



A new laboratory method for accurate measurement of the methane diffusion coefficient and its influencing factors in the coal matrix



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HIGHLIGHTS

- A new laboratory method was proposed to measure the CBM diffusion coefficient.
- The tested diffusion coefficient was the order of magnitude of 10^{-11} – 10^{-9} m²/s.
- The influences on the methane diffusion coefficient were analyzed.

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ABSTRACT

The diffusion coefficient is a key parameter for evaluating the methane diffusion properties of a coal matrix. However, all of the current measurement technologies for methane diffusion coefficients, including the particle method, the steady state method and the inverse diffusion method, have certain limitations. For example, the pore structure of the sample is easily destroyed, and the test time is long. Moreover, the results from different methods lack comparability under the various test principles and conditions. In this work, a new laboratory method for measuring the methane diffusion coefficient in coal matrixes was proposed, where coal matrix flakes instead of coal particles were used as the samples to be measured, and the factors influencing methane diffusion coefficients such as gas pressure, coal rank and moisture content were also analyzed. The results indicate that this new method can keep the intrinsic space structure of the coal matrix and yields methane diffusion coefficients on the order of 10^{-11} – 10^{-9} m²/s in coal matrix, which are close to realistic reservoir values. Under the same temperature, methane diffusion coefficients increase with increasing gas pressure regardless of the water saturation and coal rank. Additionally, the adsorption capacity increases when the metamorphic degree of coal increases, while the methane diffusion coefficient in coal matrixes exhibit a trend of first dropping and then rising (“U” shape) with an increase in coal rank. Moreover, moisture has an important effect on the methane diffusion coefficient. Increasing moisture can reduce the matrix adsorption capacity for methane, which makes it difficult to form a larger concentration gradient because the diffusion coefficients of dry samples are higher than those of the saturated water samples.

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1. Introduction

It is of great significance for the methane gas diffusion coefficient estimation since it controls the late production rate of the coal and shale reservoirs [1–5]. Diffusion refers to the phenomenon where molecules migrate from high concentration areas to low concentration areas until they are evenly distributed [6–8]. For coal reservoirs, methane cannot simultaneously be in contacted

with and adsorbed onto all of the pores and fissure surfaces because of the nonpolar characteristics of the methane molecules, which results in concentration and pressure gradients in the coal body. Therefore, methane adsorption in the coal reservoir is an integrated process of seepage, diffusion and adsorption [2,9]. The production rate of coalbed methane (CBM) is mainly controlled by diffusion in the matrix and the permeability within the cleat system [10]. Thus, the understanding of methane diffusion mechanism in coal matrix and the accurate determination the methane diffusion coefficients are of great significance for CBM production planning and economical reserve estimation [11].

The diffusion coefficient is the one of the key parameters to control the gas transport dynamics in of coal matrix [12]. The methane

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diffusion coefficient determination methods include the particle method, the steady state method and the inverse diffusion method [10,11,13–15]. Many scholars have extensively reviewed the influencing parameters on the diffusion coefficient. Zhao [16], through measuring diffusion coefficients in coal micropores, discovered that reservoir pressure, methane concentration and water saturation substantially affect methane diffusion coefficients. Shi and Durucan [1] thought that the diffusion coefficients depend on the gas concentration and the adsorption state of the inner coal surface. Through studying the diffusion rates of coal particle samples of different ranks, Clarkson and Bustin [17] speculated that the absorption capacity differences of coal samples are mainly due to the different proportions of macropores, mesopores and micropores, while the pore size distribution is related to the coal rank [18]. Furthermore, Busch et al. [19] demonstrated that diffusion coefficient decreases with increasing of particle size. Wang et al. [20] analyzed the diffusion dynamics in the process of CBM desorption under the different coal ranks and moisture contents. Xu et al. [21] revealed the influence of CBM bearing boundary conditions on methane diffusion among coal particles. Additionally, the diffusion coefficients of different gas composition are different under the same conditions, and the sensitivities to various influencing factors are also different. Saghafi et al. [22] determined the carbon dioxide and methane diffusion coefficients of the coals from the Sydney Basin by concentration difference experiments, and demonstrated that the carbon dioxide diffusion coefficient is about twice as large as that of methane. Pan et al. [10] studied the relationship between the diffusion of carbon dioxide and diffusion of methane under different moisture contents, which further indicated that the methane diffusion coefficient is more easily affected by moisture than that of carbon dioxide and that the influences of pore size on the diffusion coefficient are not identical. With the coals from Lorraine Basin in France, Charriere et al. [23] compared the carbon dioxide and methane diffusion coefficients, which indicated that temperature and pressure are the two key factors controlling the diffusion coefficient.

