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Original Paper

Impact of dissolution and precipitation on pore structure in CO₂ sequestration within tight sandstone reservoirs

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A R T I C L E I N F O

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ABSTRACT

Complex physical and chemical reactions during CO_2 sequestration alter the microscopic pore structure of geological formations, impacting sequestration stability. To investigate CO_2 sequestration dynamics, comprehensive physical simulation experiments were conducted under varied pressures, coupled with assessments of changes in mineral composition, ion concentrations, pore morphology, permeability, and sequestration capacity before and after experimentation. Simultaneously, a method using NMR T_2 spectra changes to measure pore volume shift and estimate CO_2 sequestration is introduced. It quantifies CO_2 needed for mineralization of soluble minerals. However, when CO_2 dissolves in crude oil, the precipitation of asphaltene compounds impairs both seepage and storage capacities. Notably, the impact of dissolution and precipitation is closely associated with storage pressure, with a particularly pronounced influence on smaller pores. As pressure levels rise, the magnitude of pore alterations progressively increases. At a pressure threshold of 25 MPa, the rate of change in small pores due to dissolution reaches a maximum of 39.14%, while precipitation results in a change rate of -58.05% for small pores. The observed formation of dissolution pores and micro-cracks during dissolution, coupled with asphaltene precipitation, provides crucial insights for establishing CO_2 sequestration parameters and optimizing strategies in low permeability reservoirs.

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1. Introduction

The utilization of CO_2 geological sequestration technology stands as a pivotal approach in reducing carbon emissions, representing a key facet in achieving the carbon peak and carbon neutrality objectives (Alper and Orhan, 2017; Assen et al., 2021; Chai et al., 2022; Liu and Rui, 2022; Zhang et al., 2023a). This technology, when applied to geological reservoirs such as depleted reservoirs, deep saline aquifers, unmineable coal seams, and geothermal reservoirs, facilitates the long-term or permanent isolation of CO_2 from the Earth's atmosphere, thereby substantively ameliorating the greenhouse gas effect. Notably, reservoirs possess several advantageous attributes, including distinct boundaries, substantial storage capacity, effective containment by surrounding rock formations, comprehensive ground infrastructure, and costeffectiveness for storage purposes (Alfi and Hosseini, 2016; Liu et al., 2022a, 2024; Tang et al., 2021). Consequently, they have increasingly emerged as focal points for CO₂ sequestration initiatives globally (Hao et al., 2024; Khather et al., 2019; Ozotta et al., 2022). The safety and efficacy of CO_2 geological sequestration predominantly hinge upon the macro-geological structure and micro-structure of the reservoir geological body (Berrezueta et al., 2023), with the latter primarily shaped by the interactions involving CO₂, fluids, and rock materials, encompassing dissolution and precipitation phenomena (Gauteplass et al., 2020; Li et al., 2021). Thus, an in-depth exploration of the dissolution and precipitation dynamics inherent to the CO₂ geological sequestration





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process assumes critical importance in advancing our understanding of this pivotal technology within the realm of CO_2 geological sequestration.

During the CO₂ sequestration process, CO₂ undergoes interactions with the rock formations and fluid constituents (Cheng et al., 2020a; Giammar et al., 2005; Liu et al., 2022b, 2022c). This interaction leads to concurrent dissolution and precipitation phenomena, consequently alternating the mineral composition of the rocks, the microscopic pore structure, and the macroscopic physical parameters (Dewhurst et al., 2020; Foroutan et al., 2021; Rathnaweera et al., 2017; Wang et al., 2023a, 2023c; Zhang et al., 2020b). These alterations exert a notable influence on the CO₂ injectivity and sequestration efficacy, significantly heightening the associated geological sequestration risks. Presently, investigations into CO₂ dissolution and precipitation primarily center on cap rock and saline aquifers concerning geological sequestration (Cui et al., 2018; Jayasekara et al., 2020a; Li et al., 2019; Ma et al., 2023; Zhao et al., 2022). In the case of reservoirs, the research emphasis leans towards examining the controlling influence of reservoir dissolution and precipitation on CO₂ displacement (Gao et al., 2022a; Sun et al., 2023; Zanganeh et al., 2018). While prior studies have undertaken comparisons and assessments of changes in rock mineralogy, fluid characteristics, pore structures, and physical properties pre and post experimentation (Ao et al., 2017; Cheng et al., 2020b; Li et al., 2020, 2022; Sun et al., 2021; Zhou et al., 2020), the mobility of rock fluids has remained relatively unexplored.

Upon contact with formation water, CO₂ initiates the formation of a mildly acidic solution, predominantly characterized by the dissolution of carbonate and aluminate minerals. This process concurrently engenders the deposition of secondary minerals, including carbonate, clay minerals, and sulfate (Kweon and Deo, 2017; Luquot et al., 2016; Tutolo et al., 2015; Wang et al., 2016, 2023d; Zhang et al., 2020a). Notably, mineral dissolution triggers the detachment of insoluble particles from the rock surface, accompanied by discernible fluctuations in the concentrations of Na⁺, K^+ , Ca²⁺, Mg²⁺, HCO₃, and CO₃²⁻ within the formation fluid (Berrezueta et al., 2016; Bowers et al., 2017; Cheng et al., 2020c; Yang et al., 2011). These alterations in ion concentrations corroborate the geochemical reactions induced by the injection of CO₂ (Tang et al., 2021). The dissolution and precipitation induced by CO₂ occur simultaneously, yet the intricate nature of mineral precipitation and dissolution is particularly pronounced within tight sandstone formations, characterized by complex micro- and nanoscale pore structures. This complexity leads to irregular variations in formation porosity and permeability (Aminu et al., 2018; Foroutan et al., 2022; Shi et al., 2019), thus yielding considerable heterogeneity in the impact of CO₂ injection on geological body sequestration. A study by Ma et al. (2023) posited that protracted CO₂-brine-rock interactions would result in carbonate mineral precipitation, directly diminishing the permeability of overlying rock layers while enhancing their inherent self-sealing capacity. In addition, it is observed that in the short term, mineral dissolution surpasses secondary mineral precipitation (Jayasekara et al., 2020b). Tang et al. (2021) further elucidated that mineral dissolution within reservoirs engenders pore enlargement, while subsequent precipitation and particle agglomeration precipitate a significant reduction in permeability. Chen et al. (2022) conducted experimental investigations on CO₂ geological sequestration and proposed a new concept of threshold pressure gradient (TPG) to assess the permeability of reservoir fluids. Concerning microscopic pore changes, mineral precipitation and the detachment of skeleton particles effectively shift larger pores into smaller ones, thus augmenting the proportion of small pores in tight sandstone (Wang et al., 2023b). Importantly, the specific patterns of large pore content variation differ across various sandstone types, primarily attributable to dissimilar degrees of dissolution or precipitation occurring within distinct pore spaces (Yang et al., 2011). The introduction of CO₂ into geological strata induces alterations in mineral geochemistry. When CO₂ is injected into the overlying layer, it leads to mineral precipitation, diminishing permeability, and fortifying the layer's sealing capacity. Conversely, CO₂ injection into reservoirs prompts mineral precipitation and particle detachment, resulting in a decreased permeability, thereby detrimental to CO₂ injection and sequestration. Notably, CO₂ injection into reservoirs in conjunction with crude oil interaction triggers the extraction of light hydrocarbon components from the crude oil, while heavy hydrocarbon components, in the form of organic matter, deposit and accumulate on pore surfaces (Mahdavifar et al., 2023; Mousavi et al., 2020; Nascimento et al., 2021; Zhang et al., 2015, 2023b). This asphaltene precipitation during CO₂ sequestration within reservoir geological bodies induces a substantial reduction in rock porosity and permeability, with organic matter plugging exerting a more detrimental impact on permeability than organic matter adsorption. Consequently, oil and gas biphasic flow are predominantly governed by asphaltene plugging (Mohammed et al., 2021; Wang et al., 2022). The extent of asphaltene plugging is notably more pronounced in cases where the microscopic pore structure and physical properties exhibit lower quality (Huang et al., 2022). In comparison to larger pores, smaller pores are more susceptible to the effects of precipitated asphaltene (Wang et al., 2017). However, contrasting viewpoints posit that asphaltene tend to obstruct larger pores (Lin et al., 2021; Wei et al., 2020a). The severity of asphaltene deposition directly correlates with a discernible decrease in core permeability (Khurshid and Afgan, 2021). Moreover, as geological depth increases, the quantity of asphaltene precipitation diminishes in tandem with rising stratigraphic temperatures (Espinoza Mejia and Li, 2023).

