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Co-injection of CO_2 and Nanoparticles Enhance Sequestration Potential in Subsurface Aquifers

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ABSTRACT

CO_2 storage in deep saline aquifers is a promising technique for carbon neutrality. Accelerating the dissolution rate of dissolution trapping is key to improving storage efficiency and providing long-term storage security. In this study, density-driven convection processes were conducted using magnetic resonance imaging (MRI) with two analog fluid pairs as equivalents of the CO_2-brine fluid system. Superhydrophilic SiO_2 NPs with different mass fractions (0 wt%, 0.1 wt%, and 1 wt%) were added to the dense fluid to investigate the optimum mixing ratio. The basic fluid interface instability is identified. The addition of nanoparticles shortens the onset time of instability and increases the finger numbers in each axial section. It shows that dense fluid with NPs leads to more stable interface behavior. Furthermore, experimental results confirm that a reduction in surface tension between NPs and CO_2 would enhance fluid miscibility and the mass transfer during convection. This study can expand the perspective on accelerated dissolution rates and CCS security in the substance.

Keywords: CO_2 storage, dissolution trapping, nanoparticles, interfacial instability, CCS security

NONMENCLATURE

Abbreviations	
CCS	CO_2 Capture and Storage
NPs	Nanoparticles
MRI	Magnetic Resonance Imaging
Symbols	
N_{max}	The maximum number of fingers
Ra	Rayleigh number

1. INTRODUCTION

Global warming, which is the main manifestation of climate change, has been caused by greenhouse effects, and the primary anthropogenically emitted greenhouse gas is carbon dioxide (CO_2)[1]. CO_2 capture and storage (CCS) is an effective method of reducing the amount of CO_2 emitted into the atmosphere[2, 3]. As sites of CO_2 geological storage, deep saline aquifers have demonstrated great potential because of their large storage spaces and existence in many locations around the world. Dissolution trapping in saline aquifers is considered one of the most effective and safe methods of CO_2 geological storage[4].

Under ambient temperature and pressure conditions in a typical aquifer, CO_2 dissolves into the aqueous phase (brine) and creates a dense interfacial layer The CO_2 -saturated brine in a saline aquifer is 0.1% to 1% denser than natively formed brine, depending on the pressure, temperature, and salinity. Gravitational instability or unstable stratification triggers the convection of fluids, which is commonly referred to as density-driven convection in the literature[5, 6]. As the CO_2 dissolves into the underlying brine, the CO_2-saturated brine becomes denser than the native brine, and the density difference causes instability and triggers convection. Dense CO_2-saturated brine migrates downward, while light native brine rises, and the natural convection of these two fluids generates convective fingers[7]. The downward migration of the CO_2-saturated brine significantly enhances the mass transfer of CO_2 and reduces the time required for total dissolution. More importantly, relative to the native brine that fills the pore spaces, the negative buoyancy of the CO_2-saturated brine may reduce the risk of leakage along geological faults, which is of considerable concern relative to long-term sequestration security[7]. Recently, a method has been proposed to accelerate the dissolution of CO_2 and brine and reduce the risk of leakage with co-injection of CO_2 with nanoparticles (NPs). Low-level nuclear waste containing NPs was suggested to be an economical method through a fully mixing with CO_2 before injection[8, 9]. While the interfacial properties of the accelerated dissolution processes have rarely been studied.

In this study, we visualized and quantitatively measured the density-driven convection in porous media by using magnetic resonance imaging (MRI). An analog fluid pair system of 80% glycerite (dense fluid)/$MnCl_2$

(light fluid) was used to simulate the brine with a dissolved $ScCO_2$-formation brine system. The impact of NPs fluids added into CO_2 on fluid-fluid interfacial instability was analyzed. Superhydrophilic SiO_2 NPs with different mass fractions (0 wt%, 0.1 wt%, and 1 wt%) were added to the dense fluid to investigate the optimum mixing ratio.

2. EXPERIMENTAL SECTION

2.1 Experimental materials

A handmade glass sand BZ02 was used to simulate the structure of porous media with an average particle size of 200 um, a porosity of 0.354, and a permeability of $13.3*10^{-12}$ m^2.

There are many challenges in convective mixing of the reservoir in situ temperature and pressure, and simplified experimental methods are often used in the laboratory. The method of analogous fluid pairs has been widely reported in previous studies to mimic the dissolution migration in CO_2-brine systems at the lab scale. This study was conducted using 80 wt% glycerol (as the dense fluid) and a 0.05 wt% $MnCl_2$ solution (as the light fluid). The fluid selection in this test meets the following three principles: ease of identification with MRI equipment, the satisfaction of convection conditions, and fluid characteristics (such as the density) similar to those of reservoir fluids. The related density properties of fluids can be found in previously published studies[10]. NPs with a different mass fraction (0 wt%, 0.1 wt%, and 1 wt%) were added to the dense fluid (80 wt% glycerol) as a variable to observe the convection mixing instability process.

2.2 Experimental procedures

The dissolution process was conducted at 25°C and 0.1 MPa. Firstly, tightly packed sand in a cylindrical vessel considering made of polyimide. Then, the sand was put at a vacuum pump to saturate with the light fluid (0.05 wt% $MnCl_2$) for 6 h. The vessel was removed after fully saturating the sand and injected the dense fluid (80 wt% glycerol with variable concentrations of SiO_2) up to the middle of the vessel. Both ends of the vessel were tightened with rubbers, Rapid rotation of the vessel by 180°, and placed into the MRI detection chamber. Start a pre-set MRI imaging sequence to continuously acquire images in progress.

