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Dynamic evolution of the CO_2-brine interfacial area during brine imbibition in porous media

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ABSTRACT

Explicit knowledge on the two-phase interface evolution during CO_2 dissolving in brine provides accurate predictions on the subsurface behavior of long-term CO_2 storage. In this research, the interfacial areas of CO_2-unsaturated brine were dynamically measured during multiphase flow using 3D quantitative analyses. The two-phase interfaces during brine imbibition were divided into three terms based on their attributes, i.e., ganglia, cluster and singlet. The evolution terms of the interfaces were interesting, as their fates showed wide evolution patterns due to the diverging effects of the Reynolds number (fluid velocity × length scale/fluid viscosity) and gravity. The brine bypassed the CO_2, and the interface evolved with the development of a priority path under a heterogeneity impact. Relying on the approach of the slice-averaged and volumetric measurement, the effects of forces and heterogeneity on the CO_2-unsaturated brine interface were evaluated on different directions. Linear regression of the clouded data points exploited the validity of the power-law distribution from number of trapped cluster to frequency of interfacial area, and the max interfacial areas and variance decreased, while the mean interfacial area increased with brine saturation. Slice-averaged CO_2-brine interfacial areas normalized by volume or geometric surface area decreased linearly with the brine saturation at different Reynolds numbers.

1. Introduction

To reduce the CO_2 emission into the atmosphere, CO_2 geological sequestration is an economic and valid strategy among the existing technologies [1]. For CO_2 geological storage, the optimal designs of injection wells request not only the geological information, but also those of fluid dynamics, such as the dissolution rate, which concerns with the local geo-energy conservations [2–6]. CO_2 storage security can be increased either by reducing the leakage probability or accelerating the CO_2 dissolution in reservoir brine. CO_2 can be immobilized by capillary gas trapping, dissolution in reservoir fluids (brine) or subsequent geochemical reactions.

Based on Noyes Whitney equation and the Fick's second law, the rate of change in concentration of dissolved material with time. It is directly proportional to the concentration difference between the two sides of diffusion layer. Based on the mass balance equation:

$$\frac{dm}{dt} = A_{eff} K_e (C_s - C_0) \tag{1}$$

where K_e (m/s) is the effective mass transfer coefficient, A_{eff}, the effective interfacial area between CO_2 and brine in porous media, and C_s and C_0 are CO_2 solubility and the CO_2 concentration in brine, respectively.

This time-dependent mass transfer is difficult to quantify, since the evolution of the interfacial area during the mass transfer should be quantitative descripted [7,8]. Determining the interfacial areas between CO_2 and brine is important for interpreting the mass transfer kinetics, and the characterization of the flow dynamics at the interface is closely related to CO_2 storage security. The interfacial area measurements for liquid-liquid pairs in porous media have matured, but the features that are critical in CO_2 storage are different for gas-liquid interfacial areas [9–11]. Researchers have typically measured the specific interfacial area as a lumped parameter to express the interphase mass transfer because of the inability to directly measure the gas–liquid interface [12,13]. The techniques mainly used to determine interfacial domains have been interfacial tracers and non-invasive imaging technologies [11,14–16]. Comparisons among previous measurements have shown that the liquid-liquid interfacial areas that are measured

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using interfacial tracers are larger than the areas measured using non-invasive imaging technologies due to the favorable adsorption of the tracers in porous media.

Recently, X-ray micro-tomography, which is a non-invasive technology, has been widely used to identify the fluid morphology in porous media. Researchers from Suekane lab, Tokyo Institute Technology, Japan, quantitatively measure the dissolution and convection of CO_2 in porous media by using micro CT [17,18]. This image-based measurement provides explicit knowledge on which interfacial domains are represented by the measured interfacial areas. Specifically, CT can capture both the meniscus area and the thin film area while explicitly excluding the interfacial micro-morphology [10,19]. Thus, the interfacial areas extracted from the CT image stacks represent the total "smooth" interfaces as an upscale definition for two or three phases [20]. Additionally, when the average diameter of the grain particles is consisted of less than 10 pixels in CT images, the medium segmentation is less accurate and difficult to qualitatively assess due to the small particle and pore sizes [21]. However, in former studies, the dynamic measurements rarely received sufficient consideration, and the factors that influence the fluid distribution substantially affected the interfacial area measurements under complex flow conditions [22], including studies addressing the impact of X-ray microtomography image resolution and/or segmentation technics on the identification of brine and CO_2 phases and interfacial areas. This should be taken into consideration for the current study. Furthermore, the CO_2-brine interface evolution during forced imbibition of brine has not been observed and measured. As the CO_2 dissolves into the groundwater, the CO_2-brine interface gradually shrinks and disappears in the flooding brine, and using CT observations, the continuous change can be monitored to quantify and extend the confidence range of the results.

