CO_2-Water-Sandstone Reactions and Mineralization Sequestration Mechanism in the South Yellow Sea Basin under Temperature-Pressure Conditions

Introduction

Nature possesses an inherent carbon absorption mechanism. However, excessive anthropogenic CO_2 emissions disrupt the dynamic balance between carbon sources and sinks (Marak et al. 2025), exacerbating the greenhouse effect and driving global warming. In response, the 2015 Paris Agreement set the goal of limiting the global average temperature rise to within 1.5°C above pre-industrial levels (IPCC 2014). Consequently, CCUS technology has become a critical component of the carbon neutrality system (Zhang et al. 2023). Among these, CO_2 mineral storage has garnered widespread attention as one of the most stable carbon storage methods (Bachu 2008; Benson et al. 2012). Internationally, two major demonstration projects—CarbFix in Iceland and Wallula in the USA—have confirmed the rapid mineralization capacity of CO_2 in basalts, with mineralization rates exceeding 95% and 60% respectively over two-year monitoring periods (Aradottir et al. 2011; Gislason et al. 2018; Matter et al. 2009; McGrail et al. 2011).

Compared to basalts, feldspar minerals in clastic rock reservoirs are not the most favorable for mineralization storage. However, clastic rock reservoirs predominate in China's offshore sedimentary basins. These sandstone layers, characterized by suitable burial depth, significant thickness, wide distribution, and young geological age, generally exhibit high porosity and permeability, potentially meeting large-scale storage requirements (Mohsin 2024; Ringrose and Meckel 2019). Domestic research on sandstone mineralization storage has primarily focused on reaction mechanisms, numerical simulation, and potential assessment (Mo et al. 2014; Bao et al. 2024; Liu et al. 2024). Most studies utilize high-temperature and high-pressure reactors with continental sandstone cores from basins such as Ordos and Songliao. Under simulated in-situ temperature, pressure, and fluid conditions, these studies investigate mineral dissolution sequences, secondary mineral formation, and their impact on reservoir properties (Liu 2018; Tao 2013; Wu and Liu 2025; Zhang et al. 2025).

However, previous research has paid relatively little attention to CO_2-water-rock mineralization processes in clastic reservoirs of marine sedimentary basins. China's coastal regions face high carbon emission intensity and significant pressure for emission reduction (Yuan et al. 2025). Furthermore, while onshore sedimentary basins are scarce, offshore sedimentary basins along the coast offer substantial CO_2 storage potential due to their large area and thick reservoirs. Therefore, this study selected sandstone samples from well CZ35-2-1 in the South Yellow Sea Basin to conduct high-temperature and high-pressure CO_2-water-rock simulation experiments. The objectives are to elucidate the kinetic mechanisms of CO_2-water-rock reactions in sandstone reservoirs, quantitatively characterize the control of different temperature and pressure conditions on mineralization storage, and thereby evaluate the long-term storage potential and safety of the reservoir. The results will provide an experimental basis for optimizing temperature and pressure conditions and predicting storage potential for CO_2 geological storage sites.

Method and/or Theory

This study used a high-temperature and high-pressure CO_2-water-rock reaction apparatus independently developed by the Qingdao Institute of Marine Geology. The reactor has a volume of 600 mL, a maximum pressure of 30 MPa, and a temperature range of 0–200°C. An external real-time sampling system allowed micro-sampling during the reaction. This equipment enables full-process monitoring of CO_2-water-rock interactions under high-temperature and high-pressure conditions. A schematic diagram of the apparatus is shown in Figure 1.

The rock samples used in the reaction were divided into two types: block samples suitable for electron microscopy analysis, and powder samples ground into fine particles. For each experimental group, 15.00 g of powder sample was used. The gas source was CO_2 with 99.99% purity. The aqueous medium

in the reactor was synthetic formation water prepared based on the ion concentrations measured in actual geothermal water wells from the Yancheng Formation in the Subei Basin. The solid-to-liquid ratio was set at 1:30. The formation temperature was approximately 40°C, and the formation pressure was about 11 MPa. To investigate the effects of different temperature and pressure conditions on the CO_2-water-rock reaction process, additional conditions of 80°C and 21 MPa were also set. A total of four experimental groups were designed, as shown in Table 1.

