



Two-phase convective mixing under a buoyant plume of CO₂ in deep saline aquifers



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ARTICLE INFO

Article history:

Received 2 September 2014
Received in revised form 25 November 2014
Accepted 25 November 2014
Available online 4 December 2014

Keywords:

Two-phase
Capillary transition zone
Solubility trapping
Saline aquifers
CO₂ sequestration
Numerical simulation

ABSTRACT

The storage of carbon dioxide (CO₂) in deep saline aquifers has been suggested as a promising method for stabilizing the atmospheric concentration of CO₂. An accurate evaluation of the CO₂ trapping mechanisms, such as convective mixing, is crucial for estimates of storage capacity and security. We recently investigated the gravitational stability of the diffusive boundary layer underneath a capillary transition zone by performing a linear stability analysis, which provides a quantitative description of the onset of convection for the two-phase, buoyancy-driven flow in the presence of the capillary transition zone (Emami-Meybodi and Hassanzadeh, 2013). In this paper, we further examine the effect of the capillary transition zone on the onset of convection and subsequent convective mixing using direct numerical simulations. We describe key features of the two-phase convective mixing for systems with low Rayleigh numbers ($Ra \leq 1000$) and the measurement of several global quantities, such as the total CO₂ dissolution, Sherwood number, swelling factor, and interface velocity. We show that the commonly used assumption of a sharp CO₂–brine interface with constant CO₂ concentration at the top of an aquifer (i.e. single-phase system) may lead to erroneous estimates of not only the onset of convection, but also of the rate and magnitude of CO₂ dissolution. The significant effect of the capillary transition zone on the dissolution of CO₂ under a buoyant plume in saline aquifers is explained; and, the link between the capillary transition zone and the volume change, due to CO₂ dissolution and the interface velocity over the mixing process, is demonstrated. Compared to the single-phase system, a crossflow through the interface of the diffusive boundary layer with the capillary transition zone, as well as the upward advance of the interface motion, may enhance the convective mixing early in the period of natural convection. The decrease in the onset time and stronger mass flux may be more profound in the two-phase system than in the previously reported single-phase models. Furthermore, we report several scaling relationships that characterize the mixing process in the presence of the capillary transition zone. Our findings provide further insight into the understanding of the two-phase mixing features and the long-term fate of the injected CO₂ in deep saline aquifers.

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1. Introduction

Natural convection in porous media plays a key role in a wide range of engineering and earth science applications. Environmental concerns have led to an increased interest in the natural convection process in subsurface formations, such as deep saline aquifers. Natural convection may influence a variety of geological systems, including transport of contaminants or disposed wastes [2–4] and dissolution of halites or other soluble minerals in groundwater [5–7].

Recently, there has been increasing interest in the subject of solutal natural convection in porous media with respect to the application of long-term geological storage of carbon dioxide (CO₂) [8–12]. With the rising global energy demand, CO₂ storage in geological formations, particularly in deep saline aquifers, may help reduce anthropogenic emissions of greenhouse gases [13,14], leading to more sustainable use of fossil fuels.

In the carbon capture and storage (CCS) process, CO₂ is captured at emission sources, compressed and injected into deep porous rocks located under an impermeable sealing rock at least 1 km underground [15]. In siliciclastic aquifers, brine and supercritical CO₂ are typically the wetting and the non-wetting phases, respectively. Under subsurface conditions, supercritical CO₂ is buoyant relative to the formation brine; and, driven by buoyancy, CO₂ rises through the aquifer and spreads under a sealing rock. A portion of

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this buoyant CO₂ plume may flow upward and leak through any imperfections in the geological seal (e.g. fractures) or artificial penetrations (e.g. abandoned wells), which is of great concern in the security of long-term storage [16,17].

The dissolution of injected CO₂ into formation brine is one of the primary mechanisms in the stability of long-term geological CO₂ storage. The process of gradual CO₂ dissolution creates a dense boundary layer of CO₂-rich brine (i.e. a diffusive boundary layer) that can lead to natural convection. The process of convective dissolution of CO₂ provides a very significant mechanism for the transport of CO₂ by enhancing its mass transfer and accelerating its mixing through saline aquifers [18]. Once CO₂ is dissolved in the formation brines, the risk of leakage may be safely eliminated. Convective dissolution reduces the time required for the total dissolution of the CO₂ plume and controls the long-term risk of CO₂ leakage from the geological formation and increases the security of storage [19].

Convective mixing in the geological storage of CO₂ has been studied for over two decades in theoretical settings [8,20–28], laboratory experiments [9,29] and field-scale studies [18,30]. In most studies, the two-phase capillary transition zone between the injected CO₂ and the formation brine has been ignored, and a sharp CO₂-brine interface has been assumed at a constant CO₂ equilibrium concentration on the top of the aquifer. In our previous studies [31,32], we developed mathematical models to study single-phase convective mixing induced by steady and transient buoyancy-driven flows. Our models quantify the mixing process and predict mixing of a solute (a diffusive species such as CO₂) for both steady and transient single-phase convective systems.

