Interactions between gravity currents and convective dissolution

M.T. Elenius [a,*], D.V. Voskov [b], H.A. Tchelepi [b]

[a] *Department of Civil and Environmental Engineering, Tufts University, 200 College Ave, Medford, MA 02155, USA*
[b] *Department of Energy Resources Engineering, Stanford University, 397 Panama Mall, Mitchell Building 101, Stanford, CA 94305-2210, USA*

ARTICLE INFO

ABSTRACT

Geological storage of carbon dioxide (CO_2) is a promising technology for reducing atmospheric emissions. The large discrepancy in the time- and length-scales between up-dip migration of buoyant supercritical CO_2 and the sinking fingers of dissolved CO_2 poses a challenge for numerical simulations aimed at describing the fate of the plume. Hence, several investigators have suggested methods to simplify the problem, but to date there has been no reference solution with which these simplified models can be compared. We investigate the full problem of Darcy-based two-phase flow with gravity-current propagation and miscible convective mixing, using high-resolution numerical simulations. We build on recent developments of the Automatic Differentiation - General Purpose Research Simulator (AD-GPRS) at Stanford. The results show a CO_2 plume that travels for 5000 years reaching a final distance of 14 km up-dip from the injection site. It takes another 2000 years before the CO_2 is completely trapped as residual (40%) and dissolved (60%) CO_2. Dissolution causes a significant reduction of the plume speed. While fingers of dissolved CO_2 appear under the propagating gravity current, the resident brine does not become fully saturated with CO_2 anywhere under the plume. The overall mass transfer of CO_2 into the brine under the plume remains practically constant for several thousands of years. These results can be used as a benchmark for verification, or improvements, of simplified (reduced-dimensionality, upscaled) models. Our results indicate that simplified models need to account for: (i) reduced dissolution due to interaction with the plume, and (ii) gradual reduction of the local dissolution rate after the fingers begin to interact with the bottom of the aquifer.

1. Introduction

Separation of carbon dioxide (CO_2) from other gases at large point sources, followed by compression and injection into deep geological formations, has been suggested as one viable strategy to reduce atmospheric CO_2 emissions [20,30]. Valuable experience in this technology has been gained from injection for enhanced oil recovery since the early 1970s [27], and from injection for the purpose of storage since 1996 [1]. The injected CO_2 is immiscible with and less dense than the resident brine (saline water) in the subsurface. Trapping in the formation occurs as structural trapping in the form of an impermeable caprock, residual trapping due to capillary imbibition, and dissolution. When CO_2 dissolves in the brine, the brine density increases by up to 1% [10] and thereby, the risk of upward leakage of CO_2 is reduced once it is dissolved. This small density increase can cause gravitational instability with fingers of denser brine migrating downward away from the overlying CO_2 plume [23]. The downward fingers are replaced by the upward migration of undersaturated brine. In this way, the dissolution rate is greatly enhanced, compared with purely diffusive transport of the dissolved CO_2. Even with the help of convective mixing, it may take thousands of years before the plume is completely trapped; therefore, it is important to have accurate estimates of the plume migration dynamics and trapping behaviors for long periods after injection stops. In this paper, we deal with the spreading and trapping of a CO_2 plume during the long post-injection period.

The physics of the problem involves length scales ranging from meter-wide fingers to kilometer-wide plumes of supercritical CO_2, and time scales ranging from months of convection onset times to thousands of years of plume propagation. This poses challenges for direct numerical simulations. Different strategies have been applied to cope with these difficulties. Solutions to the full set of equations using relatively coarse grids can give insight into some physical features, although quantitative results remain uncertain [8,32]. At the other end of the spectrum, analytical models can fully capture a reduced-physics problem [17,21,26] (e.g., no dissolution), but it is difficult to assess the predictive capabilities of such models. Previous numerical simulations [8] as well as results from semi-analytical methods [14,25] have indicated that dissolution with convective mixing can have a first-order impact on plume behavior, and

* Corresponding author. Tel.: +16176273513.
  E-mail address: maria.elenius@tufts.edu (M.T. Elenius).

that existing analytical models are unable to capture the effect of dissolution on large-scale migration. There is a large amount of experimental [22,40], as well as, analytical and numerical [4,11,31,33–35] work related to convective mixing in small domains (10–100 m) without plume migration. The results from these small-scale investigations have been used as input to semi-analytical models that solve for the plume migration and treat dissolution as a sink term [14,25]. Another interesting simplification that reduces the CPU time of numerical simulations is the use of a pair of miscible analog fluids that can reproduce some features of a CO_2 gravity current with convective mixing [19]. While this method looks promising, accurate modeling of the detailed evolution of CO_2 plume migration and trapping is still needed.

This paper presents solutions to the problem of plume migration in a confined sloping aquifer, accounting for residual and dissolution trapping including convective mixing. The main focus is on dissolution trapping, which is a difficult task for direct numerical simulation of the full problem. Here, we improve on previous two-phase simulations by using much higher spatial resolutions, and on previous high-resolution miscible simulations by including two-phase flow effects (capillary pressure and relative permeability). We restrict the modeling to two spatial dimensions, so that we can sufficiently resolve the length and time scales associated with the unstable flow. Line symmetry is therefore assumed in the direction perpendicular to the aquifer slope. This could represent an array of injection wells in that direction, cf. [25]. For convective mixing, Pau et al. [31] showed that the dissolution rate in 3D is 25% larger than in 2D, which is a rather small difference compared to the difference between cases studied here. We do not account for mineral trapping. We are taking advantage of recent developments in the Automatic Differentiation-General Purpose Research Simulator (AD-GPRS) [37]. In addition to modeling the migration of large CO_2 plumes, three small-scale problems are investigated, and the small-scale results are compared with the large-scale simulation results. Our work highlights important processes and challenges that upscaled models must account for and provides a benchmark for simplified models.

