Energy Proceedings
Vol 56, 2025
ISSN 2004-2965

Study on Phase Changes and Migration Characteristics of CO_2 during Leakage in Deep Saline Aquifers for Carbon Sequestration

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ABSTRACT

In response to global climate change, carbon capture and storage (CCS) has become a key strategy, opening a new chapter in the use of deep underground space. Deep saline aquifers, with their extensive distribution and substantial storage potential, are ideal for CO_2 storage. However, the risks of geological storage, including CO_2 leakage and potential environmental impacts, cannot be ignored. This study aims to investigate the migration behavior, distribution patterns, and phase changes of CO_2 in saline aquifers and their cap rocks through reservoir numerical simulation. A two-dimensional reservoir model was constructed, incorporating a highly permeable pathway to simulate a fault as a leakage channel, in order to study the phase change and longitudinal migration characteristics of CO_2 during the leakage process. The simulation results indicate that during the upward leakage process along the fault, CO_2, under the influence of buoyancy, tends to enter the upper strata. As it migrates upward, some of the CO_2 is affected by rock adsorption and becomes trapped at the interface between the fault and the overlying dense rock of the saline aquifer, distributing stably. It is noteworthy that during the leakage process, CO_2 primarily migrates in a supercritical state; however, when it reaches a critical depth, it transitions to a liquid phase. This phase change from supercritical state to liquid state can impact the storage capacity and pressure, thereby affecting the stability of the formations.

Keywords: CO_2 storage, Saline aquifer; Phase change; Migration; Leakage

1. INTRODUCTION

The massive emission of CO_2 is one of the main factors contributing to global warming, which poses a serious threat to human life and property. Therefore, reducing the concentration of CO_2 in the atmosphere has become a critical scientific issue to address. In this regard, CO_2 capture and storage (CCS) technology is considered one of the key technologies to combat global climate change. Furthermore, this emerging technology has also sparked widespread interest in the development and utilization of deep underground spaces. Deep saline aquifers, due to their wide distribution and enormous storage potential, are regarded as important reservoirs for implementing CO_2 sequestration [1,2].

Although CO_2 sequestration has many benefits, the risks and potential hazards associated with its leakage cannot be ignored. Human production activities can compromise the integrity of rock formations, and the presence of unsealed faults, natural fractures, and other factors can create pathways for CO_2 to migrate upwards. Injected CO_2 in saline aquifers may escape through these leakage pathways due to various influences, such as buoyancy, leading to a certain degree of leakage risk associated with CO_2 sequestration.

Research on the deep underground utilization of CO_2 geological sequestration started early internationally, resulting in industrial or demonstration projects such as Norway's North Sea Sleipner project [3,4,5] and Australia's CO_2CRC Otway demonstration project, aimed at monitoring and research [6]. China's first deep saline aquifer CO_2 geological sequestration project, the National Energy Group Ordos Carbon Capture and Storage (CCS) Demonstration Project (also known as the Shenhua CCS Demonstration Project), integrates the previously proposed concept model for CO_2 geological sequestration underground utilization space evaluation

This is a paper for International CCUS Conference 2024 (ICCUS 2024), June 20-22, 2024, Beijing, China.

[7] and the latest insights into site characterization and CO_2 underground migration monitoring and prediction of the Ordos CCS demonstration project [8]. This study aims to provide reference for the approval and management policy formulation of underground utilization space for CO_2 sequestration projects by investigating the deep saline aquifer CO_2 geological sequestration underground utilization space evaluation methods.

In 2005, Hassanzadeh et al. studied the impact of different formation parameters on CO_2 sequestration in saline aquifers [9]. In 2008, Behzadi et al. simulated a quasi-one-dimensional model to validate the influencing factors and provided examples illustrating the mechanisms controlling reservoir leakage rates and fluid attenuation through shallower formations [10]. In 2011, Zeidouni et al. proposed an analytical model to evaluate the pressure changes in the overlying aquifer caused by leakage from the storage aquifer [11]. Oruganti et al. introduced the concept of overpressure critical lines (CoP) as a simple metric for leakage risk assessment and mapped these on the reservoir to identify risk areas [12].

In 2012, Lu et al. developed a computational model to simulate the behavior of leakage faults connecting saline CO_2 storage reservoirs and overlying freshwater aquifers [13]. Salahshoor et al. proposed a new pressure control method based on a nonlinear model predictive control scheme to reduce the risk of CO_2 re-entering the atmosphere due to the failure of cap rock integrity [14]. In 2019, Buscheck et al. utilized downhole pressure and TDS monitoring to capture the impact of leakage depth along legacy wells, regional groundwater flow, and aquifer heterogeneity on leakage detection [15]. Ma et al. investigated the feasibility of using the ensemble Kalman filter (EnKF) data assimilation framework to estimate the hydraulic properties of storage formations and predict CO_2 plume migration from monitoring measurements, including instantaneous pressure and saturation data from branch wells and time-lapse seismic data modeled as vertically averaged saturation differences [16]. Onishi et al., as participants of the US Department of Energy's National Risk Assessment Partnership (NRAP), developed a science-based approach to quantify the risk profile of CO_2 geological sequestration sites [17].

