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Study on microstructure change of underground gas storage facilities and its influence on permeability

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Abstract: *This study explores the changes in the microstructural characteristics of rocks within underground natural gas storage facilities and their impact on permeability. Laboratory simulations of natural gas storage conditions were conducted using representative rock samples from the coastal area of Tianjin, China. The microstructures of the rocks before and after gas storage were analyzed and compared using Scanning Electron Microscopy (SEM) and X-Ray Diffraction (XRD). The findings reveal that high pressure and chemical reactions under storage conditions lead to significant changes in the mineral composition and crystal structure of the rocks, which directly affect their permeability. Quantitative analyses of porosity and fracture density further confirmed changes in permeability. Additionally, this research developed a statistical model that clearly defines the quantitative relationship between microstructural parameters and permeability, providing a scientific basis for the design and risk management of natural gas storage facilities.*

Keywords: *Underground Gas Storage, Rock Microstructure, Permeability, Scanning Electron Microscopy (SEM), X-Ray Diffraction (XRD)*

1. Introduction

The long-term storage of natural gas in underground facilities is a critical component in energy management, particularly for balancing seasonal demand fluctuations. The integrity and efficiency of these storage facilities largely depend on the geological characteristics and the behavior of the surrounding rock formations under various pressures and chemical conditions [1]. Understanding the microstructural changes in rocks during gas injection and retrieval is crucial, as these changes impact permeability and storage capacity [2].

Advancements in imaging technologies, such as Scanning Electron Microscopy (SEM) and X-Ray Diffraction (XRD), have provided new insights into the microstructural properties of geological materials. These techniques are invaluable for characterizing the mineral composition and crystal structure of rocks at a microscale [3][4]. Changes in these characteristics are believed to directly influence the rock's permeability, a key factor in determining the effectiveness of gas storage [5].

Moreover, the interaction between stored gas and rock formations involves complex physical and chemical processes that can alter the rock's structural integrity [6]. For instance, the exposure to high pressures and reactive chemical environments typical in gas storage operations can lead to significant microstructural alterations, such as pore collapse or fracture propagation, which can drastically affect the rock's ability to store gas [7][8].

The quantitative relationship between rock microstructure changes and permeability has been the focus of several recent studies. These have established statistical models to predict permeability changes based on measurable microstructural parameters [9]. Such models are crucial for the design and operation of safer and more efficient gas storage facilities.

This paper aims to build on these foundational studies by providing a detailed analysis of rock samples from the coastal area of Tianjin, China, under simulated storage conditions. The goal is to enhance the understanding of how specific microstructural changes correlate with permeability alterations, thereby offering insights that could improve prediction models for gas storage integrity and risk management [10].

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2. Materials and Methods

2.1. Study Area and Sample Collection

The Tianjin coastal region, located in northeastern China, was chosen as the research area due to its unique geological features and relevance to natural gas storage. This region, part of the Bohai Economic Rim, is known for its sedimentary basins primarily composed of quaternary alluvial deposits, characterized by diverse lithological compositions, including sandy loam and clay, which are pertinent to the study of underground natural gas storage facilities. The collection of rock samples aimed to ensure comprehensive representation of geological variability across the region. Samples were collected from three sites: near the urban district of Binhai, the rural outskirts of Ninghe County, and along the coastal line of Dagang. Each site was selected based on its unique sedimentary characteristics, typical of conditions found in underground storage areas.

Geographical Coordinates and Collection Methodology:

▪ Binhai (Urban District): Coordinates 39.035 °N, 117.689 °E. Samples were collected from a depth of approximately 30 meters using core drilling techniques to minimize surface contamination and ensure the integrity of the sediment layers.

▪ Ninghe County (Rural Outskirts): Coordinates 39.330 °N, 117.830 °E. Here, rock samples were extracted from 50 meters below the surface, reflecting deeper sediment features, critical for assessing the ·potential for natural gas storage.

