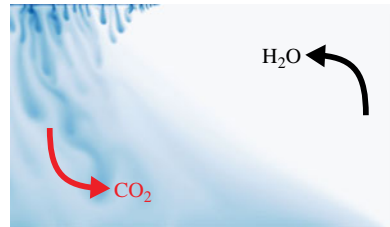# The fluid mechanics of dissolution trapping in geologic storage of CO$_2$

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Sequestration of carbon dioxide by injecting it into the deep subsurface is critical
to successful mitigation of climate change by reducing anthropogenic emissions of
greenhouse gases into the atmosphere. To achieve this we must understand how
CO$_2$ moves in the subsurface. Many interesting fluid mechanics problems emerge.
Szulczewski, Hesse & Juanes (*J. Fluid Mech.*, vol. 736, 2013, pp. 287–315) focus
on one critical aspect, namely the dissolution of CO$_2$ into the fluid resident in the
subsurface and the flow dynamics that ensue. Even for this single problem, an elegant
analysis identifies seven behavioural regimes that control the amount and timing of
dissolution.

Key words: convection in porous media, gravity currents

1. Introduction

Carbon capture and geological storage (CCS) is the process by which supercritical
carbon dioxide (CO$_2$) is injected into deep subsurface formations for long-term storage.
It is regarded as critical in viable solutions to mitigation of climate change by reducing
emissions of greenhouse gases into the atmosphere (IPCC 2005).

Depending on one's perspective, CCS is a fluid mechanician's dream or nightmare
encompassing a diverse set of complex physical phenomena. First, the flow whereby
the CO$_2$ displaces the resident fluid is a multiphase problem, which includes the
resident fluid phase, the supercritical CO$_2$ phase and the solid porous medium phase.
This results in important dynamics associated with viscosity contrasts (Nordbotten &
Celia 2006) and capillary pressures (Golding, Huppert & Neufeld 2013). The density
of supercritical CO$_2$ is less than that of the resident fluid; thus buoyancy effects
are important, with the CO$_2$ travelling as a gravity current above the resident fluid
(Dentz & Tartakovsky 2009). Further, CO$_2$ dissolves into the fluid phase (Kneafsey

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& Pruess 2010). The resulting mixture has a higher density than the resident fluid and convective instabilities can occur, with fingers of CO_2-rich fluid falling into the underlying resident fluid (Riaz *et al.* 2006). This fingering can affect the speed of the propagating boundary current (MacMinn & Juanes 2013). Dissolution of CO_2 is beneficial, since the CO_2, once dissolved, remains permanently trapped without risk of leaking into overlying geologic formations or to the atmosphere (Gilfillan *et al.* 2009). Thus, an important question is when and how quickly will dissolution occur? The paper by Szulczewski, Hesse & Juanes (2013) focuses on this problem. They elegantly consider the processes of dissolution, diffusion, buoyancy-induced instabilities and gravity currents, and identify seven distinct regimes, which can be concisely classified in a phase space.

2. Overview

Stratigraphic traps are regions of porous media with a concave-down structure and a low-permeability seal. The seal hinders upward migration of CO_2, causing it to pool beneath the trap. CO_2 can dissolve into the underlying fluid. Szulczewski *et al.* (2013) develop a simplified conceptualization of a stratigraphic trap by simulating a semi-infinite rectangular domain and imposing a constant concentration CO_2 boundary along part of the upper boundary, representing the contact between CO_2 caught in the stratigraphic trap and the resident fluid. All other boundaries are no-flux boundaries. They consider a system where initially there is no flow and no dissolved CO_2. Flow is incompressible and governed by Darcy's law with buoyancy effects incorporated; transport of dissolved CO_2 is governed by an advection–diffusion equation. For this setup all assumptions for these equations seem reasonable. The idealized representation of a stratigraphic trap is sufficiently detailed to represent the pertinent complexities in such a system, but sufficiently simple to allow a comprehensive theoretical analysis of the emerging flow behaviours.

