Reservoir Engineering To Accelerate the Dissolution of CO₂ Stored in Aquifers

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It is possible to accelerate the dissolution of CO₂ injected into deep aquifers by pumping brine from regions where it is undersaturated into regions occupied by CO₂. For a horizontally confined reservoir geometry, we find that it is possible to dissolve most of the injected CO₂ within a few hundred years at an energy cost that is less than 20% of the cost of compressing the CO₂ to reservoir conditions. We anticipate that use of reservoir engineering to accelerate dissolution can reduce the risks of CO₂ storage by reducing the duration over which buoyant free-phase CO₂ is present underground. Such techniques could simplify risk assessment by reducing uncertainty about the long-term fate of injected CO₂, and could expand the range of reservoirs which are acceptable for storage.

Introduction

Geologic storage of CO₂ is an important tool for managing global CO₂ emissions (*1*). Industrial scale CO₂ storage projects now operational or in design phase provide strong empirical support for the view that CO₂ storage can be implemented in a safe manner and at acceptable cost. Nevertheless, significant uncertainties remain regarding the risks, cost, and availability of underground storage at the scales necessary for this technology to play a significant role in managing global emissions. Managing one-third of current global CO₂ emissions with carbon capture and storage (CCS), for example, would require disposal of CO₂ at an average rate of ~15 km³/year at reservoir conditions, a rate approximately 5000 times the total volume of CO₂ currently injected in dedicated CCS projects. This quantity is similar to the scale of the entire current oil and gas industry, the technological foundation for CO₂ storage, which now produces ~5 km³/year of oil and ~15 km³/year of gas at reservoir conditions (*2*).

As assessments of geologic storage began in the early 1990s, attention naturally focused on oil and gas reservoirs (*3, 4*). Although it was recognized that deep aquifers offered very large potential storage capacity, they were typically viewed as offering less certain and less secure storage than did oil and gas reservoirs (*5, 6*). Less certain because deep aquifers have seen far less exploration than oil and gas reservoirs due to the absence of an economic incentive and less secure because the confining formations "aquitards" or cap-rocks have not been proven to hold buoyant fluids as

in the case for the cap-rocks that have confined buoyant oil and gas for geologic time scales.

The CO₂ injected into a deep aquifer is typically 10–40% less dense than the resident brine. Driven by density contrasts CO₂ will flow horizontally spreading under the cap-rock and may flow upward, leaking through any high permeability zones or artificial penetrations such as abandoned wells. After injection, free-phase CO₂ (gas or supercritical fluid) slowly dissolves in the brine (*7*). The resulting CO₂-rich brine is slightly denser than the preexisting brine, making it negatively buoyant thus eliminating the most important driving force for upward migration.

Natural convection processes can accelerate the dissolution beyond that which would occur with diffusion alone. Convection may occur when dense CO₂-rich brine formed under the "bubble" of free-phase CO₂ overlies lower density undersaturated brines. The time scale for the onset of convection can range from years to centuries. The time needed for the CO₂ to dissolve completely is typically much longer, on the order of $10^2 - 10^4$ years depending on the vertical permeability (*7–10*).

By eliminating the role of buoyancy in driving possible leakage of CO₂, the dissolution of CO₂ in brines confers a significant advantage to aquifers with respect to security of storage. Considering both aquifers and oil and gas reservoirs that contain water as "wet reservoirs", it is now plausible to argue that, contrary to earlier assumptions, such systems offer both greater capacity and, due to dissolution, superior storage security than do dry reservoirs in which CO₂ would remain buoyant indefinitely.

As a conceptual framework for assessing storage security, we adopt the view that the only relevant risk of leakage arises from mobile free-phase CO₂, that is, CO₂ that remains in the gas (or supercritical fluid) phase and which is not immobilized by residual gas trapping. Storage performance thus depends on two factors: (i) the likelihood that free-phase CO₂ will leak out of the reservoir or alternatively the time scale over which significant leakage is expected to occur and (ii) the rate at which free-phase CO₂ is immobilized by residual gas trapping, dissolution in the reservoir fluids, or subsequent geochemical reactions. Storage security can be increased either by reducing the probability of leakage or by increasing the rate at which CO₂ is immobilized within the aquifer. Hereafter, we use "reservoir" to describe a CO₂ storage site which might be an aquifer or a hydrocarbon reservoir with significant water saturation. We use "brine" to denote aqueous fluids of any salinity.

