

Dynamics of convective dissolution from a migrating current of carbon dioxide



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ABSTRACT

During geologic storage of carbon dioxide (CO₂), trapping of the buoyant CO₂ after injection is essential in order to minimize the risk of leakage into shallower formations through a fracture or abandoned well. Models for the subsurface behavior of the CO₂ are useful for the design, implementation, and long-term monitoring of injection sites, but traditional reservoir-simulation tools are currently unable to resolve the impact of small-scale trapping processes on fluid flow at the scale of a geologic basin. Here, we study the impact of solubility trapping from convective dissolution on the up-dip migration of a buoyant gravity current in a sloping aquifer. To do so, we conduct high-resolution numerical simulations of the gravity current that forms from a pair of miscible analogue fluids. Our simulations fully resolve the dense, sinking fingers that drive the convective dissolution process. We analyze the dynamics of the dissolution flux along the moving CO₂-brine interface, including its decay as dissolved buoyant fluid accumulates beneath the buoyant current. We show that the dynamics of the dissolution flux and the macroscopic features of the migrating current can be captured with an upscaled sharp-interface model.

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1. Introduction

The injection of carbon dioxide (CO₂) into deep saline aquifers is a promising tool for reducing anthropogenic CO₂ emissions [1–4]. After injection, the buoyant CO₂ will spread and migrate laterally as a gravity current relative to the denser ambient brine, increasing the risk of leakage into shallower formations through fractures, outcrops, or abandoned wells.

One mechanism that acts to arrest and securely trap the migrating CO₂ is dissolution of CO₂ into the brine [5]. Dissolved CO₂ is considered trapped because brine with dissolved CO₂ is denser than the ambient brine, and sinks to the bottom of the aquifer. In addition to providing storage security by hindering the return of the CO₂ to the atmosphere, this sinking fluid triggers a hydrodynamic fingering instability that drives convection in the brine and greatly enhances the rate of CO₂ dissolution [6–9].

Although this process of convective dissolution is expected to play a major role in limiting CO₂ migration and accelerating CO₂ trapping [4], the interaction of convective dissolution with a migrating gravity current remains poorly understood. This is due primarily to the disparity in scales between the long, thin gravity current and the details of the fingering instability. Resolving these

simultaneously has proven challenging for traditional reservoir simulation tools [10]. Upscaled theoretical models [11,12] and laboratory experiments [13,14] have recently provided some macroscopic insights, but by design these capture only the averaged dynamics of the dissolution process.

Here, we study the impact of convective dissolution on the migration of a buoyant gravity current in a sloping aquifer by conducting high-resolution numerical simulations of a pair of miscible analogue fluids. Our simulations fully resolve the small-scale features of the convective dissolution process. We define an average dissolution flux and use it to study the dynamic interactions of the fingering instability with the migrating current. We then compare these results with the predictions of an upscaled theoretical model to investigate the degree to which this simple model can capture the macroscopic features of the migrating current.

2. Analogue fluids

For simplicity, and to focus on the role of convective dissolution, we neglect capillarity and assume that the two fluids are perfectly miscible. We adopt constitutive laws for density and viscosity that are inspired by a pair of miscible analogue fluids that have been used to study this problem experimentally [15,16,13,14]. This system captures three key features of the CO₂-brine system: (1) a density contrast that stratifies the pure fluids and drives the migration

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of the gravity current, (2) an intermediate density maximum that triggers and drives convective dissolution (discussed below), and (3) a viscosity contrast between the pure fluids that influences the shape and propagation speed of the gravity current.

We write the dimensionless density ρ and viscosity μ as functions of the local concentration c of the buoyant fluid. We scale the concentration c by the solubility so that $c \in [0, 1]$. Since the analogue fluids have different densities ($\rho(c=1) < \rho(c=0)$), the buoyant one will “float” and migrate above the denser one. Since they are perfectly miscible, they will be separated by a transition zone that forms and grows through diffusion, and within which the local concentration transitions from $c=0$ to $c=1$ and the local density and viscosity vary accordingly.

To trigger convective dissolution, the essential feature of the density law is that it must be a non-monotonic function of concentration with an intermediate maximum (Fig. 1). This shape introduces a neutral concentration $c=c_n$ for which the density of the mixture is equal to the density of the ambient fluid. Fluid with concentration $c > c_n$ (i.e., to the right of c_n) is less dense than the ambient and tends to float, whereas fluid with concentration $c < c_n$ (i.e., to the left of c_n) is denser than the ambient and tends to sink. The contour of neutral concentration within the transition zone therefore emerges as a natural “interface” between buoyant and sinking fluids: the fluid above is buoyant and stably stratified (density decreasing as concentration increases from $c=c_n$ to $c=1$), the fluid below is dense and unstably stratified (density decreasing as concentration decreases from $c=c_n$ to $c=0$), and diffusion continuously transfers fluid from the stable region to the unstable region.

The concentration $c=c_m$ at which the density maximum occurs plays the role of a solubility in this system since the density of the underlying fluid increases toward this value as dissolved buoyant fluid accumulates. Convective dissolution stops entirely when diffusion at the interface is no longer able to generate a mixture that is denser than the fluid below it.

