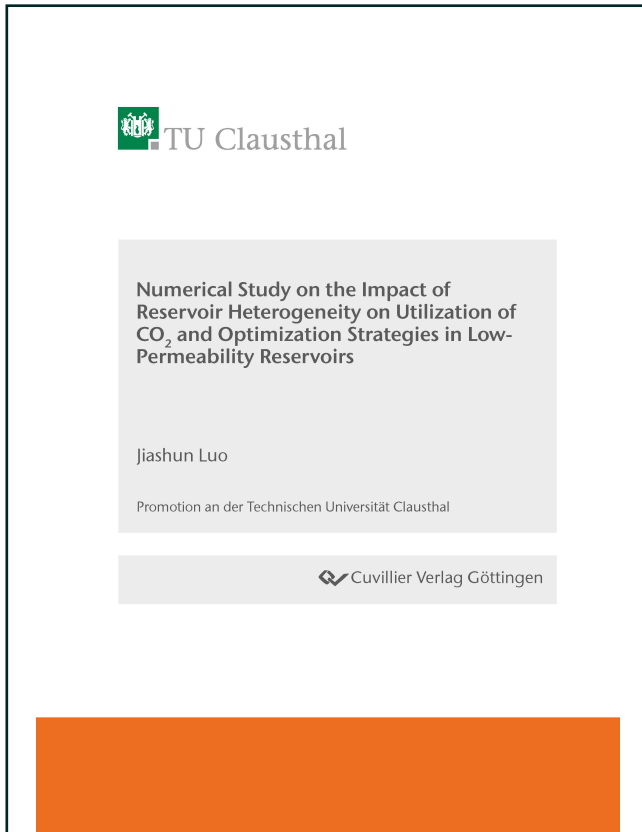




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# Numerical Study on the Impact of Reservoir Heterogeneity on Utilization of CO<sub>2</sub> and Optimization Strategies in Low-Permeability Reservoirs



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## 1. Introduction

As the dominant greenhouse gas, carbon dioxide (CO<sub>2</sub>) emission was the major cause that accounted for global warming and climate change. Figure 1-1(a) shows that the amount of carbon dioxide in the atmosphere has increased along with human emissions over the past two hundred years (Lindsey, 2023). The amount of CO<sub>2</sub> in the atmosphere has indeed increased along with human emissions since the start of the Industrial Revolution in the 18th century. Emissions of CO<sub>2</sub>, primarily from the burning of fossil fuels such as coal, oil, and natural gas, have risen steadily over time. The increase in CO<sub>2</sub> in the atmosphere is a major contributor to climate change and global warming. The Intergovernmental Panel on Climate Change (IPCC) estimates that human activities, primarily the burning of fossil fuels, are responsible for about 78% of the increase in CO<sub>2</sub> in the atmosphere since 1750 (Al - Ghussain, 2019). The rise in CO<sub>2</sub> emissions has been particularly steep since the mid-20th century, with emissions increasing from about 5 billion tons per year in the mid-20th century to more than 35 billion tons per year by the end of the century (Levine and Steele, 2021). Figure 1-1(b) shows the total fossil energy consumption since the 1850s (Gilmore et al., 2022; Höök et al., 2012). The concentration of CO<sub>2</sub> in the atmosphere was relatively stable at around 280 parts per million (ppm) for thousands of years before the Industrial Revolution. However, since the start of the Industrial Revolution in the 18th century, human activities such as the burning of fossil fuels and deforestation have resulted in a significant increase in CO<sub>2</sub> emissions. As a result, the concentration of CO<sub>2</sub> in the atmosphere has risen from below 300 ppm in pre-industrial times to over 410 ppm in recent years (Drag et al., 2020). This increase in CO<sub>2</sub> concentration is a major contributor to climate change and global warming, as CO<sub>2</sub> acts as a greenhouse gas, trapping heat in the atmosphere and leading to an increase in average global temperatures. It can be seen that the global warming is closely related to human activities, especially the massive use of fossil fuels, which leads to a continuous increase in atmospheric CO<sub>2</sub> concentration. Noticeably, emission reduction has drawn more and more attention in the international community during the past decades.

