

Geologic Storage of Greenhouse Gases: Multiphase and Non-isothermal Effects, and Implications for Leakage Behavior

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INTRODUCTION

Storage of greenhouse gases, primarily CO₂, in geologic formations has been proposed as a means by which atmospheric emissions of such gases may be reduced (Bachu et al., 1994; Orr, 2004). Possible storage reservoirs currently under consideration include saline aquifers, depleted or depleting oil and gas fields, and unmineable coal seams (Baines and Worden, 2004). The amount of CO₂ emitted from fossil-fueled power plants is very large, of the order of 30,000 tonnes per day (10 million tonnes per year) for a large 1,000 MW coal-fired plant (Hitchon, 1996). In order to make a significant impact on reducing emissions, very large amounts of CO₂ would have to be injected into subsurface formations, resulting in CO₂ disposal plumes with an areal extent of order 100 km² or more (Pruess et al., 2003). It appears inevitable, then, that such plumes will encounter imperfections in caprocks, such as fracture zones or faults, that would allow CO₂ to leak from the primary storage reservoir. At typical subsurface conditions of temperature and pressure, CO₂ is always less dense than aqueous fluids; thus buoyancy forces will tend to drive CO₂ upward, towards the land surface, whenever adequate (sub-)vertical permeability is available. Upward migration of CO₂ could also occur along wells, including pre-existing wells in sedimentary basins where oil and gas exploration and production may have been conducted (Celia et al., 2004), or along wells drilled as part of a CO₂ storage operation.

Concerns with leakage of CO₂ from a geologic storage reservoir include (1) keeping the CO₂ contained and out of the atmosphere, (2) avoiding CO₂ entering groundwater aquifers, (3) asphyxiation hazard if CO₂ is released at the land surface, and (4) the possibility of a self-enhancing runaway discharge, that may culminate in a "pneumatic eruption" (Giggenbach et al., 1991). The manner in which CO₂ may leak from storage reservoirs must be understood in order to avoid hazards and design monitoring systems.

MODEL SYSTEMS FOR LEAKAGE

We have studied the behavior of idealized CO₂ leakage systems by means of numerical simulation. For typical crustal conditions with a geothermal gradient

of $|\nabla T| \approx 30$ °C/km and a hydrostatic gradient of $|\nabla P| \approx 100$ bar/km, subsurface conditions at around 700 m depth will be close to the critical point of CO₂ ($T_{\text{crit}} = 31.04$ °C, $P_{\text{crit}} = 73.82$ bar). This suggests that upward migration of CO₂ from greater depth may be subject to strong non-isothermal effects (Pruess, 2005a). Our initial emphasis is on understanding the coupling between fluid flow and heat transfer under multiphase flow conditions of CO₂ and water (or brine), and idealized models with regular geometry and uniform hydrologic properties have been used. Future work will address more realistic hydrogeologic conditions, employing natural analogs where feasible (Shipton et al., 2004). Fig. 1 shows a permeable conduit that would enable CO₂ to migrate from a deep storage reservoir to the land surface. Such a conduit could be formed by an intersection of faults, or may be provided by an abandoned well that would be intercepted by a CO₂ plume. Fig. 2 shows an arrangement of faults that could enable CO₂ to escape from the primary storage reservoir and form a secondary accumulation at shallower depth, that would eventually leak to the ground surface.

All simulations reported here were performed with our general-purpose TOUGH2 simulator, augmented by various modules for specialized fluid systems and flow processes (Pruess, 2004a).

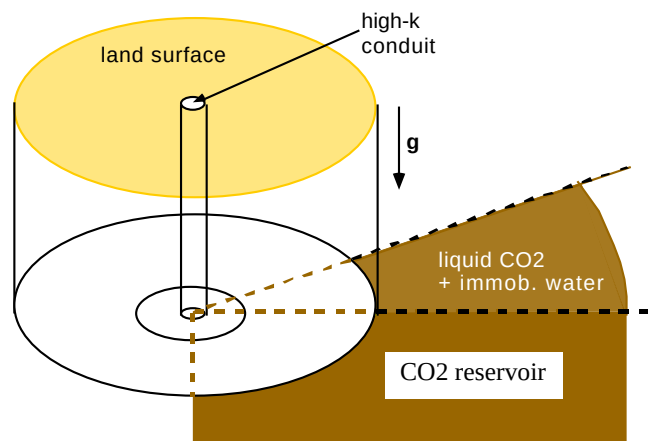


Figure 1. High-permeability conduit providing a leakage path from a deep storage reservoir.