In contrast to much of the previous work on geologic storage which has focused on site selection, monitoring, risk assessment, or remediation, our focus is on active reservoir engineering to accelerate dissolution and also to reduce the maximum pressure within the reservoir. Here we use physically based scaling arguments and numerical simulations to assess the technical and economic feasibility of accelerating the dissolution of CO₂ in aquifers.

Methods To Accelerate Mixing

Dissolution of CO₂ in aquifer brine could, in principle, be accomplished either within the reservoir or at the surface.

Surface. Surface, or ex situ, dissolution could be achieved within a pipeline operating at the pressure of the target reservoir into which CO₂ is to be injected, (Figure 1, left panel). The generation of CO₂ bubbles or droplets sufficiently small to achieve rapid dissolution might rely on turbulent two-phase flow within the pipe (*11*).

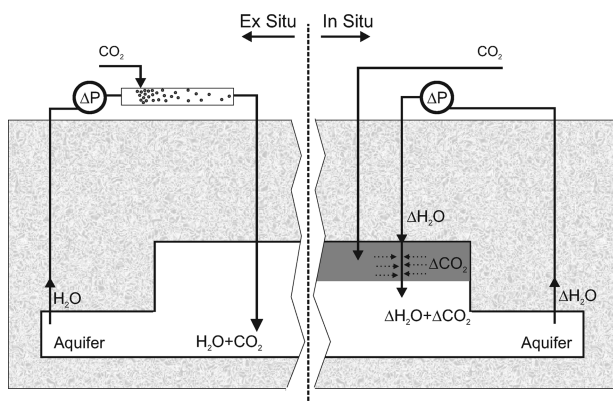* Corresponding author phone: (403) 220-6154; e-mail: keith@ucalgary.ca.

FIGURE 1. Schematic view of ex situ and in situ dissolution of CO_2 in reservoir brine.

Ex situ dissolution would require energy to overcome pressure drops in both the mixing pipeline and the aquifer. Preliminary scaling analysis based on Dukler et al. (*12*) suggests that the required pressure drop in the mixing pipeline would not be prohibitive.

Both in situ and ex situ dissolution require energy to drive brine flow in the reservoir. For ex situ dissolution the volumetric flux of brine must be much larger than the CO_2 flux because the solubility of CO_2 in brines is only a few percent by mass. Within the reservoir the pressure drop and the pumping energy per unit volume are proportional to flux (Darcy flow), so the larger fluxes required for brine injection mean larger pumping energy. For a CO_2 solubility of 5%, for example, the brine flow rate would be 20 times larger than the CO_2 flow rate, and the pumping power required for brine injection would be 400 times larger than that required for CO_2 alone (neglecting differences in viscosity and relative permeability in two-phase flow).

In Situ. If the mixing is accomplished in situ (Figure 1, right panel), then the time scales for CO_2 and brine injection can be decoupled. If CO_2 injection occurs over 20 years, for example, and forced brine circulation over 200, then the energy required for pumping brine will be about 10% of the energy that would have been required to inject fully saturated brine in which CO_2 had been dissolved ex situ assuming the same amount of brine is required. The decoupling of time scales is the primary advantage of in situ dissolution, while the primary disadvantage is the difficulty of ensuring that injected brine efficiently dissolves the CO_2.

Energy Cost of Accelerating Dissolution. The performance of an accelerated dissolution scheme may be judged according to two figures of merit, the additional energy required and the dissolution efficiency, defined below.

