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Experimental Study of Supercritical CO₂ Injected into Water Saturated Medium Rank Coal by X-ray MicroCT

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Abstract

Carbon dioxide geosequestration into deep unmineable coal seams is a technique which can mitigate anthropogenic greenhouse gas emissions. However, coal composition is always complex, and some minerals such as calcite chemically react when exposed to the acidic environment (which is created by scCO₂ mixing with formation water). These reactive transport processes are still poorly understood. We thus imaged a water-bearing heterogeneous coal (calcite rich) core before and after scCO₂ injection in situ at high resolutions (3.43 μm) in 3D via X-ray in-situ microCT flooding system. Indeed, the calcite-coal mixed layer was partially dissolved, and absolute porosity and connectivity significantly increased. We thus suggested that such process could be used as an acidizing method in CO₂ ECBM. However, such dissolved damage also can significantly affect the rock mechanical properties and potentially induce geohazards.

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1. Introduction

CO₂ injected into deep unmineable coal seams for enhanced coalbed methane recovery and combine with carbon storage have gained substantial interest in recent years [1-6]. Technically, CO₂ migrates into coal matrix micro/nano pores and displaces the original methane, at the same time, the CO₂ traps inside the matrix by adsorption trapping mechanism and closes the potential leakage route (cleats/factures) by swelling effect. Such coal matrix – CO₂ interaction induced swelling is complex and can significantly change the physical properties of the host rock, e.g. many studies proved that the CO₂ injection reduces the coal seam permeability [7-12]. Zhang et al., 2016 [12] injected scCO₂ into coal sample with permeability dramatically reducing recorded and micro fractures closed were observed by X-ray tomography. However, most of the experiments were conducted in the dry conditions and the previous models did not care about the existed formation water influence [13-16].

Note that the injected CO₂ dissolved into formation water called CO₂ – saturated brine is acidic at reservoir conditions where the pH values could be dropped to 3-4 [17, 18]. Moreover, the coal material is always heterogenous and consists of organic carbon and inorganic materials such as carbonate, quartz and clay [19, 20]. Especially for the coal from the low to medium rank group, such inorganic materials are abundant. However, such inorganic materials (mainly carbonates) may sensitive to the acid environment and the effect to the sample and the related petrophysical properties change need to be clarified. In this study, we thus used the newly in-situ microCT flooding system [21-23] has been used to investigate how the coal microstructure change by scCO₂ injected into water-bearing coal medium rank coal (calcite rich) sample in reservoir condition.

2. Methodology

2.1. Materials

The special coal sample which contained calcite was selected in this experimental study, obtained from the Pingdingshan coal mine, China. The coal had been identified as sub-bituminous containing 36 % (± 1%) volatile matter and 54 % (± 2%) fixed carbon content by Chinese standard GB/T 212 -2008 and DL/T 1030-2006. Fig. 1 showed the SEM image of the coal microstructure. A cylindrical coal plug (5 mm diameter and 10 mm length) was drilled for the following microCT in-situ flooding test.

Fig. 1. The SEM image of the coal sample; (A) the calcite particles inside the coal matrix, (B) the coal matrix, (C) the micro fractures.
2.2. MicroCT in-situ flooding test

The microCT in-situ scanning techniques developing in recent years give us the opportunity to observe the microstructures change by fluid-rock interaction in 3D at reservoir conditions, where the resolution could achieve as high as less than 10 μm voxel size (e.g. [24-30]). Such technique successfully overcomes the shortcomings of the traditional imaging tools such as SEM which only can obtain 2D surface images at laboratory conditions. Here the cylindrical coal plug was mounted inside an novel X-ray transparent core holder [12, 22, 23], which as a part of the microCT in-situ imaging core flooding system in Curtin University, Australia, see Fig. 2. This core flooding system included two parts: the microCT instrument itself (Xradia VersaXRM instrument, a 2000 × 2000 pixel detector was used and the X-ray accelerating voltage set as 60 kV in this experiment), and the high pressure – high temperature (HPHT) flooding system. The in-situ microCT core flooding test was then conducted by the below steps (also see [21, 23]):

1. The coal plug was saturated with 5 wt% NaCl brine before the coal sample amounted into the coal holder (coal plug merged into the brine water with vacuumed 1 week).
2. After coal plug mounted into core holder, the tubes and coal plug were injected 5 wt% NaCl brine by backpressure pump (Teledyne ISCO 500D, B in Fig. 2), then the coal plug and tubing system were vacuumed for 24 hours to air removed; all flow tubes and pumps were continuously isothermally heated to 50°C (323 K) with heat jackets by continuously circulating warm water. The core holder was heated by electric tape.
3. Initially the saturated coal plug was imaged (voxel size: 3.43 μm) under a confining pressure of 5 MPa (without fluid injection, thus 5 MPa effective stress). The confining pressure was applied by compressing deionized (DI) water by pump C in Fig. 2.
4. Then the coal plug was flooded by supercritical CO₂ (scCO₂) at 10 MPa pore pressure (backpressure pump, B in Fig. 2), and a 15 MPa confining pressure was applied (i.e. the experiment was conducted at a constant effective stress of 5 MPa). The injection flow rate was 0.25 ml/min with approximately 100 pore volumes were flooded.
5. The sample was then microCT imaged again at the same condition with 15 MPa confining pressure, 10 MPa pore pressure, and temperature of 50°C / 323K.

