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Multi-scale quantitative characterization of 3-D pore-fracture networks in bituminous and anthracite coals using FIB-SEM tomography and X-ray μ-CT

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ABSTRACT

To study the three-dimensional (3-D) pore-fracture networks of bituminous (BC) and anthracite (AC) coals, a combination of focused-ion beam-scanning electron microscopy (FIB-SEM) tomography and X-ray computed micro-tomography (X-ray μ-CT) was used to characterize 3-D pore-fracture characteristics at different scales. First, as observed in the SEM images, organic-matter (OM) pores, shrinkage-induced pores and dissolution pores are developed in BC samples with pore sizes ranging from ~25 nm to 600 nm, and are independently distributed within OM and partial pores filled with minerals. In contrast, a large number of gas pores are widely developed in the AC sample with a cluster distribution in OM, and the pore widths range from ~10 nm to 2 μm. Second, we reconstructed the 3-D pore-fracture networks of the BC and AC samples, and quantitatively characterized the porosity, 3-D pore-throat characteristics and its connectivity using the Pore Network Model (PNM) in Avizo. The results indicate that the pores in the BC sample are poorly connected and isolated from each other, whereas the pores in the AC sample are well connected by the throats. There is a logarithmic correlation between the cumulative throat volume and throat size, indicating that the smaller throats make the main contribution to the throat volume. Moreover, the normalized pore size distribution obtained from FIB-SEM and X-ray μ-CT analysis shows that both BC and AC samples exhibit a three-peak structure, but the nano-scale pores are more developed in the AC sample and its 3-D pore morphologies are more complex than those of the BC sample. Furthermore, the macro-pores and micro-fractures in the two coal samples show complex spatial distribution, interconnectivity and tortuosity. The total resolved porosity is 1.108% for BC samples consisting of 0.279% of FIB-SEM and 0.829% of X-ray μ-CT, and 6.082% for AC sample composed of 4.194% of FIB-SEM and 1.888% of X-ray μ-CT, including the connected and non-connected porosity. These results show that the pore-fracture networks of the AC sample are more conducive to CBM storage and flow ability than those of the BC sample. Therefore, this finding provides an accurate method of evaluating the physical properties of coal reservoirs and assists in understanding the mechanisms of CBM storage and transport.

1. Introduction

Coalbed methane (CBM) has attracted continuous attention in recent years because of its benefits to mining safety and greenhouse gas reduction and significant commercial value [1–5]. As a complex carbon-rich porous media, coal is generally characterized by a dual-porosity system, including primary porosity consisting of a highly heterogeneous micro-/mesoporous system in the coal matrix and a secondary porosity system composed of macro-pores and natural fractures [6–8]. According to Cai et al. [3] and Hodot [9], the pore networks can be classified into four categories: micro-pores (< 10 nm in diameter), transition pores (10–10^2 nm in diameter), meso-pores (10^2–10^3 nm in diameter), macro-pores (10^3–10^4 nm in diameter) and micro-fractures (> 10^4 nm in diameter), respectively. The adsorption pores (< 100 nm) have extremely large specific surface areas which can provide much more space for CBM storage, whereas the seepage pores (> 100 nm) can provide the important permeable pathways for CBM and formation water during CBM recovery [10–12]. Hence, multi-scale quantitative characterization of pore-fracture networks is important for understanding the CBM storage and flow mechanisms and enhancing CBM recovery.

Recently, various methodologies have been applied to study the pore-fracture networks at different scales in coals, including low-pressure gas adsorption [13–15], mercury injection porosimetry [16–18], scanning electron microscopy [19], field emission scanning electron microscope [20], nuclear magnetic resonance [21–23], small-angle X-

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ray scattering and small-angle neutron scattering [24,25]. Due to the limitations of experimental conditions and the measurement accuracy of different methods, two or more techniques have usually been combined to characterize the pore types, pore size distribution and total porosity of coals. However, these methods are mainly focused on two-dimensional (2-D) pore geometry characteristics and are therefore inadequate for the study of the three-dimensional (3-D) spatial distribution of pore-fracture networks and connectivity. To address this problem, X-ray computed micro-tomography (X-ray μ-CT) and focused ion beam scanning electron microscopy (FIB-SEM) have gradually shown the advantages and application prospects of evaluating the 3-D pore-fracture morphology and its connectivity in porous media. For X-ray μ-CT, studies by Karacan and Okandan [26], Mazumder et al. [27] and Jing et al. [28] reported that it is an effective non-destructive method for analyzing the fracture/cleat density, cleat surface morphology and cleat spacing distribution. Other researchers [29–32] have used X-ray μ-CT to investigate the spatial disposition of minerals, pores and fractures, and the evolution of 3-D fracture networks during stressing. Moreover, for the quantitative characterization of nano-pores, the FIB-SEM approach has become an important and effective method to show the 3-D characteristics of adsorption pores with resolutions of a few nanometers in recent years. With regard to coal reservoirs, a study by Liu et al. [33] adopted FIB-SEM to accurately describe the interconnected pores and pore connectivity in high-rank coals. Zhou et al. [34] conducted a FIB-SEM experiment on sub-bituminous and high-volatile bituminous coals to characterize the pore-fracture spaces and their nano-scale connectivity in three dimensions. However, there have been extremely few studies which quantitatively investigate the 3-D characteristics of pore-fracture networks in coals by combining FIB-SEM and X-ray μ-CT.