At early times a thin dissolution layer forms below the constant concentration boundary with no flow in the domain (figure 1). During this first *early diffusion* regime, the flux of CO_2 into the domain can be estimated from a one-dimensional diffusion analysis and scales as $t^{-1/2}$, with t representing time. Since water with dissolved CO_2 is denser than the underlying fluid, the system is unstable. For sufficient Rayleigh number a convective instability with fingers will occur. This *fingering* behaviour is the second regime. There is an increased flux of CO_2 into the system, which attains a constant value proportional to the characteristic vertical buoyancy velocity.

The third regime, *shutdown/fingering*, begins when fingers interact with the bottom boundary. This causes recirculation of CO_2-rich fluid towards the source. The authors divide the domain into three regions: an inner region near the source (left boundary), an outer region far from this and a middle region. In the outer region fingering continues. In the inner region convective shutdown due to interaction with the bottom boundary slows dissolution. In the middle region horizontal advection and fingering occurs. Szulczewski *et al.* (2013) estimate the flux into the domain by averaging over the three regions. There is a slowdown of CO_2 flux into the domain.

The fourth regime is the *shutdown/slumping* regime. It can be understood again by breaking the domain into inner and outer regions. In the inner region convective shutdown continues to slow dissolution. In the outer region fingering continues, but the extent of the gravity current is larger than the thickness of the layer. Since the outer region has higher concentrations of CO_2 the flux carried into the outer
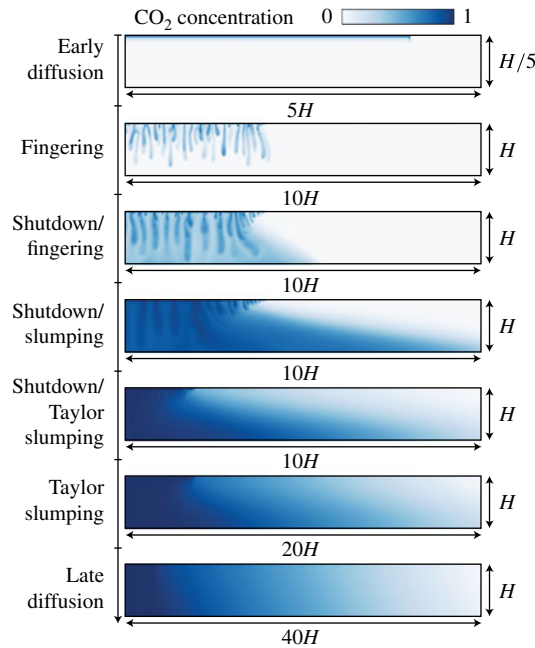
FIGURE 1. Regimes of CO_2 dissolution; modified from Szulczewski *et al.* (2013).

region decreases, slowing the CO_2 flux further. Similarly there is an inner and outer region for the fifth *shutdown/Taylor slumping* regime. Again the inner region displays convective shutdown, but there is a difference with the gravity current. A broad transition region occurs between the forward and backward currents. Diffusive mixing slows propagation. This results in a faster deceleration of the dissolution flux as the return current can no longer provide unsaturated water as quickly.

After the characteristic diffusion time across the aquifer thickness, dissolution enters the *Taylor slumping* regime. The inner region is almost saturated with CO_2 and convection nearly stops. At the edge dissolution continues due to inflow of water from the return current. In the seventh and final *late diffusion* regime, the current is so long that horizontal density gradients driving flow are small. Convective effects are negligible and diffusion dominates. The dissolution flux returns to a scaling of $t^{-1/2}$ as in regime 1, but with a different constant of proportionality.

In summary, by breaking the problem of CO_2 dissolution into seven behavioural regimes and segmenting the domain into inner and outer regions, Szulczewski *et al.* (2013) show that this complex problem can be tackled in a systematic manner. They conclude and show the utility of this methodology by applying the regime models to representative geologic systems, demonstrating that specific reservoir setups and properties control CO_2 dissolution fluxes. By integrating dissolution models into decision-making processes, stakeholders will ultimately better identify and select optimal sites for carbon sequestration.

