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Time-varying diffusion characteristics of different gases in coal particles

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ABSTRACT

Based on the analytical solution of gas diffusion in spherical coal particles with a constant diffusion coefficient, a calculation method of time-varying diffusion coefficient is proposed by constructing objective function. The time-varying diffusion behavior of methane, nitrogen and carbon dioxide in the coal particles was studied. The results show that with the increase of diffusion time, the diffusion coefficients of methane, nitrogen and carbon dioxide gas in the coal particles exhibit an attenuation characteristic, eventually approaching a limit value individually. The diffusion coefficient of carbon dioxide is larger than methane, and the diffusion coefficient of nitrogen is smallest. Significant phenomenon of limited diffusion was observed for coal of strong adsorption capability. Through the analysis of the diffusion coefficient of gases at different diffusion time, a mathematical model describing the time-varying diffusion characteristic of gases is obtained. The implementation of mixed gases to replace coal bed methane has a very important practical significance.

1. Introduction

Coal mine gas (also known as coalbed methane (CBM)) is one of main concerns threatening the safety of underground mining [1]. It is a kind of hydrocarbon gas and its main component is methane, which mainly adsorbs on the surface of coal matrix and partially exists in the pores of coal as free gas or dissolved in the seam water. Meanwhile, CBM is an unconventional natural gas. Under the pressure of environmental protection, it is desired to promote the development of CBM recovery and improve China's energy consumption structure. However, CBM resource is different from conventional gas resources. Coal bed methane resources are usually bounded in deep, high stress environment with the characteristics of low permeability, which makes it difficult to recover and thus CBM cannot be effectively exploited by using conventional oil and gas technologies. The enhancement of CBM recovery rate has been a hot topic in China to ensure its economic benefits. In order to solve this dilemma, many studies have been conducted to advance the CBM recovery with the advent of new technology. For example, technology of hydraulic fracturing [2–4], heat injection [5], coal bed acidification coal [6], nitrogen foam [7], and coalbed methane replacement technology by carbon dioxide [8–10] have been introduced to enhance the permeability of coal seam strata and CBM recovery rate. Coalbed methane replacement technology by carbon dioxide can not only achieve the purpose of increasing the production of coal-bed methane. It can also be beneficial to environmental improvements by sequestrating CO_2 in coal, which has been gradually adopted in developed countries [11,12]. Due to the stronger CO_2 adsorption capacity on coal compared to methane, the coal permeability decreases after injecting pure CO_2 gas due to coal matrix swelling. On the other hand, pure CO_2 injection into coal makes it prone to be instable. This may lead to coal or rock dynamic disasters [13]. Many researchers have proposed the technology of using mixed gases to enhance coalbed methane recovery, which refers to inject mixed gases rich with N_2 into coal seam [14,15]. For one hand, the escape of N_2 gas can enhance the coal seam permeability. For the other hand, CO_2 gas can achieve the goal of methane replacement and hence improve the production of coalbed methane. After injecting driving gases into the coal seam, micro-porosity system evolves into the fracture system [16,17]. In this process, the change of pore structure can lead to the change in gas diffusion behavior [18]. Currently, many studies investigated the characteristics of multicomponent gas under different adsorption balance pressure in the coal particles [8–17], or diffusion characteristics of gas in the coal particles [18,19], without considering the change of different gas diffusion properties during the evolution of the micro-porosity system into the fracture system.

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For this purpose, the diffusion characteristics of methane, nitrogen and carbon dioxide in the coal particles were studied. The time-varying diffusion characteristics of methane, nitrogen and carbon dioxide in the coal particles were compared and analyzed by constructing objective function minimization method. This study can provide important basic data to determine a reasonable multi-component gas ratio in place of coal bed methane.

2. Experimental methods

2.1. Preparation of coal samples

Experimental coal samples were obtained from No. 27131 working face of Yanmazhuang colliery in Henan province, China. Coal samples were crushed, screened and selected. The coal samples of particle diameter of 0.2–0.25 mm, 0.25–0.5 mm, 0.5–1 mm and 1–3 mm were prepared. The coal samples were observed and photographed using image processing technology as shown in Fig. 1. Fig. 1 shows that the larger the particle diameter range, the more obvious the difference degree of coal sample overall dimensions. Also, the shape of particle diameter of 0.2–0.25 mm tends to be nearly spherical or ellipsoidal.

Considering the influence of the preparation method of crushing and screening on experimental coal particle shape and dimension [20–22], coal sample of a particle diameter of 0.2–0.25 mm with 20.003 g in mass was used in the experimental tests.

