



## Effect of moisture on the desorption and unsteady-state diffusion properties of gas in low-rank coal

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### ABSTRACT

Currently, resource integration in coal-burning facilities and clean energy reconstruction are being strongly pushed forward in China. As a clean fossil energy, unconventional natural gas, such as coalbed methane (CBM), exhibits promise as an energy development strategy. In China, there are abundant CBM resources in the considerable reserves of low-rank coal. Therefore, the study of low-rank coal is the key to realize the high-yield and high-efficiency development of CBM resources in China. In this paper, a new water injection device was designed to prepare coal samples with a variety of moisture contents, and a novel method to measure the gas desorption properties in coal containing water was introduced. On this basis, the gas desorption law in low-rank coal with different moisture contents was tested, and the effect of moisture on the gas diffusion properties in low-rank coal was analyzed. The results indicate that the initial gas desorption volume of low-rank coal is extremely large and that the gas desorption law in the moisture-containing low-rank coal can be described by Airey's empirical equation. It is also observed that the ultimate gas desorption volume, initial gas desorption rate and initial effective gas diffusion coefficient all shift toward the direction that is beneficial to CBM development. Therefore, reducing the moisture content in low-rank coals through certain techniques can dramatically enhance the gas migration behavior and achieve the purpose of high-yield CBM development.

### 1. Introduction

In China, coal was always the main energy resource for the urban modernization in the past few decades. However, this has led to severe environmental pollution. Thus, all over the country, especially in large and medium-sized cities, resource integration in coal-burning facilities and clean energy reconstruction have been strongly pushed forward. As a clean fossil energy, unconventional natural gas, such as coalbed methane (CBM), will certainly become a bright spot for energy development strategies. The total CBM volume is approximately  $3.68 \times 10^{13} \text{ m}^3$  in China, of which the CBM volume in low-rank coal seam is around  $1.6 \times 10^{13} \text{ m}^3$ , which accounts for 43% of the total (Wang et al., 2016). For a long time, most of the research (He et al., 2010; Xu et al., 2014; Zhao et al., 2013, 2016) was concentrated on medium-rank and high-rank coals in China. In the 21st century, CBM in the low-rank coal seam from the Powder River basin of USA, Surat basin

of Australia and Alberta basin of Canada has achieved commercial development (Beaton et al., 2006; Busch and Gensterblum, 2011; Stricker et al., 1998). By comparison, the study of CBM in the low-rank coal, especially lignite, still stays in the stage of scientific research and experimental development.

In general, coal with a vitrinite reflectance  $R_0 < 0.65\%$  is called low-rank coal in China (Guo et al., 2015; Wang et al., 2016). The porosity of low-rank coal is high, its specific surface area is large and it has many polar functional groups, such as hydroxy and carboxyl; therefore, it can absorb more water than coals of other ranks (Crosdale et al., 2008; Fu et al., 2005, 2012; Wang et al., 2011). The statistical data show that in China, the moisture content of long flame coal is mostly in the range of 3%–12% and the moisture content of lignite is typically 10%–28% (Guo et al., 2015; Qiao, 2009). There are four existing forms of moisture in the low-rank coal (Allardice et al., 2003; Crosdale et al., 2008): 1) In the macropores and fractures in the form of

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free water; 2) in the capillary channel of low-rank coal; 3) adsorbed in the coal pores in the form of multimolecular layer; 4) adsorbed in the coal pores in the form of monomolecular layer.

Moisture in the low-rank coal matrix has significant impact on the gas ad-desorption properties and plays an important role in gas migration in coal (Gensterblum et al., 2013; Pan et al., 2010). In the past, studies on water-containing coal by most scholars (Day et al., 2008; Gensterblum et al., 2013; Krooss et al., 2002; Liu et al., 2012) have mainly focused on equilibrium water coals, and the influence of the variation of moisture content in coal on the CBM development is rarely taken into account. In addition, some scholars (Crosdale et al., 2008; Joubert et al., 1974; Moore and Crosdale, 2006; Xie and Chen, 2007; Zhang et al., 2009) have also studied the gas adsorption properties in coal with different moisture contents; however, due to limits of the experimental device, it was difficult to uniformly humidify the experimental coal sample and ensure accurate moisture content when coal samples with different moisture contents were prepared via artificial addition of humidity. In this paper, a water injection device that can uniformly humidify the experimental coal samples was developed to prepare coal samples with different moisture contents, and the new device and method to measure the gas desorption properties in coal containing water were introduced. On this basis, the effect of moisture on the gas migration mechanism in low-rank coal is studied to provide a theoretical basis for the CBM development in low-rank coal reservoirs.

## 2. Coal samples and experimental methods

### 2.1. Coal samples

The coal samples were collected from the northern China mining area. The samples were sealed in a package from the site and prepared for the proximate analysis, petrographic analysis and porosity tests. The basic physical parameters of the low-rank coal samples are shown in Table 1.

