Three-Dimensional Visualization of Natural Convection in Porous Media

Lei Wang a, Akimitsu Hyodo a, Shigeki Sakai a, Tetsuya Suekane a

Abstract

Carbon dioxide (CO₂) geological storage (CGS) is a preferred technique to mitigate anthropogenic climate change. In terms of CGS, density-driven natural convection dominated dissolution of CO₂ into formation brine results in a stable trapping increasing the storage security.

A full utilization of a nonlinear density property of fluid system has been applied to mimic the natural convection in saline aquifers. Three-dimensional natural convection in porous medium has been observed by employing X-ray computer tomography (CT) technology. Since the X-ray attenuation depends on the concentration of sodium iodide, the distribution of local concentration can be evaluated during the downward extension based on the CT images.

We successively visualized the process of formation and the development of Rayleigh-Bénard fingers. This process modelled natural convection between heavier CO₂ loaded brine and brine with little (or no) CO₂. High density fingers extend downward at the characteristic buoyancy velocity and coalesce with the neighbouring finger before reaching the bottom of the porous medium. The coalescence of fingers leads to a decrease in finger number and an increase in finger diameter. As the Rayleigh number increases, the finger tends to be fine and extends faster. The mass transfer rate increases with the Rayleigh number also.

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Keywords: Visualization, Natural convection, Rayleigh-Bénard fingers, Mass transfer, Rayleigh number

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1. Introduction

Geological carbon dioxide (CO₂) storage (CGS) is a preferred technique to mitigate anthropogenic climate change. The CO₂ is injected into the saline aquifers or depleted oil and gas reservoirs at the depths of 750 m or more where the CO₂ is in a supercritical state. Under the typical reservoir conditions, the density of CO₂ is somewhat lower than that of brine. Therefore, one of the major concerns with CCS is a potential migration and leakage from the reservoirs due to buoyancy. However, four mechanisms [1] are considered to trap the CO₂ in the reservoirs: 1) physical or structural trapping by the impermeable layer; 2) capillary trapping [2–4]; 3) dissolution into the formation brine [5–9] and 4) mineralization [10]. In present work, we focus to the dissolution of the CO₂ into the brine. The brine in contact with CO₂ would be saturated with CO₂ and due to the slight rise in the density, the natural convection would be induced in a reservoir scale. The unstable stratification of density drives natural convection in form of descending fingers in porous medium. In Fig. 1, the densities of the MEG solutions, containing 50 wt % ethylene-glycol, doped with 13 wt %, and 17 wt % NaI, are lower than those of 10 wt %, 12 wt % and 13 wt % NaCl solutions, respectively. The unstable stratification of density drives natural convection in form of descending fingers in porous medium. We selected this material for the porous medium for the sake of high contrast CT images.

The natural convection due to the density difference is scaled with the dimensionless number, i.e., Rayleigh number $Ra = (H\Delta \rho g k)/(\varphi D\mu)$, where $H$ is the characteristic length; $k$ the permeability; $\Delta \rho$ the density difference; $g$ gravitational acceleration; $\varphi$ porosity; $D$ molecular diffusion coefficient; and $\mu$ viscosity. In the North Sea reservoir conditions, the Rayleigh number is very large, $Ra \sim 10^3 – 10^4$ [5, 11, 12]. Onset of natural convection and the mass transport of dissolved CO₂ give high impact on the long-term behavior of injected CO₂ in geological formations. To this end, the convection process has been investigated by numerical simulations [13, 21]. Recent, rapid growth of the computer performance allows us the three dimensional numerical simulations at high Rayleigh numbers, and a flat finger structure [15] and the strong scaling of the wavenumber [20] which are peculiar to the three-dimensional natural convection. On the other hand, the experimental researches have been conducted by utilizing two-dimensional porous media [22–24] including Hele-Shaw cells [25–27], because of opaque property of porous materials, except for MRI application [28].

Neufeld et al. [11] reproduced the convective behavior of CO₂-enriched brine by applying a fluid system (solutions of methanol and ethylene-glycol mixing with water). Two-dimensional laboratory experiments in a relative high Rayleigh number and high-resolution numeral simulation in lower Rayleigh number had been conducted.

In this paper, we develop a novel technique to have time-lapse three-dimensional images by using X-ray computed tomography (CT). We propose a full utilization of a nonlinear density property of fluid system (NaCl solution, sodium iodide and a mixture of methanol, ethylene glycol) to mimic the process of CO₂ dissolving into saline aquifers. In order to create a similar geological porous medium as it is under reservoir condition, a tube with 70 mm inter diameter was filled by plastic resin particles which has an average diameter of 780 μm to 80 mm in height. In these visualization experiments, the advancement of convective fingers in porous medium has been observed. The quantitative analysis of the finger velocity, the finger number and the mass flux during natural convective can be achieved.