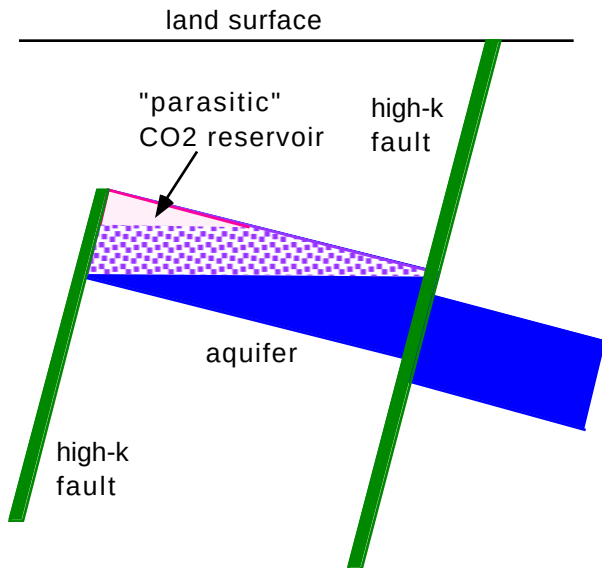


Figure 2. Schematic of fault or fracture zones that may allow CO₂ to accumulate at shallower depths and eventually be released at the ground surface.

CO₂ MIGRATION ALONG A LOCALIZED CHANNEL

Fig. 3 shows a flow system used to study CO₂ discharge behavior. A central channel of 3 m radius and 10^{-13} m² permeability is embedded in country rock with a permeability of 10^{-14} m². Initial conditions are prepared as hydrostatic equilibrium with a normal geothermal gradient, relative to ambient land surface conditions. The flow path from the primary CO₂ storage reservoir to the channel is not modeled; instead, discharge is initiated by applying CO₂ at a slight overpressure at the bottom of the channel. The injected CO₂ migrates upward, initially primarily along the channel, displacing the water and partially dissolving in it. Strong cooling effects from (nearly) adiabatic expansion draw thermodynamic conditions towards the critical point and the saturation line (Fig. 4). Additional cooling takes place as liquid CO₂ boils into gas, causing formation of three-phase conditions (aqueous phase-liquid CO₂-gaseous CO₂). Fluid mobility is reduced in the three-phase zone from relative permeability effects, which causes lateral diversion of CO₂ (sideways flow) and broadening of the plume. Over time the temperature decline associated with CO₂ boiling and expansion becomes quite strong, and the three-phase zone becomes very thick and broad. Additional information on the behavior of the system is available in (Pruess, 2004b).

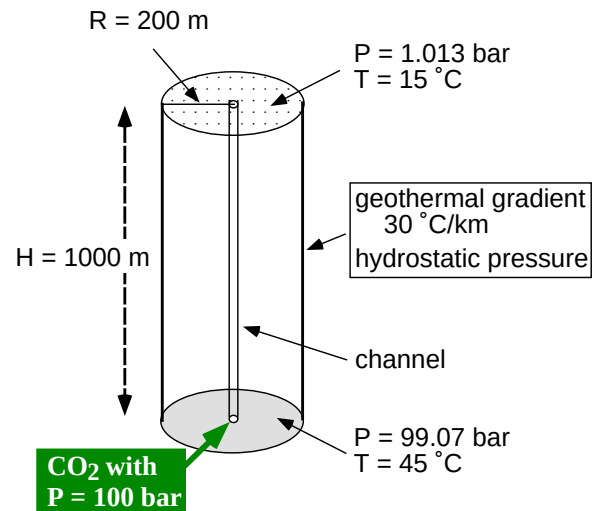


Figure 3. Specification of a localized circular channel used to study CO₂ discharge behavior.

