

## Liquid CO<sub>2</sub> injection for geological storage in deep saline aquifers

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### ABSTRACT

CO<sub>2</sub> will remain in supercritical (SC) state (i.e.  $p > 7.382$  MPa and  $T > 31.04$  °C) under the pressure ( $p$ ) and temperature ( $T$ ) conditions appropriate for geological storage. Thus, it is usually assumed that CO<sub>2</sub> will reach the aquifer in SC conditions. However, inflowing CO<sub>2</sub> does not need to be in thermal equilibrium with the aquifer. In fact, surface operations are simpler for liquid than for SC CO<sub>2</sub>, because CO<sub>2</sub> is transported in liquid state. Yet, problems might arise because of thermal stresses induced by cold CO<sub>2</sub> injection and because of phase changes in the injection tubing or in the formation. Here, we propose liquid CO<sub>2</sub> injection and analyze its evolution and the thermo-hydro-mechanical response of the formation and the caprock. We find that injecting CO<sub>2</sub> in liquid state is energetically more efficient than in SC state because liquid CO<sub>2</sub> is denser than SC CO<sub>2</sub>, leading to a lower overpressure not only at the wellhead, but also in the reservoir because a smaller fluid volume is displaced. Cold CO<sub>2</sub> injection cools down the formation around the injection well. Further away, CO<sub>2</sub> equilibrates thermally with the medium in an abrupt front. The liquid CO<sub>2</sub> region close to the injection well advances far behind the SC CO<sub>2</sub> interface. While the SC CO<sub>2</sub> region is dominated by gravity override, the liquid CO<sub>2</sub> region displays a steeper front because viscous forces dominate (liquid CO<sub>2</sub> is not only denser, but also more viscous than SC CO<sub>2</sub>). The temperature decrease close to the injection well induces a stress reduction due to thermal contraction of the media. This can lead to shear slip of pre-existing fractures in the aquifer for large temperature contrasts in stiff rocks, which could enhance injectivity. In contrast, the mechanical stability of the caprock is improved in stress regimes where the maximum principal stress is the vertical.

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### 1. Introduction

Pressure ( $p$ ) and temperature ( $T$ ) conditions of deep geological formations suitable for storing carbon dioxide (CO<sub>2</sub>) are such that this greenhouse gas remains in supercritical (SC) state, i.e.  $p > 7.382$  MPa and  $T > 31.04$  °C (e.g. Bachu, 2003). Thus, it is usually assumed that CO<sub>2</sub> will reach the aquifer in SC conditions (e.g. Pruess and Garcia, 2002). However, injecting CO<sub>2</sub> in SC state may not be the best option. Several engineering methodologies have been proposed as alternatives to the concept of injecting SC CO<sub>2</sub>. They focus on accelerating CO<sub>2</sub> dissolution to minimize the risk of leakage of free-phase mobile CO<sub>2</sub> by means of dissolving CO<sub>2</sub> at surface (Burton and Bryant, 2009; Jain and Bryant, 2011; Zendejboudi et al., 2011) or at depth (Carrera et al., 2011b), by injecting brine at some distance from the CO<sub>2</sub> injection well that mixes with the CO<sub>2</sub> plume enhancing dissolution (Hassanzadeh et al., 2009) or by injecting

CO<sub>2</sub> under temporal pressure fluctuations, which enhances CO<sub>2</sub> dissolution (Bolster et al., 2009). On the other hand, a few studies suggest that cold CO<sub>2</sub> (and therefore in liquid state) injection may have some advantageous implications for CO<sub>2</sub> storage (Rayward-Smith and Woods, 2011; Silva et al., 2011). However, these studies are approximations that do not take into account the whole coupling of the thermo-hydro-mechanical effects inherent to cold CO<sub>2</sub> injection.