In the following, we describe the small- and large-scale model problems (Section 2) and the numerical treatment (Section 3), followed by the results (Section 4) and conclusions (Section 5).

2. Model problem

2.1. Governing equations

The governing equations used to describe two-phase flow and transport are

$$\partial_t \sum_\alpha \phi S_\alpha \rho_\alpha X_\alpha^C + \nabla \cdot \sum_\alpha (\rho_\alpha X_\alpha^C \mathbf{u}_\alpha + S_\alpha \rho_\alpha \mathbf{J}_\alpha^C) = 0, \tag{1}$$

$$\mathbf{u}_\alpha = -\frac{k_{r\alpha} k}{\mu_\alpha} (\nabla p_\alpha - \rho_\alpha \mathbf{g}), \quad \mathbf{J}_\alpha^C = -\phi D_\alpha^C \nabla X_\alpha^C, \tag{2}$$

where subscript $\alpha \in \{w, n\}$ denotes the wetting phase (brine) and the nonwetting phase (supercritical CO_2), superscript $C \in \{CO_2, H_2O\}$ denotes a component, ϕ porosity, S saturation, ρ density, X mass fraction, $\mathbf{u}$ Darcy velocity, $\mathbf{J}$ Fick's diffusion flux, k_r relative permeability, k absolute permeability, μ viscosity, p pressure and $\mathbf{g}$ is the vector of gravitational acceleration.

The following assumptions are made in order to obtain a well-defined benchmark problem. The absolute permeability is homogeneous and isotropic, and the porosity and viscosities are constant.

The notation is simplified such that $X = X_w^{CO_2}$. We use a constant solubility $X_{max} = 0.03$ kg CO_2/kg brine [3], corresponding to $x_{max} = 0.0125$ mol CO_2/mol brine. We also assumed that a small amount of water ($Y_{max} = 1.0 \cdot 10^{-4}$ kg H_2O/kg CO_2, corresponding to $y_{max} = 2.44 \cdot 10^{-4}$ mol H_2O/mol CO_2 phase) can vaporize into the

CO_2 phase, and because of this, the CO_2 phase always contains some water. This solubility was modeled based on constant K-values for the {CO_2–H_2O} system. The assumptions yield the following K-values that describe the thermodynamic equilibrium

$$K_{CO_2} = \frac{1 - y_{max}}{x_{max}} = 80.0, \tag{3}$$

$$K_{H_2O} = \frac{y_{max}}{1 - x_{max}} = 2.47 \cdot 10^{-4}. \tag{4}$$

In addition, the brine density is assumed to increase linearly with the mass fraction of dissolved CO_2, that is

$$\rho_w = \rho_w^{X=0} + \Delta \rho_w X / X_{max}. \tag{5}$$

This linear dependence on CO_2 content was observed in experiments related to CO_2 storage [40]. The brine has low compressibility, which supports the assumption of constant brine density with respect to pressure and temperature. We use a constant CO_2 phase density, apart from including a very small compressibility C_n for numerical issues. The assumption of constant CO_2 phase density has been shown to be reasonable because the pressure- and temperature effects may offset each other [32]. The CO_2 density is then

$$\rho_n = \rho_n^{p_{atm}} (1 + C_n (p_n - p^{atm})). \tag{6}$$

The phase pressures are related by the capillary-pressure $p_c = p_n - p_w$ and the saturations by $S_n + S_w = 1$. The following Brooks–Corey functions relate capillary pressure and relative permeability to the effective brine saturation, S_e

$$S_e = (S_w - S_{wr})/(1 - S_{wr}), \tag{7}$$

$$k_{rw} = S_e^4, \tag{8}$$

$$k_{rn} = 0.4(1 - S_e^2)(1 - S_e)^2 - C, \tag{9}$$

$$p_c = p_e / \sqrt{S_e}. \tag{10}$$

We take the entry pressure to be $p_e = 20$ kPa. These relationships were used in a previous benchmark proposed by Dahle et al. [2] (with $C = 0$ in Eq. (9)). We do not model hysteresis. Instead, using $C = 0.0109$ leads to $k_{rn} = 0$ at $S_n = S_{nr}$, and we use $k_{rn} = 0$ for smaller values of S_n, see Fig. 1. In this way, residual trapping is modeled, although only drainage functions are used. For calculations of p_c (only), $S_e < 10^{-3}$ is replaced with $S_e = 10^{-3}$ to avoid pressures tending to infinity. Because the phase densities differ and the capillary pressure is related to the saturation, a so-called capillary transition zone (CTZ) develops in the plume, where the CO_2-phase saturation in the plume increases with height above the single-phase brine region.

2.2. Description of test cases

Three small-scale problems and one large-scale problem are investigated. The corresponding domains with initial- and boundary conditions are shown in Fig. 2. The small-scale problems (100 m × 50 m) are used mainly for comparison with the large-scale sloping aquifer simulations (20 km × 50 m). The large-scale problem is inspired by the benchmark proposed by Dahle et al. [2] as well as previous results from different groups summarized and discussed in [29]. Here, the diffusivity (applied for diffusion of both components in both phases) was increased from $1 \cdot 10^{-9}$ to $2 \cdot 10^{-9}$ m^2/s, which is still realistic but facilitates computations by increasing the wavelength of instabilities, cf. Appendix B. Some parameters that were not defined in the benchmark are chosen here according to Nordbotten and Dahle [28], who investigated the migration problem using a vertically integrated model. Parameter values of the formation and fluids that are common to all problems (a–d) are provided in Table 1. The boundary and initial conditions are described below.