2.3 MRI data processing

The bright area in the original MRI image is the 80% glycerol solution and the darker area is the 0.05 wt% $MnCl_2$ solution (Mn^{2+} is a paramagnetic substance and inhibits imaging). The main steps in image processing are as follows: 1) Edge cropping. 2) Gaussian filter processing. A 1.0-pixel Gaussian filter was used, and all images were conducted after noise reduction. 3) Colored image sequencing. The grayscale image is covered with a pseudocolor image and finally saved in TIF format. The details are shown in Fig.2.

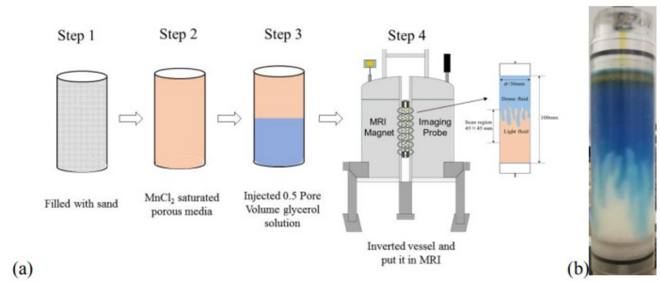

Fig 1. (a) Schematic diagram of density-driven convection mixing experiment. (b) Physical picture of convection mixing for case 1, the glycerol solution was stained with methylene blue in order to distinguish two fluids outside the MRI.

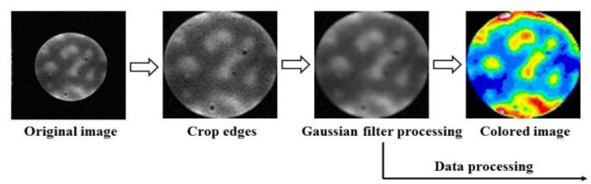

Fig 2. MRI data processing.

3. RESULTS AND DISCUSSTIONS

In this section, the basic parameters of the finger-in instability were first estimated and then visualized to confirm the improvement in interface stability.

The onset time of convection is the time at which the signal intensity shows a significant downward trend and obvious convection fingers can be observed. The earlier onset time reflects better sequestration safety. The presence of NPs shortens the onset time due to the improved interface properties. During the whole process, the convective development of fingers consists of the following stages: finger appearance, finger propagation, new finger generation, and finger coalescence. The largest finger numbers appear in the finger appearance stage, and there is a linear downward trend of the finger number after this point. Convection occurring in porous media depends not only on whether the mixture has reached the critical Rayleigh number (Ra) but also on the domain aspect ratio.

The interfacial behavior of the fluid during the convection mixing process is continuously captured. It shows that dense fluid with NPs leads to more stable interface behavior, as shown in Fig. 3. Significantly higher interface sweep area, which means greater mixed

Table 1. Maximum of fingers number in each axial section

No.	0 wt%	0.1 wt%	1 wt%
Axial-01	26	32	36
Axial-02	15	17	21
Axial-03	11	13	16

solubilities. Furthermore, experimental results confirm that a reduction in surface tension between NPs and CO_2 would enhance fluid miscibility and the mass transfer during convection. The maximum of finger numbers N_{max} in each axial section is analysis, which determines the sweet area and the fluid mass flux. It can be directly measured via the MRI images, as shown in Table 1. The highest number of finger numbers near the fluid interface, with the number of cross-sectional finger numbers decreasing as dissolution extends. This is due to the merging or dissolving disappearance of the fingers on the way down. In any axial sections, N_{max} increases with increasing SiO_2 concentration. It is indicated that the improved interfacial stability enhances mass transfer

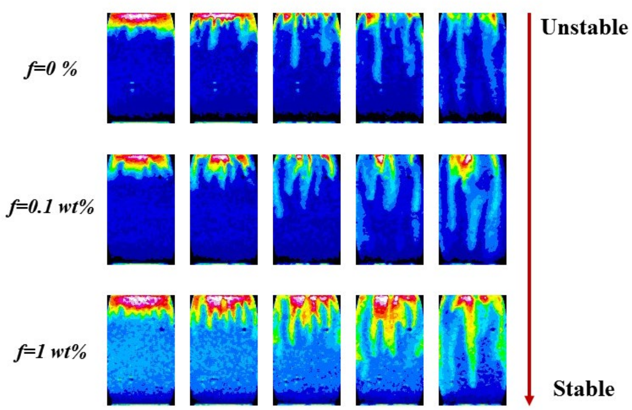

Fig 3. Finger development process with the different mass fractions of NPs.

fluxes. Compared with the traditional injection method, CO_2 containing a high concentration of nuclear waste may have a higher sweeping area and higher sequestration efficiency. These results challenge our view of carbon sequestration and dissolution efficiency in the subsurface, suggesting that co-injection of $ScCO_2$ and low-level nuclear waste containing NPs perhaps a feasible approach that can enhance sequestration potential.

4. CONCLUSIONS

In this study, the effect of NPs on density-driven convection in porous media was quantified. The spatiotemporal evolution of the fingering instability and fluid concentration distribution was visualized with high-field MRI equipment. The results were analyzed to identify the major control of the CO_2 dissolution process with the assistance of NPs. By continuously acquiring the interfacial behaviour of fluids during convective mixing, it can be found that the interfacial behaviour of dense fluids containing NPs is more stable. Reducing the interfacial tension between nanoparticles and CO_2 increases the fluid miscibility and mass transfer capacity of the convective mixing process. CO_2 containing high concentrations of nuclear waste may have a higher swept area and storage efficiency compared to conventional injection. More research could be done to apply nanoparticles to enhance storage efficiency.

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