CO₂ storage in heterogeneous aquifer: A study on the effect of temperature and mineral precipitation

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Abstract. CO₂ storage in suitable geologic media has been recognized as a major strategy taken to have a carbon free environment. This practice can be done in depleted reservoirs as well as brine aquifers where sufficient storage capacity is available to hold carbon dioxide for thousands of years. Storage in an aquifer is often achieved through four trapping mechanisms, among which capillary trapping is a rapid and effective phenomenon. Although, there have been studies pointing out the relationships of different storage related factors with capillary trapping, more studies are still required to recognize other parameters linked to this effective trapping mechanism. The aim of this paper is to evaluate the effect of temperature and mineral precipitation on trapping mechanisms of heterogeneous aquifer. A dynamic numerical simulation was run by the commercial reservoir simulator Eclipse300 to simulate 30 years of CO₂ injection. A synthetic but realistic model of a geologic formation was considered to evaluate the efficiency of trapping mechanisms under different temperature and mineral precipitation conditions. The results obtained indicated that trapping mechanisms are affected by both temperature and mineral precipitation in a short and long terms - temperature is indirectly affecting the trapping ability regardless of the precipitation effect. However, precipitation have a severe impact on injectivity as well as trapping mechanisms in the long term. Although some practical conclusions were drawn, the results obtained and presented in this study may need experimental verification before taking into serious consideration.

1. Introduction
CO₂ storage in suitable geologic media has been recognized as a major strategy taken in recent years to reduce the amount of greenhouse gas released into the atmosphere [1, 2]. The storage capacity and characteristics of saline aquifers make them a suitable candidate for CO₂ storage. A saline aquifer is often composed of sandstone and carbonates and located at the depth of 800 to 3000 m. They offer natural immobilization of CO₂ due to having favourable chemistry, porosity and subsurface conditions [3]. The subsurface temperature and pressure conditions at these depths are within 25°C to 200°C and 10 bar to 300 bar, respectively [4], which ensures that CO₂ appears in its supercritical form and occupies a lesser space for a higher volume injection [5].

Four trapping mechanisms are often generated due to injection of CO₂ in geologic structures [6]: i) structural trapping, ii) capillary trapping, iii) solubility trapping, and, iv) mineral trapping [7, 8]. These trapping mechanisms are significantly affected by subsurface pressure and temperature conditions as well as rocks and fluids characteristics [9]. Comparatively, capillary trapping is arguably recognized as a rapid, effective and safe mechanism to immobilize CO₂ in subsurface formations [10-13]. Moreover,
it may offer more CO₂ entrapment compared to other trappings such as solubility and mineral trappings [14].

During the injection period, rapid and significant changes in hydrodynamic, chemical, thermal, and mechanical aspects of subsurface formations are observed [9]. The chemical activity of CO₂ with the resident fluid and minerals creates precipitation which affects different aspects of storage sites [15, 16]. For instance, injection of CO₂ causes mineral precipitations near the wellbore region which ultimately impacts the injectivity [17,18]. Saeedi [19] briefly explained the geochemical reactions taking place during the process of mineral precipitation. These reactions might be sensitive to pressure and temperature and are often faster in carbonates compared to siliciclastic rocks [15]. Tables 1 summarises studies carried out in recent years to evaluate the effect of dissolution/precipitation on rocks properties.

The aim of this paper is to evaluate the effect of temperature and mineral precipitation on structural (free), capillary (residual) and solubility (dissolved) trapping mechanisms of heterogeneous aquifers. A numerical simulation was run for this purpose and trapping mechanisms were evaluated in terms of temperature and mineral precipitation.

| Contributors        | Remarks/Approach                                             |
|---------------------|--------------------------------------------------------------|
| Bacci et al., 2011  | Dissolution/precipitation dependent on pressure and temperature have the significant effect on injectivity/ Experimental & Simulation |
| Liu et al., 2013    | Precipitation process result permeability change/ Experimental |
| Peysson et al., 2014| Drying and mineral precipitation due to dry gas injection reduce the rock permeability/ Experimental |
| André et al., 2014  | Dissolution/precipitation result change in permeability and injectivity caused by the interplay of capillary forces and the salinity of the initial brine / Experimental & Simulation |
| Zheng et al., 2015  | Deformation of quartz–feldspar–detrital sandstone occurs by dissolution effect due to NaCl solution and CO₂–NaCl solution / Experimental |
| Ott et al., 2015    | Drying and mineral precipitation during dry CO₂ injection affect the permeability in unimodal sandstone / Experimental |

2. Simulation approach

CO2STORE dynamic numerical modeling [20, 21], which is a part of Schlumberger Eclipse300 Simulator, was used for the purpose of this study to estimate CO₂ trapping during injection. To make a realistic and static geologic reservoir sophisticated for a storage job, grid geometry, porosity and permeability properties were borrowed from Juanes Research Group [22].