The extent of rock mineral reactions is contingent upon various factors, including temperature, pressure, reservoir lithology, and the properties of formation water (Dai et al., 2022; Wu et al., 2019). Chen et al. (2023) studied the solubility of CO₂ in saline aquifers and found that temperature is the most sensitive factor affecting solubility, followed by pressure. In particular, temperature, pressure, crude oil viscosity, pore size, and heterogeneity serve as critical determinants influencing asphaltene precipitation (Fakher and Imqam, 2019). Injection pressure exerts a twofold impact on CO₂ injectability and plays a pivotal role in the CO₂ geological sequestration process. With escalating injection pressure, the dissolution effect gradually intensifies, accompanied by increased concentrations of Na⁺, K⁺, Ca²⁺, and Mg²⁺ (Wei et al., 2020b). Concurrently, precipitation and particle obstruction exhibit incremental trends. Specifically, under miscible pressure conditions, organic species precipitation exacts a more pronounced toll on core permeability (Wang et al., 2023a). The underlying cause of alterations in core porosity and permeability is attributed to asphaltene precipitation and mineral dissolution (Gao et al., 2022b; Hug et al., 2015; Wang et al., 2020). These processes induce changes in pore microstructure, ultimately affecting the seepage and storage capacities of geological bodies.

Prior research predominantly focused on assessing alterations in the physical and mineral components of geological formations resulting from dissolution or precipitation effects during CO₂ injection processes (Table 1). These assessments primarily revolved around computing changes in porosity and permeability to evaluate their impact on the effectiveness of CO₂ flooding. Nonetheless, porosity and permeability changes represent macroscopic transformations that do not offer a precise depiction of variations in the microscopic pore structure within geological formations. Currently, the conventional method for calculating CO₂ sequestration is based

Summary of previous studies on CO2 sequestration on pore structure.

| Reference | Objectives | Methods | Findings | This work |
|---------------------------------|--|--|---|---|
| Cheng et al. (2020a) | study Sc-CO ₂ effects on shale's mineral composition and pore structure | shale treated with Sc-CO ₂ (40 —80 °C, 8 MPa) | peak clay mineral dissolution at 60 °C; decrease in specific surface area and total pore volume | Previous research focused on macroscopic changes in porosity and permeability, lacking precision in understanding microscopic pore |
| Rathnaweera et al. (2017) | investigate changes in sandstones with CO ₂ injection | CO_2 /brine/rock interactions (4 -10 MPa, 35 °C, one year) | higher pressure enhances mineral dissolution and induces mechanical changes | structure variations. This study innovatively uses crude oil component testing and low-field NMR to quantitatively analyze CO ₂ dissolution, |
| Cui et al. (2018) | analyze fluid-rock interactions on reservoir properties and CO ₂ injectivity | reactive transport model used | CO ₂ injection causes mineral dissolution (anorthite, calcite) and precipitation (kaolinite) mainly in zones with higher formation water content. | asphaltene precipitation, and pore structure changes, providing a precise assessment of $\rm CO_2$ sequestration in low-permeability reservoirs. |
| Tang et al. (2021) | understand CO ₂ - brine-rock interaction on formation properties | dynamic and static experiments (8 MPa, 80 °C) | improves porosity and permeability, enhancing CO ₂ injectivity and sequestration capacity | |
| Jayasekara et al. (2020b) | study geochemical reactions of caprock upon injection of supercritical CO ₂ | geochemical reactions (40 °C, 10 MPa) | significant mineral dissolution compared to secondary mineral precipitation in the short term | |
| Huq et al. (2015) | investigate chemical composition changes and permeability during CO ₂ injection | flow-through experiments (125 °C, 50 bar) | predominant dissolution of anhydrite and calcite observed; permeability of sandstone sample doubled | |
| Alfi and Hosseini (2016) | evaluate performance of storage strategies and CO ₂ -EOR. | history matched oil and gas production rates, CO ₂ -EOR production data | CO ₂ -EOR stores more CO ₂ long-term; CO ₂ injection stores more annually but leads to earlier field abandonment | |
| Chen et al. (2022) | investigate effects of miscible degree and pore scale on fluid seepage characteristics in unconventional reservoirs | long core displacement tests for CO ₂ -soaking alternating-gas (CO ₂ -SAG) flooding and simultaneous CO ₂ flooding | supercritical CO ₂ injection significantly improves oil recovery and permeability in low-permeability reservoirs, reducing flow resistance | |
| Wang et al. (2020) | investigate formation damage during miscible CO ₂ flooding in tight sandstone reservoirs | miscible CO ₂ flooding (18 MPa, 70 °C). | asphaltene precipitation decreases permeability, reversible upon removal | |
| Huang et al. (2022) | evaluate asphaltene precipitation characteristics during CO ₂ flooding in tight light oil reservoirs | CO ₂ flooding under immiscible and miscible phases | asphaltene precipitation increases with ${\rm CO}_2$ injection ratio and pressure | |

on measuring the amount of CO₂ injected and the amount of gas released during the experimental process, requiring high precision in experimental equipment. Consequently, understanding how changes in the microscopic pore structure of geological bodies influence CO₂ storage efficiency during the sequestration process remains a critical knowledge gap. To address this gap, this study employed crude oil component testing in conjunction with lowfield NMR technology. A series of CO₂ dissolution and asphaltene precipitation experiments under varying storage pressure conditions are conducted. The study comprehensively analyzed alterations in mineral composition, pore structure, seepage capacity, and storage capacity before and after these experiments. Through quantitative characterization, this research elucidated the quantitative effects of CO₂ sequestration on the storage space and seepage capacity of geological formations. A novel methodology was devised, relying on alterations in NMR T_2 spectra, to quantitatively determine the extent of pore volume change. This approach further facilitates the calculation of CO₂ sequestration volume, culminating in a quantitative assessment of CO₂ mineralization sequestration. These findings provide invaluable theoretical support for the establishment of CO₂ sequestration parameters and the optimization of sequestration strategies within low permeability reservoir.

2. Experimental materials and methods

2.1. Pore scale classification

For tight sandstones, the T_2 relaxation is dominated by the surface relaxation time, and a longer T_2 relaxation time indicates a smaller specific surface area of the rock. The surface relaxation of the rock can be calculated by Lai et al. (2018):

$$\frac{1}{T_{2S}} = \rho_2 \frac{S}{V} = \rho_2 \frac{F_S}{r}$$
(1)

where ρ_2 is the relaxation rate, μ m/ms, *S*/*V* is the specific surface area, $1/\mu$ m, *F*_S is the dimensionless shape factor of pore throat and *r* is the pore-throat radius, μ m.