In this work, micro CT was used to measure the CO_2–brine interfacial areas for seventeen sets of experiments in synthetic porous media. Three objectives guided the work: (1) to provide a method to aggregate the terms of evolutions of interfacial area which could be validated on both 2D and 3D measurements, (2) to evaluate the influence factor on the CO_2-brine interfacial area when dissolution occurs in porous media, and (3) to develop empirical μCT-based models of the interfacial area-brine saturation relationships for sandy porous media. The utility of the empirical model was assessed by estimating the interfacial area-water saturation relationships for a variety of experimental configurations for which independent measurements were available.

2. Experimental methods

2.1. Experimental apparatus and materials

The experimental apparatus and its components that were used to observe the brine imbibition experiments are shown in Fig. 1, the same as the previous study [23]. Detail description of the experimental apparatus can be found in the previous study. An Inspexio SMX-225 CT system (Shimadzu, Japan) was used to visualize the internal fluid in the porous media. The holder (30 mm length and 6 mm inner diameter) was fabricated using Perspex and an epoxide-resin glue and had a low X-ray attenuation for normal pressure and anti-distortion. CO_2 with a 99.9% purity was used. The brine contained 3 wt% NaCl to simulate underground salinity. Also, 6 wt% potassium iodide (KI) was dissolved in the brine to enhance the contrast between the brine and gas in the CT images. To achieve well-sorted fractions of heterogeneous porous media, two packed beads, sole glass beads (GB medium with a diameter of 0.4 mm) and mixed glass beads (GB mix, weight fraction of 1:1 for glass beads with diameters of 0.2 mm and 0.4 mm), were assembled in the holder.

2.2. Experimental procedure

The flow conditions are listed in Table 1. Before each experiment, the whole system was flushed with N_2 and then vacuumed to ensure no liquid remained. Prior to the CO_2 filling, one scan (dry scan) was conducted for the porosity measurement. Then, the original CO_2 distribution was acquired via another scan (CO_2 scan) after the CO_2 filling. Fresh brine without saturated CO_2 was injected into the sample at 1 atm and room temperature, and a series of scans (wet scan) were conducted during the injection until the variation in the gas saturation was less than 1%. Each wet scan had an exposure time of 120 s, and the time-step between two scans is 40 s. The Reynolds number, Re, could represent the flow rate at a certain extent. Re is defined as fluid density × fluid velocity × a representative grain diameter for the porous media/fluid viscosity:

$$Ra = \frac{\rho V d_{30}}{\mu} \tag{2}$$

where ρ is the density of brine, v is the specific discharge, d_{30} is a representative grain diameter for the porous media (0.4 mm and 0.3 mm), and μ is the viscosity of the brine.

A relatively wide region of Re was covered to allow reasonable comparisons. In addition, brine was injected upward or downward to observe the impact of gravity on the non-equilibrium mass transfer process. We obtained 308 slice images for each scan, and the CT images with a cross section area of 512 × 512 voxels were acquired at a voxel size of 15 μm. The length of the field of view in the Z axis was 4.8 mm in the middle of the holder to prevent the end effect and to allow the fluid to fully develop. In these experiments, the scanning was performed at 90 kV and 50 μA.

2.3. Image processing

The three phase segmentation process, which directly assigns a certain voxel to the CO_2, brine, or solid phase, determines the reliability of the quantitative information derived from the CT images. The segmentation process for the CT images with CO_2 or CO_2-brine in the pores is illustrated in Fig. 2. The raw CT data was saved as the gray value images using the preset software of CT machine, in which the porous media, CO_2 and brine were attributed to the pixels of black, gray and dark gray, respectively. Each CT image for the dry and wet scans was imported into the ImageJ software (NIH image, U.S) to remove the surrounding outer regions and external walls and filtered with a Gaussian blur filter. The segmentation task for the three phases was relative simple and reliable due to the slight noise level of the raw CT data. For dry and wet scans, two sets of appropriate thresholds were applied to the images based on the visual inspection of each image. A threshold with a CO_2 distribution in contravention of the raw CT data was applied as a sensitivity analysis in Fig. 3. Such thresholds resulted in a small deviation of the final calculations of interfacial areas. The CO_2, solid and solid-brine phase were extracted from the images by creating a binary, and then, the brine phase was separated by subtracting the solid phase from the solid-brine phase. All the phase boundaries were detected, e.g., CO_2-brine, solid-brine and solid-CO_2. The CO_2-brine phase boundaries were separated from the other boundaries. The slice-averaged interfacial area was calculated by measuring the CO_2-brine boundary length in the two-dimensional slice images.