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Review Article

Experimental Methods and Progress of CO_2-Brine-Rock Interactions for CO_2 Storage in Saline Aquifers

Qian Chen, Jian Hou*, Bei Wei, Jiahe Sun, Qingjun Du

Department of Petroleum Engineering, China University of Petroleum (East China), Qingdao 266580, P.R. China

ABSTRACT

The greenhouse effect caused by excessive CO_2 emissions is a great challenge facing the world, and CO_2 geological storage in saline aquifers is an effective way to solve this problem because of its great storage potential. The injection of CO_2 into saline aquifers interacts with brine and rock, leading to complex physical and chemical reactions, which have a direct impact on the safety of CO_2 storage. This review synthesizes findings from recent researches, particularly focusing on experimental studies of CO_2-brine-rock interactions. Through a detailed review of both static dissolution and dynamic flow experiments, CO_2-brine-rock interaction mechanisms and the influence of fluid/rock interactions on properties of brine and rock are elucidated. Meanwhile, this review points out that innovative experimental methods are needed in future research to improve the accuracy of measuring parameters. It is also recommended to conduct CO_2-brine-rock interaction experiments under *in situ* conditions in the reservoir on a long-time scale, which will improve the accuracy and reliability of parameters for subsequent seepage simulations.

Keywords: CO_2 storage; CO_2-brine-rock interactions; Saline aquifers; Static dissolution; Dynamic flow

INTRODUCTION

In recent years, Carbon Capture and Storage (CCS) technology has received widespread attention because it is an effective method to reduce atmospheric CO_2 levels [1]. CO_2 storage sites mainly include saline aquifers, depleted oil and gas reservoirs, and unmineable coal seams [2]. Among them, saline aquifers are considered to be the most suitable due to their large storage capacity. The injection of CO_2 into saline aquifers interacts with brine and rock, leading to complex physical and chemical reactions.

Following the injection of CO_2 into the saline aquifers, it moves upward due to the force of buoyancy and gathers in the lower part of the cap layer at the top of the reservoir. This process is analogous to structural trapping, whereby the CO_2 is effectively storage. During its upward movement, CO_2 contacts with brine. The dissolved CO_2 is then storage in the brine by means of dissolution trapping, resulting in an increase in the density of the brine. Brine then moves downward under the action of gravity, generating convection currents. Concurrently, CO_2 diffuses into the minute pores, where it is trapped by capillary pressure and subjected to residual trapping. The dissolution of CO_2 in brine results in the generation of H_2CO_3, which subsequently reduces the pH value of the reservoir environment, while H_2CO_3 decomposes into HCO_3- and CO_3^{2-}. In different time scales, mineralization reaction with brine and rock minerals occurs, resulting in the storage of the process in the form of mineral trapping. As the reaction time is extended, chemical storage (e.g. dissolution trapping and mineral trapping) becomes the dominant process. Dissolution trapping is the most prevalent form of storage, while mineral trapping is the most secure [3].

Indoor dissolution and dynamic flow experiments are one of the most effective ways to understand the short-term performance of CO_2 storage process. The former focuses on the solubility of CO_2 in brine, the ionic composition and concentration of brine, the dissolution of rock minerals, and the generation of secondary minerals. While the latter focuses on the flow and transport laws of CO_2 in porous medium of reservoirs, as well as

Correspondence to: Jian Hou, Department of Petroleum Engineering, China University of Petroleum (East China), Qingdao 266580, P.R. China; E-mail: houjian@upc.edu.cn

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changes in the pore-throat characteristics and reservoir wettability in the process of CO_2 storage.

This paper takes the CO_2-brine-rock system as the background, systematically reviews the current research status of indoor static dissolution and dynamic flow experiments, elucidates CO_2-brine-rock interaction mechanisms, and sort out the main problems and technical bottlenecks of the current research, and puts forward the direction of future research.

LITERATURE REVIEW

Static dissolution experiments

CO_2 solubility experiments: The study of CO_2 solubility in brine focuses on its influence on temperature, pressure, and salinity. As the temperature decreases and the pressure increases, CO_2 solubility in brine increases. Scholars have obtained results on the solubility of CO_2 through experimental methods. The most widely used model for calculating the CO_2 solubility in brine is Duan and Sun's solubility model, but the model ignored the influence of binary interaction parameters [4]. Ratnakar et al. developed a method based on machine learning to calculate the solubility of CO_2 in brine under different conditions and determined key characteristics of brine components that make significant contributions [5].