Therefore, the pore throat radius r and the relaxation time are linearly connected, setting the conversion factor $C = \rho_2 F_S$, the T_2 relaxation time can be described by the following equation:

$$r = T_2 \cdot C \tag{2}$$

Using the pore size conversion method proposed by Gao et al. (2015), the optimal conversion coefficient *C* value for each core can be obtained by transforming the results of high-pressure mercury pressure and NMR tests. Based on the results of NMR tests (Fig. 1), 8 µm and 1 µm were used as the thresholds between large and small pores in artificial and natural cores, respectively. Therefore, the defined pore size range of small pores in artificial cores is 0.001 µm < r < 1 µm, and for large pores r is 1 µm < r < 1000 µm. In natural cores, the pore size range for small pores is 0.001 µm < r < 8 µm, and for large pores, r is 8 µm < r < 1000 µm.

2.2. Calculation of the pore change degree

The quantitative assessment of dissolution and precipitation effects on the pore structure of experimental core samples, as well as their impact on the seepage capacity of geological formations, was carried out through the meticulous comparison of T_2 spectra amplitude variations pre- and post- CO₂ sequestration



Fig. 1. Schematic diagram of experimental cores pore classification.

experiments. The schematic representation elucidating the alterations in pore characteristics is elucidated in Fig. 2. Herein, S_a denotes the initial volume of saturated fluid within the pore throats, whereas S_b signifies the volume of saturated fluid subsequent to the CO₂ sequestration process. The quantification of the alteration degree of pore volume, denoted as ε , can be accomplished via:

$$\varepsilon = \frac{S_{\rm a} - S_{\rm b}}{S_{\rm b}} \times 100\% \tag{3}$$

where ε is the porosity change, %, S_b is the envelope area of the T_2 spectrum before the experiments, and S_a is the envelope area of the T_2 spectrum after the experiments.

2.3. Experimental samples

In order to study the effects of dissolution and precipitation on the microporous structure of tight sandstone reservoirs during the CO_2 sequestration process, samples earmarked for dissolution



Fig. 2. Schematic diagram of calculating the degree in core porosity change.

within the framework of geological CO₂ sequestration were procured from the tight sandstone deposits within the Ordos Basin. Meanwhile, experiments focusing on asphaltene precipitation in the context of CO₂ geological sequestration were conducted employing artificial cores fashioned from quartz sand and epoxy resin cement. Essential parameters pertaining to the experimental cores are detailed in Table 2. The crude oil sample, characterized by a viscosity of 1.7 mPa·s and an asphaltene content of 1.25%, was evaluated at 25 °C and atmospheric pressure measuring 101.35 kPa. Detailed information regarding its components is provided in Fig. 3. Additionally, the minimum miscibility pressure of the crude oil, as determined through the slim tube experiment, stood at 20.8 MPa. The simulated formation water employed in the experimentation was prepared using CaCl₂, yielding a salinity of 25,000 mg/L. The CO₂ utilized in the experimental setup boasted a purity level of 99.99%, and the experimental temperature was rigorously maintained at 40 °C.

2.4. Experimental setup

High-temperature and high-pressure displacement flow instruments and an NMR instrument are employed in the CO₂ geological sequestration experiment. Notably, the displacement pump, an advection pump sourced from Beijing Xingda Technology Development Co., Ltd., boasts a pressure operating range spanning from 0 to 40.0 MPa, complemented by a flow rate capacity spanning from 0.001 to 10 mL/min. Additionally, the core holder, furnished by China Nantong Huaxing Petroleum Instrument Co., Ltd., accommodates pressures within the range of 0–40.0 MPa. The NMR analyzer utilized in these experiments, bearing the model number PQ001 and originating from Suzhou Niumag Analytical Instrument Co., Ltd., exhibits a magnetic field strength spectrum encompassing 0.28 to 0.03 T, with magnet uniformity at 30 ppm. Its frequency source spans from 1 to 30 MHz, underpinned by a frequency control precision of 0.1 Hz and a pulse accuracy of 10 ns. A schematic representation of the experimental flow system is delineated in Fig. 4 for reference.

2.5. Experimental procedures

2.5.1. CO₂ dissolution experiment

- (1) Natural core specimens underwent a rigorous cleaning regimen involving immersion in a benzene and alcohol solvent for a duration of 7 days at a temperature of 100 °C, followed by desiccation within a drying oven set at 80 °C for a 24-h period.
- (2) Subsequent to the drying process, measurements of core permeability were undertaken, and the ensuing outcomes are meticulously documented in Table 1.
- (3) The core samples were carefully positioned within the coreholder. Concomitantly, simulated formation water was introduced, and under a confining pressure of 8 MPa, the cores were systematically saturated with the simulated formation water at a constant injection rate of 0.05 mL/min, reaching a saturation level of 3–6 pore volumes (PV). The rock core's porosity was computed based on the discernible mass differential before and after the saturation process, as outlined in Table 1. Furthermore, NMR testing was executed on the rock cores.
- (4) To establish the initial distribution of formation fluids, kerosene samples were introduced into the core at a controlled flow rate of 0.05 mL/min. This continued until the effluent stream consistently yielded 100% oil-containing liquid. Volumes of the outflow were meticulously recorded,

Physical properties parameters of the cores.

| ID | Length, cm | Diameter, cm | Porosity, % | Permeability, $10^{-3} \ \mu m^2$ | Conversion factor | Storage pressure, MPa | Category |
|-----|------------|--------------|-------------|-----------------------------------|-------------------|-----------------------|-----------------|
| 1 | 5.22 | 2.47 | 10.03 | 0.0331 | 1.03 | 10 | Natural core |
| 2 | 5.23 | 2.49 | 7.80 | 0.0312 | 1.18 | 15 | |
| 3 | 5.45 | 2.51 | 10.51 | 0.0294 | 1.09 | 20 | |
| 4 | 5.68 | 2.50 | 8.80 | 0.0301 | 1.15 | 25 | |
| A-1 | 4.22 | 2.54 | 7.65 | 0.2592 | 1.11 | 10 | Artificial core |
| A-2 | 4.20 | 2.54 | 8.29 | 0.2603 | 1.14 | 15 | |
| A-3 | 4.23 | 2.54 | 8.43 | 0.2597 | 1.13 | 20 | |
| A-4 | 4.23 | 2.54 | 7.49 | 0.2600 | 1.13 | 25 | |



Fig. 3. Compositional analysis of the crude oil samples.

and the T_2 spectrum of the saturated kerosene cores was systematically measured.

- (5) Physical simulation experiments focusing on CO_2 sequestration were executed under controlled conditions of 40 °C and varying pressures spanning 10, 15, 20, and 25 MPa, respectively. Concurrently, the differentials in inlet and outlet pressures were regulated within a tight range of 0.1–0.2 MPa via adjustments to back-pressure valves. These experiments were conducted over a duration of 168 h.
- (6) Following the conclusion of the experiments, the fluids generated were collected, and the rock cores underwent post-experiment NMR analysis.
- (7) A repetitive sequence encompassing Steps (1) through (3) was enacted, facilitating a comparative evaluation of the disparities in T_2 spectra between the natural rock cores saturated with simulated formation water before and after CO₂ injection. This comprehensive analysis was instrumental in elucidating the impact of CO₂-brine-rock interactions on the pore structure of tight sandstone.



Fig. 4. Schematic diagram of the experimental setup.