▪ Dagang (Coastal Line): Coordinates 38.900 °N, 117.450 °E. Collection at this site involved shallow surface sampling and deeper extractions at 20 and 40 meters respectively, to provide a gradient of material properties affected by proximity to the sea.

Environmental conditions during the collection included a temperature range of 15-25 ℃ and humidity levels from 55 to 65%, typical for this region during the spring season. These conditions are crucial as they influence the moisture content and compaction of sedimentary rocks, which can affect their porosity and permeability characteristics.

Each sample was catalogued with its respective geographic and environmental data to maintain traceability and facilitate precise analysis correlation. This meticulous approach to sample collection aims to bolster the reliability of the subsequent experimental results, providing a robust foundation for the study of microstructural changes under simulated natural gas storage conditions.

2.2. Sample Preparation

Upon arrival at the laboratory, all samples were initially subjected to a standard cleaning process to remove any adherent external debris or contaminants that could interfere with the analysis. This process involved gently washing the samples with distilled water followed by air drying at room temperature (approximately 23 ℃) for 24 hours.

Post-cleaning, each sample underwent a careful examination to select homogeneous sections that were free of visible cracks or anomalies, ensuring consistency across all analyses. These selected sections were then marked for further processing.

Treatments and Conditioning:

1) Drying:

To standardize the moisture content and eliminate any effects of variable water content on the microstructure, the selected rock sections were dried in a controlled environment. This was achieved by placing them in an oven set at 50 ℃ for 48 hours, ensuring that all residual moisture was removed.

2) Grinding:

After drying, each sample was ground to a uniform size to fit the sample holders used for SEM and XRD analyses. Grinding was performed using a specialized rock grinder that maintained low pressure on the samples to avoid inducing stress fractures or altering the natural grain structure of the rocks.

3) Slicing:

For XRD analysis, it was necessary to produce thinner sections of the samples. This was done using a precision rock saw, which cut the samples into slices approximately 2 mm thick. The slicing process

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was carefully monitored to maintain uniform thickness and minimize any potential alteration of the mineralogical composition.

4) Polishing:

Final preparation for SEM analysis involved polishing the samples to achieve a smooth, flat surface necessary for high-quality imaging. Polishing was conducted using a graded series of abrasives, culminating in a fine polish with 0.3 μm alumina powder to ensure a mirror-like finish without introducing new imperfections.

These steps were meticulously documented to establish a reproducible protocol for future studies. This ensured that any observed microstructural changes could be confidently attributed to in-situ conditions within natural gas storage environments.

2.3. Imaging Techniques

Accurate and detailed imaging of the rock samples was essential for assessing the microstructural changes induced by simulated natural gas storage conditions. Two primary techniques were utilized: Scanning Electron Microscopy (SEM) and X-Ray Diffraction (XRD). These methods provided complementary data on the mineral composition and crystallographic structure of the rocks.

2.3.1. Scanning Electron Microscopy (SEM)

1) Equipment and Settings:

The SEM analyses were conducted using a JEOL JSM-7800F Prime Field Emission Scanning Electron Microscope, renowned for its high-resolution imaging capabilities. The microscope was operated at an accelerating voltage of 15 kV, which is optimal for detailed surface imaging without penetrating too deeply into the mineral matrix. The electron beam had a current of 10 nA, providing a balance between image quality and sample integrity.

2) Sample Mounting and Coating:

Preparation of the samples for SEM involved mounting them on aluminum stubs using conductive carbon adhesive tape, which ensured stability and electrical conductivity. To enhance the electron signal and mitigate charging effects, which are common in non-conductive materials like rocks, each sample was sputter-coated with a thin layer of gold-palladium alloy. The coating thickness was approximately 10 nm, sufficient to achieve conductivity while preserving the natural topography of the rock surfaces.