It can be conjectured that injecting CO<sub>2</sub> in liquid state is energetically more efficient than doing so in SC state and more optimal from a storage engineering point of view because liquid CO<sub>2</sub> is denser than SC CO<sub>2</sub>. Therefore, for a given mass of CO<sub>2</sub>, a smaller volume of formation fluid will be displaced, leading to a lower overpressure in the reservoir. More importantly, the increased weight of liquid CO<sub>2</sub> in the injection well implies that a far lower pressure is required at the wellhead. Additionally, CO<sub>2</sub> is usually transported in liquid state (pressure above 8.5 MPa and ambient temperatures (Fig. 1)) (McCoy and Rubin, 2008). Thus, it can be injected at the conditions in which it arrives to the wellhead, without having to perform throttling or heating operations. In fact, since pressure at the wellhead is reduced, it may be smaller than transport pressure, which may allow recovering some energy from the incoming CO<sub>2</sub>.

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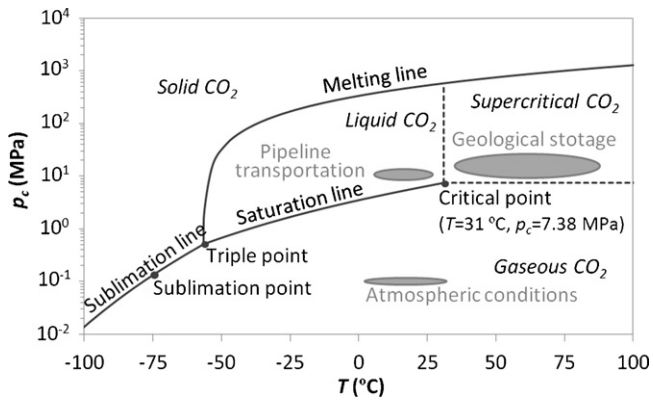


Fig. 1. CO<sub>2</sub> phase diagram. CO<sub>2</sub> is a gas in the atmosphere. Pipeline transportation is done in liquid CO<sub>2</sub> conditions and geological storage stays in supercritical CO<sub>2</sub> conditions.

Furthermore, if pressure needs to be increased, a smaller compression work has to be done to inject liquid CO<sub>2</sub> because liquid CO<sub>2</sub> is less compressible than SC CO<sub>2</sub>. This compression can be performed by means of pumps without having to use compressors, which are much harder to operate. Despite these apparent advantages, liquid CO<sub>2</sub> injection has not been considered in the scientific literature and it has not been attempted in practice except for the case of Snøhvit, where CO<sub>2</sub> is injected in liquid state at the wellhead (at 4 °C because the wellhead, placed on the seabed at 300 m below the sea surface, thermally equilibrates with the sea), but reaches the reservoir, placed at 2700 m below the seabed, in SC conditions because CO<sub>2</sub> thermally equilibrates with the geothermal gradient (at 98 °C in the reservoir) (Estublier and Lackner, 2009). This may reflect the fact that so far industrial operations have been associated to oil industry, where CO<sub>2</sub> is obtained in gas form. It may also reflect fear to phase transitions in the injection equipment or in the formation, or to thermal (thermo-mechanical) stresses associated to a cold fluid injection.

Hydro-mechanical, but not thermo-mechanical, effects have been widely investigated in the context of geological storage of CO<sub>2</sub> (e.g. Rutqvist et al., 2007; Ferronato et al., 2010; Vilarrasa et al., 2010b; Goerke et al., 2011; Rutqvist, 2012). The main concern is to guarantee that the mechanical stability of the caprock will not be compromised in order to prevent CO<sub>2</sub> leakage. Nimtz et al. (2010) argue that, when injecting liquid CO<sub>2</sub>, the overpressure at the bottom of the well will be too high because CO<sub>2</sub> pressure at the wellhead has to be enough to ensure liquid conditions; and the hydrostatic pressure in the well will be also high because liquid CO<sub>2</sub> has a density around 900 kg/m<sup>3</sup>. However, they do not perform any hydro-mechanical simulation to confirm their hypothesis. Moreover, they do not consider reducing temperature, which ensures liquid conditions with moderate pressures. Note that an excessive overpressure can induce microseismicity (Phillips et al., 2002; Guglielmi et al., 2008; Cappa and Rutqvist, 2011), which may open up migration paths for CO<sub>2</sub>. However, since liquid CO<sub>2</sub> is colder than the formations where it will be injected, liquid CO<sub>2</sub> injection implies a combination of hydro-mechanical and thermo-mechanical effects that should be studied simultaneously to properly evaluate the caprock mechanical stability.