Earlier studies on two-dimensional (2D) single-phase convective systems have focussed on examining the dynamics of the system and the solute flux into the system as described by the Sherwood number, both of which are functions of the Rayleigh number (Ra). The Rayleigh number is a measure of the buoyancy-driven flows as the ratio of diffusive and convective time-scales. Natural convection depends on the fluid and rock physical properties. In steady convective systems, for $Ra < 1300$, the flow forms convection rolls that undergo several mixing regimes. Over this range of Rayleigh numbers, although the dynamics of the flow varies from low to higher Rayleigh numbers, the convection rolls do not completely break down [29,33,34]. On the other hand, at higher Ra , the convective rolls are broken down due to the vigorous nature of the flow, forming vertical columnar mega-plumes [35].

There have been a number of numerical and experimental studies for aquifers of finite thickness that examine the dynamics and evolution of convective flows before density-driven fingers reach the bottom of the domain. On the theoretical side, we observed the behavior of transient convective flow for $Ra < 1000$ and quantified the corresponding mixing [26,31]. Theoretical studies by Hewitt et al. [35] for $1300 < Ra < 40000$ and Hidalgo et al. [24] for $5000 < Ra < 30000$ provided behavior of the convective mixing and dynamics of flow after the onset of convection. Recently, Slim [36] characterized the temporal regimes for solutal convection from almost first contact to high dissolved solute concentration in a 2D ideal porous medium for $100 < Ra < 50000$. Elenius and Johannsen [37] analyzed the timescales associated with unstable fingering induced by the density contrasts in miscible displacement in porous media by performing numerical simulations. Numerical simulations by Pau et al. [38] demonstrated the effects of convective instability on geologic sequestration of CO₂.

In experimental studies, Kneafsey and Pruess [39] performed experiments in a Hele-Shaw cell and quasi 2D porous medium for $40 < Ra < 370$. Slim et al. [29] presented results for $100 < Ra < 1700$ in a Hele-Shaw cell and categorized the evolution of the convective flow. Neufeld et al. [9] provided the

measurement of the convective flux for $50,000 < Ra < 600,000$ in a quasi 2D porous medium; and, Backhaus et al. [40] investigated the flow behavior for $6000 < Ra < 90,000$ in a Hele-Shaw cell. Although some deep saline aquifers generate strong convective systems with high Rayleigh numbers, e.g. the Sleipner field in the North Sea [41], many of them are categorized otherwise, such as most of the deep saline aquifers in Western Canada [42]. In the present study, we analyze two-phase convective systems with low Rayleigh numbers, $Ra \leq 1000$.

Despite its significance, relatively little is known about the highly nonlinear dynamics of the two-phase natural convection systems in porous media with application to CO₂ geosequestration. Many studies of natural convective flows have assumed a sharp interface between the CO₂ and the brine before the evolution of the diffusive boundary layer. This assumption decouples the buoyant plume of CO₂ from the underlying formation brine and instead imposes a constant concentration and impermeable boundary conditions for the flow and transport on top of the single-phase domain, respectively. In the dissolution of CO₂ in deep saline aquifers, there is the additional complication in the formation of a two-phase region, where a capillary transition zone is formed between the free-phase CO₂ and the underlying brine.

Most studies on the convective dissolution of CO₂ in saline aquifers have considered a single-phase system by assuming a sharp CO₂-brine interface at a constant CO₂ equilibrium concentration on top of the aquifer. This assumption not only neglects the presence of the capillary transition zone, but also the correct CO₂ mass influx into the underlying brine. Furthermore, this assumption ignores the crossflow of the formation brine across the interface and swelling of water upon CO₂ dissolution.

Hidalgo et al. [43] showed that neglecting the volume change due to solute mass influx leads to erroneous solutions, especially with Dirichlet boundaries for transport and impervious boundaries for flow. As a result of the volume change due to CO₂ dissolution, the interface between the CO₂ phase and the brine moves upward with time. The maximum possible volume change corresponds to the time when the brine is fully saturated with CO₂ at the equilibrium concentration. This swelling maybe significant at certain conditions, based on the subsurface pressure and temperature, and can reach as high as 1.07 of the original volume [44].

A few recent studies have independently examined the effects of a capillary transition zone and the interface movement on the onset of buoyancy-driven convection, mostly based on linear stability analyses [1,45–48]. Meulenbroek et al. [47] found that the advance of the interface can lead to a more than tenfold decrease in the onset of convection, whereas Myint and Firoozabadi [46] reported a maximum 10% reduction in the onset time of convection. Elenius et al. [45] considered a crossflow between the capillary transition zone and the diffusive boundary layer and by performing LSA showed that the onset of instabilities can occur up to five times faster in the presence of a capillary transition zone. In another study, they showed that the capillary transition zone may reduce the onset time of instability by a factor of two and may enhance the rate of dissolution up to twice [49].

In a previous work [1], we presented a quantitative description of the onset of convection in the presence of this transition zone. More precisely, we studied the role of the capillary transition zone solely on the onset of the instability of the diffusive boundary layer using LSA. However, the LSA only determines the onset of the marginal instability and cannot be used to evaluate the behavior CO₂ dissolution beyond the onset of convection.

In the present study, we investigated the effect of a capillary transition zone on the onset of convection and the subsequent mixing process and dissolution timescales using high-resolution direct numerical simulations (DNS). We also analyzed how the

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