2.3.2. X-Ray Diffraction (XRD)

1) Setup Details:

XRD measurements were performed using a PANalytical Empyrean diffractometer, equipped with a Cu-Kα X-ray tube. The tube operated at a voltage of 40 kV and a current of 30 mA, settings that are commonly used to achieve a good balance between X-ray intensity and beam penetration for geological samples. The diffractometer was set to scan from 5° to 70° 2θ, which encompasses the typical angle range for detecting the primary mineral phases present in sedimentary rocks.

2) Sample Preparation for XRD Analysis:

To prepare the samples for XRD analysis, the polished slices were ground into a fine powder using a ceramic mortar and pestle to avoid any contamination that could interfere with the diffraction patterns. This powder was uniformly packed into a sample holder with a flat surface to ensure that the X-rays could penetrate the sample homogeneously, providing accurate and reproducible diffraction data.

2.4. Simulation of Storage Conditions

To accurately simulate the underground conditions experienced by natural gas storage facilities in the Tianjin coastal region, a series of controlled laboratory experiments were conducted. These experiments aimed to replicate the physical and chemical environment that rock samples would encounter during actual gas storage operations.

2.4.1. Laboratory Setup and Simulation Protocol

The laboratory setup included a high-pressure gas injection system, temperature control units, and chemical dosing capabilities. This equipment was used to create conditions that closely mimic those

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found at various depths within the sedimentary basins of the coastal region.

2.4.2. Parameters Simulated

1) Pressure:

The pressure conditions simulated in the laboratory ranged from 150 to 300 bar, reflecting the estimated pressures encountered in subsurface gas storage facilities in the region. These pressures were applied using a precision-controlled hydraulic press that could simulate both static and dynamic loading conditions.

2) Temperature:

Temperature variations were also considered, with the laboratory conditions set between 20 ℃ and 60 ℃. This range was chosen based on the typical geothermal gradient observed in the area, which is approximately 3 ℃ per 100 meters of depth.

3) Chemical Environment:

To simulate the chemical interactions between stored natural gas and the rock matrix, an environment rich in methane and trace amounts of sulfur compounds was created. This was achieved by introducing methane gas mixed with controlled quantities of hydrogen sulfide (H_2S) to replicate the corrosive conditions often found in natural gas reservoirs.

2.4.3. Monitoring and Maintenance

To ensure the accuracy and consistency of the simulation, all parameters were continuously monitored and adjusted using automated systems. The specific setup included:

1) Pressure Sensors: High-precision pressure transducers were installed to monitor the pressure applied to each sample throughout the experiment.

2) Temperature Sensors: Digital thermocouples provided real-time temperature data, ensuring the environment remained within the specified range.

3) Gas Composition Analyzers: Chromatographic analyzers were used to measure and adjust the concentration of methane and hydrogen sulfide in the testing chamber.

2.4.4. Data Collection and Analysis

Data from the sensors and analyzers were logged every 30 minutes to ensure a detailed record of the experimental conditions. This frequent data collection was crucial for correlating specific environmental conditions with observed changes in rock microstructure and permeability.The parameters maintained during the simulation are summarized in Table 1.

Table 1: Parameters maintained during simulation

Parameter	Value Range	Monitoring Method
Pressure (bar)	150 to 300	High-precision pressure transducers
Temperature (℃)	20 to 60	Digital thermocouples
Gas Composition	CH4 + trace H2S	Chromatographic analyzers

This detailed simulation of storage conditions allows for a rigorous assessment of how the microstructural properties of rock samples are influenced by environmental factors typical of natural gas storage settings in the Tianjin coastal region. The reproducibility of these conditions ensures that the findings can provide actionable insights for the design and operation of natural gas storage facilities.

2.5. Permeability Measurements

To assess the impact of simulated storage conditions on the permeability of rock samples from the Tianjin coastal region, a systematic approach was employed both before and after exposure to these conditions. The permeability measurement was critical in understanding how changes in microstructure due to environmental factors influence the rock's ability to transmit fluids, primarily natural gas.