Additional Energy. Forced brine circulation uses energy beyond that required for a conventional CO_2 capture and storage operation. This energy requirement is crucial to determining the overall cost of accelerating dissolution. The figure of merit, *E,* is the ratio of additional pumping work done to the additional mass of CO_2 dissolved. For convenience, we normalize this by 400 kJ/kg which is the approximate energy requirement to compress CO_2 from standard conditions to typical reservoir conditions (*1*), shown as follows:

$$E = \frac{\Delta \text{ pumping work (kJ)}}{\Delta \text{ } CO_2 \text{ dissolved (kg)}} \times \frac{1}{400 \text{ (kJ/kg)}}$$

Dissolution Efficiency. The second figure of merit is the dissolution efficiency ϵ, the ratio of the additional mass of dissolved CO_2 due to force of brine circulation to the mass of brine circulated, normalized by solubility of CO_2 in brine at mean reservoir conditions, α.

$$\epsilon = \frac{\Delta \text{ } CO_2}{\Delta \text{ } H_2O} \times \frac{1}{\alpha}$$

The solubility of CO_2 in brine at typical reservoir conditions (pressure $\sim$ 100–150 bar; temperature = $\sim$50–75 °C; salinity 0–10% of solid by weight) is about 3–7% by mass. So for α = 5% at 100% dissolution efficiency (ϵ = 1) the mass of brine would need to be 20 times the mass of CO_2.

Numerical Simulations

There is no doubt that sufficiently aggressive brine circulation can accelerate CO_2 dissolution in aquifers; the question is, can acceleration be accomplished with sufficiently low energy cost and high efficiency in realistic reservoirs with sufficiently inexpensive injection well geometries? We explored the effectiveness with which brine injection can accelerate dissolution in a suite of numerical experiments using idealized reservoir geometries in which injection rates and the geometry of injection wells was varied in order to assess the sensitivity of energy cost, *E*, and dissolution efficiency, ϵ, to reservoir parameters and injection system design. The experiments used reservoir properties typical of aquifers that might be available for CO_2 sequestration (*13*).

Figure 2 presents results from simulations of an idealized "top-hat" reservoir geometry such as that shown schematically in Figure 1, which one may think of as a crude representation of an anticlinal petroleum reservoir. The horizontal step in the cap rock that confines the buoyant CO_2 is circular with a radius of 1.2 km and thickness of 60 m. The closed aquifer of radius 6 km and a maximum thickness of 100 m is homogeneous, isotropic, and isothermal with permeability of 0.2×10^{-12} m^2 (200 mD), porosity of 25%, and rock compressibility of 1.45×10^{-5} 1/bar. Initial conditions include temperature (50 °C), pressure (150 bar), salinity (4% of NaCl by weight), water saturation (1), and gas saturation (0). See Supporting Information Section 1. These reservoir properties are very similar to those used in the Berkeley Laboratory intercomparison study (*14*). We consider the applicability of this idealized reservoir geometry to real world cases in Results and Discussion.

Simulations used a Computer Modeling Group Ltd. (CMG) (*15*). Commercial simulator which was validated against TOUGH-2 simulator (*16*); the maximum differences on Problem 3 of the intercomparison study (*14*) were 3, 5, and <1% for pressure, CO_2 saturation, and dissolved CO_2, respectively. The simulations in Figure 2 used a 3-D radial nonlinear grid with a resolution of 60 × 80 × 39 (radius × height × angle; see Supporting Information). Numerical discretization was tested using various grids and the residual discretization errors in the quantity of dissolved CO_2 were less than 5% (Section 2 of Supporting Information). While it is common to use compositional simulators for CO_2 storage modeling, we employ a "black oil" simulator (CMG IMEX). For a two-component problem the black oil and compositional formulations are mathematically equivalent, while the black oil one is substantially more computationally efficient. The black oil parameters (brine as an oil and CO_2 as a gas) simulate the CO_2−brine system using high-accuracy approximations to the equation of state (*17*).