In 2020, Liu et al. proposed a method for predicting porosity and permeability using borehole observations and surface geophysical data [18]. In 2021, Luther et al. studied the effect of stratification on the onset time of convective instability and the flow patterns beyond the onset time during CO_2 sequestration [19]. Yahaya-Shiru et al. used systematic and process-based seismic and well log data sets to describe the structure and stratigraphic framework of sandstone reservoirs within oilfields to determine their capacity for effective CO_2 sequestration [20]. Gan et al. conducted a system-level risk assessment of China's Shenhua CO_2 sequestration formations using the National Risk Assessment Partnership Integrated Assessment Model for Carbon Sequestration (NRAP-IAM-CS) [21]. Zhou Yinbang et al. suggested that high-quality monitoring data can effectively reduce potential leakage risks and that multiple monitoring combinations facilitate long-term safe geological CO_2 sequestration [22].

Currently, saline aquifers as potential storage media for CO_2 sequestration have received widespread attention and research. Extensive field investigations, laboratory simulations, and numerical modeling studies have been conducted to explore the characteristics and potential of saline aquifers as CO_2 sequestration reservoirs. Despite the certain sequestration capacity and stability of saline aquifers as CO_2 storage reservoirs, several issues and challenges remain in practical research.

Firstly, the characteristics of saline aquifers are highly complex, including variations in reservoir pore structure, permeability, and salinity, posing challenges for modeling and predicting the sequestration process. Secondly, the long-term stability of saline aquifers is a critical consideration, requiring assessment of the long-term retention of CO_2 post-sequestration and the effectiveness of the sequestration system. Additionally, evaluating and monitoring the leakage risk during saline aquifer sequestration is an important topic, necessitating research on leakage mechanisms, pathways, and rates to ensure the safety and sustainability of the sequestration process. Therefore, further research and exploration are necessary to optimize saline aquifer sequestration technology and provide a more comprehensive scientific basis for CO_2 geological sequestration.

Overall, research on saline aquifers as CO_2 sequestration reservoirs has made progress, but further studies are required to address existing issues and challenges to ensure the feasibility and sustainability of saline aquifer CO_2 sequestration technology. This study aims to predict the migration and leakage of CO_2 sequestered in saline aquifers, considering phase changes, by deeply investigating formation migration patterns and leakage mechanisms. The goal is to provide scientific decision-making support for CO_2 geological sequestration projects, ensuring the safety of the sequestration process.

2. METHODOLOGY

Based on the existing research foundation and the practical engineering applications predominantly focused on CO_2 sequestration in deep saline aquifers, this study utilizes CMG, a widely applied reservoir numerical simulation software, to build a typical deep saline aquifer geological model. By examining the CO_2 sequestration process in saline aquifers, we analyze the characteristics of CO_2 migration and distribution during sequestration, the storage capacity of CO_2, and the types and extents of CO_2 sequestration mechanisms. This analysis aims to provide reference and basis for hypothesizing potential leakage scenarios during the sequestration process.

2.1 Model description

The numerical simulation software selected for this study is CMG-GEM, a mature and powerful commercial software known for its ease of operation. This study focuses on the conceptual model of CO_2 sequestration in saline aquifers, selecting a set of saline aquifers with typical geological characteristics for analysis, and emphasizing the migration of CO_2 between different layers. By establishing a conceptual model, it is possible to deeply investigate the migration characteristics of CO_2 in saline aquifers, including migration pathways, rates, and directions between different layers. As subsequent studies focus on the vertical leakage and migration of CO_2 within the aquifer, a 2D vertical model is constructed, as shown in Figure 1.

The model is based on the actual CO_2 injection engineering of the Shenhua CCS Demonstration Project. A CO_2 injection well is set up on the left side of the model and a production well is set up on the right side of the model to balance formation pressure. The model grid is 50×1×70 in *x*, y and *z* directions, respectively, and each grid cell measures 20 × 20 × 20 m. The lateral length of the model is 1000 m, whereas the vertical span is 1400 m. The top and bottom depths of the reservoir are 600 m and 2000 m, respectively.