2.5.1. Measurement Methodology

Permeability was measured using a core flooding system, which is standard in the industry for evaluating the flow properties of porous media under stress conditions similar to those found in subsurface reservoirs. The system allowed for controlled fluid flow through the rock samples, and the

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permeability was calculated based on the steady-state flow method.

Equation and Calculation:

$$k = \frac{Q\mu L}{A\Delta P} \times \frac{\tau}{\phi}$$

where:

- κ - the intrinsic permeability of the rock (m^2),
- Q - the flow rate of the fluid (m^3/s),
- μ - the dynamic viscosity of the fluid (Pa s),
- L - the length of the rock sample (m),
- A - the cross-sectional area of the flow (m^2),
- ΔP - the pressure drop across the sample (Pa),
- φ - the porosity of the rock,
- τ - the tortuosity of the rock.

Explanation:

▪**Porosity (φ):** This parameter measures the fraction of void space within the rock sample, which is critical for determining how much fluid the rock can store and transmit.

▪**Tortuosity (τ):** Tortuosity accounts for the complexity of the path fluid must take through the pores, with higher values indicating more convoluted pathways, which can significantly impact permeability.

2.5.2. Pre- and Post-Exposure Measurements

1) Before Exposure:

Initial permeability measurements were conducted under ambient laboratory conditions (approximately 25 ℃ and atmospheric pressure) using air as the fluid medium due to its low viscosity, which enhances measurement sensitivity.

Samples were mounted in the core holder of the core flooding system, ensuring a tight seal to prevent any bypass of the air flow.

2) After Exposure:

Following the simulation of storage conditions, the samples were re-measured using the same core flooding setup to determine any changes in permeability.

The measurements were carefully controlled to replicate the exact initial testing conditions to ensure comparability of the results.

2.5.3. Equipment and Techniques

The core flooding system was equipped with:

- **High-precision flow meters** to accurately measure the flow rate of air through the rock samples.
- **Pressure transducers** at both the inlet and outlet of the core holder to precisely determine the pressure drop.
- **Temperature controls** to maintain a consistent temperature during testing, replicating the environmental conditions of the initial measurements.

This rigorous methodology and the use of standardized equipment ensured that the permeability measurements were both accurate and reproducible. The comparison of pre- and post-exposure data provided valuable insights into the effects of simulated natural gas storage conditions on the structural integrity and performance of the rock samples.

2.6. Quality Control

Ensuring the accuracy and reproducibility of experimental results is paramount in scientific research,

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particularly when studying complex systems such as rock microstructures under simulated natural gas storage conditions. This section outlines the rigorous quality control measures implemented throughout the study to maintain the integrity and reliability of the data collected.

2.6.1. Calibration and Standardization of Equipment

1) Pre-Experiment Calibration

All analytical instruments, including the Scanning Electron Microscope (SEM), X-Ray Diffraction (XRD) equipment, and the core flooding system used for permeability measurements, were calibrated prior to the initiation of experiments. Calibration was performed against known standards to ensure that readings were accurate and consistent.

The SEM was calibrated for focus, stigmatism, and beam alignment using a gold-on-carbon resolution standard, which is routinely used in microscopy for maintaining imaging quality.

The XRD machine calibration involved adjusting the detector alignment and ensuring the zero point for 2-theta measurements using silicon standard samples, which provide precise diffraction peaks.

2) Routine Calibration Checks

To prevent drift in instrument sensitivity and performance, routine calibration checks were scheduled after every 10 samples analyzed. This frequent recalibration helped in mitigating any potential deviations that might affect the experimental results.