In the results shown in Figure 2, a single circular horizontal injection well is located directly below the cap-rock with a radius of 1.1 km. As we have shown elsewhere (*18*), this choice of injection well radius roughly maximizes dissolution rate for the conditions chosen here. The circular geometry is an artifact of the circular reservoir geometry; results for other well configurations are shown in the Supporting Information. Brine injection uses the same horizontal well, starting when CO_2 injection stops 20 years into the simulation. Brine is produced using three vertical brine producer wells placed
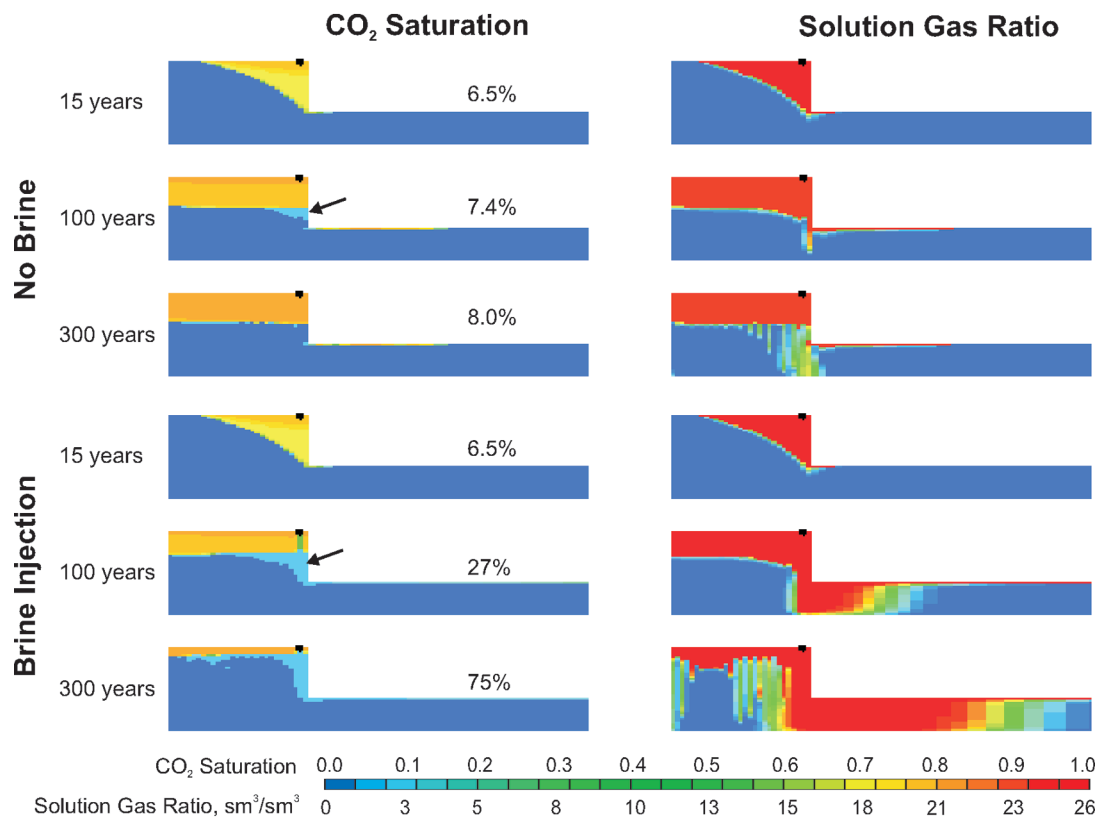
FIGURE 2. Simulation results in a "top-hat" reservoir geometry. Each subplot shows 100 m in the vertical and 3.5 km in the radial direction. The left column shows CO₂ saturation, the fraction of pore volume occupied by free-phase CO₂, while the right column shows the amount of dissolved-phase CO₂ in the brine. The black dot in each subplot shows the location of the injection well. The percentages given on the CO₂ saturation plots indicate the fraction of CO₂ that is dissolved at the specified time. Black arrows mark the plume of residually trapped CO₂ that accelerates the onset of convection. An animated version of this simulation can be found in Supporting Information (video file).

symmetrically 5 km from the center of the reservoir, far enough to avoid breakthrough of high-CO_2 brine during the simulations.

Injection of both brine and CO_2 is at constant 1 Mt/year flux, while the brine production wells maintained at a constant bottom hole pressure allowing brine to flow to the surface without pumping. At year 20 when injection switches from CO_2 to brine, the brine wells are producing at a rate of 1.5 Mt/year (because CO_2 has lower density than brine at reservoir conditions) which quickly declines to match the 1Mt/year brine injection rate. The composition of injected brine is constant matching the initial brine composition in the reservoir which is consistent with that which would be produced using continuous reinjection since the CO_2-rich brine does not reach the production wells during the 300 year simulation period.