2. Experiment method and equipment

In this study, we employed a new experimental analogue that mimics natural convection in three-dimension. Since the X-ray attenuation depends on the concentration of sodium iodine (NaI), NaI was dissolved into the mixture of methanol and ethylene-glycol solution (MEG), then merging with NaCl solutions. Initially, the solution of MEG doped with NaI (NaI + MEG) is less dense than NaCl solution, but the density of the mixture (MEG+NaI and NaCl solution) may exceed that of NaCl solution, depending on the fraction of MEG+NaI. In Fig. 1, the densities of the MEG solutions, containing 50 wt % ethylene-glycol, doped with 13 wt %, 15 wt %, and 17 wt % NaI, are lower than those of 10 wt %, 12 wt % and 13 wt % NaCl solutions, respectively. The mixing of NaI + MEG with NaCl solution leads to an increase in density that resulting in the formation of convection fingers. With a decrease of MEG+NaI in content, the density of the mixture gradually decreases to the density of NaCl solution after reaching the maximum equilibrium density. The increasing of density is the key parameter that governs the strength of natural convection and determines the mass transfer rate of CO₂ dissolving. The unstable stratification of density drives natural convection in form of descending fingers in porous medium.

We used a packed bed of melamine resin particles (Ube Sand Industries, TPS 18 – 30) with diameter from 530 μm to 1030 μm as porous medium. We selected this material for the porous medium for the sake of high contrast CT images. In this study, we employed a new experimental analogue that mimics natural convection in three-dimension. Since the X-ray attenuation depends on the concentration of sodium iodine (NaI), NaI was dissolved into the mixture of methanol and ethylene-glycol solution (MEG), then merging with NaCl solutions.

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images. The permeability of packed bed was estimated by measuring the pressure drop for various flow rates of NaCl solution. The permeability was \( k = 2.60 \times 10^{-10} \text{ m}^2 \) and the porosity was \( \phi = 0.49 \), which was estimated on the basis of the weight change before and after saturation with NaCl solution.

![Figure 1](image1.png)

**Fig. 1** The density of MEG doped with NaI mixed with different weight percentage NaCl solution (a) 10 wt%; (b) 12 wt%; (c) 13 wt%.

Experimental conditions are summarized in Table 1.

<table>
<thead>
<tr>
<th>Case</th>
<th>Particle diameter (μm)</th>
<th>NaI concentration (wt %)</th>
<th>NaCl concentration (wt %)</th>
<th>Density difference (g/cm³)</th>
<th>Rayleigh number (-)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>530-1030</td>
<td>17</td>
<td>12</td>
<td>0.025</td>
<td>5000</td>
</tr>
<tr>
<td>2</td>
<td>15</td>
<td>10</td>
<td></td>
<td>0.017</td>
<td>3500</td>
</tr>
<tr>
<td>3</td>
<td>17</td>
<td>13</td>
<td></td>
<td>0.014</td>
<td>2700</td>
</tr>
</tbody>
</table>

In this work, fingers formed by the density-driven natural convection will not appear immediately at the beginning of dissolving NaI + MEG. High density fingers will occur when the density difference reaches a certain value.

With the reconstruction software, the CT values were adjusted in order that the CT values of an acrylic resin tube and surrounding air remain same among the scans. After that, the CT values were transformed into the NaI concentration with the calibration curve which was taken in advance.

![Figure 2](image2.png)

**Fig. 2** The relation between CT value and NaI + MEG weight percentage.

From the horizontal-cross sectional images of the initially formed fingers, the CT value of fingers can be obtained. Based on the relation between CT value and weight percentage of NaI + MEG as shown in Fig.2, the weight percentage of dissolved NaI + MEG for three cases in Table 1 were from 64 wt% to 72 wt%. Thereby, we
used the density at 70 wt% of NaI + MEG to evaluate the Rayleigh number in Table 1 as a typical density difference between the fingers and NaCl solution.

The experiments were conducted under the room temperature condition and applied by the following procedures. First, the NaCl solution was pulled into the packed bed by using a vacuum chamber. Next, after removing the top part of the packed bed, the particles saturated MEG and NaI solution were put on the top of tube above the particles saturated NaCl 30 mm in height (Fig. 3). Then the packed bed was scanned by the X-ray CT scanner (Comscantechno, ScanXmate-RB090SS).

A whole packed bed was scanned by the X-ray CT scanner. The reconstructed images consist of 496 slice images of 496 × 496 pixels at the resolution of 193μm/pixel in all directions. It takes approximately 60 seconds to scan one image. Until natural convection completes, the scan continues every four minutes.