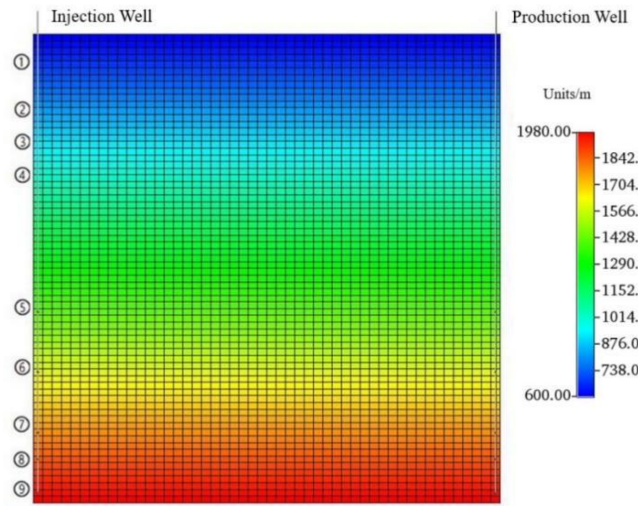

Figure 1: 2D Model of a Saline Aquifer

The basic geological data for the model were collected and organized from published literature and publicly available engineering data, with reference to similar conditions from the Shenhua Ordos sequestration project [23]. Other parameters of the model are listed in Table 1.

Table 1. Reservoir Parameters in the Model

Reservoir Parameter	Value	Reservoir Parameter	Value
Surface Temperature, °C	8.2	Depth of Gas-Water Contact, m	100
Geothermal Gradient, °C·(100m)$^{-1}$	2.99	Rock Compressibility, kPa^{-1}	4.5×10^{-7}
Reference Depth, m	2000	Reference Pressure, MPa	20.3
Maximum Injection Rate, m^3·d^{-1}	250000	Injection Temperature, °C	50

The entire simulation process involves injecting stratified CO_2 into the saline aquifer with a bottomhole temperature of 50°C. By controlling the opening and closing of different intervals of injection well and production well, the effect of stratified injection well is achieved. The injection process lasts for three years and three months, after which the injection well is closed. The simulation monitors the changes of CO_2 in the formation, simulating its migration, physicochemical reactions, and sequestration mechanisms underground. Monitoring continues until 217 years after the cessation of injection.

The interlayers between each saline aquifer are separated by caprocks with a porosity of 0.01, permeability of 0.001×10^{-3} μm^2, and a vertical/horizontal permeability ratio of 0.1. The geological parameters for each saline aquifer are detailed in Table 2. Saline aquifers 1 through 4 are shallow aquifers and serve as distribution locations for upward CO_2 leakage. Saline aquifers 5 through 9 are target injection layers where geological parameters, referencing actual formation conditions [23], comply with the anti-rhythm rule, with permeability gradually increasing from bottom to top.

Table 2: Geological Parameters of Saline Aquifers in the Model

Saline Aquifer	Top Depth (m)	Bottom Depth (m)	Porosity	Horizontal Permeability (10^{-3} μm^2)	Vertical Permeability (10^{-3} μm^2)	Thickness (m)
1	660	700	0.3	1000	100	40
2	800	840	0.3	1000	100	40
3	900	940	0.3	1000	100	40
4	1000	1040	0.3	1000	100	40
5	1400	1440	0.3	1000	100	40
6	1580	1620	0.3	850	85	40
7	1740	1800	0.3	800	80	60
8	1860	1880	0.3	700	70	20
9	1940	1980	0.3	500	50	40

In the model, there is an open fault that spans 9 saline aquifers with a width of 60 m. The fault's top is at 660 m and bottom at 2000 m, with a porosity of 0.15 and permeability of 5000×10^{-3} μm^2. The vertical/horizontal permeability ratio is 0.1. This fault provides a pathway for upward migration of CO_2. Figure 2 shows the distribution of porosity in the leakage model, illustrating the shape of the fault as depicted in Figure 2.

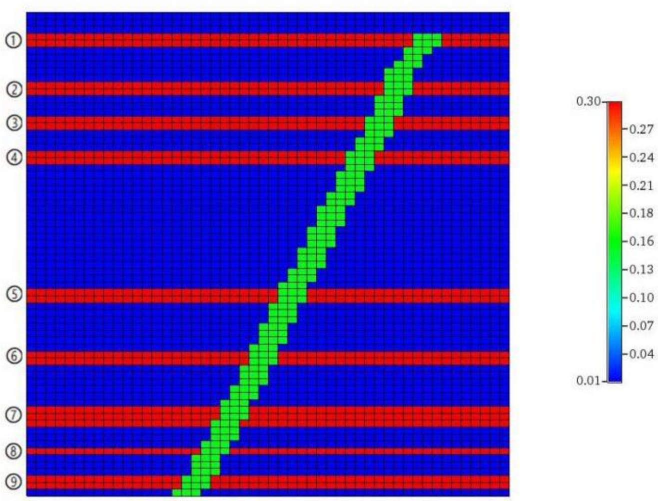

Figure 2. Porosity distributions of leakage model

2.2 *CO_2 injection capacity and storage capacity*

During the sequestration process, the injection rate and the total amount of CO_2 injected are among the most critical issues in the study. These factors directly affect the CO_2 sequestration potential and injection capacity of the model.

