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Numerical analyses of the effects of nitrogen on the dissolution trapping mechanism of carbon dioxide geological storage

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ABSTRACT

Numerical simulations are carried out to investigate the effects of impurity on geological storage of carbon dioxide (CO_2), in the context of carbon capture and storage which has been considered as one of the primary options for significantly reducing anthropogenic emissions of greenhouse gases into the atmosphere. The CO_2 streams captured from power plants or other large industrial processes contain a variety of impurities. This study investigates the effects of nitrogen (N_2) on the dissolution trapping mechanism, which occurs when the injected CO_2 mixture dissolves into the formation fluids. The density of the formation water/brine at the two-phase interface would increase in response to the dissolution of CO_2. At favorable conditions, convection would be triggered and could greatly accelerate the dissolution rate of CO_2. This density-driven convection process is beneficial for both storage security and permanence. However, N_2 would lead to a density reduction of the aqueous phase when dissolved into the formation water/brine. The onset of convection would be delayed and the dissolution rate may be affected when co-injecting CO_2 with N_2. In addition, the spatial distribution of CO_2 in the aqueous phase would also be different with varying amounts of N_2 in the CO_2 streams.

1. Introduction

There are growing concerns over the global warming due to the increase of the concentration of greenhouse gases in the atmosphere, especially CO_2. Carbon capture and storage (CCS) has been identified as one of the major methods to reduce CO_2 emissions from industrial processes, especially from large-scale stationary power plants [1–3]. The carbon capture and storage technology is a three-stage process, which involves the capture of CO_2 from major CO_2 point sources, the transport through the pipeline and the long-term sequestration in the geological storage sites [4]. At the present time, there are three main carbon capture technologies applicable to large-scale power plants, i.e., post-combustion, pre-combustion and oxy-fuel combustion [5]. All these three technologies produce CO_2 dominant streams with a certain amount of impurities [6]. It is not technically difficult to provide CO_2 streams with high purity from the fossil-fuel-fired power plants [7]. However, high purity requirements, reducing the number of non-CO_2 species and/or the level of impurities, would increase the costs for CCS including the energy use. The high capture cost is currently one of the major challenges to large-scale CCS deployment. Co-injection of CO_2 with a certain amount of impurities into the geological structures might be a cost-effective way to reduce the total cost of CCS by dramatically lowering the requirements on the capture side [8]. Furthermore, one of the barriers to operate CO_2 injection projects on a large-scale is forming regulations or guidelines regarding the purity of the CO_2 streams [9].

Permitting the existence of different impurities (e.g., SO_x, H_2S, N_2, O_2) in the CO_2 streams, while lowering the capture cost, has been indicated to have undesirable effects on transport and underground sequestration storage. The effects of impurities on the pipeline transport have received some attention, e.g., [10–12], but there are less studies focusing on the effects of impurities on (short- and long-term) storage. Impurities in the CO_2 streams are indicated to influence all types of geological storage mechanisms, including the static trapping, residual-gas trapping, adsorption trapping, dissolution trapping and mineral trapping mechanisms [1,3], and the effects are often negative. Among the various trapping mechanisms, the dissolution trapping occurs when the injected CO_2 mixture dissolves into the existing subsurface fluids. It is suggested that the dissolution of CO_2 leads to an increase in the density of the aqueous phase at the two-phase interface that, depending on salinity, is typically ranging from 1% (for pure water) to 0.1% (for saturated brine) [13]. The density increase leads to a gravitational instability. Coupled with the heterogeneity in the formation

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Nomenclature

c_1	constant for the onset time
c_2	constant for the critical wavelength
D	diffusivity of gases in aqueous phase (m^2/s)
D_{nm}	distance between meshes n and m (m)
g	vector of gravitational acceleration (m/s^2)
k	absolute permeability of the formation (m^2)
K	equilibrium constant
K_H	Henry's constant (MPa)
M	molecular weight
n	normal vector
P	pressure (MPa)
q	Darcy velocity (m/s)
S_n	surface area of connected mesh interface
t	time (s)
t_c	critical onset time
T	temperature (K)
$v_{\max}$	maximum velocity in the vertical direction
V_n	mesh volume (m^3)
V_i	partial molar volume of dissolved gas (m^3/mol)
x	component mole fraction in aqueous phase
X, Z	Cartesian coordinates
y	component mole fraction in gas phase
z	component mole fraction in the total system