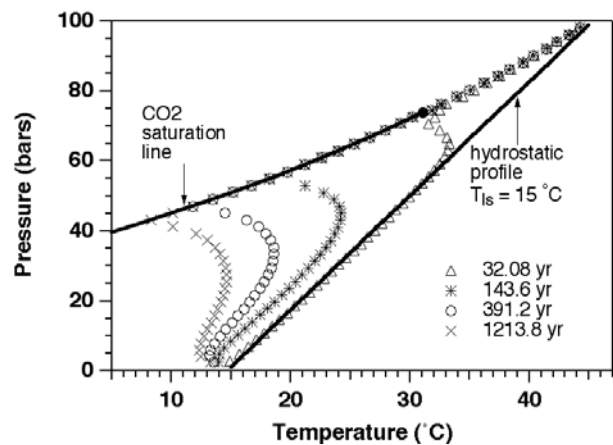


Figure 4. Temperature-pressure profiles in the center of the upflow channel at different times.

UPFLOW IN AN IDEALIZED FAULT (FRACTURE) ZONE

We idealize a fault zone as a homogeneous permeable medium embedded in country rock of negligibly small permeability (Fig. 5). The flow system as depicted in Fig. 5 is two-dimensional, except that important heat transfer effects may take place in the third dimension, perpendicular to the fault plane. Transient heat conduction in the third dimension is modeled by means of the semi-analytical technique of Vinsome and Westerveld (1980). The flow path connecting the primary storage reservoir to the fault zone is not modeled; instead we proceed as in the previous example and introduce CO₂ directly into the permeable feature. The flow system is again prepared with geothermal/hydrostatic initial conditions relative to land surface conditions of (T, P) = (15 °C, 1.013 bar), and leakage is initiated by applying CO₂ at a

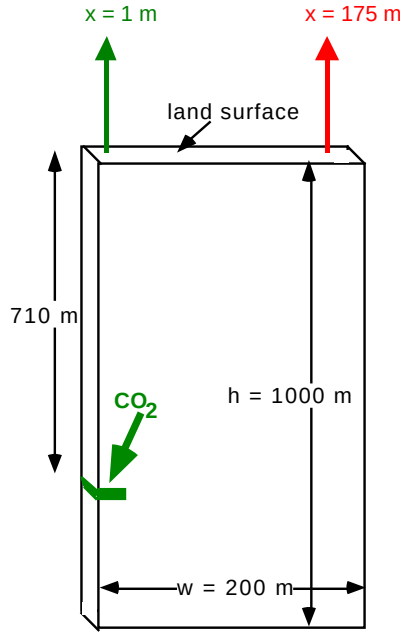


Figure 5. Idealized fault zone for studying CO_2 discharge behavior. Land surface monitoring points at 1 and 175 m distance from the left boundary are also shown.

pressure of 80 bar over a 6 m wide section of the fault at 710 m depth, where initial hydrostatic pressure is approximately 70.5 bar.

The evolution of thermodynamic conditions in the leakage system is similar as for the circular channel studied in the previous section, in that thermodynamic conditions are drawn towards and are controlled by the CO_2 saturation line. However, whereas in the previous example the volume of the three-phase zone and rates of CO_2 discharge at the land surface increased monotonically with time, the fault zone shows non-monotonic behavior with quasi-periodic variations in three-phase volume and surface discharge (Fig. 6). There is an anticorrelation between discharge fluxes at the $x = 1$ m and 175 m monitoring points, and the flux at $x = 175$ m varies in phase with cyclic variations in the volume that is in three-phase conditions. This behavior is due to an interplay of multiphase flow in the fracture plane with heat conduction perpendicular to it. As CO_2 upflow above the injection point increases, cooling effects occur from boiling and expansion. This gives rise to evolution of three-phase conditions with reduced fluid mobility, and CO_2 increasingly gets diverted sideways, away from the left side of the fault, to flow

