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Dynamics and design of systems for geological storage of dissolved CO₂

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ABSTRACT

The standard approach for geologic storage of CO₂ consists of injecting it as a supercritical CO₂ phase. This approach places stringent requirements on the caprock, which must display: (1) high entry pressure to prevent the buoyancy driven upwards escape of CO₂; (2) low permeability to minimize the upwards flux of brine displaced by the CO₂; and (3) high strength to ensure that pressure build up does not cause caprock failure. We propose an alternative approach for cases when the above requirements are not met. The approach consists of extracting brine from the storage formation and then re-injecting it so that it mixes with CO₂ at depth in the injection well. Mixing at depth reduces the pressure required for brine and CO₂ at the surface. This CO₂-saturated brine will sink to the aquifer bottom because it is denser than resident brine, which eliminates the risk of buoyant escape of CO₂. The method is particularly favorable when the aquifer dips, because CO₂-saturated brine will tend to flow downslope. We perform two- and threedimensional numerical simulations to study how far upslope the extraction well needs to be located to ensure a very long operation without CO2 ever breaking through. Several sets of simulations were carried out to evaluate the effect of slope, temperature, pressure and CO₂ concentration, which is significantly reduced if flue gas (i.e., without capture) is mixed with the brine. We analyze energy requirements to find that the system requires high permeability to be viable, but its performance is improved by taking advantage of the thermal energy of the extracted brine.

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1. Introduction

Large volumes of carbon dioxide (CO_2) should be injected in deep saline aquifers to reduce significantly its emissions to the atmosphere. The standard approach consists of injecting CO_2 as a free phase in supercritical conditions, such as in Sleipner [1] or In Salah [2]. Supercritical CO_2 injection in deep saline aquifers forms a CO_2 -rich phase which is characterized by gravity override [e.g., 3–6]. CO_2 will remain in the upper portion of the aquifer provided that no leakage occurs across the caprock. While the concept is simple and robust, actual application is hindered by numerous difficulties. First, CO_2 storage at the industrial scale will involve large volumes of fluid injection, which will displace the resident brine, possibly contaminating shallower drinking water aquifers [7,8]. Furthermore, CO_2 injection induces a pressure buildup that can become a limiting factor for the suitability of certain aquifers for storing CO_2 [9,10], can reactivate fractures or faults [11–13] and

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even induce seismic events [14,15]. Last but not least, supercritical CO_2 is less dense than resident brine and may leak through faults [16,17] or wells [18,19].

Since the structural trapping can fail, other trapping mechanisms may be needed to provide additional storage security. CO_2 can get trapped by capillarity, but only partially [20,21]. This is especially true in sloping aquifers where CO_2 travels long distances [22] even if trapped by capillarity in its tail [23]. Mineral trapping in most rocks will only trap a small percentage of the injected CO_2 and for time scales in the order of hundreds to thousands of years [24]. Dissolution trapping acts much faster than either capillary or mineral trapping and it allows a safe storage because brine with dissolved CO_2 is denser than resident brine and tends to sink. Dissolution generates an unstable situation, with a denser brine on top of a lighter brine which accelerates dissolution [25–28]. Still, the onset of fast (convective) dissolution may take some years and full dissolution may take thousands. That is, all permanent trapping processes take very long.

These difficulties have motivated alternative strategies to the standard supercritical CO_2 injection. [29,30] introduce the Active CO_2 Reservoir Management concept, which combines brine extraction at the bottom of the aquifer at several distances from the CO_2



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injection well, desalination and residual brine re-injection with CO_2 in saline formations. The approach increases CO_2 storage capacity and offsets pressure buildup. An improvement of the caprock mechanical stability due to thermo-mechanical effects can also be achieved by injecting liquid (cold) CO_2 instead of supercritical CO_2 [31]. Nevertheless, the risk of leakage persists with these two injection strategies because CO_2 will still float to the top of the aquifer.

To minimize the risk of leakage, CO_2 dissolution can be accelerated by injecting CO_2 under temporal pressure fluctuations [32], alternating CO_2 and brine injection [33] or by injecting brine at some distance from the CO_2 injection well that mixes with the CO_2 plume [34,35]. These options accelerate dissolution, but a significant portion of the injected CO_2 continues in free-phase for decades and even centuries.