Without brine injection, 6.5% of the CO_2 is dissolved during the 20 years of CO_2 injection and only an additional 1.5% dissolves over the remaining 300 years of simulation (Figure 3), whereas with brine injection at a rate of 1 Mt/year, 71% of the CO_2 is dissolved within 300 years. Over this period, the figures of merit for additional energy and dissolution efficiency are $E = 0.16$ and $\epsilon = 0.89$.

With or without brine injection, convection develops within the brine during the later half of the simulation driven by the density contrast that arises when dense CO_2-rich brine created by contact with the free-phase CO_2 overlie the less-dense unsaturated brine. Interestingly, convection is stronger in the case with brine injection. This appears to be due to the fact that in the injection case the residually trapped CO_2 left below the retreating bubble of free-phase CO_2 (see arrows in Figure 2, left panel) produces a plume of saturated brine

which is larger than the layer of saturated brine produced in the base case, creating a stronger density instability and thus a more rapid initiation of convection.

Dissolution would be slow without brine injection. Analytical results derived elsewhere (*19*) suggest that for the case presented in this paper it would take ∼500 years for convection to become fully developed (to reach the maximum Sherwood number) in the brine directly underneath the CO_2 gas-cap, resulting in the dissolution of ∼8% of the CO_2. After equilibration of the brine below the gas-cap, the rate of dissolution should slow dramatically as it became limited by radial convection outward into the thinner regions of the reservoir.

Figure 3 presents the results for dissolution vs time for different brine injection rates (solid lines). It also shows the results for the nonconfined case (dashed lines), which is infinite reservoir with the depth of 100 m and it has the same reservoir conditions as described for the confined reservoir. Brine injection is substantially more effective in accelerating dissolution in the confined geometry in comparison with a nonconfined one because the buoyancy force acting on the CO_2 brings it into closer contact with the injected brine. In the case of the nonconfined reservoir, brine injection pushes the CO_2 away from the injection point reducing the effectiveness of further injection.

The dissolution rate is proportional to the brine flow rate at the beginning of injection, but the rate declines with time due to the declining thickness of the CO_2 plume which reduces the contact between injected brine and CO_2 (Figure 3). At first, the brine flows downward through a two-phase region where it can be efficiently mixed with the existent free-phase CO_2. The "knee" in the cumulative dissolution curves occurs
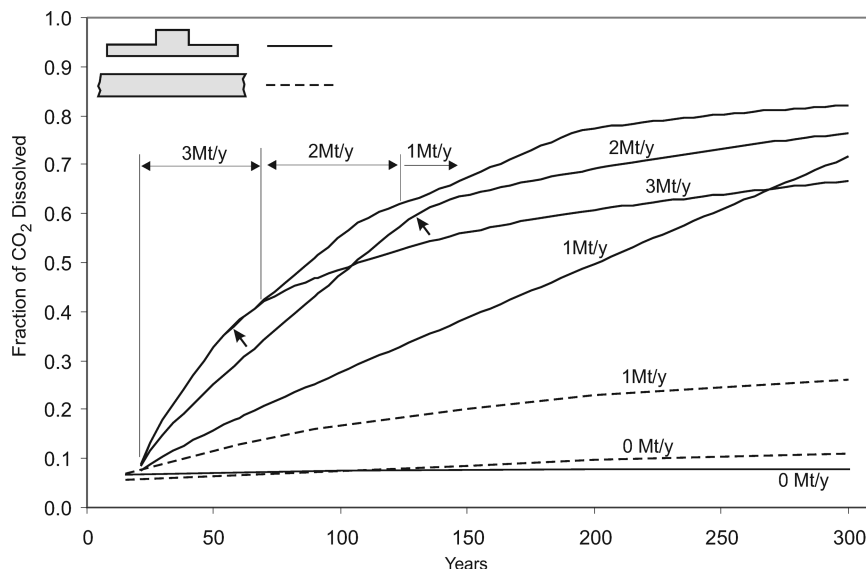
FIGURE 3. Dissolved fraction as a function of time. Solid lines show results from the top-hat geometry, while dashed lines are for an infinite slab geometry. Brine injection rates are indicated in megatons per year. Arrows denote the "knee" in the curves discussed in the text.

when the CO_2 saturation in the downward flowing brine declines to zero, so that the brine flows downward through a single (brine) phase region. Dissolution then depends on diffusion of CO_2 from the surrounding two-phase region into the brine "jet" reducing dissolution efficiency. For the 2 Mt/year brine injection, ϵ drops from ~0.9 at simulation year 130 to 0.15 after the knee in the curve at year 150. Comparison of the 1 and 3 Mt/year cases in Figure 3 shows that lower injection rates may provide more efficient dissolution. The uppermost curve shows results with an injection rate that declines with time to increase the dissolution rate while maintaining efficiency.