Over 3 years, CO_2 was continuously injected at a rate of 250,000 m³/day, with a total of 2.88×10^8 m³ of CO_2 injected. The maximum injection pressure is 35 MPa, the maximum injection rate is 250,000 m³/d, and the injection temperature is 50°C. Under these injection conditions, the injected CO_2 is in a supercritical state. After the injection stopped, the distribution and migration of CO_2 within the formation were simulated over a period of 217 years.

To optimize the CO_2 injection process, this study employs a layered injection-production technique and real-time monitoring of the gas production rates in the production wells. When gas production is detected in the production wells, the geological CO_2 sequestration efficiency significantly decreases. Therefore, by monitoring the gas production rates in the production wells, adjustments to the production layers can be made in time before gas breakthrough occurs, thereby improving sequestration efficiency.

3. ***RESULTS AND DISCUSSION***

In this study, a typical deep saline aquifer geological model is established and simulated using CMG-GEM. By examining the process of CO_2 leakage through an open fault in the saline aquifer, the study analyzes the migration characteristics and phase changes of CO_2 during the leakage and migration process.

3.1 *CO_2 migration characteristics*

In this section, CO_2 saturation and the mole fraction of CO_2 in the formation water are used as indicators to analyze the migration of CO_2 within the model. Faults typically exhibit relatively high permeability and connectivity, allowing CO_2 to migrate more rapidly through fault areas. In the leakage model, due to the presence of the fault, all saline aquifers are interconnected, providing a migration pathway for CO_2.

As shown in Figure 3, the injected CO_2 migrates upward due to the presence of the fault. The CO_2 saturation and solubility in the lower 7th, 8th, and 9th saline aquifers on the right side of the fault significantly decrease compared to a non-leakage scenario. The CO_2 that should have been stored in the 7th, 8th, and 9th saline aquifers migrate upward along the intersections of the fault and the saline aquifers.

