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# Cost-Effective Carbon Sequestration: Evaluating Liquid vs. Supercritical CO<sub>2</sub> in Deep Aquifers

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## Abstract

The transportation and injection of  $CO_2$  are critical components in the carbon capture and storage (CCS) chain. While transporting  $CO_2$  in the liquid phase allows for higher volumes, supercritical  $CO_2$  (SC-CO<sub>2</sub>) is generally preferred for pipeline transportation due to its unique (combined) properties of SC-CO<sub>2</sub>, reducing energy requirements for its injection. However, alternatively, injecting cold, liquid  $CO_2$  can offer potential benefits, including enhanced brine displacement efficiency and reduced  $CO_2$  buoyant migration through the caprock due to its higher density, but this would require additional energy for cooling. This study aims to study the implications of injecting liquid vs SC-CO<sub>2</sub> based on economic and technical points of view. We evaluate and compare the cost and performance implications of  $CO_2$  injection for economic feasibility at three injection temperatures: i) 5 °C, ii) 25 °C, iii) 35 °C. For this, we take into account that the average energy prices in Louisiana are 0.161 USD/kWh according to the U. S. Bureau of Labor Statistics. The technical point of view is performed by conducting numerical simulations using TOUGH3 to analyze the injection process and subsurface behavior.

## Introduction

The International Energy Agency (IEA) emphasizes that CCS technology plays a vital role in the global effort to counteract the effects of greenhouse gas emissions, stating that achieving "net" zero targets would be virtually impossible without it [1, 2]. However, in CCS operations,  $CO_2$  leakage from the storage sites is one of the biggest challenges [3] mainly due to the complexity of the injection process itself. Therefore, to address this challenge, engineers must take all the necessary measures to prevent  $CO_2$  from escaping the storage site and thereby minimize the artificial emission of  $CO_2$  into the atmosphere [4]. Villarasa et al. proposed injecting liquid  $CO_2$  rather than supercritical. Their studies were based on the assumption that  $CO_2$  is often stored in vessels at very low temperatures (about -20 C) [5]. Therefore, more energy would be required for compression and heating to reach the supercritical condition.

#### Methods

a) Model Description

A two-dimensional radial grid is employed for the numerical simulation to model the  $CO_2$  injection process into a 50-meter-thick deep aquifer. The aquifer has a porosity and horizontal permeability of 0.3 and 500 mD, respectively. The storage formation is overlain by a 50-meter-thick caprock with porosity and horizontal permeability of 0.05 and 0.05 mD, respectively. Above the caprock lies a uniform overburden extending from a depth of 2400 meters to the surface. The well is considered vertical with a diameter of 0.15 (~6 in) and a depth of 2500 m.

The model consists of 3401 grid blocks. To accurately capture detailed flow dynamics and thermal effects, the grid is refined near the wellbore and progressively coarsened using a logarithmic increment toward the outer radial direction. A uniform discretization in the vertical direction is employed, with layer thicknesses of 50 meters for the overburden, 5 meters for the caprock, and 5 meters for the reservoir. Thermal conductivities equal to 1.72 W/(m\*°C) and 2.51 W/(m\*°C) are assigned to the caprock and reservoir layers, respectively. A rock-grain specific heat capacity of 1000 J/(kg\*°C) is assigned to both caprock and reservoir layers. The model applies the Dirichlet boundary condition, maintaining constant pressure at the outermost grid blocks. The geothermal gradient is set equal to 30 °C/km, and the surface pressure and temperature are equal to 1 atm and 25 °C, respectively. The reservoir pressure and temperature are 445 bar and 100 °C, respectively. CO<sub>2</sub> is injected at a constant mass injection of 1.5 Mt/y (~48 kg/s).

The injection model strategies are simulated utilizing TOUGH3 for thermal and compositional simulation. To properly handle the  $CO_2$  phase transition during its injection, the ECO2M tabulated Equation of State (EOS) module of TOUGH3 is used. Also, this work uses coupled wellbore-reservoir modeling to capture the interaction between fluid flow inside the well and within the reservoir. This integrated approach enables a more realistic evaluation of how each injection strategy impacts injectivity and storage performance under different temperature and pressure conditions.

### b) Economic Evaluation Methodology

In this work, an economic analysis is performed to calculate the yearly cost of each injection. For this, we take into account that the average energy prices in the state of Louisiana are 0.161 USD/kWh according to the U.S. Bureau of Labor Statistics. Costs related to  $CO_2$  injection come with heating/cooling and compression/pumping. Assuming that  $CO_2$  arrives at the storage site at 30 °C and 80 bar, it needs to be heated 5 °C for Tinj = 35 °C, and cooled down 10 °C and 25 °C for Tinj = 20 °C and Tinj = 5 °C, respectively.