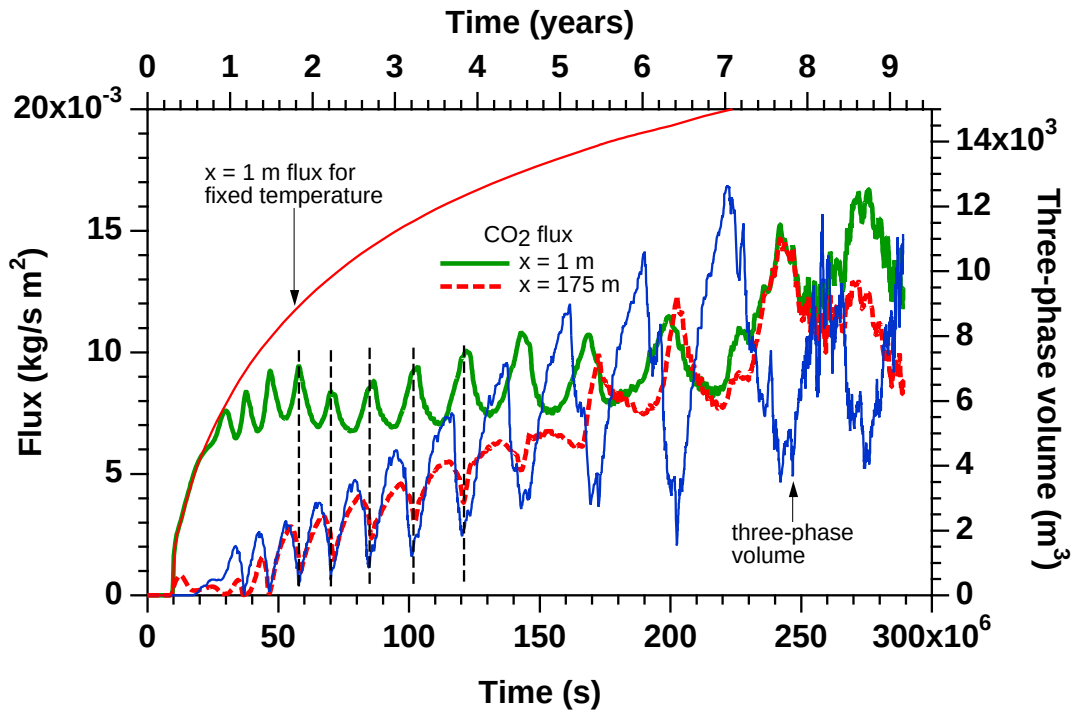


Figure 6. Temporal variation of CO_2 leakage fluxes at two different positions at the land surface. Total flow system volume with three-phase conditions is also shown. The vertical dashed lines are drawn to highlight the anticorrelation between leakage flux at $x = 1$ m on the one hand, and leakage flux at $x = 175$ m and three-phase volume on the other. The curve marked "fixed temperature" shows the simulated evolution of leakage fluxes in a system without heat transfer limitations.

around the three-phase zone. This reduces upflow above the injection point, and simultaneously increases CO_2 fluxes at laterally offset locations. The reduced upflow above the injection point reduces cooling rates there, and allows heat conduction from the wall rocks to catch up and evaporate a considerable fraction of the liquid CO_2 . Fluid conditions then return to two-phase aqueous-gas, which enhances fluid mobility and allows CO_2 fluxes to rebound, starting another cycle. Fig. 6 also shows $x = 1$ m fluxes for a problem variation where temperatures are held fixed. Under these conditions there are no heat transfer limitations, no three-phase zones develop, and CO_2 fluxes increase monotonically with time (Pruess, 2005b). This behavior makes it plain that it is the heat transfer limitations that limit CO_2 discharge fluxes.

DISCHARGE OF WATER/ CO_2 MIXTURE FROM A WELL

The final study presented here is concerned with discharge of CO_2 -laden water from a well (Fig. 7). A wellbore of 20 cm diameter extending to 250 m depth is subjected to inflow of water with 3.5 % CO_2 by weight, which is slightly below the CO_2 solubility limit for prevailing temperature and pressure conditions at 250 m depth. The well discharges to atmospheric conditions of $(T, P) = (15^\circ\text{C}, 1.013 \text{ bar})$.