Mineral dissolution and precipitation experiments: Due to the unstable chemical properties of many rock minerals (calcite, dolomite, feldspar, etc.) and cementing materials in the reservoir, they will dissolve under the action of chemical reactions when in contact with weakly acidic fluids formed by CO_2 and brine. Scholars used high-temperature and high-pressure reactors to simulate CO_2-brine-rock interaction process under actual reservoir temperature and pressure. The results indicate that during the reaction process, silicate minerals and carbonate minerals mainly composed of potassium feldspar and calcium feldspar underwent dissolution, accompanied by the formation of new mineral precipitates such as calcite and iron calcite. Dissolution and precipitation lead to changes in physical parameters such as reservoir porosity and permeability [6]. In addition, Navarre Sitchler et al. used stacked electron scanning microscope images, 3D pore reconstruction, and small angle neutron dispersion techniques to study and discuss the changes in porosity and permeability of mudstone after reaction under CO_2 brine solution conditions [7].

Dynamic flow experiments

Core flooding experiments: Different from static dissolution experiments, core flooding experiments focus more on the characteristics of changes in reservoir porosity, permeability, and wettability [8]. Many scholars have conducted relevant experimental work, and the comprehensive results show that CO_2 has a strong dissolution phenomenon on rock minerals under the action of brine. The number of dissolution pores increases, the original pore size becomes larger, and the porosity and permeability of the core increase [9].

Sand pack experiments: Sand pack experiments are conducted on steel pipes with sufficient pressure levels pre-filled with glass beads or sand particles under different lengths. Compared to cores, the length of sand pack is more flexible and the material is easy to obtain. Therefore, sand pack experiments are often used to study reservoir porosity, permeability heterogeneity, and other issues. Plug and Bruining presented a method with which static drainage and imbibition capillary pressures can be measured continuously as a function of saturation based on unconsolidated sand-distilled water-CO_2 system. Jiang et al. used a glass packed bed to provide a quantitative investigation of CO_2-liquid mass transfer.

Microscopic visualization experiments: Microscopic visualization model is a transparent pore network used for laboratory research on the microscopic distribution and transport of fluids in porous media. This model can intuitively observe CO_2 flow patterns and occurrence states o under different phase states and flow rates. Kim et al. used transparent engineered silica micromodels to study the wetting behavior of $scCO_2$ by directly visualizing the phase displacement processes under conditions relevant to geological CO_2 storage. A visualization test is also carried out in a Helee Shaw cell for the observation of convective instabilities and their effect on the dissolution rate.

CT scanning experiments: X-ray Computed Tomography (CT) is increasingly being used by researchers to obtain fluid distribution in the pores and throats, as it does not damage the core and has repeatability. Øren used a micro-CT to characterize the pore structure of core samples and studied CO_2 residual trapping. Supercritical CO_2 cluster captured in sandstone under high-temperature and high-pressure conditions were imaged and the distribution of captured CO_2 cluster was measured. Chen et al. used CT scanning technology to in situ measure the contact angle of the reservoir under the CO_2-brine-rock system to monitor the wettability of the reservoir.

DISCUSSION

Indoor experiments are an effective way to quickly understand CO_2-brine-rock interactions under reservoir conditions. The study of CO_2-brine-rock interaction mechanisms is mainly carried out through static dissolution and dynamic flow experiments. In general, current experimental researches mainly use mineral composition analysis, scanning imaging, fluid composition analysis, and other methods to monitor changes in mineral composition, pore throat changes, and ultimately changes in core porosity and permeability before and after the reaction or during the reaction process.

However, current researches lack targeted characterization methods to characterize the dynamic processes and characteristics of CO_2-brine-rock interactions. Therefore, it is necessary to innovate relevant experimental devices and methods to enrich the understanding of CO_2-brine-rock interaction mechanisms and provide reliable theoretical support. For example, online dynamic flow experiments using CT scanning devices can be employed to elucidate CO_2 migration patterns in porous medium. Long-term static dissolution experiments using visualized high-temperature and high-pressure reaction vessels can be conducted to monitor the changes in brine components and mineral dissolution at different reaction times.

CONCLUSION

After CO_2 is injected into saline aquifers, it will interact brine and rock to undergo complex physical and chemical reactions. Clarifying CO_2-brine-rock interaction mechanisms under reservoir conditions is crucial for the safety of CO_2 storage. This review systematically summarizes the research results of indoor static dissolution and dynamic flow experiments, and also points out possible future research directions.

Static dissolution experiments are conducted to elucidate CO_2 solubility in brine, changes in brine components, and the dissolution and precipitation of rock minerals. Dynamic flow experiments are carried out to monitor the pore-throat characteristics and wettability changes of rocks during CO_2 storage process. These experiments are one of the most effective ways to understand the short-term performance of CO_2 storage process.

In future research, it is essential to innovate the time scale, experimental methods, and other aspects to quantitatively characterize relevant experimental parameters, so as to more accurately elucidate CO_2-brine-rock interaction mechanisms and provide theoretical basis and technical support for the safe implementation of CCS technology.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interest regarding the publication of this paper.

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