### 2.2. Experimental methods

In general, coal sampled from the mining area is in a pulverized or broken state; that is, most coal samples are in particulate form. Therefore, the gas desorption and diffusion properties in coal samples are usually measured via desorption or sorption methods (Guo et al., 2016b; Zhang, 2008). In this paper, the desorption method was used. To prepare coal samples with different moisture contents, a water injection device that can uniformly humidify experimental coal samples was developed. Using the device, we can inject water into the coal sample after vacuum degassing or achieving gas adsorption equilibrium and then stir the experimental coal sample to humidify it uniformly. Meanwhile, in this paper, the new experimental device and method to measure the gas desorption properties were introduced to accurately obtain experimental results. The schematic diagram of the experimental device is shown in Fig. 1.

The experimental steps and methods for the analysis are as follows:

- 1) The coal sample was crushed in a pulverizer and screened to obtain the desired sample size of 1–3 mm. Then, the specimens were placed in a vacuum drying oven at 50 °C to completely dry for 72 h to

remove the moisture in coal. The density ( $\rho$ ) of the coal sample was measured using an Ultracyc 1200e Automatic Gas Pycnometer for Density from Quantachrome; the instrument has a resolution and accuracy of 0.0001 g/mL and 0.02%, respectively. Then, the coal samples (weighing  $m_{\text{coal}}$ ) were placed in the coal sample tank. The volume of gas molecules adsorbed onto the pore surfaces of coal matrices is extremely small, so it can be ignored. Thus, the volume of the coal sample in the coal sample tank can be calculated using Eq. (1):

$$V_{\text{coal}} = \frac{m_{\text{coal}}}{\rho} \quad (1)$$

- 2) Combining with the volume ( $V_{01}$ ) of the coal sample tank, the volume ( $V_f$ ) of the free gas in the coal sample tank at adsorption equilibrium can be obtained using the following equation:

$$V_f = V_{01} - V_{\text{coal}} \quad (2)$$

Thus, the free gas volume ( $V_{f0}$ ) under the standard conditions can be obtained using the following equation (Guo et al., 2016b):

$$\frac{P_{\text{eq}} V_f}{T_{\text{eq}}} = \frac{P_0 V_{f0}}{T_0} \quad (3)$$

where  $P_{\text{eq}}$  is the adsorption equilibrium pressure in the coal sample tank, MPa;  $T_{\text{eq}}$  is the temperature of the coal samples within the coal sample tank, K;  $P_0$  and  $T_0$  are the standard pressure (0.101325 MPa) and temperature (273.15 K), respectively.

- 3) The temperature of the thermostatic oil bath was adjusted to  $(60 \pm 0.1)$  °C, and the coal sample tank and coal samples were outgassed for approximately 48 h under high vacuum to remove air and other impurities in them. Then, the valve of the coal sample tank and the switch of the vacuum pump were closed successively.
- 4) The temperature of the thermostatic oil bath was changed to  $(30 \pm 0.1)$  °C, and then the constant-flux pump and the stirring apparatus were opened to inject the distilled water into the coal samples in the coal sample tank. The volume of injected water was calculated in advance according to the requirement for the experiment. After the water injection was completed, the switch of the constant-flux pump and the corresponding valve of the coal sample tank were closed. It should be noted that to uniformly humidify the experimental coal sample, the stirring apparatus should keep continuous stirring for 45–50 min before it is closed (Guo et al., 2016b). The coal sample in the coal sample tank was equilibrated for 48 h after the stirring apparatus being closed to allow the injected water to fully humidify the coal sample.
- 5) The coal sample tank was filled with methane (> 99.9% purity) and the gas intake valve was adjusted to let the gas pressure stabilize at 2 MPa when the gas adsorption equilibrium state was achieved. It was considered that adsorption equilibrium had been reached after 6 h of unchanged pressure in the coal sample tank.
- 6) To test the gas desorption law in coal, the first step was pressure relief when the gas desorption law in coal was tested. As shown in Fig. 1, the valve (m) was opened to link the gas sample bag to the coal sample tank, making the pressure reduce to the atmospheric pressure. Then, the valve (m) was turned to connect the coal sample tank with the gas desorption measuring cylinder as soon as possible.

**Table 1**

Basic physical parameters of the low-rank coal samples.

Proximate analysis (wt%)			Petrographic composition (vol %)				Maximum vitrinite reflectance ( $R_o$ )/%	Porosity/%
$A_{\text{ad}}$	$V_{\text{daf}}$	$F_{\text{cad}}$	Vitrinite content	Inertinite content	Liptinite content	Mineral content		
9.28	49.53	32.47	95.17	0.59	0	4.24	0.41	17.82

$A_{\text{ad}}$  = ash content on air-dried basis;  $V_{\text{daf}}$  = volatile matter content on dry ash-free basis;  $F_{\text{cad}}$  = fixed carbon content on air-dried basis.

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