2.5.2. CO₂ precipitation experiment

- (1) Following a thorough cleaning and drying process, synthetic core samples were meticulously weighed, measured, and assigned unique identifiers. Subsequently, gas permeability tests were executed, and the detailed outcomes are presented in Table 1.
- (2) Simulated formation water was meticulously prepared, and under a confining pressure of 8 MPa, the core specimens were securely placed within the core-holder. A systematic saturation process ensued, involving the injection of simulated formation water at a controlled flow rate of 0.05 mL/ min until a saturation level exceeding 5 PV was attained. Following this, a comprehensive T_2 spectrum sampling was undertaken on the fully saturated rock cores, followed by precise mass measurements. The porosity was then diligently computed, relying on the discernible mass differential before and after saturation with formation water, as thoughtfully presented in Table 1.
- (3) Employing MnCl₂ solution at a concentration of 30,000 mg/L, a consistent injection rate of 0.05 mL/min was maintained to displace the simulated formation water within the core. This displacement process continued until the injected volume reached 5 PV. Subsequently, an NMR T_2 spectrum scan of the core was meticulously executed to monitor the signal elimination effect of the formation water. In cases where saturation proved inadequate, additional injections of manganese water were performed until signal elimination was unequivocally observed in the T_2 spectrum.
- (4) At a controlled flow rate of 0.05 mL/min, simulated oil was injected into the core specimens until no further fluid was observed at the outlet. This step established the original oilwater distribution within the cores. The volumes of output fluid were conscientiously recorded, and the ensuing samples underwent meticulous NMR measurements.
- (5) The core samples underwent CO₂ injection under carefully regulated conditions, including a temperature of 40 °C and pressure settings of 10, 15, 20, and 25 MPa, respectively. Stringent control over back-pressure valves was maintained to uphold an inlet and outlet pressure differential of 0.1–0.2 MPa. The experiment continued for a duration of 168 h.
- (6) At each stage of sequestration, specifically at 1, 2, 4 PV, and 6 PV, outflow oil volumes were meticulously recorded, and oil samples were systematically collected. NMR sampling was thoughtfully conducted to scrutinize the fluid distribution characteristics within the rock cores.
- (7) A comprehensive four-component test was conducted on the produced oil samples to accurately calculate the rate of asphaltene precipitation.
- (8) The cores employed in the CO₂ sequestration experiments were subjected to a drying process at a consistent temperature following their treatment with n-heptane reagent at 100 °C. Steps (1) through (4) were then iterated to facilitate a comparative analysis of the disparities in the NMR T_2 spectrum curves, specifically pertaining to the initial oil-water distribution, both before and after the artificial core experiment. This rigorous examination aimed to elucidate the influence of asphaltene precipitation on the pore structure.

3. Results and discussion

3.1. Changes in petrophysical properties of geological body

3.1.1. Changes in porosity and permeability

The measured results pertaining to the porosity and permeability of the cores subsequent to the experimental procedure have been presented in Table 3. In the context of CO₂ sequestration, the physical attributes of natural cores undergo enhancements through dissolution processes. In the case of reservoirs, the presence of asphaltene precipitates tends to obstruct the core pore spaces because of CO₂ injection, thereby leading to a deterioration in the physical characteristics of the artificial cores. Fig. 5 illustrates the notable improvement in the physical properties of natural cores with increasing storage pressure, contrasting with the significant deterioration observed in the physical properties of artificial cores. The dissolution induced by CO₂ is limited under low storage pressures, thus the alterations in the physical properties of natural cores is not significant. As the storage pressure escalates, the CO₂ density increases concomitantly, resulting in an augmented dissolving capacity. Under a pressure magnitude of 20 MPa, the rates of alteration in porosity and permeability for natural cores are quantified at 8.16% and 24.15%, respectively. Notably, as the pressure ascends from 20 to 25 MPa, the rates of change in porosity and permeability reach their zenith, registering values of 10.09% and 29.24%, respectively. Upon the injection of CO₂ into the reservoir, the extent of interaction between CO₂ and crude oil is found to be relatively limited under low storage pressures. Consequently, the quantity of asphaltene precipitation remains modest. However, as the storage pressure mounts, the solubility of CO₂ in crude oil experiences a concomitant increase. Simultaneously, supercritical CO₂ consistently extracts lighter components from crude oil, leading to an augmented deposition of asphaltene in the artificial cores. This phenomenon serves to occlude the pore spaces and throats within the cores, thereby exacerbating the degradation of their physical properties. Notably, it is at the point of reaching the miscible pressure, particularly evident at a pressure level of 25 MPa, where the asphaltene precipitation phenomenon attains its maximum manifestation. Under these conditions, the permeability experiences a pronounced reduction of 36.58%, and the porosity undergoes a significant decline of 11.35%. It is discernible that asphaltene precipitation exerts the most profound deleterious impact on the permeability of geological formations under such circumstances.

3.1.2. Change in movable fluid parameters

As depicted in Table 4, the alterations in movable fluid parameters and the corresponding transformations in pore structure within both natural and artificial cores, engendered by processes of dissolution and asphaltene precipitation, are elucidated herein. These insights are drawn from meticulous analysis of NMR data, from which movable fluid parameters are rigorously deduced (Gao and Li, 2015). The calculations thus derived underscore that following dissolution, the aggregate proportion of mobile fluid within the natural core experiences an augmentation, concomitant with a diminishment in the $T_{2cutoff}$ value. Furthermore, it is discernible that the extent of these changes exhibits an escalation in tandem with the elevation of sequestration pressure. This phenomenon is intricately linked to the heightened solubility of CO₂ under increasing storage pressures, thereby intensifying the dissolution processes. This, in turn, amplifies mineral dissolution, augments pore connectivity, and consequently results in a reduction in the natural core's $T_{2cutoff}$ value. Consequently, there is a

Porosity and permeability after the experiments.

| - | | - | | | | | | |
|-----|---------------|-------------|-------|--------------------|---|--------|------------------------|-----------------|
| ID | Pressure, MPa | Porosity, % | | Porosity change, % | ity change, % Permeability, 10 ⁻³ μm ² | | Permeability change, % | Category |
| | | Before | After | | Before | After | | |
| 1 | 10 | 10.06 | 10.50 | 4.37 | 0.0331 | 0.0382 | 15.41 | Natural core |
| 2 | 15 | 10.08 | 10.64 | 5.56 | 0.0312 | 0.0372 | 19.23 | |
| 3 | 20 | 10.05 | 10.87 | 8.16 | 0.0294 | 0.0365 | 24.15 | |
| 4 | 25 | 10.11 | 11.13 | 10.09 | 0.0301 | 0.0381 | 29.24 | |
| A-1 | 10 | 7.65 | 7.31 | -4.44 | 0.2592 | 0.2207 | -15.12 | Artificial core |
| A-2 | 15 | 8.29 | 7.79 | -6.03 | 0.2603 | 0.2037 | -21.65 | |
| A-3 | 20 | 8.43 | 7.6 | -9.85 | 0.2597 | 0.1829 | -29.65 | |
| A-4 | 25 | 7.49 | 6.64 | -11.35 | 0.2600 | 0.1649 | -36.58 | |



Fig. 5. Variation curves of porosity and permeability with storage pressure.

notable rise in the proportion of movable fluid and a concurrent enhancement in fluid mobility within the pore network, as exemplified in Fig. 6. As the sequestration pressure incrementally rises, the deposition of asphaltene compounds within the core becomes increasingly pronounced, leading to a progressive and more severe pore plugging phenomenon. This, in turn, precipitates a discernible decline in the proportion of movable fluid within the core postexperimentation, coupled with a significant elevation in the lower threshold T_{2cutoff} value for mobile fluid (as illustrated in Fig. 6). Remarkably, it is under conditions of a sequestration pressure reaching 25 MPa that the magnitude of variation in the $T_{2cutoff}$ value for both natural and artificial cores attain its zenith. Fig. 7 supplements this narrative by presenting the NMR T_2 spectrum distribution for saturated water, along with the core's centrifugation data, both pre- and post-experimentation. It is based on these observations that the *T*_{2cutoff} values for the core are meticulously

determined, enabling the subsequent computation of core fluid parameters.