In the present study, FIB-SEM tomography and X-ray μ-CT scanning were combined to accurately characterize the multi-scale characteristics of 3-D pore-fracture networks in BC and AC samples. First, the morphology of pore-fractures was qualitatively identified in 2-D SEM images. Based on FIB-SEM and X-ray μ-CT slices, 3-D reconstruction volumes and pore-fracture networks were revealed using Avizo software. Using the Avizo Pore Network Model (PNM), we qualitatively demonstrated the 3-D spatial distribution of pore fractures and quantitatively characterized the porosity, interconnectivity and pore size distribution of coal samples. The effects of 3-D pore-fracture networks on CBM storage and transportation were further discussed, which is of great importance for understanding its mechanisms.

2. Material and methods

2.1. Samples and coal analyses

The two coal samples used in this study were collected from the Gujiao and Yangquan mine areas of the Qinshui basin, China. Following China National Standards GB/T 6948-2008 and GB/T 8899-2013, the maximum vitrinite reflectance ($R_{o,max}$) and maceral composition analyses were performed on polished slabs using a Leitz MPV-3 photometer microscope. The results indicated that the sample from Gujiao was a bituminous coal with $R_{o,max}$ of 1.71% and the sample from Yangquan was an anthracite with $R_{o,max}$ of 2.61%. An Automatic Proximate Analyzer 5E-6600 was used to analyze the moisture, volatile matter, ash yield, and fixed carbon contents. Table 1 shows the $R_{o,max}$, maceral composition, and the proximate analysis of coal samples.

2.2. Porosity and permeability analyses

Helium porosity and air permeability analyses were performed on the core plugs with a diameter of 2.5 cm and a length of 5–6 cm using a high-pressure gas permeameter/porosimeter OPP-1 (TEMCO, USA), following the Chinese Oil and Gas Industry Standard (SY/T) 5336-1996. The porosity was measured using the helium expansion method and calculated by dividing the pore volume by the total volume. The air permeability was analyzed using a bubble flowmeter with air flowing through the core plug and was determined using Darcy's equation [35]. The porosity and permeability of the coal samples are listed in Table 1.

2.3. FIB-SEM imaging and X-ray μ-CT scanning

Prior to the FIB-SEM imaging experiments, the cuboidal-shaped coal samples about 0.5 × 1 × 1 cm^3 were polished using dry emery paper to create a flat surface and then polished by argon-ion. Subsequently, the polished coal samples were dried in an oven at 65 °C for 12 h and then coated with carbon to prevent electrostatic charging. Each sample was inserted into a FEI Helios NanoLab 650 FIB-SEM dual-beam system for imaging. A series of SEM images about the freshly ground coal surface were obtained at a high resolution of 2.5 nm with an acceleration voltage of 2 kV and a working distance of 4 mm. The experimental procedure is described in detail in our previous research [34] and Hemes et al. [36].

For the X-ray μ-CT scanning measurements, two coal pillars with a diameter of ~2.0 mm and a height of ~5.0 mm were drilled from the block samples. In order to avoid the influence of water dissipation on the experimental results, the coal pillar was sealed in wax [37]. The X-ray μ-CT scanning experiments were then performed using a Nano Voxel-2000 X-ray 3-D microscope (Sanying Precision Instruments Co., Ltd., China) with a 20× lens detector. The detector resolution was set to 1024 × 1024 pixels and 900 projections were obtained during a rotation of 360°, corresponding to a rotation step of 0.4°. The sample was scanned with the X-ray of 80 kV source voltage and a current of 95 μA, which resulted in a maximum resolution of 0.5 μm. In total, more than 900 2-D images with an interval spacing of 1 μm were obtained.

2.4. 3-D tomography data analysis

In this study, the 3-D FIB-SEM and X-ray μ-CT data were analyzed using the Avizo 9.0.1 software. The 2-D slices are stacked and then an image enhancement filter is applied on the image-stack, which includes noise reduction, contrast enhancement and sharpening of pore edges. Subsequently, the stack of slices is aligned to reconstruct a continuous 3-D volume, which can be read with optional *z*- values for each slice. The sub-volumes are extracted and smoothed with a non-local means filter. For image segmentation, the local thresholding method adopted in this work mainly utilizes the spatial covariance of the image in conjunction with indicator kriging to determine object edges, which makes the thresholding local and guarantees smoothness in the threshold surface [38]. Moreover, the different pore structure parameters of a separate pore space can be accessed by the *Pore Network Model* (PNM) in Avizo, including pore volume, pore area, pore equivalent diameter, pore size distribution, and resolved porosity. The PNM used in this study is based on the maximal ball algorithm [39–41]. The pore size distribution of coal samples can then be plotted using the *Distribution Analysis* module. To check for the hypothesized power-law behavior of pore-size distributions, power-law exponents (D) can be derived from the slopes of the linear fits to the log-log pore-area distributions [42]:

$$\log(N_i/(b_i S_{area})) = -D\log(S_i) + \log(C) \quad (1)$$

where, N_i is the pore-size frequencies per bin, b_i is the bin-sizes, S_{area} is the size of the area analyzed, S_i is the centers of the respective bins, and C is a constant of proportionality.

3. Results and discussion

3.1. Morphology of pore-fractures from FIB-SEM images

Due to the difference in coal-forming environments and