The geometry of the 3D model was consisted of a dome in the center and five layers of fluvial sands and shales. The average reservoir thickness was 15 m and the model was discretized into 19x28x5 grid blocks, of which 1761 blocks were active. The X and Y dimension of each block was 180 meter and the permeability anisotropy ratio was about 3. The injection well was assumed to be located in the middle of the model. Two cases with and without precipitation were considered having an average porosity and permeability of 20% and 100 mD, respectively. The porosity distribution map obtained is given in figure 1. For the sake of simplicity and to ensure that CO₂ appears under supercritical condition, top depth of the reservoir was set to be 840m. The rock compressibility used in this study was 5 ×10⁻⁴ bar⁻¹.
The liquid phase in both cases was having H₂O (0.9258 mole fraction), CO₂ (0 mole fraction), NaCl (0.0741 mole fraction) and CaCl₂ (0.0001 mole fraction). NaCl and CaCl₂ were present in the aqueous and the solid phases according to following reactions [20, 21]:

\[
\begin{align*}
H_2O & \rightleftharpoons H^+ + OH^- \quad (1) \\
CO_2 + H_2O & \rightleftharpoons HCO_3^- + H^+ \quad (2) \\
HCO_3^- & \rightleftharpoons CO_3^{2-} + H^+ \quad (3) \\
CaCl^+ & \rightleftharpoons Ca^{2+} + Cl^- \quad (4) \\
NaCl(s) & \rightleftharpoons Na^+ + Cl^- \quad (5) \\
CaCl_2(s) & \rightleftharpoons CaCl^+ + Cl^- \quad (6)
\end{align*}
\]

Before injection, the initial pressure was set to be 278 bar (4032 psia) at the top of the structure while the temperature was varied from 49 to 148°C, ensuring that CO₂ appears under supercritical state. Three phases of CO₂ rich (gas), H₂O rich (water phase) and a solid were taken into consideration. The diffusion function was also introduced to cover the effect of CO₂ interaction with formation water. In particular, this diffusion results in transportation of CO₂ in each grid block causing more CO₂ dissolution within the medium. It should be noticed that the simulator generally calculates the mutual solubility of CO₂ and H₂O to match it with experimental data under a typical storage temperature and pressure condition. The effect of mineral and CO₂ was considered using the Ezrokhi’s method (Eq. (7)) to calculate the water density [20, 21].

\[
\log_10(\rho) = \log_10(\rho_o(P,T)) + \sum_i A_i(T) w_i \quad (7)
\]

In Eq. (7) \( w_i \) is the weight fraction of the non-water component \( i \), \( \rho_o(P,T) \) is the density of pure water, \( A_i(T) \) is the temperature coefficients determined by Eq. (8) in °C.
\[ A_f(T) = a_{0f} + a_{1f}T + a_{2f}T^2 \] (8)

The functions to consider the effect of solid concentration on mobility in case of precipitation effect near the wellbore and overburden pressure effect were introduced. The relative permeability and capillary pressure curves were generated using the Corey and van Genuchten correlations (i.e., Eq. (9-11)) [23].

\[ k_{rw} = \left( \frac{S_w - S_{wr}}{1 - S_{wr}} \right)^4 \] (9)

\[ k_{rg} = k_{rg\text{-max}} \left( 1 - \frac{S_w - S_{wr}}{1 - S_{wr} - S_{gr}} \right)^2 \] (10)

\[ P_c = P_o \left( \frac{S_w - S_{wr}}{1 - S_{wr}} \right)^{\frac{1}{2} - 1} - 1 \] (11)

where \( k_{rw} \) and \( k_{rg} \) are the water and gas relative permeability, respectively, \( S_w \) is the water saturation, \( S_{wr} \) is the residual water saturation, \( S_{gr} \) is the residual gas saturation, \( k_{rg\text{-max}} \) is the maximum gas relative permeability, \( P_c \) is the capillary pressure, \( P_o \) is the capillary entry pressure and \( \lambda \) is the capillary pressure exponent.

In both cases, supercritical CO\(_2\) was injected into the four layers of the storage formation at 5.66 million sm\(^3\)/day (standard cubic meter per day) equivalent to 200 million standard cubic feet per day for 30 years. The CO\(_2\) was injected at the pressure below the facture pressure of the seal (i.e., 500 bar) and maximum bottom-hole pressure (i.e., 483 bar). The values of the parameters used in this studies are reported in table 2.