In order to deal with these challenging threats, human society worldwide is actively responding and taking relevant measures. As a vital positive result and guideline for action, the Paris Agreement was reached and proposed to control the temperature increase within 1.5-2 °C by reducing carbon emissions and achieving climate and carbon neutrality (Rogelj et al., 2016; Stua et al., 2022). In order to fulfill the commitments, carbon reduction techniques such as carbon capture, utilization, and carbon storage (CCUS) have received significant interest



storage. The mechanisms of CO<sub>2</sub> storage in terrestrial geologic formation include structural trapping, residual trapping, solubility trapping, and mineral trapping (Figure 1-2) (Luo et al., 2023) (Alanazi et al., 2023). Structural traps refer to the accumulation of carbon dioxide in deep saline formations and depleted oil and gas reservoirs. The injected CO<sub>2</sub> usually moves upward, however, due to the existence of caprocks (Knopf and May, 2017), CO<sub>2</sub> leakage can be effectively avoided. As a physical trapping of CO<sub>2</sub> in the pores, structural trapping is the mechanism that traps the greatest amount of CO<sub>2</sub>. Residual trapping involves the transport of carbon dioxide and water (Ge et al., 2022). As a response to the CO<sub>2</sub> injection, the water saturation of the pores will decline while the CO<sub>2</sub> saturation rises, leading the carbon dioxide to be trapped in the pore and pore throat space between the rock grains. In solubility trapping, a portion of the injected CO<sub>2</sub> will dissolve into the brine water leading to solubility trapping, which depends on the water salinity, reservoir temperature and pressure (Gilfillan et al., 2009). It should be pointed out that since the buoyancy is caused by density contrast, CO<sub>2</sub> could migrate upward in the terrestrial storage reservoirs consequently, hence an impermeable sealing layer is indispensable, such as cap rock. Mineral trapping is a geochemical reaction process, in which dissolved CO<sub>2</sub> initiates reactions with the formation rock minerals leading to the formation of carbonate minerals (Al-Khdheawi et al., 2023; Xu et al., 2004).

## 1.1 Properties of CO<sub>2</sub>

Pure CO<sub>2</sub> is a colorless, odorless, inert, and non-combustible gas (Cho, 2015). The molecular weight at standard conditions is 44.010 g/mol, which is one and a half times higher than air (Saravanan et al., 2021). CO<sub>2</sub> is a naturally occurring compound that exists in three different states of matter: solid, liquid, and gas. Its pressure and temperature determine the phase transitions of CO<sub>2</sub>, and the phase diagram for CO<sub>2</sub> illustrates the conditions under which each state of matter exists. At low temperatures and pressures, CO<sub>2</sub> exists in a solid state, commonly known as dry ice. As the temperature increases, the solid CO<sub>2</sub> will eventually transition to a liquid state. This process is known as sublimation, and it occurs at a temperature of -78.5°C and a pressure of 0.518 MPa (Yamasaki et al., 2017). As the temperature and pressure continue to increase, the liquid CO<sub>2</sub> will eventually transition to a gaseous state. This process is known as vaporization, and it occurs at a temperature of -0.4°C and a pressure of 0.1013 MPa (Zhao and Lvov, 2016). The phase diagram for CO<sub>2</sub> is a graph that shows the conditions under which each state of matter exists. It is typically represented as a pressure-temperature plot, with lines separating the solid, liquid, and gaseous regions. The triple point of CO<sub>2</sub> is at a temperature of -78.5°C and a pressure of 0.518 MPa, where all three states of matter exist in equilibrium. It's