In view of these problems, it is not surprising that [36] argue for storage protocols based on injecting dissolved CO₂, which had been proposed by Burton and Bryant [37], Jain and Bryant [38]. The system consists of extracting brine from the storage formation, mixing it with CO₂ at the surface, and then injecting the CO₂-saturated brine into the subsurface formation. Increased levels of sophistication can be achieved by generating appropriate flow conditions to accelerate CO₂ dissolution [39]. The approach sounds reasonable but raises several questions. First, surface dissolution requires compressing the brine at high pressure (say over 100 bars) in order to obtain a significant dissolution of CO₂, which may lead to unaffordable energetic and economic costs. Therefore, it would be desirable to seek an alternative concept to reduce compression costs. Second, the injected CO₂-saturated brine will tend to flow towards the pumping well, which would ruin the system. Therefore, the dynamics of the system need to be elucidated for proper eventual design and construction. Finally, huge volumes of brine need to be mobilized so that, even if technically feasible, the system might not be economically viable. Therefore the energy requirements of the system have to be evaluated.

The aim of the present work is to assess the feasibility of dissolved CO_2 injection by providing responses to these three questions. First, to reduce compression, we propose producing the mixing at depth, so that gravity ensures a high pressure of brine and helps pressurizing the CO_2 . Second, to elucidate the dynamics of the proposed system, we model the system numerically in order to, first, obtain realistic representations and predictions of its behavior, and, second, to identify the optimal location of the wells and their pumping rates. Third, we use the results to assess the energy cost for the proposed system.

2. Dissolved CO₂ injection system

The basic idea of the system is described in Fig. 1. The goal is to reduce compression of brine and CO_2 at the surface, so that most

pressurization is performed by Earth's gravity. The advantages of the proposed system with respect to the conventional supercritical CO_2 injection and storage systems are: (1) The risk of CO_2 leakage is reduced because CO_2 -saturated brine is denser than resident brine and will sink to the aquifer bottom, which relaxes the requirements for the caprock formation, (it is no longer needed), and (2) The extraction of brine from the same permeable geological formation prevents fluid pressure buildup at a regional scale. This not only reduces the risk of contaminating the nearest water bodies, but also reduces compression costs.

The method is favorable when the aquifer dips, because locating the extraction well upslope can ensure a very long operation without CO_2 ever breaking through into the extraction well. If the aquifer does not dip, the method is still applicable, but the distance between pumping and injection wells needs to be much larger to ensure a long operation prior to breakthrough.

An important additional advantage of the system is that CO_2 does not need to be pure. Specifically, the system can be applied directly to combustion gases without the need of capturing pure CO₂. This eliminates capture costs and allows obtaining credits by storing other greenhouse gases (CH₄, SO_x, NO_x). Efficiency of dissolution is reduced in the case of injecting a mixture containing low solubility gases (LSG, e.g., N₂) because the least soluble components will float back to the wellhead, which prevents full dissolution of CO₂ and other soluble gases (a gas phase will be always present). Even worse, the dissolved concentration is controlled by the partial pressure of CO₂, which is proportional to its molar fraction and is low in typical flue gas (around 15%). Notice that the problem lies not so much on the need for uselessly compressing the LSGs (their residual pressure can be recovered when they return to the wellhead), as on the fact that the concentration of CO₂ in the injected brine would be reduced, thus wasting storage space. This problem can be partly overcome by perforating the well deeper than the storage formation, so that mixing occurs at an increased pressure.

The proposed concept is outlined in Fig. 2. It consists of the following steps:

- -Step 1: Brine (salt water) is pumped from a well.
- -Step 2: The brine is conditioned at the surface. In practice, this may involve several operations. First, brine should be cooled down. This will yield some energy. The theoretical energy drop is comparable to the cumulative pumping and compression work. More important, for our purposes, the solubility of CO₂ increases dramatically when temperature is lowered (see Table 1). CO₂ concentration should not increase above its saturation at aquifer pressure and temperature, but mixing at low temperature facilitates dissolution. Conditioning may also imply adding alkalinity, to increase CO₂ solubility



Fig. 1. Schematic description of the dissolved CO₂ injection system. In favorable cases (a or b), gravity will drive the CO₂ saturated brine away from the pumping well. Further gravity potential difference can be gained from partially penetrating or horizontal wells (c). If the aquifer is horizontal (d), the distance between pumping and injection wells must be such that CO₂ will take long to arrive at the injection well.

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