### Table 2. Scenarios adopted in simulations cases

| Case. No. | CASE A | CASE B |
|-----------|--------|--------|
|           | Temperature effect |        |
|           | Without precipitation | With precipitation |
| 1         | 49\(^\circ\)C (Base Case) | 49\(^\circ\)C |
| 2         | 60\(^\circ\)C | 60\(^\circ\)C |
| 3         | 71\(^\circ\)C | 71\(^\circ\)C |
| 4         | 82\(^\circ\)C | 82\(^\circ\)C |
| 5         | 93\(^\circ\)C | 93\(^\circ\)C |
| 6         | 104\(^\circ\)C | 104\(^\circ\)C |
| 7         | 115\(^\circ\)C | 115\(^\circ\)C |
| 8         | 126\(^\circ\)C | 126\(^\circ\)C |
| 9         | 137\(^\circ\)C | 137\(^\circ\)C |
| 10        | 148\(^\circ\)C | 148\(^\circ\)C |

Out of two cases, a base case (Case A) was developed without considering the effect of precipitation. This case was simulated at the temperature of 49\(^\circ\)C and the pressure of 278 bar with a 5.66 million sm\(^3\)/day injection rate. Nine subcases were then generated by changing the temperature from 49\(^\circ\)C to 148\(^\circ\)C. In the second scenario (Case B), similar subcases were simulated under the same conditions by
taking the precipitation effect into consideration. At the end, the results obtained from this scenario were compared with those of the Case A to evaluate the importance of precipitation and temperature on structural, capillary and solubility trappings.

3. Results Analysis
The results obtained from running the simulation for both cases indicated the effect of the temperature on the injection rate and pressure buildup as seen in figures 2. In the Case A, the injection rate stabilizes for 20 years and then declines rapidly. In fact, the temperature revealed to have an indirect relationship with the injection rate after 20 years. The reservoir pressure was increasing because of the continuous injection of CO₂ and ultimately affecting the trend of the injection rate. It was also found that temperature is changing by the variation of the reservoir pressure. This might be due to changes in the density of CO₂. A very same trend was observed for the injection rate and pressure build-up of the Case B. It should be noticed that the injection rate begins to decline after 11 to 15 years in different cases depending on the temperature.

Figure 2. Effect of temperature on injection rate (FGIR) and on pressure buildup (FPR) for Case A and Case B.
Considering the relationship of temperature with the free, residual and dissolved \( \text{CO}_2 \) saturations in the absence and presence of precipitation, it can be concluded that a medium with a higher temperature would trap lesser amount of \( \text{CO}_2 \) compared to a low-temperature medium (See figure 3a, b). In fact, temperature is showing an inverse effect on capillary and solubility trapings. This effect becomes significant after five years of injection and has not been observed in any of the earlier studies. However, precipitation reduces the trapping ability in later stages.

**Figure 3.** a) Effect of temperature on trapping mechanisms considering no precipitation effect (Case-A-left); b) Effect of temperature on trapping mechanisms considering precipitation effect (Case-B-right).
This was aligned with the statement of Metz et al. [24] where it was indicated that low-temperature gradient basins are more favourable than the high-temperature ones due to achieving a high density CO₂. It was also found that the ganglia of CO₂ is left behind in the pore space and surrounded by water during the capillary trapping. It might be possible that the temperature variation is affecting the water movement and lowering the capillary trapping ability causing the free gas saturation to be enhanced. It could also be due to the inverse effect of temperature on CO₂ solubility in brine which causes CO₂ plume to flow at a high rate with a lesser solubility trapping [25].

Comparing the results obtained from the Case A at two different temperatures (49°C and 148°C) with those of the Case B revealed a quantitative impact of precipitations on the capillary trapping including structural and solubility trappings as shown in figure 4. It was also revealed that the capillary and solubility trappings would be in their maximum and minimum range at low temperatures while the structural trapping is more active at high temperatures (See figure 5). However, it was found that the residual and dissolved gas saturations of CO₂ significantly reduce due to mineral precipitation after 50% of injection. A minor impact was observed on the free gas saturation as well. On the other hand, it was concluded that precipitation may have impacts on the near wellbore injectivity due to the variation of permeability and capillary pressure forces [17, 18]. It was also noticed that precipitations may change capillary, structural (free) and solubility trappings of the storage site in the long term.

![Graphs showing trapping mechanisms](image-url)

**Figure 4.** Comparison of trapping mechanisms at 49°C and 148°C with and without precipitation effect.
Figure 5. Comparison of CO₂ volume in total injected and different phases considering temperature and precipitation effects

4. Conclusions
This study investigated the effect of temperature and precipitation on trappings of CO₂ during a storage practice in aquifers. The results obtained indicated that temperature is indirectly affecting the trapping ability regardless of the precipitation effect. Precipitation, however, may have a severe impact on rock properties, injectivity, as well as trapping mechanisms in the long term. It is suggested to have the precipitation effect included in the dynamic modelling to mimic the real scenario of CO₂ injection.

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