phases coexist in equilibrium. A technique for constructing the CO<sub>2</sub> phase diagram would include charting the pressure and temperature conditions at which the three phases of matter (solid, liquid, and gas) exist in equilibrium. This is possible by using the Gibbs free energy and Clausius-Clapeyron equation. The Gibbs free energy is a thermodynamic quantity that indicates a system's work-doing capacity. In the case of CO<sub>2</sub>, the Gibbs free energy is dependent on the system's pressure and temperature, and it varies during a phase transition. The Clausius-Clapeyron equation is a connection between the Gibbs free energy and the system's pressure and temperature. It asserts that the change in Gibbs free energy per unit volume is proportional to the change in both temperature and pressure. The slope of the phase boundary on the pressure-temperature phase diagram may be calculated using this equation. To produce the CO<sub>2</sub> phase diagram, it would be necessary to measure the system's pressure and temperature at many sites and then utilize these measurements to depict the phase boundaries. The Clausius-Clapeyron equation may then be utilized to compute the slope of the phase boundaries, which can be used to connect the data points and finish the phase diagram. It is also feasible to forecast the phase transitions of CO<sub>2</sub> using theoretical calculations, such as Density Functional Theory or Molecular Dynamics, and then create a phase diagram based on these predictions. It is crucial to remember that the phase diagram for CO<sub>2</sub> is a graph that depicts the conditions under which each form of matter exists and that it may alter under different situations, such as impurities or varying pressure and temperature ranges.

## **1.2 Types of geological utilization of CO<sub>2</sub>**

CO<sub>2</sub> utilization involves the direct conversion of CO<sub>2</sub> into valuable products, such as fuels or chemicals, and its use as a displacement/energy exchange medium in the geologic energy industry (Huang and Tan, 2014; Zhu, 2019). For example, CO<sub>2</sub> is used as a working fluid and injected into underground formations such as oil and gas reservoirs, aquifers, and hot dry rocks to enhance the production of underground energy reserves through displacement, dissolution, heat transfer, and fracture generation, while simultaneously achieving underground CO<sub>2</sub> storage (Li and Zhiwei, 2021; Xie et al., 2014). Consequently, CCUS technology in the conventional geological energy industry mainly includes CO<sub>2</sub>-EGR (such as natural gas, coalbed methane, and shale gas), CO<sub>2</sub>-EOR, CO<sub>2</sub>-enhanced water recovery (CO<sub>2</sub>-EWR), CO<sub>2</sub> fracturing, CO<sub>2</sub> geothermal systems, and CO<sub>2</sub> in-situ uranium leaching (CO<sub>2</sub>-IUL) (Figure 1-4).







be seen that the viscosity and density decrease with the increase in  $\text{CO}_2$  concentration (Hou et al., 2021a). Rezk and Foroozesh studied the phase behavior and mutual interactions between light crude oil and  $\text{CO}_2$  under high pressures and high temperatures conditions. The swelling factor (SF) measurements demonstrated an increasing trend with pressure up to a specific extraction pressure. Beyond this point, the SF began to decrease, even reaching values below one. Furthermore, the measurements of oil viscosity revealed that the dissolution of  $\text{CO}_2$  in the oil sample led to a significant reduction in the mixture's viscosity, by up to 61% (Rezk and Foroozesh, 2019). Mansour et al. conducted a study on the application of ( $\text{CO}_2$ ) miscible flooding in Egyptian oil fields using swelling experiments. In Figure 1-6 (Mansour et al., 2019), the results of ( $\text{CO}_2$ )-oil viscosity versus ( $\text{CO}_2$ )-saturation pressure at reservoir temperature are shown for ten wells: S.ID (1), S.ID (2), S.ID (3), S.ID (4), S.ID (5), S.ID (6), S.ID (7), S.ID (8), S.ID (9), and S.ID (10). Each solid line in the figure represents a sample, illustrating the variation in ( $\text{CO}_2$ )-oil viscosity with different percentages of carbon dioxide ( $\text{CO}_2$ ) from saturation pressure (bubble-point pressure) to minimum miscibility pressure (MMP). The findings indicate that the addition of carbon dioxide ( $\text{CO}_2$ ) significantly reduced the ( $\text{CO}_2$ )-oil viscosity in each sample. Wei et al. conducted a series of experiments on cyclical  $\text{CO}_2$  injection into tight reservoirs in Lucaogou, Jimusar Basin. With PVT experiments and minimum miscibility pressure (MMP) measurement, they observed that as the  $\text{CO}_2$  concentration increased,  $\text{CO}_2$  dissolving in tight oil tended to be saturated, and the density and viscosity of tight oil decreased slowly (Wei et al., 2017). Furthermore, they suggested that even if the pore pressure was higher than MMP, the injected  $\text{CO}_2$  may not be able to form a miscible phase with crude oil due to the extremely tight porous media.