Results and Discussion

Forced dissolution is applicable to a range of reservoir geometries. The infinite slab geometry is a particularly unfavorable case for accelerating dissolution because most water bypasses the unconfined CO_2 plume, resulting in high E and low ϵ. The top-hat case presented here is more favorable, but it is by no means the most favorable circumstance. More favorable conditions for accelerating dissolution would include (i) a geometrically similar reservoir in which the CO_2 gas-cap has a higher thickness-to-width ratio, (ii) a geometry in which the cap-rock had an anticlinal curvature or a pronounced dip caped by an unconformity; or finally, (iii) a reservoir with higher permeability.

In results presented elsewhere we have obtained high dissolution efficiencies and low dissolution energy using several different reservoirs and injection well geometries and different reservoir anisotropy and inhomogeneity (*18*) (see also Section 4 of Supporting Information). Similar results are obtained in infinite-acting reservoir geometries so long as there is a step in the cap-rock to provide some horizontal confinement of the CO_2 (*18*). Use of cyclic injection of water or CO_2 can also improve the efficiency of dissolution (*18, 20*). The consistency of results across reservoir and injection well geometries suggests that similar engineering techniques could be applied to real-world reservoirs. While the optimal geometry would depend on the specifics of the target reservoir our results suggest that for a reservoir with relatively sharp horizontal confinement, use of a horizontal well (or wells) that roughly encircle the periphery of the initial CO_2 plume might provide a favorable geometry.

In addition to accelerating dissolution, brine production enables management of reservoir pressure which can increase
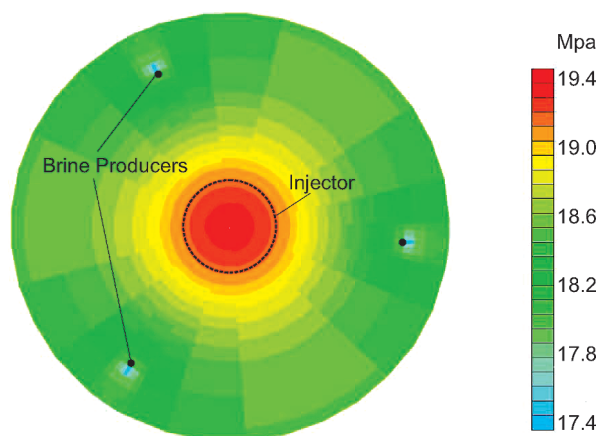

FIGURE 4. Pressure field in the top-hat reservoir geometry showing location of the brine production wells. Data are plotted to a maximum radius of 6 km. Data are for the bottom of the reservoir at the end of the CO_2 injection (20 years).

available storage capacity. In a closed (volumetric) reservoir without brine production the bottom hole pressure may approach fracture pressure forcing CO_2 injection to stop long before the available storage capacity has been utilized. In the Western Canada Sedimentary Basin, for example, fracture pressure varies from 1.5 to 3 times hydrostatic pressure (*21*). Taking the lower limit and using the top-hat reservoir described above, CO_2 injection would need to be terminated as early as year 7 when the bottom hole pressure exceeded 22.5 MPa. Whereas when brine production is allowed, the maximum bottom hole pressure reached at the end of CO_2 injection (model year 20) is under 19 MPa. Figure 4 shows the pressure field at the bottom of the reservoir at year 20.

In addition to increasing the effective storage capacity, brine management may reduce the risk of leakage by reducing the driving force for the CO_2 transport (*22*) and by reducing geomechanical stresses that might activate fractures. It also lowers the energy required for CO_2 injection by reducing required injection pressures. In some cases, the energy saved in CO_2 injection can be more than the energy required for brine pumping to accelerate dissolution.