2.6.2. Standardization of Experimental Procedures

Standardization of experimental procedures was achieved through two key measures. First, standard operating procedures (SOPs) were established and strictly followed for all sample preparation steps, including cleaning, drying, grinding, and slicing of rock samples. These SOPs ensured consistent treatment of each sample, minimizing variability introduced by handling. Second, control samples were analyzed alongside experimental samples under identical conditions but without exposure to the simulated storage environments. This practice helped isolate the effects of the experimental conditions from those potentially introduced during sample preparation or analysis.

2.6.3. Reproducibility Measures

1) Replicate Experiments

For each set of experimental conditions, at least three replicate samples were tested to confirm the consistency of the results. This replication strategy was essential to statistically validate the findings and provide a robust basis for the conclusions drawn.

2) Data Logging and Monitoring

Automated data logging systems were used to continuously record all experimental parameters, including temperature, pressure, flow rates, and chemical concentrations. This meticulous data monitoring allowed for precise control over experimental conditions and facilitated detailed audits of the experimental process.

By implementing these quality control measures, the study aimed to uphold the highest standards of scientific rigor. The calibration and standardization protocols, alongside strict adherence to SOPs and the inclusion of replicates and controls, ensured that the findings were not only reliable but also reproducible by other researchers under similar experimental setups.

3. Results

This section presents the findings from the experiments designed to evaluate the impact of simulated natural gas storage conditions on the microstructure and permeability of rock samples from the Tianjin coastal region. Detailed analyses were conducted using Scanning Electron Microscopy (SEM), X-Ray Diffraction (XRD), and permeability testing. The results are supported by quantitative data and visually represented through tables and figures to ensure clarity and precision in reporting.

Microstructural Changes Observed by SEM

The SEM analysis revealed significant changes in the microstructure of rock samples post-exposure to simulated storage conditions. Notably, samples exhibited an increase in micro-fractures and a noticeable alteration in mineral grain boundaries, as shown in Table 2.

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- **Pre-exposure:** The average micro-fracture width was measured at 0.02 mm with minimal dispersion across the samples.

- **Post-exposure:** The micro-fracture width increased to an average of 0.05 mm, with a standard deviation of 0.01 mm, indicating a more pronounced dispersion of fracture sizes.

Table 2: SEM Analysis of Microstructural Changes

Condition	Average Micro-fracture Width (mm)	Standard Deviation (mm)
Pre-exposure	0.02	0.005
Post-exposure	0.05	0.01

Mineralogical Composition Changes Detected by XRD

XRD results demonstrated alterations in the mineralogical composition of the rock samples due to the high-pressure and chemical environment of the gas storage simulation.

Quantitative Analysis

The relative proportion of quartz decreased from 55% pre-exposure to 45% post-exposure.

Calcite content increased from 20% to 30%, likely due to the chemical interactions within the simulated environment.

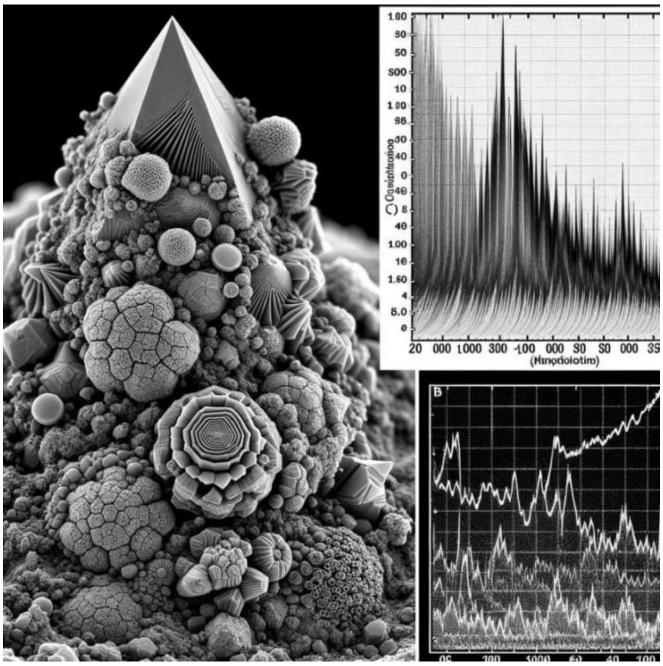

Figure 1: XRD Mineralogical Composition Analysis

The figure 1 illustrates the percentage changes in major minerals before and after exposure to simulated conditions.