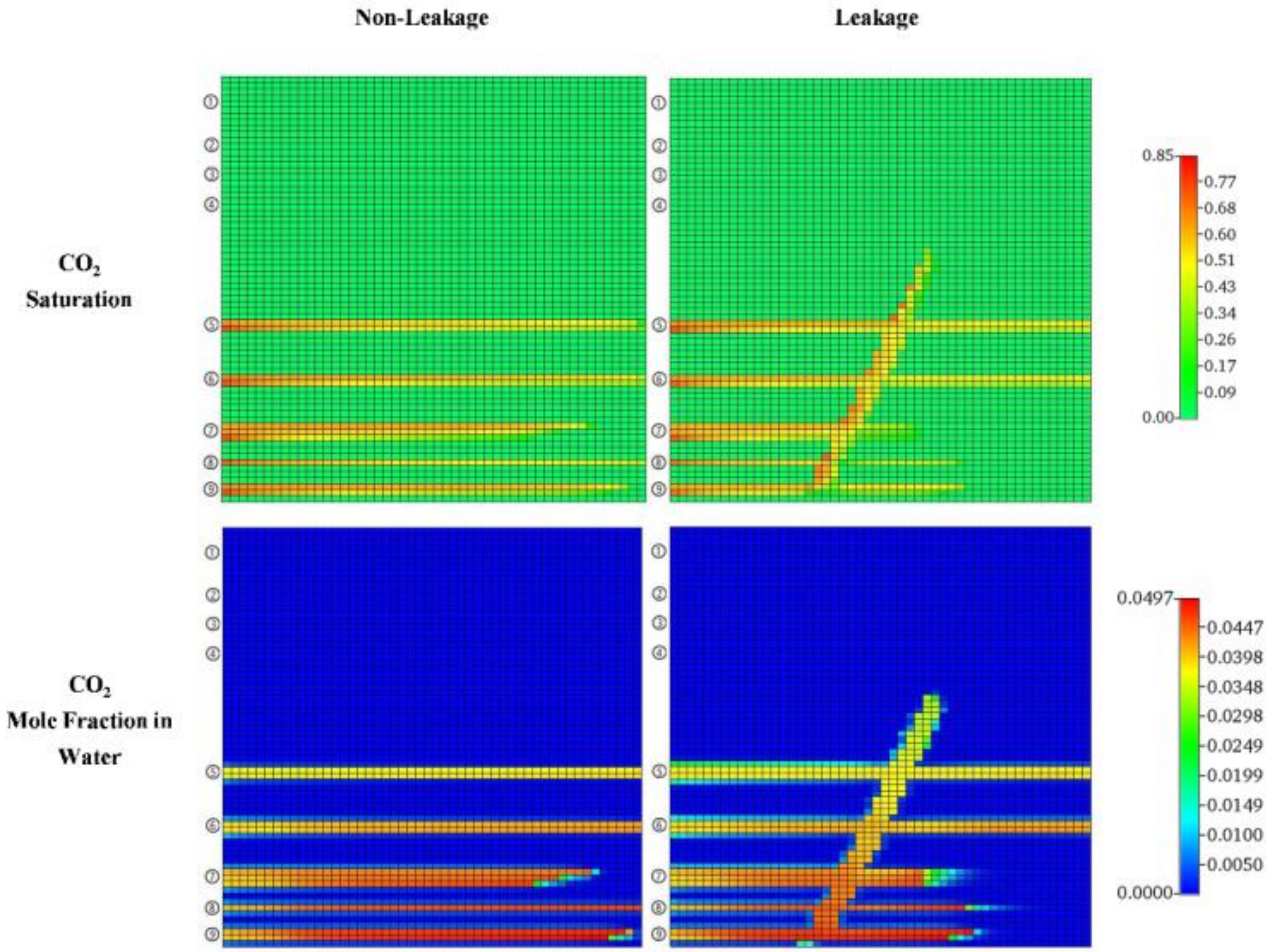

Figure 3. Distribution of CO_2 under different conditions at the end of injection

Faults typically have higher permeability, allowing CO_2 to be transported through the fault at a relatively high velocity. When injection ceases, the CO_2 has migrated above the 5# saline layer and is close to a stratigraphic level of about 1200 meters. During the injection process, the driving force for CO_2 comes from the pressure difference between the injection well and the shallow saline layer connected to the fault. Due to the stratigraphic span exceeding 1000 meters, the significant pressure difference results in the rapid transport of CO_2 to the depth of 1200 meters in the formation. Inferred from the geothermal gradient, when CO_2 migrates upward to about 940 meters, it transitions from a supercritical state to a liquid state. At this point, the phase composition of CO_2 saturation includes both supercritical and liquid states.

After 5 years of cessation of injection (Figure 4a, Figure 4b), it can be seen that the CO_2 has migrated upward by about 840 meters through the formation and has traversed the 3# and 4# saline layers. Observations of the underlying saline layers reveal that the CO_2, which originally migrated to the right through the fault, has a tendency to migrate leftward after the injection stops. The CO_2 saturation in the area where the fault intersects with the saline layer on the right side has decreased.

Comparing Figure 4c with Figure 4a, the CO_2 in the fault has risen further, but without subsequent CO_2 replenishment, the discontinuous distribution of supercritical CO_2 in the formation becomes more pronounced. As seen in Figure 4e, after 27 years of cessation of injection, the CO_2 has migrated upward to the 1# saline layer and has also undergone lateral migration. At this point, the migration of CO_2 is influenced by buoyancy and diffusion. By comparing Figure 4d with Figure 4f, it can be observed that when CO_2 enters the upper 1, 2, 3, and 4# saline layers, it spontaneously diffuses from areas of high concentration to areas of low concentration. At this time, a concentration gradient exists between the fault and the shallow saline layer, and CO_2 will be transferred from high-concentration areas to low-concentration areas through diffusion in the pore space. After 17 years, the upper saline layers clearly show that when a certain amount of CO_2 is dissolved in the saltwater, the density of the saltwater increases, causing the denser saltwater to migrate under the influence of gravity and allowing lighter saltwater to rise, thus achieving convective mixing within the layer and increasing the CO_2 solubility in the lower part.

