

1. INTRODUCTION

Carbon dioxide (CO₂) capture and sequestration (CCS) is one of the viable techniques to mitigate greenhouse gases in the atmosphere^[1, 2]. CO₂ storage in saline aquifers has the highest potential global capacity for long-term CO₂ storage^[3]. When CO₂ is injected into saline formations, it reaches supercriticality, the positive selection and peer-review under responsibility of the scientific committee of the 11th Int. Conf. on Applied Energy (ICAE2019).
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buoyancy of scCO₂ (supercritical CO₂) relative to the ambient brine causes the CO₂ to rise beneath an impermeable cap rock^[4]. CO₂ dissolution and diffusion in the brine increases its density, CO₂-dissolved brine is 0.1% to 1% denser compared to native brine, depending on pressure, temperature, and salinity^[5]. The density difference leads to gravitational instability and results in convective mixing between CO₂-dissolved brine and native brine, which could significantly enhance the CO₂ dissolution rate^[6, 7].

Many studies have been reported in this area to investigate the natural convection driven by density difference^[8]. The numerical simulation of the natural convection process is complicated by the coupling of multi-dimensional parameters with uncertainties in geological parameters. Despite recent improvements in reservoir simulation technology made by researchers, it is still difficult to accurately predict and quantify the convective mixing process of CO₂ dissolution in saline aquifers^[9, 10].

The subsurface geological conditions are not readily simulated in laboratory, so scholars' interest in experimental investigation for natural convection is growing. Hele-Shaw cell is the preferred experimental device for visualization of the dissolution process, and it is usually used to study convective instability. The common optical techniques for Hele-Shaw cell include schlieren method^[11], interference method^[12] and Indicator

discoloration method^[13]. Another experimental setup is pressure-volume temperature (PVT) equipment, it usually consists of a cylindrical cell used to measure mass transfer and dynamic dissolution rate due to natural convection between CO₂ and brine^[14, 15].

2. EXPERIMENTAL SECTION

2.1 Apparatus

The experimental configuration is shown in Fig. 1. The images were captured using an MV-E800M image processing charge-coupled device (CCD) camera (Microvision Digital Image Technology Co., Ltd). It was placed in a camera bellows accompanied by a monochromatic light filter and a uniform light source as a backlight. The camera was directly connected to a computer to facilitate advanced image processing.

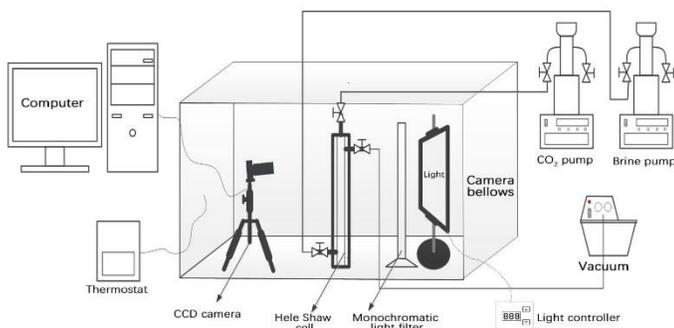


Fig 1 Schematic diagram of CO₂/brine convective mixing experiment

Natural convection experiments were conducted in a high-pressure Hele Shaw cell (Fig. 2). The Hele Shaw cell is equipped with two flat glass windows, which are maintained together vertically in a gravity field and separated by a spacer with a thickness of $l = 1.0$ mm. The maximum allowable pressure and temperature of the high-pressure cell is 15 MPa and 80 °C.

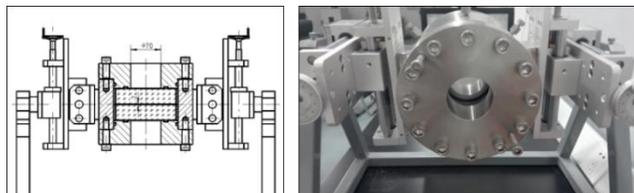


Fig 2 High-pressure Hele Shaw cell.