3.2. Changes in the pore structure

Table 5 provides an overview of outcomes derived from experimental investigations aimed at elucidating alterations in the extent of pores at various scales. The ensuing calculations shed light on the dynamic evolution of pore volume within natural cores during CO₂ sequestration, primarily attributed to the dissolution phenomenon. Notably, a discernible augmentation in pore volume within natural cores is manifest as a direct result of CO₂ dissolution. Furthermore, it is noteworthy that the dissolution capacity of CO₂, in tandem with the overall rate of pore change, exhibits a marked amplification in response to heightened sequestration pressures. Under divergent sequestration pressures, it becomes evident that the rate of change in smaller-scale pores significantly outpaces that observed in larger counterparts. This observation underscores the predominant influence of dissolution mechanisms on smaller pores during the CO₂ sequestration process. Consequently, the volume of smaller pores within natural cores experiences a substantial increment, which is notably reflected in the amplification of the NMR T_2 spectrum curve, as exemplified in Fig. 8(a)–(d). Upon attaining a sequestration pressure of 25 MPa, a zenith is reached in the overall rate of pore change, marking an impressive value of 32.74%. Simultaneously, the alteration rates for smaller and larger pores ascend to their peaks, registering values of 39.14% and 21.63%, respectively. However, the introduction of asphaltene precipitation during the CO₂ sequestration endeavor instigates pore plugging phenomena within natural cores. Consequently, the core's pore volume undergoes a reduction, accompanied by a diminished magnitude of the NMR T₂ spectrum after subsequent re-saturation with oil, as evident in Fig. 8(e)–(h). Notably, the extent of pore plugging is positively correlated with the sequestration pressure, and the rates of change in smaller pores consistently exceed those observed in larger counterparts. This underscores that smaller pores are predominantly subject to blockage following asphaltene

Table 4

Changes of movable fluid in pores at different scales pre- and post-experimentation.

| ID | Pressure, MPa | Percentage of moving fluid before, % | | | Percentage of moving fluid after, % | | | $T_{2 \text{cutoff}}$ before | T _{2cutoff} after | Category |
|-----|---------------|--------------------------------------|------------|---------|-------------------------------------|------------|---------|------------------------------|----------------------------|-----------------|
| | | Large pore | Small pore | Overall | Large pore | Small pore | Overall | | | |
| 1 | 10 | 40.60 | 12.95 | 23.43 | 52.99 | 15.62 | 30.27 | 16.63 | 14.88 | Natural core |
| 2 | 15 | 56.79 | 13.61 | 27.57 | 48.81 | 18.80 | 27.93 | 16.40 | 13.57 | |
| 3 | 20 | 33.89 | 12.95 | 18.84 | 44.23 | 10.35 | 18.89 | 21.21 | 16.32 | |
| 4 | 25 | 55.82 | 12.57 | 27.86 | 60.88 | 12.76 | 28.35 | 19.56 | 14.12 | |
| A-1 | 10 | 27.13 | 26.61 | 26.98 | 31.29 | 11.76 | 25.94 | 9.43 | 10.79 | Artificial core |
| A-2 | 15 | 30.48 | 9.70 | 24.02 | 27.06 | 5.72 | 21.10 | 9.59 | 11.15 | |
| A-3 | 20 | 36.88 | 18.15 | 31.83 | 32.26 | 6.07 | 27.44 | 9.53 | 11.62 | |
| A-4 | 25 | 34.18 | 5.58 | 25.10 | 25.42 | 7.53 | 21.51 | 9.54 | 11.97 | |



Fig. 6. Relation curves of core $T_{2cutoff}$ rate change and sequestration pressure after the experiments.

precipitation, and this blockage becomes markedly pronounced with escalating sequestration pressures.

The analysis underscores a dual mechanism at play: firstly, as sequestration pressure rises, the solubility of CO_2 in crude oil. Upon entering the miscible flooding phase, CO_2 achieves full dissolution in the crude oil, thereby intensifying its capacity to extract lighter components from the crude oil. This results in a concomitant

elevation in asphaltene precipitation, thereby accentuating the degree of pore blockage. Secondly, heightened pressures facilitate the migration and rapid accumulation of deposited asphaltene within smaller pores, exacerbating the degree of pore occlusion within this subset. Particularly noteworthy is that when the sequestration pressure approaches 25 MPa, the comprehensive pore alteration rate for natural cores peaks at 36.51%, with the change rates for smaller and larger pores similarly reaching their zeniths at 58.05% and 27.01%, respectively.

3.3. Analysis of mineral dissolution

Owing to the dissolution processes encountered during CO_2 sequestration, the reservoir's diverse mineral constituents are subject to intricate physicochemical transformations. Of particular significance in this regard are the feldspar and carbonate minerals, which undergo the most pronounced dissolution effects. This transformative journey engenders a dual consequence: an enhancement in the intrinsic properties of the rock, concomitant with alterations in the microscopic morphology of the core sample. The salient observation is that, as the sequestration pressure escalates, the dissolution capacity of CO_2 within the core augments, thereby accentuating the changes witnessed in the micromorphology of the core.

Prior to the initiation of the experiment, intergranular pores within the core were already well-developed, and the surface of feldspar minerals exhibited a predominantly flat and seemingly unaltered appearance, devoid of any overt signs of dissolution. However, as illustrated in Fig. 9, noticeable dissolution phenomena became increasingly evident under varying sequestration pressures. Notably, at a sequestration pressure of 10 MPa, feldspar



Fig. 7. Distribution of NMR *T*₂ spectra of saturated and centrifuged cores pre- and post-experimentation. (a) *T*₂ distribution of saturated and centrifuged core sample 4 before experiment, (b) *T*₂ distribution of saturated and centrifuged core sample 4 after experiment, (c) *T*₂ distribution of saturated and centrifuged core sample A-4 before experiment, (d) *T*₂ distribution of saturated and centrifuged core sample A-4 after experiment, (c) *T*₂ distribution of saturated and centrifuged core sample A-4 after experiment.

Data table of pore change at different scales of experimental core.

| ID | Pressure, MPa | Rate of small pore change, % | Rate of large pore change, % | Overall rate of change, % | Category |
|-----|---------------|------------------------------|------------------------------|---------------------------|-----------------|
| 1 | 10 | 12.66 | 7.36 | 10.52 | Natural core |
| 2 | 15 | 24.19 | 13.61 | 20.65 | |
| 3 | 20 | 33.79 | 16.76 | 28.79 | |
| 4 | 25 | 39.14 | 21.63 | 32.74 | |
| A-1 | 10 | -19.59 | -14.74 | -16.11 | Artificial core |
| A-2 | 15 | -22.01 | -16.05 | -17.77 | |
| A-3 | 20 | -50.30 | -19.21 | -27.30 | |
| A-4 | 25 | -58.05 | -27.01 | -36.51 | |



Fig. 8. *T*₂ relaxation distribution of saturated water/hydrocarbon in the core pre- and post-experiments. (**a**) *T*₂ signals that before and after water saturated in core sample 1, (**b**) *T*₂ signals that before and after water saturated in core sample 2, (**c**) *T*₂ signals that before and after water saturated in core sample 4, (**e**) *T*₂ signals that before and after oil saturated in core sample A-2, (**g**) *T*₂ signals that before and after oil saturated in core sample A-2, (**g**) *T*₂ signals that before and after oil saturated in core sample A-2, (**g**) *T*₂ signals that before and after oil saturated in core sample A-3, (**h**) *T*₂ signals that before and after oil saturated in core sample A-4.