The injection of a cold fluid induces a thermal contraction of the rock, leading to a reduction of the effective stresses (Segall and Fitzgerald, 1998), which tends to bring the stress state closer to failure conditions. Thermo-mechanical effects have been studied specially in geothermal reservoir stimulation (Ghassemi et al., 2007; Majer et al., 2007). The thermo-mechanical effects of injecting CO<sub>2</sub> at a colder temperature than that of the reservoir have been investigated at the In Salah injection project (Algeria), where

CO<sub>2</sub> is injected in supercritical conditions, but significantly cooler than the formation (Bissell et al., 2011; Preisig and Prévost, 2011; Rutqvist, 2012). Additionally, non-isothermal CO<sub>2</sub> flow simulations have been performed, but without considering the mechanical coupling and always in supercritical conditions (Han et al., 2010; Singh et al., 2011). Therefore, the thermo-mechanical effects of liquid CO<sub>2</sub> injection remain to be studied.

We propose to inject CO<sub>2</sub> in liquid state as a new engineering methodology for minimizing energy costs and phase changes in the capture–transport–injection chain, and improving the short- and long-term storage efficiency of CO<sub>2</sub>. This injection concept will be tested at the pilot site of Hontomín (Carrera et al., 2011a), Burgos, Spain, which is the injection site of the CO<sub>2</sub> storage Technology Demonstration Plant (TDP) of the Compostilla OXYCFB300 project (EU funded: European Energy Programme for Recovery), operated by Fundación Ciudad de la Energía (CIUDEN). Hontomín is a dome-like structure with a dolomitized reservoir located at 1450 m depth, which is overlaid by a caprock made of marls. Several experiments are planned both for site characterization and for injection technology development (Carrera et al., 2011a).

The objective of this work is to analyze liquid CO<sub>2</sub> injection into a deep aquifer in terms of (1) the energetic efficiency and (2) caprock mechanical stability. This represents a first step toward the design of the liquid CO<sub>2</sub> injection test that will be performed at the Hontomín pilot test. We calculate CO<sub>2</sub> flow in both the injection well and the reservoir. We perform simulations of non-isothermal two-phase flow in a deformable porous media to evaluate mechanical stability of the caprock.

## 2. Mathematical and numerical methods

We first solve CO<sub>2</sub> injection in a vertical injection well and afterwards in a saline formation. The geometry of the problem consists in a homogeneous 100 m thick horizontal aquifer that is overlaid and underlain by a seal. The system is axisymmetric and extends 20 km laterally. The nature of the outer hydraulic boundary condition does not affect the results because the radius of the pressure perturbation cone is smaller than the radius of the domain for the injection time scales presented here. Therefore, the model behaves as an infinitely acting aquifer. The top of the aquifer is located at a depth of 1500 m, which corresponds to the depth of the reservoir at the Hontomín test site. The seals that overlay (caprock) and underlie the aquifer have a thickness of 200 m. We assume that the caprock is covered by a 1300 m thick overburden of such a low shear stiffness that does not need to be included in the model. An injection well with a radius of 0.15 m is placed in the center of the domain. This radius was initially planned at Hontomín, but has now been reduced.

### 2.1. Non-isothermal flow in the injection pipe

Flow of CO<sub>2</sub>, or any fluid, and its mixtures in non-isothermal wells involves solving the partial differential equations (PDE) that express energy, mass and momentum conservation. These PDEs are coupled through the equations of state (EOS) governing fluid and thermodynamic properties. Several authors describe numerical procedures to solve these equations (Lu and Connell, 2008; Paterson et al., 2008; Pan et al., 2009; Han et al., 2010).

Here, we adopted the approach of Lu and Connell (2008). They presented a methodology to solve steady state non-isothermal multiphase flow of CO<sub>2</sub> in an injection well, in which the flow equations are based on the averaged-flow model (e.g. Brill and Mukherjee, 1999; Hasan and Kabir, 2002). We assume that the steady state assumption describes reasonably well the operation after the initial stages. This leads to a system of one dimensional

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