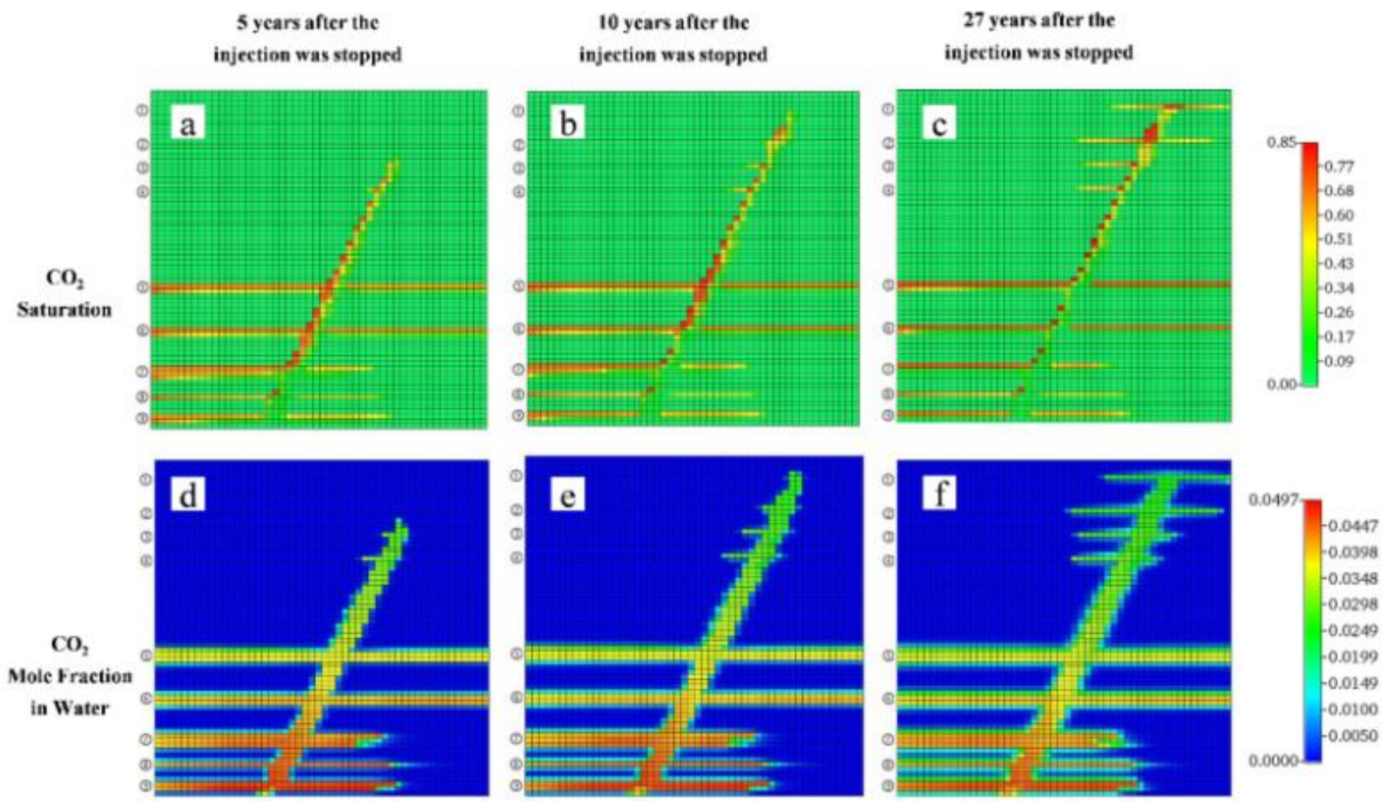

Figure 4 Distribution of CO2 under different conditions from 5 years to 27 years after stopping injection

By comparing Figure 4c with Figure 4e, it can be seen that there is a discontinuous distribution of CO_2 in the fault. This indicates that CO_2 is separated and confined to local areas within the fault, unable to freely flow and diffuse to other places. This confined state is mainly influenced by the adoption action of the rock. On the surface of the fault rock and within the pores, CO_2 may undergo adsorption, fixing CO_2 molecules to the rock surface or pore walls, thereby limiting their free movement and diffusion.

Figure 5a shows that at this stage, CO_2 is mainly influenced by buoyancy, migrating upward to the saline layers or above the fault. Therefore, the CO_2 saturation below the cap layer increases, while the CO_2 saturation in the lower part of the saline layers decreases. The CO_2 in the upper saline layers continues to migrate laterally, and the discontinuously distributed CO_2 below the fault is bound in place by the adsorption action of the rock, with no movement occurring.

Comparing Figure 5b with Figure 4f, it is found that under the influence of the concentration gradient, the phenomenon of CO_2 diffusing and escaping from high-concentration saline layers to the overlying low-concentration strata becomes more evident. Combined with Figure 5a, it can be seen that the amount of CO_2 diffused out is small and the escape rate is slow, which is due to the modest injection volume of the model and the stratigraphic levels controlled by the fault.