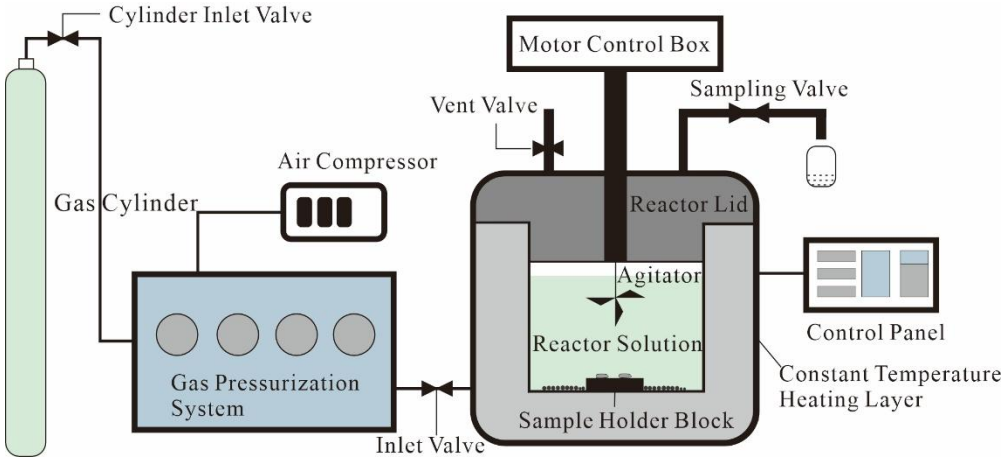

Figure 1. Schematic diagram of the experimental apparatus

Table 1. Experimental design

Experimental Group	Temperature (°C)	Pressure (MPa)	Time (h)
Experimental Group	40	11	552
High-Temperature Low-Pressure Group	80	11	552
High-Pressure Low-Temperature Group	40	21	552
High-Temperature High-Pressure Group	80	21	552

The experimental procedure was as follows: (1) Solid samples were weighed; (2) The reactor was assembled and heated to the target temperature, then CO_2 was injected to the set pressure, with timely replenishment during the experiment to maintain constant pressure; (3) Samples were collected at regular intervals and labeled; (4) After the experiment, the remaining solid samples were weighed; (5) Dissolution features and newly formed minerals on the block samples were observed and analyzed using SEM and EDS; (6) The pH value, major cation concentrations, and HCO_3^- concentration of the liquid samples were measured.

Results

In this study, increasing the temperature from 40°C to 80°C, despite reducing the solubility of CO_2 in the formation water, significantly enhanced the chemical reactivity of minerals and the ionization degree of water. This led to a simultaneous increase in H^+ and HCO_3^- concentrations in the system, resulting in an overall increase in acidity. In these two experimental groups, silicate minerals (primarily feldspars) showed a substantial response to temperature, with their dissolution rates controlled by multiple factors including temperature, pH, and fluid chemical composition (Schott et al. 2024).For carbonate minerals, on one hand, the same acidic conditions clearly promoted the dissolution of magnesium-bearing minerals (such as dolomite); on the other hand, however, high temperature altered the dissolution equilibrium of calcite, resulting in weaker dissolution intensity in the high-temperature low-pressure group compared to the low-temperature group.

When the system temperature was constant, high-pressure conditions significantly enhanced CO_2 solubility, increasing the total amount of CO_2 participating in dissolution and ionization per unit volume. This continuously replenished H^+, lowering the system pH and thereby significantly promoting mineral dissolution. Experiments showed that under high-pressure conditions, the degree of albite dissolution was greater than that of K-feldspar. However, due to passivation effects and differences in the initial mineral content of the reaction samples, K^+ release was hindered. Simultaneously, magnesium-bearing minerals (such as ankerite) and calcite exhibited enhanced dissolution under high pressure, releasing more Ca^{2+}, Mg^{2+}, and Fe^{2+}/Fe^{3+}. Due to secondary precipitation, the concentrations of these ions in the solution fluctuated throughout the experiment.

In CO_2 geological storage, the reservoir environment is a coupled system where temperature and pressure interact. When both temperature and pressure increased simultaneously, the CO_2-water-rock reaction was instead inhibited, indicating that the two variables do not simply superimpose to enhance the reaction. Overall, both temperature and pressure conditions intensified acidification, but in this experiment, the influence of temperature was more pronounced. For silicate minerals, the synergistic effect of temperature and pressure resulted in significant leaching of Na^+, K^+, and Si ions, which maintained a state of dissolution throughout the experimental period. However, the impact on Ca^{2+} and Fe ion leaching was not significant, with their concentration changes depending more strongly on pressure conditions.

Conclusions

(1) Temperature and pressure exert distinct effects on mineral dissolution. At constant pressure, increasing temperature significantly promotes the dissolution of silicate minerals (albite, K-feldspar) and the release of Na^+ and K^+. At constant temperature, rising pressure enhances CO_2 solubility and system acidity, driving the dissolution of calcite and iron-bearing minerals and accelerating Ca^{2+} and Fe release.

(2) The coupled effect of temperature and pressure is not a simple superposition. When both increase simultaneously, the total dissolution is lower than that under high pressure alone; the release of Na^+, K^+, and Mg^{2+} is significantly enhanced, whereas the promotion of Ca^{2+} and Fe release is similar to that under high pressure alone.

(3) Microscopic observations show that with increasing temperature and pressure, dissolution pits, honeycomb-like pores, and passivation of mineral edges develop on sample surfaces, with their extent closely related to temperature and pressure conditions.

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