Although the fluid feeding the well is just a single aqueous phase, two-phase conditions develop as rising fluid encounters lower pressures and CO_2 exsolves. We model the two-phase flow by means of the "drift flux" model (Zuber and Findlay, 1965). This model

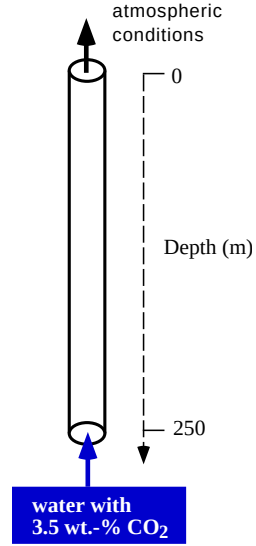


Figure 7. Schematic of wellbore discharging a water- CO_2 mixture.

considers the liquid-gas mixture as a single fluid phase with volumetrically averaged properties, but accounts for slip between gas and liquid arising from non-uniform velocity profiles, as well as from buoyancy forces. Fig. 8 shows the simulated discharge behavior for a constant injection rate of 0.2 kg/s at the base of the well. It is seen that after an initial incubation period of approximately 22000 s, the discharge goes through regular cyclic variations with a period of approximately 1600 s, i.e., the well behaves as a geyser. The geysering is due to an interplay between different flow velocities for gas and liquid, and associated changes in the average density

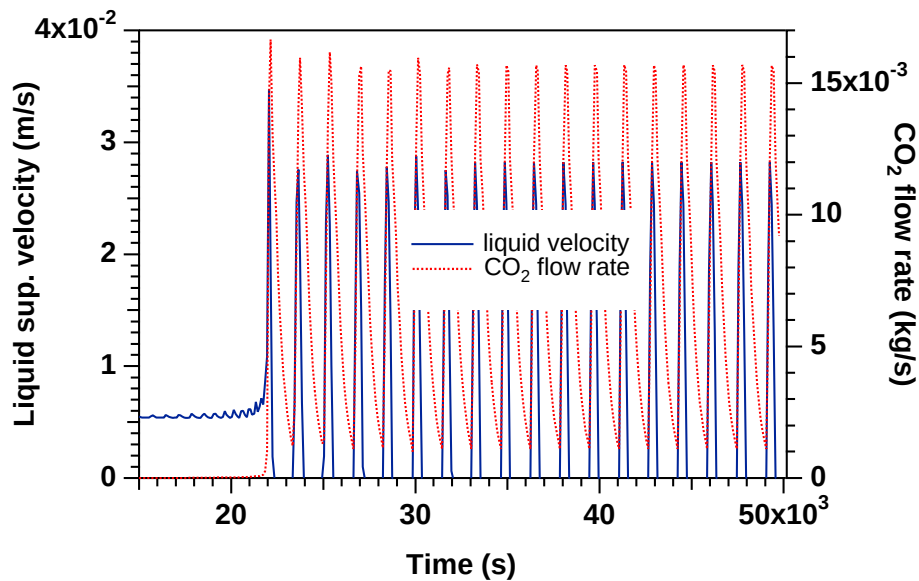


Figure 8. Discharge behavior of a well producing from a reservoir of water with dissolved CO_2 .

of the two-phase mixture and corresponding hydrostatic pressures. Discharge is enabled by CO₂ gas coming out of solution, but the preferential upflow of CO₂ also depletes the fluid of gas. This produces alternate cycles of self-enhancement and self-limitation.

In natural systems CO₂ venting usually occurs in a diffuse manner, but there are examples of "cold" geysers that are entirely driven by the energy released when high-pressure CO₂ expands (Shipton et al., 2004).

CONCLUDING REMARKS

Our numerical simulation experiments have shown that upflow of CO₂ from depth is subject to strong coupling between different fluid phases, and between fluid flow and heat transfer. CO₂ has physical properties that provide a potential for self-enhancing flows, including smaller density and viscosity than water, and much larger compressibility. Heat transfer limitations and flow interference between different phases tend to limit CO₂ upflow rates. Discharge of water with dissolved CO₂ from a wellbore is prone to instabilities, and may give rise to geysering.

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