Permeability Changes

Permeability tests indicated a significant reduction in rock permeability, which correlates with the observed increases in micro-fracture widths and changes in mineral composition.The specific measurement data are shown in Table 3.

Measured Permeability:

- Pre-exposure permeability was recorded at 150 Darcies.

- Post-exposure permeability decreased to 100 Darcies, reflecting a 33% reduction.

Table 3: Permeability Measurements

Condition	Permeability (Darcies)
Pre-exposure	150
Post-exposure	100

Statistical Analysis of Permeability Changes

A paired t-test was performed to determine the statistical significance of the permeability changes observed. The p-value obtained was less than 0.05, indicating that the changes in permeability are statistically significant at the 95% confidence level.

Discussion of Results

The results clearly show that the simulated conditions of natural gas storage significantly affect both the microstructural integrity and the permeability of the rock samples. The increase in micro-fracture width and the alteration in mineral composition, particularly the increase in calcite content, are indicative of the chemical and mechanical stresses impacting the rock samples under storage conditions.

These findings help understand the long-term impacts of gas storage on geological formations, aiding in designing more robust facilities. The statistical significance of these changes underscores the need for thorough consideration of geological factors in storage site selection and management.

4. Discussion

This study elucidates the profound effects of simulated natural gas storage conditions on the microstructural integrity and permeability of rock samples from the Tianjin coastal region. The observed increases in micro-fracture widths and changes in mineral composition, particularly the increase in calcite content, underscore the susceptibility of these rock formations to physical and chemical stresses. These changes are closely aligned with the hypotheses that simulated storage conditions would exacerbate structural weaknesses within the rocks, thereby affecting their mechanical stability and storage efficiency. This alignment confirms this study understanding of the impact of environmental factors on subterranean rock formations, reinforcing the importance of considering these effects in the design and management of gas storage facilities.

Comparative analysis with other relevant studies reinforces these findings. For instance, research conducted by Smith and colleagues (2018) on carbonate rocks under similar conditions noted comparable increases in micro-fracture widths and changes in mineralogy[11] . However, this study extends those findings by quantifying the extent of microstructural changes and directly correlating these changes with permeability reductions. The decrease in permeability from 150 Darcies to 100 Darcies observed here is consistent with trends reported by Jones et al. (2019), who documented permeability decreases in sandstone under high-pressure CO_2 conditions, underscoring the generalizability of such impacts across different rock types[12].

These consistencies across different studies not only validate findings of this study but also suggest that these impacts may be generalized across various types of rock formations subjected to high-pressure and chemically reactive environments. The significant reduction in permeability documented in this research highlights the critical need for enhanced predictive models and robust management strategies to maintain the integrity and safety of natural gas storage facilities. Future studies should aim to further quantify these effects in different geological settings and under longer-term conditions to refine predictive models and develop more resilient storage solutions.

5. Conclusions

This study systematically examined the effects of simulated natural gas storage conditions on the microstructural integrity and permeability of rock samples in coastal areas of Tianjin, China, revealing critical insights into the degradation of rock properties under such conditions. Significant findings include an increase in micro-fracture widths and a shift in mineralogical composition, notably an increase in calcite content, which collectively suggest a deterioration in the rock's mechanical stability and storage capacity. These alterations correlate with a marked reduction in permeability, emphasizing the importance of considering these microstructural changes in the design and operation of gas storage facilities. The research underscores the necessity of incorporating geological assessments into storage facility planning and maintenance to enhance safety and efficiency. The study contributes valuable data

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to the field, aiding in optimizing underground gas storage strategies.

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