To estimate the costs related to heating or cooling of CO<sub>2</sub>, first, we need to calculate the amount of energy required to achieve the desired injection temperature, using the heat equation as seen in (1). Then, we must account for compression (SC-CO<sub>2</sub>) or pumping (liquid CO<sub>2</sub>). To calculate the amount of energy required for pumping, we use the following equation as seen in

(2). In contrast, we use the equation in (3) to estimate the energy required for compression.

$$Q = mc_p \Delta T \tag{1}$$

$$W_{pumping} = \frac{m\Delta p}{\eta \rho} \tag{2}$$

$$W_{compression} = \frac{m\Delta h}{\eta} \tag{3}$$

Results

When CO<sub>2</sub> flows from the surface to the storage formation through the well, it experiences several thermodynamic changes, with convection being the dominant mechanism [6]. Since CO<sub>2</sub> is injected at a lower temperature than the reservoir, it experiences a significant temperature drop as it travels downwards. This cooling effect can have an impact on storage efficiency since it will determine the phase at which the CO<sub>2</sub> reaches the reservoir. Figure 1 shows the bottom hole pressure and temperature evolution with time for all three cases. For Tinj = 35 °C, the CO<sub>2</sub> reaches the reservoir in its supercritical state (BHT = 52 °C). In comparison, at Tinj = 5 °C, the CO<sub>2</sub> reaches the storage formation in its liquid state since the temperature is below the CO<sub>2</sub> critical value (BHT = 11 °C). However, as seen from Figure 1, when injecting CO<sub>2</sub> at 20 °C, it reaches the reservoir near its critical state (BHT = 30 °C).



Figure 1 Pressure and temperature evolution at the wellhead and bottomhole for different injection temperatures

As the CO<sub>2</sub> flows through the porous media, the temperature in the near wellbore-area decreases due to the combined effects of the Joule-Thomson effect and water vaporization. However, as the CO<sub>2</sub> travels further from the wellbore into the reservoir, it gradually heats up again due to heat exchange with the surrounding rock, eventually reaching the initial reservoir temperature. The CO<sub>2</sub> will also experience a small heating effect due to its exothermic dissolution into the resident brine (about 1-5 °C) [7, 8]. This behavior can be observed in Figure 2.



Figure 2 Temperature distribution along the reservoir (TOUGH3)

Figure 2 shows that, after 10 years of injection,  $CO_2$  remains in liquid state at the top and bottom layer of the reservoir, reaching approximately 45 m and 170 m, respectively for an injection temperature of 20 °C. In contrast, for Tinj = 5 °C, liquid CO<sub>2</sub> reaches a distance away from the wellbore of about 110 m at the top layer and 230 m at the bottom. For the case of Tinj = 35 °C, as observed, CO<sub>2</sub> will always be in the supercritical state. However, when the injection temperature is 20 °C and 5 °C, the CO<sub>2</sub> initially exists in liquid state in the proximity of the well, transitioning to a supercritical state further from the wellbore. When CO<sub>2</sub> is in liquid state, its density is higher compared to supercritical CO<sub>2</sub>. Therefore, the buoyancy effects are minimized.

As seen from Figure 3, CO<sub>2</sub> escapes through the caprock due to its buoyancy. In all cases, regardless of its injection temperature, there is a minor CO<sub>2</sub> breakthrough (about 0.1 saturation) along the entire radial extent of CO<sub>2</sub> migration in the top reservoir layer. Additionally, for the case of Tinj = 35 °C, it can be

noticed that, after 10 years of injection, the CO<sub>2</sub> ascends 15 meters vertically above the caprock-reservoir contact and migrates radially, reaching a saturation level of about 0.35. In comparison, for injection temperatures of 20 °C and 5 °C, the CO<sub>2</sub> travels shorter vertical distances of 14 m and 12 m above the top reservoir layer, respectively, and reaches a radial distance of about 28 meters with a saturation of 0.3 m.



Figure 3 CO<sub>2</sub> saturation distribution along the reservoir (TOUGH3)

From the technical point of view, it would be best to inject CO<sub>2</sub> at 5 °C to prevent a significant amount of CO<sub>2</sub> from escaping through the caprock. However, this decision must come along with economic feasibility. As stated by the Global CCS Institute, SC-CO<sub>2</sub> is generally preferred for transportation through pipelines due to its liquid-gas combined unique properties [9]. Therefore, injecting at 20 °C or 5 °C would require extra energy to cool the transported CO<sub>2</sub> down. Assuming that CO<sub>2</sub> arrives at the storage site at 30 °C, and considering 0.161 USD/kWh as the average energy price in Louisiana according to the US Bureau of Labor Statistics [10], the total costs for injecting CO<sub>2</sub> at 5 °C, 20 °C, and 35 °C over 10 years at a constant injection rate of 1.5 Mt/y are 90.93, 36.19, and 28.47 million USD.

#### Discussion

When it comes to sequestering  $CO_2$  underground, understanding the hydro-thermal phenomena the  $CO_2$  undergoes is vital to minimize any risk of leakage. These phenomena, including phase transitions, heat transfer with the surroundings, influence the  $CO_2$  behavior in the storage formation. One critical aspect is the role of injection temperature. This study shows that reducing the injection temperature can help limit the  $CO_2$  breakthrough and vertical migration, but it might also cause hydrate formation, affecting the injectivity, and an increment in operational costs.

## Conclusions

This study demonstrates that reducing  $CO_2$  breakthrough is possible by lowering the injection temperature. However, it is important to note that reducing the injection temperature at the surface also results in the bottomhole temperature reduction. Therefore, care must be taken to avoid excessive cooling of the  $CO_2$  at the surface since it could lead to hydrate formation (which can negatively impact injectivity) and geomechanical issues (which can lead to fracturing the rock). Furthermore, reducing further the injection temperature can significantly increase operational costs. As a result, determining the optimal injection strategy is a complex decision that must balance both technical and economic considerations. On the technical part, several factors must be accounted for, including reservoir heterogeneity, fractures, which can serve as leakage pathways for the  $CO_2$ . Striking a balance between reducing leakage risks and maintaining operational feasibility is key to successful  $CO_2$  long-term underground sequestration.

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