Economics

Although the reservoir engineering necessary to accelerate dissolution will be strongly site-specific, we nevertheless describe how the simulation results might be applied to a more realistic reservoir and provide a rough estimate of the costs and compare them to the costs of CO_2 capture and storage in order to gauge the potential applicability of the technique.

While the cylindrically symmetric reservoir geometry described here is of course unrealistic, the required ~7 km total length of the horizontal injector well is roughly indicative of the length of a horizontal well that would be needed to surround the periphery of a similar volume reservoir with a more realistic geometry assuming a reservoir thickness of 100 m which is typical of the high injectivity formations often considered for CO_2 storage. There is a trade-off between the number and length of brine production and injection wells and the pressure drop and thus the energy and operating costs of brine injection. The horizontal injection wells would be drilled using a few drill pads each with two opposite directed horizontal legs of a few kilometers (see Figure 1S in Supporting Information).

The costs of accelerating dissolution arise from the cost of energy required for brine pumping and the capital costs of the pumps and any additional wells. Considering the base case described above (Figures 2 and 4), the additional well requirements would include three brine injectors and the horizontal segment of the injection well which could otherwise have a simple vertical completion. The current cost for the required drilling and completing in the Alberta basin would be of the order of $10 million. (All figures in current U.S. dollars. It should be noted that drilling costs are at historic highs and that such costs are strongly site-specific (23).) The capital cost of the required pumps and associated controls would be less than $50 thousand, insignificant in comparison to the cost of wells. Assuming a brine injection rate of 1 Mt/year and a pump efficiency of 70%, electrical power requirement would be ~90 kW. Assuming an electricity cost of $100/MWh (relatively high for a North American industrial customer), the annual operating cost would be approximately $80 thousand/year.

These costs may be compared to the costs of a full CCS project. Assume that the CO_2 originates from a new power plant with capture which has an overall capital cost of $2000/kW and an efficiency of 38%, which is a capital cost of $450 million for each 1 Mt/year of CO_2 that must be sequestered (typical values from the IPCC study (1) of IGCC plants with capture [IPCC SRCSS, Table 3.10]). In this case, the capital cost of accelerated dissolution is ~2% of the capital cost of the full project and the electricity required to run the brine injection pumps over 300 years is 0.8% of the net electricity produced by the power plant over the 20 year operating period considered here.

Alternatively, the costs may be expressed as dollars per ton of CO_2 stored. Given the financial assumptions above and discounting future electricity costs at 5%/year, the total cost to force the dissolution of ~80% of the injected CO_2 over 300 years is ~$0.08/(ton of CO_2), less than 0.2% of the typical costs of reducing CO_2 emissions using CCS.

Production and reinjection of brine within an aquifer can substantially accelerate the dissolution of stored CO_2 at a capital and energy cost that is a small fraction of the overall costs of CO_2 capture and storage. In addition, such reservoir engineering can also decrease reservoir pressures (and pressure gradients) near the point of injection which will likely further reduce the risk of leakage. Methods similar to those explored here might be used to increase the volumetric storage efficiency or to steer large-scale movement of the free-phase CO_2 "bubble" to direct it away from a spill point or a known weakness in the cap-rock.

Most analysis of CO_2 storage has focused on site selection, monitoring, and risk assessment. While these activities are necessary for managing CO_2 storage, the possibility of more active reservoir engineering such as the brine injection explored here should not be ignored. Such methods might play an important role in geologic storage for three distinct reasons. First, active reservoir engineering can reduce the risk of leakage. Second, such engineering may increase available storage capacity by increasing the range of aquifers in which CO_2 can be safely stored. Third, by shortening the time scale over which free-phase CO_2 remains in the reservoir, such methods might facilitate risk analysis and reduce regulatory and other uncertainties related to storing mobile CO_2 underground for long durations.

Supporting Information Available

Detailed description of reservoir geometry, wells placement, numerical discretization, and different reservoir anisotropy and inhomogeneity and a video file of simulation results animation. This information is available free of charge via the Internet at http://pubs.acs.org.

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