All the obtained grayscale tomograms were then filtered by 3D non-local means filter [31] method to image denoising, and the watershed algorithm [32] was used for phase segmentation according to their (different) relative radio-densities. Finally, the different phase could be extracted in 3D for the further qualitative and quantitative analysis [24]. All the images processing used the Avizo 9.2 software which provided by the Pawsey Supercomputing Centre, Australia.
3. Results and discussion

The microCT images showed a very highly heterogogenous morphology for such coal sample. In the water saturated tomography (see Fig. 3), it can be distinguished as three parts: the coal matrix (dark grey – low CT number), the calcite mineral (white – high CT number), and calcite-coal mixed layer (grey – medium CT number). Not that, this CT image did not show any micro fractures/cleats system which were presented in the former studies for dry coal sample [33, 34] and also the dry SEM image (Fig. 1), this may due to the micro fractures/cleats are closed due to water adsorption by coal matrix swelling effect [24].
After scCO$_2$ injected into the water-bearing coal sample, the chemical–dissolved effect was significant (see Fig. 4). The dissolved wormhole clearly presented in the 3D images (B in Fig. 4). Obviously, the CT scanning results by scCO$_2$ flooded into water-bearing coal were totally different with the previously dry coal – scCO$_2$ injection (see [12] where scCO$_2$ induced swelling effect on same coal sample at dry condition); the significant dissolution happed from our experiments. The calcite minerals chemical reacted during the carbon geosequestration by the following equations [35, 36]:

\[
\begin{align*}
H_2O + \text{scCO}_2 & \leftrightarrow H_2O + CO_2(aq) \\
H_2O + CO_2(aq) & \leftrightarrow H_2CO_3 \\
H_2CO_3 & \leftrightarrow HCO_3^- + H^+ \\
HCO_3^- & \leftrightarrow CO_3^{2-} + H^+ \\
CaCO_3 + H^+ & \leftrightarrow Ca^{2+} + HCO_3^- \\
CaCO_3 + H_2CO_3 & \leftrightarrow Ca^{2+} + 2HCO_3^- \\
\end{align*}
\]

The dissolved area showed the heterogenous characteristics where most located in the calcite-coal mixed layer. This could be explained by the less consolidated of this phase in nature. The 3D segmented dissolved hole presented in Fig. 5, which represented 2.78% volume fraction.

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Fig. 4. The microCT image (3.43 μm voxel size) for the coal sample after scCO$_2$ flooding where the dissolved area is black (lowest CT number), coal matrix is dark grey (low CT number), the calcite mineral is white (high CT number), and calcite-coal mixed layer is grey (medium CT number); (A) is the 2D slice, (B) is the 3D cut view.
In summary, such dissolved area largely increasing the porosity and improved the connectivity of the coal sample. We thus suggested such process could be an environmental friendly acidizing method in the CO$_2$ ECBM. At the same time, such damaged can significant affect the rock mechanical properties of the coalbed – may cause geohazards, e.g. the layer collapse and fault reaction – which has been seriously considered at the carbon geosequestration in carbonate reservoir.

4. Conclusion

Carbon geosequestration in deep geological formations has been suggested as the most efficient ways to mitigate climate change with reducing the CO$_2$ concentration from the atmosphere [37-40]. The deep unmineable coal seams are some of the main targets; however, the injected CO$_2$ with formation water induce an acidic environment – may impact some acidic sensitive minerals (such as calcite) in coal seam, which are still poorly investigated.

We thus investigated such interactions in pore-scale by 3D microCT in-situ core flooding experiments, the scCO$_2$ was injected into a heterogeneous water-bearing bituminous coal at reservoir conditions (15 MPa confining pressure, 10 MPa pore pressure, and 323 K). The coal’s microstructure (calcite-coal mixed layer) partially dissolved after the flooding test. We thus concluded that such dissolved area largely increasing the porosity and improved the connectivity of the coal seam. We also suggested such process could be an environmental friendly acidizing method in the CO$_2$ ECBM in some calcite rich coal seams. Moreover, such dissolved damage also can significant affect the rock mechanical properties of the coalbed which can induce potential geohazards, e.g. the layer collapse and fault reaction.

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