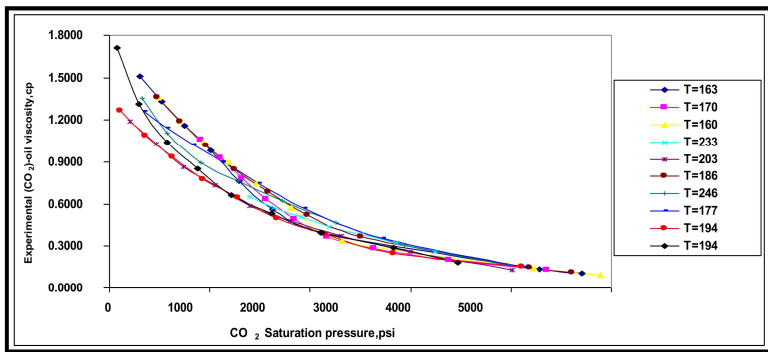


Figure 1-6. ( $\text{CO}_2$ )-Oil viscosity and ( $\text{CO}_2$ )-saturation pressure.

## (2) Oil swelling

CO<sub>2</sub> injection exhibits a notable swelling effect on crude oil (Sohrabi et al., 2009). Following CO<sub>2</sub> injection, as it dissolves in the oil, the specific volume, reservoir volume factor, and compressibility coefficient of the crude oil increase. The expansion of the oil occurs due to CO<sub>2</sub> molecules entrapping tiny air bubbles within its molecular structure, creating narrow pathways that improve viscosity and facilitate flow. As a result, the compressibility of the crude oil and oil well productivity are enhanced.

Oil swelling also leads to the breakdown of the oil structure, contributing to viscosity reduction (Salleh et al., 2019). The relatively weak intermolecular forces holding oil molecules together are weakened by the introduction of CO<sub>2</sub> molecules, leading to a decrease in oil viscosity. The extent of swelling is directly proportional to the amount of injected CO<sub>2</sub>, reservoir pressure and temperature, and the oil's composition. Oils with more polar or polar functional groups exhibit greater expansion compared to nonpolar oils. Furthermore, increasing injection pressure and/or temperature also enhances the magnitude of swelling. The rate at which the oil expands is influenced by various variables, including the oil's composition, pressure, temperature, and CO<sub>2</sub> injection rate. Habibi et al. visualized the CO<sub>2</sub>-oil interface at 50 °C and 2000 psi in the visual cell to investigate the CO<sub>2</sub>-oil interactions at reservoir conditions. Figure 1-7 presents the CO<sub>2</sub>-oil interface at the specified reservoir temperature. In panel (a), the initial setup involved 235mL of oil with a molar quantity of 0.78, placed in a cell at atmospheric pressure, without the presence of CO<sub>2</sub>, and maintained at 50 °C. Subsequently, in panel (b), 6.4 mol of CO<sub>2</sub> was injected into the system to increase the pressure from 0 to 13.8 MPa. Notably, as depicted in panel (c), after a duration of 5 hours, further CO<sub>2</sub> dissolution into the oil phase resulted in the significant swelling of the oil (Habibi et al., 2017). These observations provide compelling evidence of CO<sub>2</sub>'s remarkable ability to dissolve into and expand the oil under reservoir conditions, which is of great importance for understanding the behavior of CO<sub>2</sub>-oil interactions in such settings. Su et al. investigated the effects of CO<sub>2</sub> on crude oil and found that increasing the mole fraction of CO<sub>2</sub> from 0 to 0.45% resulted in a considerable increase in specific volume, reservoir volume factor, and compressibility coefficient of the crude oil (Yang et al., 2022). Similarly, Zhang et al. injected CO<sub>2</sub> into crude oil, leading to an expansion coefficient increase from 1.00 to 1.19 after adding 45% CO<sub>2</sub>. Additionally, Pu et al. conducted visual tests on the CO<sub>2</sub> huff-n-puff process for enhanced oil recovery in tight reservoirs, affirming that the oil swelling factor and CO<sub>2</sub> solubility both increased with pressure (Pu et al., 2016). These studies collectively provide valuable insights into the significant effects of CO<sub>2</sub> on crude oil expansion and its implications for enhanced oil recovery in various reservoir conditions.