Fig. 9. Variation in microscopic morphology of natural core at the different sequestration pressure.

minerals primarily manifested bay-like dissolution patterns, which represented a discernible departure from their nreexperimentation state. As the sequestration pressure was incremented to 15 MPa, the dissolution effect became more pronounced, with discernible dissolution pits emerging on the particle surfaces, resulting in a more prominent dissolution pattern compared to the 10 MPa scenario. The scope of dissolution expanded significantly when the sequestration pressure reached 20 MPa, characterized by the emergence of microcracks on the mineral surfaces. Finally, under a pressure magnitude of 25 MPa, the post-experiment examination unveiled a marked reduction in the degree of rock cementation, accompanied by the presence of flaky debris adhering to the core surface when observed at lower magnification. A closer inspection at increased magnification revealed a pronounced dissolution of feldspar minerals, thereby resulting in uneven surfaces, an extensive proliferation of microfractures, and the establishment of numerous interconnected pores at cementation junctures. These developments notably accentuated the interconnectedness of pore networks within the rock core.

Supplementing the findings derived from X-ray diffraction analysis, the principal mineral composition of the natural core employed in the experimental framework is delineated as follows: quartz (42.7%), potassium feldspar (17.0%), plagioclase feldspar (11.5%), calcite (12.5%), clay minerals (10.6%), with a minor presence of dolomite (5.7%). Notably, the clay mineral component predominantly comprises illite (19.76%), mixed-layer illite (22.29%), kaolinite (42.15%), and chlorite (15.8%). Subsequent to CO₂ sequestration, discernible shifts in the content of feldspar and carbonate minerals are observed, with concomitant increases in the quantities of quartz and clay minerals. Particularly striking is the pronounced alteration in mineral composition witnessed within the core post-conclusion of the 25 MPa sequestration pressure experiment (as exemplified in Fig. 10). In this context, the content of potassium feldspar diminishes by 8.3%, plagioclase by 9.1%, calcite by 10.6%, and dolomite by 5.3%. Furthermore, comparative scrutiny of the X-ray energy spectrum pre- and post-experimentation corroborates the dissolution of feldspar and carbonate minerals, thereby aligning with the insights gleaned from SEM observations. Indeed, SEM investigations affirm the conspicuous manifestation of dissolution phenomena within the feldspar minerals encapsulated within the core, thereby providing additional validation to the dissolution process following the experiment.



Fig. 10. Relative content changes of core mineral components pre- and post-experimentation.

Subsequent to the ingress of CO₂ into the geological formation for sequestration, a cascade of chemical reactions ensues. CO₂ interfaces with the formation water, giving rise to the generation of carbonic acid, as delineated in Eq. (4). Carbonic acid, in turn, undergoes ionization, resulting in the formation of H⁺, HCO³⁻, and CO_3^{2-} ions, as expounded in Eqs. (5) and (6). Under the aegis of an acidic milieu, feldspar minerals enter into an association with H⁺ ions, vielding the formation of kaolinite, as elucidated in Eqs. (7) and (8). This process manifests as an augmentation in the kaolinite content within the formation. Moreover, in the aftermath of CO₂'s interaction, clay minerals, such as chlorite, are susceptible to transformation into quartz and kaolinite. In parallel, carbonate minerals, exemplified by calcite and dolomite, engage in a chemical liaison with H⁺ ions within the acidic milieu. This union precipitates the dissolution of carbonate minerals, thereby resulting in a diminution in the content of calcite and dolomite, as encapsulated in Eqs. (9) and (10).

$$\mathrm{CO}_2 + \mathrm{H}_2\mathrm{O} \leftrightarrow \mathrm{H}_2\mathrm{CO}_3 \tag{4}$$

$$H_2CO_3 \leftrightarrow HCO_3^- + H^+$$
 (5)

$$HCO_3^- \leftrightarrow CO_3^{2-} + H^+$$
 (6)

$$2 \underset{(Potassium feldspar)}{KAlSi_{3}O_{8}} + 2H^{+} + 9H_{2}O \rightarrow Al_{2}SiO_{3}(OH)_{4} + 2K^{+}$$

$$+ 4H_{4}SiO_{4}$$
(7)

 $2 \underset{(\text{Sodium feldspar})}{\text{NaAlSi}_3\text{O}_8} + 3\text{H}_2\text{O} + 2\text{CO}_2 \rightarrow \text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4 + 2\text{Na}^+ + 4\text{SiO}_2$

 $+ 2HCO_{3}^{-}$

$$\begin{array}{c} \text{CaCO}_3 + \text{H}^+ \rightarrow \text{Ca}^{2+} + \text{HCO}_3^{-} \\ \text{(Calcite)} \end{array} \tag{9}$$

$$\begin{array}{c} \text{CaMg}(\text{CO}_3)_2 + 2\text{H}^+ \rightarrow \text{Ca}^{2+} + \text{Mg}^{2+} + 2\text{HCO}_3^{-} \\ \text{(Dolomite)} \end{array} \tag{10}$$

Fig. 11 illustrates alterations in cationic content within the resultant liquid under varying sequestration pressures. Post-



Fig. 11. The ionic composition of the produced liquid changed pre- and postexperimentation.

sequestration, discernible elevations in the concentrations of Na⁺, K^+ , and Ca^{2+} ions manifest within the produced solution. Additionally, the analysis, in conjunction with Eqs. (7)–(10), elucidates the origins of these cations. It is evident that the dissolution processes induce an increase in the concentrations of Na⁺, K⁺, and Ca²⁺ ions within the effluent, attributable to the dissolution of sodium feldspar, potassium feldspar, as well as calcite and dolomite. respectively. Furthermore, the presence of Mg^{2+} ions in the produced solution is attributed predominantly to the dissolution of dolomite and chlorite, among other minerals encapsulated within the core. Notably, the dissolution effect intensifies with escalating sequestration pressures, as evidenced by the heightened concentration of Mg^{2+} ions detected in the effluent. It becomes apparent that as sequestration pressure increases, the dissolution effect grows in magnitude, consequently leading to a more substantial presence of Mg^{2+} ions in the output fluid. In summation, this CO_2 sequestration dissolution experiment unveils differential dissolution propensities between feldspar minerals and carbonate minerals within the core. It is notably discerned that the dissolution degree of carbonate minerals surpasses that of feldspar minerals. Additionally, a comparative analysis underscores that the increment in Na⁺ ion content exceeds that of K⁺ ions, stemming from the greater susceptibility of sodium feldspar to dissolution compared to potassium feldspar under acidic conditions.