However, these results lack comparability with each other under the different test principles and conditions [24], and these methods have the following disadvantages: (1) Although the particle method is the most widely adopted test technology in the CBM industry, the coal briquettes are smashed into tiny particles which results in the particles being easily accumulated and leading to the formation of intergranular pores. Meanwhile, the specific space structures of the coal matrix are destroyed. (2) With the steady state method, a cylindrical coal sample is chosen to measure the diffusion coefficient under a very small differential pressure. However, the requirements of the sample are demanding (i.e., the sample must be without fractures) and the test time is too long. Moreover, the results are sensitive to the environment temperature, which limits the wide application of the method. Additionally, it is difficult to ensure that only diffusion without seepage exists during the test process. (3) The inverse diffusion method can ensure that the experimental process is not influenced by pressure differences. Nevertheless, the diffusion coefficient of the high-pressure environment cannot be measured. Hence, it is necessary to explore a suitable measurement and analysis technology for methane diffusion estimation.

In this work, we took into account that the space structure of the coal matrix cannot be damaged, the instrument can measure the methane diffusion instead of the adsorption and desorption, also can simulate the actual underground temperature and pressure condition, and the experiment can be conducted under safe conditions and no other gases interfere. A new laboratory method, which can overcome the deficiencies of current methods was proposed to measure the methane diffusion coefficient in coal matrixes. Furtherly, because gas pressure, coal rank and moisture

content are essential factors for the adsorption, desorption and diffusion properties, their influences on methane diffusion coefficient in the coal matrix were analyzed, which is great importance for the CBM production.

2. Experimental work

2.1. Sample preparation and experimental setup

The bulk coal samples with different metamorphic degree were obtained from three active mine areas (Yimin, Hancheng and Jincheng) located in the Hailar, Ordos and Qinshui Basins of China, respectively. A total of six active coal mines (Liaoyuan, Shengjie, Nangou, Xiangshan, Wumuchang and Tang'an) were investigated in this research (Fig. 1).

Prior to the start of the matrix diffusion coefficient test, the flake coal samples were polished and prepared from the bulk coals. The size (length \times width \times thickness) is approximately 20 mm \times 15 mm \times 3 mm (Fig. 2) and the largest fracture width must be less than 3 μ m. Then, the flake coal samples were cleaned with an ultrasonic cleaner and the volumes were measured by the water drainage method. Moreover, the thicknesses of the samples were also measured with a vernier caliper. Then, the samples were dried completely in a constant temperature chamber at 110 °C. The dried samples were used to measure the coal matrix diffusion coefficient. To compare the diffusion coefficients of the dry samples and the water saturated samples, all the dry samples were put into distilled water for saturation before they were tested. In this work, the moisture treatment for each sample was processed for 48 h. This situation is much closer to actual underground condition. Then, the diffusion coefficients under the wetted condition were obtained. The test gas used in this work was methane.

Additionally, broken coal samples from the three mine areas were collected, and some were ground to 60–80 mesh for adsorption isotherm experiments and the others were measured for vitrinite reflectance, proximate analysis and coal composition.

Fig. 3 is a schematic illustration of the experimental setup for measuring methane diffusion in the coal matrix, including the coal matrix, the scaffold, the seal diffusion chamber, the thermostat, the constant pressure device, the gas source (methane), the counting device and the evacuating device. The functions of these devices are described as follows:

- (1) The constant pressure device is composed of a metering pump, which is connected to the seal diffusion chamber and the gas source through the sealing catheter. This device is used for detecting the gas pressure in the diffusion chamber at each time and judging whether the gas pressure is the tested pressure (0.1–10 MPa). If the gas pressure is lower than the tested pressure, the extra gas will be injected into the seal diffusion chamber with the sealing catheter until the gas pressure is the same as the tested pressure.
- (2) The thermostatic device is made up of a water pool and is connected to the seal diffusion chamber and is used to keep the seal diffusion chamber at the rated temperature (20–60 °C).
- (3) The scaffold is used for supporting the coal matrix to ensure that the coal sample is hung in the seal diffusion chamber.
- (4) The counting device is composed of a data acquisition card and a computer. The data acquisition card, which is used to record the injection volume of gas into the seal diffusion chamber at each time interval and send it to the computer, is connected to the constant pressure device. According to the tested pressure, rated temperature and the quantity of the injected gas which is recorded by the data acquisition

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