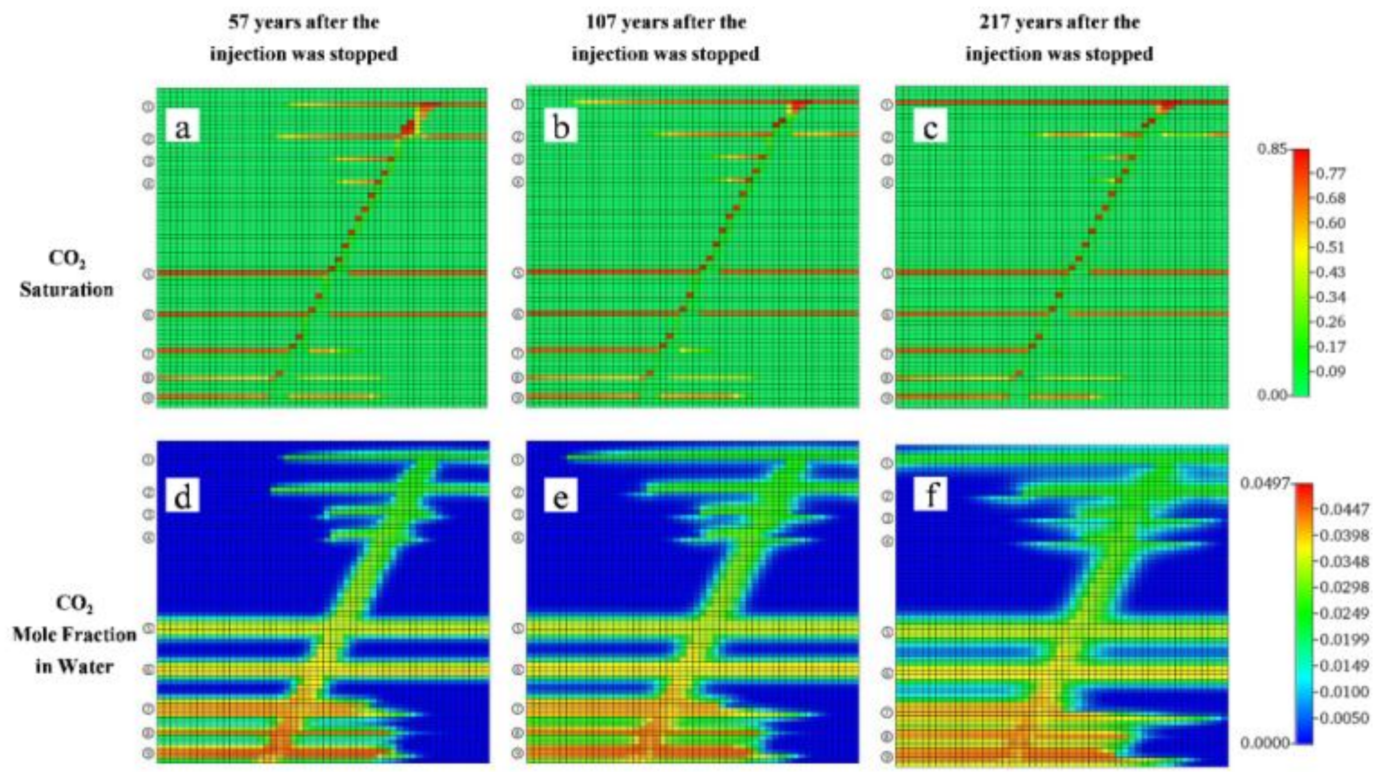

Figure 5 Distribution of CO_2 under different conditions from 57 years to 217 years after stopping injection

Figures 5b, 5d, and 5f collectively illustrate that CO_2 continues to diffuse within the strata, with dissolution adsorption and density-driven convection occurring. CO_2 can migrate by dissolving in the water and adsorbing onto the rock within the saline layers, thereby diffusing and transporting laterally within the medium. At the same time, CO_2 has a higher density, making it denser than the saline layers. This density difference leads to convection of supercritical CO_2 to move downward. Since the density of supercritical CO_2 is greater than that of the surrounding saline layers, it will sink and move towards lower areas, forming density-driven convective flow. Therefore, at the end of the CO_2 plume, the solubility below will increase and diffuse into the surrounding areas.

3.2 *Phase changes of CO_2 during leakage*

As CO_2 leaks upward along the fault to the shallow saline layer, there may be a change in its phase state, transitioning from the original supercritical state to liquid and gaseous CO_2. These phase state changes are primarily influenced by the formation pressure, temperature, and the physical properties of CO_2. Variations in the geological conditions can lead to transitions between phases, with the pressure and temperature of CO_2 determining whether it remains in a liquid, supercritical, or gaseous state. During the upward leakage process along the fault, changes in the phase state of CO_2 can significantly affect its migration characteristics and the effectiveness of sequestration. This is of critical importance for the design and assessment of carbon sequestration projects.