3.4. Analysis of precipitation effect

In the context of CO₂ sequestration, the interplay between CO₂ and crude oil engenders a discernible alteration in the composition of the latter. When CO₂ is sequestered within geological formations, it transitions into a supercritical state, thereby facilitating the extraction of lighter fractions from the crude oil. Concurrently, as sequestration pressure increases, a substantial quantity of CO₂ dissolves into the crude oil, resulting in an augmented deposition of resin and asphaltene constituents. As depicted in Fig. 12, this phenomenon unfolds progressively, with the content of resin and asphaltene in the recovered hydrocarbons registering a continuous decline. This trend signifies an escalating accumulation of asphaltene within the pore network. Notably, once the sequestration pressure surpasses the 10 MPa threshold, there is a conspicuous upswing in the mass fraction of asphaltene deposition, ultimately culminating at 16.0%. Upon reaching a pressure magnitude of 20 MPa, CO₂ transitions into a near-miscible state within the crude



Fig. 12. The content changes of four components of crude oil pre- and post-experimentation.

oil. This transformation is accompanied by a surge in the quantity of CO_2 dissolved in the crude oil, thereby precipitating a sharp escalation in the mass fraction of asphaltene deposits, which reaches a pinnacle of 28.8%. Consequently, an extensive accumulation of asphaltene within the core's pore spaces occurs, leading to blockages. As the sequestration pressure escalates to 25 MPa, CO_2 continues to exhibit a heightened propensity for dissolution in the crude oil, causing the mass fraction of asphaltene deposition to surge to 33.6%. This exacerbates the phenomenon of pore plugging, thereby accentuating the severity of the issue.

4. Mechanism of pore structure changes in the $\ensuremath{\text{CO}}_2$ sequestration

4.1. Calculation of CO₂ sequestration capacity

During the CO₂ sequestration process, soluble minerals such as potassium feldspar, sodium feldspar, calcite, and dolomite in the core are dissolved resulting in the increase of pore volume. In order to quantitatively evaluate the CO₂ sequestration amount, a method based on NMR T_2 spectra is proposed to calculate the degree of pore volume change and subsequently calculate the CO₂ sequestration amount. This method can be used to quantitatively assess the amount of CO₂ required for mineralization sequestration of soluble minerals during CO₂ sequestration process.

Initially, based on Eq. (3), the degree of pore change ε can be obtained, which is considered that the changes in pore volume are attributable to the dissolution of soluble minerals, and the reaction volume of soluble minerals can be ascertained:

$$V_{\rm S} = \frac{\pi d^2}{4} \cdot L \cdot \epsilon \tag{11}$$

where V_S represents the reaction volume of soluble minerals, cm³, *d* is the core diameter, cm, and *L* is the core length, cm.

The density of soluble minerals during CO_2 dissolution is expressed as the average value of soluble minerals involved in the reaction and is calculated from Eq. (12):

$$D_{\rm S} = \frac{D_1 + D_2 + D_3 + D_4}{4} \tag{12}$$

where the symbol D_S is the average density of the soluble minerals, g/cm³, D_1 , D_2 , D_3 , and D_4 correspond to the densities of potassium feldspar, sodium feldspar, calcite, and dolomite respectively, g/cm³.

According to Eqs. (11) and (12), the mass of soluble minerals involved in the reaction can be obtained as follows:

$$m_{\rm S} = D_{\rm S} \cdot V_{\rm S} \tag{13}$$

where $m_{\rm S}$ is the mass of soluble mineral, g.

To calculate the mass of each type of soluble mineral participating in the reaction based on its proportion in the core, the calculation method is shown in Eq. (14) as follows:

$$m_i = \frac{\alpha_i}{\alpha_1 + \alpha_2 + \alpha_3 + \alpha_4} \cdot m_{\rm S}, (i = 1, 2, 3, 4)$$
(14)

where m_i is the mass of each soluble mineral, g, α_i is the content of each soluble mineral as a percentage, %.

After obtaining the mass of each type of soluble mineral participating in the reaction, the amount of H^+ consumed by the reaction of potassium feldspar can be calculated using the chemical reaction Eqs. (4)–(6). The amount of CO₂ consumed in the reaction of sodium feldspar can be determined from Eq. (8), the amount of H^+ consumed in the reaction of calcite can be calculated from Eq.

(9), and the amount of H^+ consumed in the reaction of dolomite can be determined from Eq. (10). Therefore, the amount of CO_2 participating in the mineralization reaction can be calculated using the following equation:

$$n_{\rm C} = \frac{n_1 + n_3 + n_4}{2} + n_2 \tag{15}$$

in the formula, $n_{\rm C}$ represents the amount of substance of CO₂ involved in the mineralization reaction, mol, the symbols n_1 , n_3 , and n_4 correspond to the amount of substance of H⁺ consumed by potassium feldspar, calcite, and dolomite, respectively, mol, n_2 is the amount of substance of CO₂, mol.

Consider the compressibility of gases under varying temperature and pressure conditions, the volume of CO_2 sequestered under specific temperature and pressure conditions can be derived using the actual gas state equation (Eq. (16)).

$$PV_{\rm C} = Zn_{\rm C}RT \tag{16}$$

where *P* is the sequestration pressure, Pa, V_C is the volume of CO₂, m³, *Z* is the CO₂ compression factor, *R* is the gas constant with a value of 8.31 [/(mol·K), *T* is the sequestration temperature, K.

In summary, the calculation method for CO_2 sequestration can be summarized in two steps. Firstly, the mass expression of the soluble minerals involved in the reaction is calculated based on the change in pore volume, as presented in Eq. (17) part ①. Secondly, the consumed CO_2 is calculated according to the corresponding chemical reaction equation, as indicated in part ② of Eq. (17). The CO_2 sequestration amount can thus be obtained, and the calculation results along with relevant parameters are presented in Table 6.

$$\begin{cases} m_{\rm i} = \frac{\pi d^2 L \varepsilon}{16} \cdot \frac{\alpha_{\rm i} \cdot (D_1 + D_2 + D_3 + D_4)}{\alpha_1 + \alpha_2 + \alpha_3 + \alpha_4} & \textcircled{0} \\ V_{\rm C} = \frac{Z R T \cdot (n_1 + 2n_2 + n_3 + n_4)}{2P} \cdot 10^6 & \textcircled{2} \end{cases}$$
(17)

in the equation: $V_{\rm C}$ represents the CO₂ sequestration amount, mL.

4.2. Mechanism of pore structure changes

In the course of CO_2 sequestration, a dichotomy emerges in its effects on geological formations. Firstly, dissolution processes transpire as CO_2 reacts with formation water, giving rise to the formation of carbonic acid. This event confers a dual impact, enhancing the physical attributes of the core while concurrently expanding its pore structure. However, upon the amalgamation of CO_2 with crude oil, a scenario ensues wherein asphaltene precipitation occurs. This process instigates the accumulation of organic matter deposits within pore throats, ultimately resulting in the blockage of smaller pores and the consequent exacerbation of damage. The integration of CO_2 with formation fluids engenders an acidic milieu within the geological formation, as elucidated in Fig. 13(a). In this acidic environment, carbonate minerals and feldspar components within tight sandstone exhibit a propensity to dissolve. Moreover, with an increased dissolution of CO_2 within

| Tab | le 6 | | | | |
|-----|-------------|---------|--------------------|---------|---------|
| The | calculation | results | of CO ₂ | sequest | ration. |

crude oil, the deposition of asphaltene within the pores experiences an upsurge, with a substantial accumulation occurring at the throats of smaller pores, thereby yielding core pore impairment, as portrayed in Fig. 13(b). As sequestration pressure intensifies, the interaction dynamics between CO2, reservoir fluids, and rock minerals undergo enhancement. Consequently, there is an amplification in both dissolution processes and asphaltene deposition within the core. In parallel, mineral particles and asphaltene molecules engage in migration, ultimately occupying pore spaces within the geological formation. This intricate interplay exerts a discernible influence on the reservoir performance of the geological body, as exemplified in Fig. 13(c). Within the framework of CO_2 sequestration, the cementation that previously filled the interstitial spaces between pores, as depicted in Fig. 13(d), experiences dissolution. This dissolution, while augmenting pore connectivity, introduces a counteracting effect, wherein asphaltene molecules obstruct minute pore throats. This obstruction, in turn, curtails the overall sequestration and seepage capacity of the geological formation