3. Future

Given the diverse regimes that arise during dissolution of CO_2, interesting questions emerge relating to mixing between the fluid rich in CO_2 and the resident fluid. Since

the flux of CO_2 controls the amount of mixing that can occur, understanding how this impacts actual mixing is important. Hidalgo *et al.* (2012) considered mixing during the fingering regime to show that, at sufficiently high Rayleigh number, the rate of mixing is constant and independent of Rayleigh number. Future work should consider mixing in the other regimes of flow. A reason why mixing is important is that the chemical makeup of CO_2-rich and CO_2-poor water is different. The mixtures can be highly reactive with the permeable medium, resulting in interesting feedbacks between flow and reactions.

One assumption made by Szulczewski *et al.* (2013) is that transport of dissolved CO_2 is with a constant diffusion coefficient. Although a reasonable starting assumption that facilitates analysis, dispersion in porous media is rarely constant: it depends on local flow speeds and can be highly anisotropic. Hidalgo & Carrera (2009) showed that time scales associated with fingering can be accelerated by hydrodynamic dispersion. Similar effects may play a role in emerging flows. Additionally, transverse dispersion is a dominant driver of mixing. Thus anisotropy could play a role in emerging phenomena. Such effects are unlikely to affect the qualitative nature of emerging flows, but could influence when and how they occur. Other effects worthy of study, not incorporated in Szulczewski *et al.* (2013), might include a radial geometry, sloping boundary in the aquifer and a natural background flow in the aquifer, which could alter the emerging regimes.

References

DENTZ, M. & TARTAKOVSKY, D. M. 2009 Abrupt-interface solution for carbon dioxide injection into porous media. *Trans. Porous Med.* **79** (1), 15–27.

GILFILLAN, S. M. V., LOLLAR, B. S., HOLLAND, G., BLAGBURN, D., STEVENS, S., SCHOELL, M., CASSIDY, M., DING, Z., ZHOU, Z., LACRAMPE-COULOUME, G. & BALLENTINE, C. J. 2009 Solubility trapping in formation water as dominant CO_2 sink in natural gas fields. *Nature* **458**, 614–618.

GOLDING, M. J., HUPPERT, H. E. & NEUFELD, J. A. 2013 The effects of capillary forces on the axisymmetric propagation of two-phase, constant-flux gravity currents in porous media. *Phys. Fluids* **25**, 036602.

HIDALGO, J. J. & CARRERA, J. *et al.* 2009 Effect of dispersion on the onset of convection during CO_2 sequestration. *J. Fluid Mech.* **640**, 441–452.

HIDALGO, J. J., FE, J., CUETO-FELGUEROSO, L. & JUANES, R. 2012 Scaling of convective mixing in porous media. *Phys. Rev. Lett.* **109** (26), 264503.

IPCC 2005 *Special Report on Carbon Dioxide Capture and Storage* (ed. B. Metz), Cambridge University Press.

KNEAFSEY, T. J. & PRUESS, K. 2010 Laboratory flow experiments for visualizing carbon dioxide-induced, density-driven brine convection. *Trans. Porous Med.* **82**, 123–139.

MACMINN, C. W. & JUANES, R. 2013 Buoyant currents arrested by convective dissolution. *Geophys. Res. Lett.* **40**, 2017–2022.

NORDBOTTEN, J. M. & CELIA, M. A. 2006 Similarity solutions for fluid injection into confined aquifers. *J. Fluid Mech.* **561**, 307–327.

RIAZ, A., HESSE, M., TCHELEPI, H. A. & ORR, F. M. Jr 2006 Onset of convection in a gravitationally unstable, diffusive boundary layer in porous media. *J. Fluid Mech.* **548**, 87–111.

SZULCZEWSKI, M. L., HESSE, M. A. & JUANES, R. 2013 Carbon dioxide dissolution in structural and stratigraphic traps. *J. Fluid Mech.* **736**, 287–315.

VILARRASA, V., BOLSTER, D., OLIVELLA, S. & CARRERA, J. 2010 Coupled hydromechanical modelling of CO_2 sequestration in deep saline aquifers. *Intl J. Greenh. Gas Control* **4** (6), 910–919.