Greek symbols

β	mole fraction of gas phase in the system
ϕ	porosity
λ_c	critical wavelength
γ	fugacity coefficient
ρ	molar density (mol/m^3)
ρ_o	mass density (kg/m^3)
μ	dynamic viscosity (Pa s)

Subscripts

aq	aqueous phase
i	component index
n, m	grid block index
nm	harmonic weighting at the interface between meshes n and m
w	water

properties such as the permeability heterogeneity, this instability will give rise to "convective mixing" that transports CO_2-rich aqueous phase downward from the two-phase interface while driving underlying water/brine with lower CO_2 concentration upwards. During this process, a pattern of the CO_2 concentration that looks like fingers will form, and the phenomenon is therefore called fingering [14]. Such density-driven convection could greatly accelerate the dissolution rate of CO_2 into the aqueous phase [15]. The dissolution of CO_2 in the formation water/brine not only reduces the risk of leakage through faults or fractures, but also enhances geochemical reactions of CO_2 with possible minerals in the rock matrix (mineral trapping).

The density-driven convective mixing has been widely studied due to the practical importance for CO_2 storage. Linear stability analysis and numerical simulation are the two frequently used methods. Without limitations of numerical resolution and inaccuracy, linear stability analysis can detect very small instabilities under simplified conditions. Thus, it has been used to determine the onset time of convection and the critical wavelength of the convective instabilities [16–21]. However, linear stability analysis is valid only at the early stages before the occurrence of convective mixing. It cannot be used to evaluate the nonlinear mixing behaviors after the convection takes place [22]. To partially validate the linear theory and further investigate the nonlinear evolution of the instability at larger times, numerical simulation method has been used rather extensively. In addition, numerical simulations can be combined with optimization schemes to guide industrial practices [23]. Optimization and parallel algorithms are also available to improve computation performance, e.g., [24,25]. For CO_2 storage, both the short-term and long-term behaviors of the density-driven convection have been investigated. The existing investigations on the dissolution trapping mechanism are extensive. Some of them are briefly discussed in the following.

It is known that the density-driven convection would not happen in all the potential storage sites. The Rayleigh number is an important parameter that depends both on the characteristics of the porous media and the fluid properties [19] and it compares the convective rate with the diffusive rate. It has been suggested that only if the Rayleigh number of a certain aquifer was greater than the classical critical Rayleigh number, that is, $4\pi^2$, should the convection be expected [26]. For instance, the calculated Rayleigh number of the Glauconitic sandstone aquifer in the Alberta basin was only 14, and simulation results have confirmed that convective mixing would not take place in such an aquifer [27]. Besides, dissolution of CO_2 at high temperature (>120 °C) may lead to a reduction of the density of the formation brine. Thus, formations with higher temperature or geothermal reservoirs were also indicated to suppress the dissolution trapping mechanism [28]. At favorable conditions, density-driven convection would take place and the mass transfer of the injected CO_2 would be enhanced. The effect of the convective mixing was indicated to increase with increasing Rayleigh number [29]. To predict the dissolution rate of the injected CO_2, scaling analysis based on direct numerical simulations of the convective mixing has been used to find simple relationships for the estimation of the onset time of convection, the initial wavelength of the convective instabilities, the degree of mixing and the Sherwood number, which was defined as the ratio of convective to diffusive mass transport [30].

The dissolution rate of CO_2 can be affected by a number of factors. It was indicated that the geochemical reactions would alter the permeability of the formation and thus have an impact on the dissolution rate. Simulation results revealed that the coupling of Ca^{2+} and Mg^{2+} concentrations to formation water density could accelerate the dissolution rate of CO_2 [31]. In addition to the influence on the dissolution of CO_2, the geochemical reactions and mineral compositions also played a part in the dissolution of other species. Both could affect the formation water density. In the case of the mineral alteration and then the permeability variations, the onset of convection predicted would be delayed in simulations without consideration of geochemical reactions [32]. The heterogeneity was demonstrated to be important for the density-driven convective mixing in porous media. Heterogeneous medium with the same effective permeability had a shorter onset time for convection than the homogeneous medium [33]. The dissolution rates increased with increasing heterogeneity of the permeability within a formation. Furthermore, heterogeneity could affect the character of convective mixing. It was indicated that there were three flow regimes, i.e., fingering, dispersive, and channeling, for the density-driven natural convection flow in heterogeneous media [34]. Although some of the simulations mentioned above employed a rather coarse grid resolution, the results did give an insight into