To make the density law dimensionless, we shift it by the brine density and scale it by the height of the density maximum so that the dimensionless brine density is always $\rho(c=0)=0$ and the dimensionless density maximum is always $\rho(c=c_m)=1$. We represent the density law with a polynomial of degree three, $\rho(c)=6.19c^3-17.86c^2+8.07c$, which has neutral concentration $c_n=0.56$, a density maximum at $c_m=0.26$, and a dimensionless CO₂ density of $\rho(c=1)=-3.6$. This density law is qualitatively

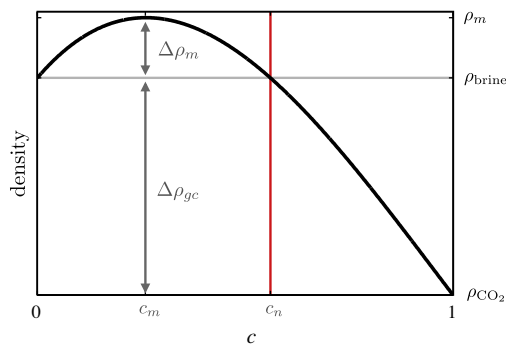


Fig. 1. Non-monotonic density law (dimensional) inspired by miscible analogue fluids [15,16]. The density has a maximum at $c=c_m$. The contour of neutral concentration $c=c_n$ (red line) acts as an interface: mixtures with $c < c_n$ (left of the red line) are denser than the ambient brine and will sink, whereas those with $c > c_n$ (right of the red line) are buoyant relative to the ambient brine and will rise. $\Delta\rho_m$ is the characteristic density difference that drives convective dissolution and $\Delta\rho_{gc}$ is the one that drives the migration of the buoyant gravity current. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

and quantitatively similar to the true density law for mixtures of propylene glycol ($c=0$, brine analogue) and water ($c=1$, CO₂ analogue) [16].

We choose an exponential constitutive law for the dimensionless viscosity, $\mu(c)=\exp[R(c_m-c)]$, where we have scaled $\mu(c)$ by characteristic viscosity μ_m so that $\mu(c=c_m=0.26)=1$. The parameter $R=\ln \mathcal{M}$, where $\mathcal{M}=\mu_{\text{brine}}/\mu_{\text{CO}_2}=\mu(c=0)/\mu(c=1)$ is the mobility ratio. This viscosity law is qualitatively and quantitatively similar to the true viscosity law for mixtures of propylene glycol and water for $R \approx 3.7$ [16].

Since these analogue fluids are perfectly miscible, our results do not incorporate the various impacts of capillarity, including residual trapping, the development of a capillary fringe, and capillary pressure hysteresis. The absence of capillarity is a limitation in the sense that these analogue fluids cannot capture every aspect of the CO₂-brine system, but it is also an advantage in the sense that it allows us to isolate and study convective dissolution as a transport process without these additional complications [15,16,13,14].

Capillarity may impact the dynamics of the gravity current. For example, the gravity current will shrink due to residual trapping along its trailing edge [17–19]. The formation of a capillary fringe between the CO₂ and the brine may change the shape and reduce the propagation speed of the gravity current [20–22]. Capillary pressure hysteresis may also reduce the propagation speed of the gravity current and even arrest its migration [23,24]. All of these effects can be incorporated into upscaled models for CO₂ migration, but incorporating them into our 2D simulations is less straightforward. These effects would impact the total dissolution rate by changing the length of the “interface” between the two fluids, and by reducing the amount of ambient fluid available for “storing” dissolved CO₂. However, we would not expect them to change the dynamic interactions of migration and dissolution as described here.

Capillarity may also have a quantitative impact on the onset and subsequent rate of convective dissolution [25–27]. These effects have never been studied experimentally and are not well understood, but we expect the same qualitative behavior of the dissolution flux (diffusion, onset, convection). Although miscible analogue fluid systems may feature quantitatively different fluxes, they are useful for studying the dynamics of the dissolution flux and its impact on migration.

3. Mathematical model

We consider a two-dimensional aquifer in the x - z plane, with dimensional length L_x and uniform dimensional thickness L_z . The aquifer is tilted by an angle θ relative to horizontal. This can be viewed as a cross-section of a sedimentary basin taken perpendicular to a line-drive array of injection wells [28,4]. We assume that the aquifer is homogeneous and with isotropic permeability.

We use the classical model for incompressible fluid flow and advective–dispersive mass transport under the Boussinesq approximation, modeling hydrodynamic dispersion as a Fickian process with a velocity-independent diffusion–dispersion coefficient. The governing equations for this model in dimensionless form are [29]

$$\nabla \cdot \mathbf{u} = 0, \quad (1)$$

$$\mathbf{u} = -\frac{1}{\mu(c)} (\nabla p - \rho(c) \hat{\mathbf{e}}_g), \quad (2)$$

$$\frac{\partial c}{\partial t} = -\mathbf{u} \cdot \nabla c + \frac{1}{\text{Ra}} \nabla^2 c, \quad (3)$$

where p is the scaled pressure deviation from a hydrostatic datum, \mathbf{u} is the scaled Darcy velocity, and $\hat{\mathbf{e}}_g = (-\sin \theta, -\cos \theta)$ is the unit vector in the direction of gravity. $\rho(c)$ and $\mu(c)$ are the dimensionless

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