The experimental temperature was controlled by a temperature controller (Autolise AT72AAS4R), it can maintain a stable temperature point from 25 °C to 80 °C. Two syringe pumps (ISCO 260D, Teledyne Technologies Inc.), were used for brine and CO₂ injection,

respectively. The digital signals of the temperature, injection flow rate, and injection pressure were monitored using the computer.

2.2 Material and method

CO₂ with a specified purity of 99.99% (instrument grade) was supplied by Dalian Date Gas Co., LTD.. The brine samples used in the experiments was prepared by bromocresol green, distilled water and pure NaCl crystal. Bromocresol green was used as acid-base color indicator, its concentration 2.4×10^{-4} mol/L. Table 1 shows the summary of the density driven natural convection experiments condition. The spectrophotometric method for measuring dissolved CO₂ in brine was described in detail in a previous article^[16].

Tab 1 Schematic diagram of density driven natural convection experiment

Temperature(°C)	Pressure(MPa)	Salinity(g/L)
33	9	10
38	10	20
43	11	30

2.3 Procedures

Experiments were conducted according to the following procedure. First, the Hele Shaw cell was filled with colored brine to the experimental pressure, and the temperature controller was used to control the temperature. Second, the CCD camera recorded images without interruption. Third, CO₂ was introduced through the top of the high-pressure cell using a tube connected to a CO₂ pump that was operated at constant pressure to ensure that the CO₂ mixed with the brine. Fourth, images were captured until the gray value remained constant, which indicated that natural convection was complete and the system had equilibrated. Lastly, pressure was released from the system, and the high-pressure cell was cleaned for the next experiment. The steps mentioned above were repeated, and new experiments were started.

3. RESULTS AND DISCUSSION

3.1 Fingering morphology in natural convection

A region of interest (ROI) in the images was selected to analyze the convective mixing process. Each ROI was 50 mm high, and 49 mm wide. The convective fingers images of the density-driven natural convection in Hele Shaw cell were shown in Fig. 3. The dark means the brine without dissolved CO₂ (native brine); the gray means CO₂-dissolved brine, and the bright means CO₂.

The rate of CO₂ dissolution is high under in-situ pressure, the density difference between native brine and CO₂-dissolved brine formed in a short time. The instability was triggered because the denser CO₂-dissolved brine was located above the less dense native brine. Natural convection onset time was much shorter than the CCD camera shooting interval(1s), therefore, we cannot obtain the specific onset time.

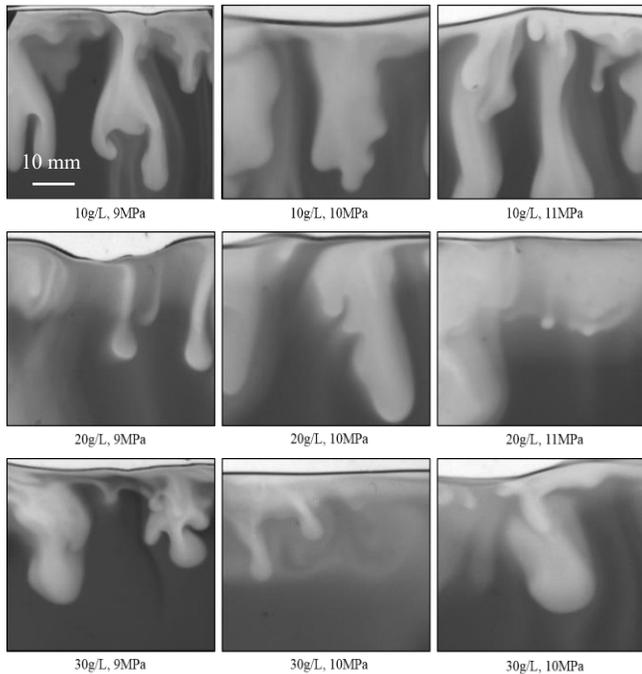


Fig.3 Convective fingers images of the density-driven natural convection experiments at 10th sec and 38°C

Compared with the CO₂/brine convective mixing experiments under atmosphere pressure^[17], the fingers width increased and the fingers number decreased. The migration rate of the fingers is fast that can reach 5mm/s, fingers propagation period last 12-15 sec on average, it hit the bottom of the ROI quickly.