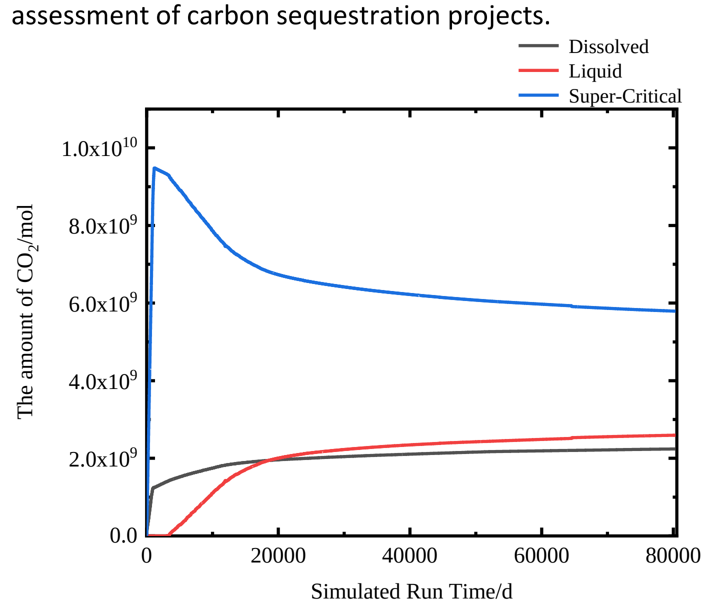

Figure 6 The phase distribution of CO_2 in the leakage model

The distribution patterns of CO_2 in the formation are shown in Figure 6. As indicated in the figure, the forms of CO_2 presence in the leakage model are supercritical, dissolved, and liquid states. Most of the supercritical CO_2 in the formation is kept stable by geological sequestration effects, but it is essentially in a free state. If there is a change in the geological structure, the CO_2 stored geologically can migrate. In the leakage model, the supercritical CO_2 migrates upward along the fault. As the degree of migration increases, both the temperature and pressure of the formation decrease. Once they fall below the critical point, where either the temperature or pressure drops below the critical value, the CO_2 will transition to a liquid state.

The critical depth in this model is set at 940 meters. Referencing this critical temperature, the leakage model is divided into upper and lower parts. Using the amount of substance of CO_2 as an indicator, the changes in CO_2 content in various regions over different times are analyzed. The distribution maps of CO_2 in different areas are shown in Figure 7.

Comparing Figure 6 with Figure 7, it is observed that the change curve for the area above the critical depth in Figure 7 is similar to the CO_2 liquid state change curve in Figure 6, with both showing a significant increase starting and following the same trend of change at the same time. This indicates that after that time, CO_2 leaked to a depth above the critical point and transitioned from a supercritical state to a liquid state, altering its properties and sequestration stability. The density of supercritical CO_2 is higher, allowing for more CO_2 to be stored per unit volume compared to liquid CO_2. This implies that when the phase state changes as described, the storage capacity of the saline aquifer will decrease. Additionally, when CO_2 transitions from a supercritical state to a liquid state, the decrease in density leads to an increase in the volume of CO_2, which may cause changes in the pressure of the saline aquifer and affect the stability of the sequestration system.

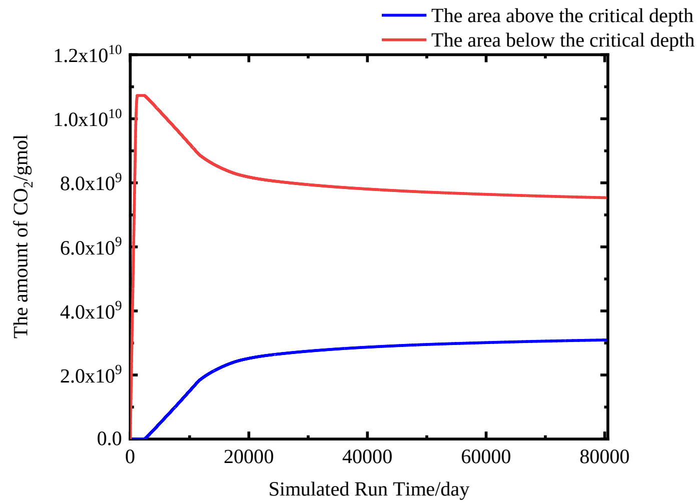

Figure 7. Variation of CO_2 content in different regions of the leakage model

4. CONCLUSIONS

Based on existing theoretical foundations and using actual on-site sequestration data and geological information, this work constructed a leakage model containing a fault through reservoir numerical simulation methods. This work focused on analyzing the phase state changes of CO_2 during the leakage process and explored the potential impacts. Conclusions can be drawn as follows:

During the leakage process, CO_2 is subject to a variety of mechanisms. It is driven by the pressure difference between formations and the net buoyancy of CO_2 itself, while capillary forces, adsorption resistance, and viscous forces impede its migration. As CO_2 migrates upward along the fault, the pressure difference decreases, reducing the driving force, leading to some CO_2 being bound in local areas, unable to freely flow and diffuse to other places.

When leakage occurs, CO_2 transitions from a supercritical state to a liquid state during the upward migration process. When liquid CO_2 leaks into shallow saline layers, it can cause a series of hazards, posing risks to human safety and environmental property. When selecting sites for CO_2 sequestration projects in saline aquifers, areas with faults or fractures should be avoided to ensure the long-term and stable sequestration of CO_2.

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