![Fig.3 Experimental equipment.](image)

### 3. Results and discussion

#### 3.1. Fingers of natural convection

High concentration fingers appear on the interface and extend straight downward. The development of natural convection in the particle pack is shown in Fig. 4 at the Rayleigh number of 3500. The three-dimensional images are shown in Fig.5 at the Rayleigh number of 5000.

![Fig.4 Vertical cross-sectional X-ray CT images at the Rayleigh number of 3500.](image)
During the processing of natural convection, the fingers extend downward and coalesce with neighbor fingers before arriving at the bottom of the packed bed. From the distance of proceeding finger tips the finger velocity is estimated for each Rayleigh number as shown in Fig. 6. The development of fingers occurs more quickly with an increasing in Rayleigh number. The finger extension velocity agrees with the characteristic velocity \( U = \frac{\Delta \rho g k}{\mu} \), which defined by the Darcy velocity of finger extension with the initial density difference, with the coefficient \( c \) of 0.34.

As a following procedure, the number of fingers is counted from the horizontal cross-sectional images. First, high concentration pixels with a radius of lower than 10 are removed from the images as noise. Then, a Gaussian blur filter is applied to images. Finally, a local maximum in NaI concentration is detected as a finger.

Figure 7 illustrates the finger cell distribution at an early stage of finger formation under different Rayleigh number conditions. The number of the fingers is in a certain range, between 150 and 200 around the interface position, and increases with Rayleigh number.

Figure 8 shows the time evolution of the finger number density at each cross-section at \( Ra = 3500 \). For all experiments, similar results were observed. Initially, the finger appears on the interface at high finger number density. Then the position of the maximum finger number density shifts downward with the finger velocity shown in Fig. 6, with reducing the number density. Figure 9 shows the horizontal-cross sectional images 5 mm below the initial interface. Initially, fine finger appears on the interface (Fig. 9a), then the coalescence of fingers leads to a decrease in finger number and an increase in finger diameter (Fig. 9b–d). Among the cross-sectional images Fig. 9b–d, the structure of the fingers remain same, even though the finger diameter grows. It suggests that the Rayleigh–Bénard cell structure is formed during the natural convection and the position of upward and downward flowing regions is immobile with time. Finger diameter would grow with time, because of the mixing between the upward and downward flowing regions.
Fig. 7 Vertical cross-sectional X-ray CT images (top) and horizontal images (bottom) for the Rayleigh number of (a, d) 5000, (b, e) 3500, and (c, f) 2700, respectively.

Fig. 8 The change of finger number density with time for Rayleigh number is 3500. The initial interface located at the height of 30 mm.

Fig. 9 Horizontal-cross X-ray images 5mm below the initial interface at (a) 0 s, (b) 240 s, (c) 480 s, and (d) 720 s respectively for the Rayleigh number of 3500. Brightness of images is proportional to the NaI concentration as shown in Fig. 2.
Fig. 10 shows the finger number density increases with an increase of Rayleigh number. During the process of finger extending downward, the finger number density decreases with time.

![Graph showing finger number density vs. dimensionless time](image1)

Fig.10 The change in finger number density with dimensionless time at the position 5 mm below the initial interface.

### 3.2. Mass flux of natural convection

The quantification of CO₂ mass transfer into brine is of great importance to determine the rate of solubility trapping. In this experimental study, in order to calculate the mass transfer of MEG+NaI dissolving into NaCl solution, the packed bed was divided into two cylinder regions, a top cylinder region and a bottom cylinder region at the plane where the initial interface located. Based on the local concentration of NaI solution, the mass of NaI in the top and bottom cylindrical regions was evaluated for each time step. A decrease in mass in the top region agreed well with an increase of that in the bottom region. From the change of the mass, the maximum of mass flux $F$ for each Rayleigh number were estimated as shown in Fig. 11. It indicates that the mass flux increases with an increase in Rayleigh number.

![Graph showing mass flux vs. Rayleigh number](image2)

Fig.11 The relation between Rayleigh number and mass flux

### 4. Conclusion

This study describes experimental research on density-driven natural convection due to the density difference between NaI + MEG and NaCl solution in porous medium. The process of the finger development and formation has been visualized by using an X-ray CT scanner.
During process of convective mixing, high density fingers descend downwards and the change of NaI + MEG concentration in fingers has been observed. The finger number diminishes with the time. At early stage, the maximum finger number density appears at the interface. The position of the maximum finger number density moves with finger tips. The fingers coalescence with each other leads to the decrease of finger number and the increase of finger diameter.

As the Rayleigh number increases, the finger number increases and the finger extends more quickly. The mass transfer rate increases with the Rayleigh number as well.

Acknowledgements

This work was supported by JSPS KAKENHI Grant Numbers 25281036 and 26630463.

References