In summary, within the realm of CO₂ geological sequestration, CO₂ interacts intricately with both formation fluids and geological substrata. On one facet, CO₂ undergoes dissolution in formation water, resulting in the production of carbonic acid. This acid subsequently engages in reactions with feldspar, leading to the disintegration of the mineral framework. Concurrently, heightened CO₂ sequestration pressures prompt the detachment and migration of clay particles through the rock matrix. This phenomenon amplifies the interconnectedness of pores, collectively culminating in a noteworthy augmentation of the geological body's permeability and porosity. These cumulative effects play a pivotal role in instigating transformations within the pore structure of the geological formation. Conversely, under elevated sequestration pressures, CO₂ dissolves extensively in crude oil, thereby inducing the release and subsequent deposition of asphalt constituents from the crude oil into the pores of the geological formation. This eventuality precipitates the occlusion of pore spaces within the rock, thereby detrimentally impacting the seepage and sequestration capacity of the geological body. In a comprehensive appraisal, it is evident that with the escalation of CO₂ sequestration pressure, the effects of dissolution and precipitation of organic matter are intensified. Both effects collectively contribute to alterations in the microscopic pore structure of the geological body, thereby exerting implications on the stability of CO₂ sequestration within the geological formation. Consequently, when embarking upon CO₂ sequestration endeavors, it is imperative to meticulously account for the geological context and judiciously select an appropriate sequestration pressure. This ensures not only the efficacy of sequestration but also underscores the imperatives of safety and stability in the sequestration process.

4.3. Further discussion

CO₂ geological sequestration is a prolonged and intricate process, making it challenging to fully comprehend the mechanisms involved. This technology encompasses four primary methods: structural sequestration, mineral sequestration, dissolution sequestration, and adsorption sequestration (Hosseininoosheri

| ID | P, Pa | <i>Т</i> , К | Ζ | D_1 , g/cm ³ | D_2 , g/cm ³ | D ₃ , g/cm ³ | D_4 , g/cm ³ | <i>m</i> ₁ , g | <i>m</i> ₂ , g | <i>m</i> 3, g | <i>m</i> 4, g | V _C , mL |
|----|---------------------|--------------|--------|---------------------------|---------------------------|------------------------------------|---------------------------|---------------------------|---------------------------|---------------|---------------|---------------------|
| 1 | 1.0×10^7 | 313.5 | 0.4804 | 2.54 | 2.61 | 2.71 | 2.84 | 0.2536 | 0.1761 | 0.1902 | 0.0845 | 4.80 |
| 2 | 1.5×10^{7} | 313.5 | 0.4025 | | | | | 0.3937 | 0.2734 | 0.2953 | 0.1312 | 5.10 |
| 3 | 2.0×10^7 | 313.5 | 0.3250 | | | | | 0.7859 | 0.5458 | 0.5894 | 0.2620 | 2.75 |
| 4 | 2.5×10^7 | 313.5 | 0.2689 | | | | | 0.7744 | 0.5377 | 0.5808 | 0.2581 | 2.20 |



Fig. 13. Schematic diagram of dissolution and precipitation mechanism.

et al., 2018; Lyu et al., 2021; Xie et al., 2021). In this study, due to the short reaction time, CO_2 primarily demonstrated adsorption sequestration. The interaction mechanisms between CO_2 and rock vary among these methods, presenting a challenge for a single model to adequately explain the changes in pore structure within CO_2 sequestration reservoirs. Therefore, comprehensive analysis and simulation are necessary to explore optimization strategies for different sequestration methods and to ensure their long-term stability (Jiang, 2011). This study preliminarily elucidated the mechanisms of pore structure changes in tight sandstone during CO_2 sequestration. Although the experiment lasted only 168 h, which is short compared to the actual timescales of hundreds or even thousands of years for CO_2 sequestration, it effectively reflects the short-term characteristics of sequestration following CO_2 injection into the reservoir.

Future research should integrate longer-term experiments and numerical simulations to further validate these findings. Numerical simulations can assess the stability and effectiveness of CO₂ sequestration over extended periods, accounting for complex processes such as multiphase fluid flow, rock-fluid interactions, and mineral dissolution and precipitation. This approach will enable the simulation of the long-term CO₂ sequestration process. Additionally, future work should include multiscale analyses to comprehensively understand the fluid dynamics and rock responses during CO₂ sequestration. This approach should be combined with on-site CO₂ sequestration projects to develop new methods and technologies to address related challenges.

5. Conclusions

The primary determinants shaping alterations in the permeability and sequestration capacity of geological formations during the CO₂ geological sequestration process hinge upon the phenomena of dissolution and precipitation. This study, leveraging NMR technology, has undertaken a comprehensive exploration of CO_2 geological sequestration dissolution and asphaltene precipitation. It delves into the transformation of mineral composition, micro-pore architecture, and fluid mobility under varying CO_2 sequestration pressures. Moreover, it quantitatively elucidates the shifts in the seepage and sequestration capabilities of the geological body during CO_2 sequestration. Additionally, the research introduces a method based on changes in the NMR T_2 spectrum to calculate the degree of pore volume change. Consequently, this calculation is employed for a quantitative assessment of CO_2 sequestration effectiveness, providing valuable insights into the amount of CO_2 sequestered. The findings culminate in the following key conclusions.

- (1) In the context of CO_2 geological sequestration, dissolution processes wield a diverse influence, imparting variable degrees of enhancement to the porosity and permeability of tight sandstone. This manifests through the diminution of the core's $T_{2cutoff}$ and an escalation in the percentage of mobile fluid. Concurrently, dissolution instigates alterations in the core's mineral composition, signified by diminished levels of carbonate and feldspar minerals. Notably, at a pressure magnitude of 15 MPa, scrutiny under lowmagnification SEM reveals the presence of numerous dissolution pores on the core's surface. Upon closer examination, higher magnification underscores the emergence of harborlike dissolution features within feldspar minerals, with select dissolution pores harboring debris.
- (2) Asphaltene precipitation, an integral facet of the CO_2 geological sequestration process, precipitates deposits within the pore spaces, leading to a discernible augmentation in the core's $T_{2cutoff}$. Simultaneously, its ushers in reductions in the percentages of movable fluid and movable fluid porosity. These consequences exert a direct influence on the core's sequestration and permeability capacities. Notably, with an escalation in CO_2 sequestration pressure, the

precipitation process assumes a substantially intensified character. This culminates in more pronounced asphaltene deposition and a marked escalation in the extent of pore transformations.

(3) Throughout the trajectory of the CO₂ geological sequestration process, the dissolution effect emerges as a force that dissolves select minerals, consequently amplifying the geological formation's sequestration and permeability capabilities. Conversely, the precipitation effect instigates asphaltene deposition and thereby resulting in a deterioration of the geological body's physical properties. This phenomenon translates into a diminished sequestration capacity, constricted sequestration space, and a reduction in inter-pore connectivity.

CRediT authorship contribution statement

Hui Gao: Writing – review & editing, Conceptualization. Kai-Qing Luo: Writing – original draft, Methodology, Investigation. Chen Wang: Methodology, Formal analysis. Teng Li: Investigation, Data curation. Zhi-Lin Cheng: Writing – original draft, Investigation. Liang-Bin Dou: Visualization, Formal analysis. Kai Zhao: Visualization, Data curation. Nan Zhang: Writing – review & editing. Yue-Liang Liu: Writing – review & editing, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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