It can be observed from Fig 3 that the fingers growth velocity increase with the pressure and decrease with increased salinity. However, the pressure and salinity variation impact on fingers growth is relatively weak, because the changing gradient of pressure and salinity is too small.

3.2 Concentration of dissolved CO₂

The concentration of dissolved CO₂ during the density driven natural convection process was measured by the spectrophotometric method^[16]. The gray value variation of images was used to determine the dissolved CO₂ concentration. The end of the convective mixing corresponds to the fingers become blurred and the gray

value remained constant. Figure 4 shows the measured dissolved CO₂ concentration for the CO₂/brine convective mixing experiment.

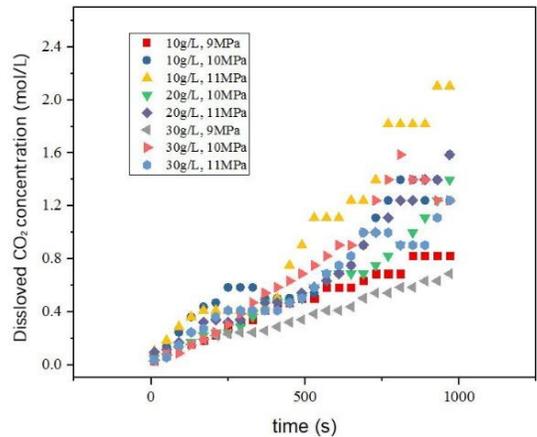


Fig 4 Average value of dissolved CO₂ concentration for different conditions

At the initial stage of experiments, the dissolved CO₂ concentration of 10 g/L, 11MPa (yellow triangle) was highest than others, and the 30 g/L, 9 MPa (gray triangle) was the lowest. It means that pressure increase can promote CO₂ dissolution into brine, while salinity increase can inhibit it. For the first 15 second of the finger's propagation period (Fig 5), we focused on the effect of temperature on CO₂ concentration. Although temperature increase reduces the solubility of CO₂, the absolute solubility of CO₂ in brine is much higher than the experiments conducted under atmospheric pressure. The temperature increase leads to the greater mass transfer coefficient and the lower viscosity, therefore, the higher the temperature, the faster fingers migration rate and the higher the CO₂ concentration.

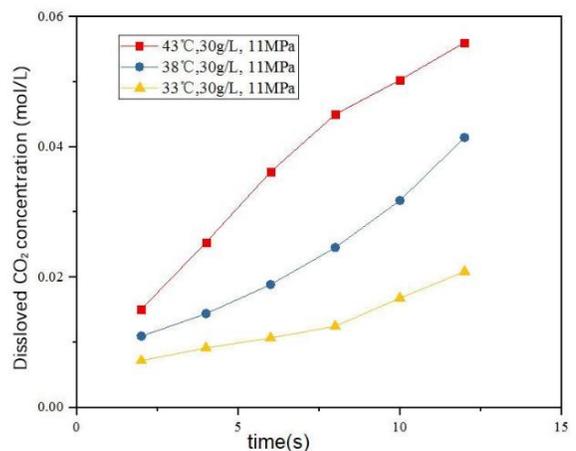


Fig 5 Effects of temperature on the increasing rate of dissolved CO₂ concentration

4. CONCLUSION

The setup in this study can be carried out visual quantitative analysis of CO₂/brine density driven natural convection under reservoir condition. The influence of pressure, salinity and temperature change on the natural convection process have also been analyzed. The increase in pressure and temperature could improve the dissolved CO₂ concentration, while the increase in salinity could reduce it. Based on calculations, we concluded that the instability of density driven natural convection results in a faster increase in dissolved CO₂ concentrations. The proposal of using spectrophotometric method to measure the concentration of dissolved CO₂ has a guiding significance for the quantitative study of natural convection in the CO₂ dissolution process in the future.

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