2.2. Experimental set-up and procedure

The experimental equipment through the process of coal gas adsorption testing is HCA high pressure adsorption measuring device, independently developed by Chongqing Research Institute of China Coal Technology Engineering Group. The profession standard of experimental implementing steps can be found in standard practice of GB/T19560-2008. In the test, degassing is firstly conducted. The coal samples were placed into coal sample tank and the roots vacuum pump is turned on to degas from the tank. The degassing time is 3–4 h. Second step is inflation and adsorption. After degassing, a certain amount of the test gas was filled. The test gases referred to the paper are methane, nitrogen and carbon dioxide and the purity is 99.99%. The adsorption time is 72 h. The last step is to determine the amount of gas diffusion in coal samples by connecting the adsorption tank and the high-pressure gasbag. When the gas pressure in the tank is 0, record the time of this process. In this process, the volumes of gas diffusion is based on the volume of the tank and the density of coal samples. Then connect the adsorption tank to the flow measuring device so that the amount of gas diffusion of coal samples can be continuously determined. Determination process of gas in the coal particles was conducted using internationally accepted vent valve. In other words, the coal adsorption tank during adsorption and diffusion of the gas is placed in thermostated water bath by setting the temperature of 30 °C.

3. Calculation method of the diffusion coefficients

The adsorption constants and industrial analysis parameters of methane, nitrogen and carbon dioxide were measured based on the standard practice of GB/T19560-2008 before the experiment. Industrial analysis of and the adsorption constants of different test gases are shown in Tables 1 and 2.

The shapes of coal particles used in this paper are cylindrical or ellipsoidal. Coal particles in the calculation process of the diffusion coefficient are considered as a homogeneous sphere, which is consistent with the literature [20]. It is assumed that after gas adsorption equilibrium of coal particles, the diffusion coefficient of the test gases inside the coal particles follows a time varying function. Therefore at a given time when the gas diffuses through a certain cross section, the product of the diffusion flux, the concentration gradient and the diffusion coefficient at that time are still proportional. The analytical solution taking the Laplace transformation under the steady diffusion coefficient was considered as the basis for the calculation of diffusion coefficient [20].

$$\frac{Q_t}{Q_\infty} = 1 - \frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{n^2}\exp\left(-\frac{n^2\pi^2 D}{r_0^2}t\right) \tag{1}$$

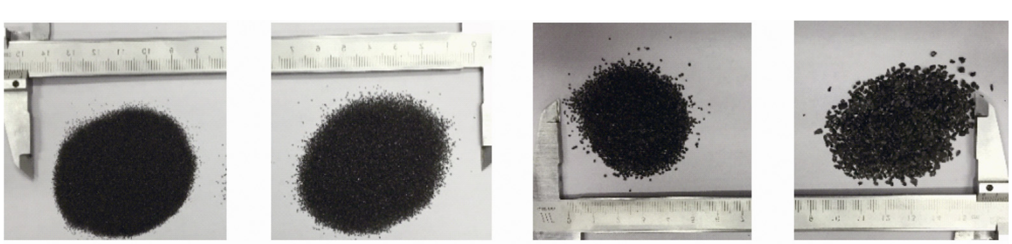
where Q_t is the cumulative diffusion amount of gases at time t, cm^3/g; Q_∞ is the limiting diffusion amount of coal particles and its value equals the difference of between the initial gas content and gas content at the final state, cm^3/g; D is the diffusion coefficient, cm^2/s; and r_0 is the radius of coal particles, cm. The calculation of Q_∞ is combined with Langmuir equation according to the parameters shown in Tables 1 and 2.

Since n values can be up to infinity in Eq. (1), it is set to 1 for simple purpose [20,23]. Thus, Eq. (1) can be transformed as follows:

$$\ln\left(1 - \frac{Q_t}{Q_\infty}\right) = -\lambda t + A \tag{2}$$

where λ is the curve slope of the function [ln $(1 - Q_t/Q_\infty) - t$], and $\lambda = \frac{\pi^2 D}{r_0^2}$; A is the intercept of the function curve and the longitudinal axis, and $A = \ln\frac{6}{\pi^2}$.

Handling of Eq. (2) may be indicative to the variation characteristics of gas diffusion coefficient throughout the diffusion process to a large extent. Here, only the case of $n = 1$ is considered. Thus there are some deviations between the calculation accuracy and the actual process. Therefore, the author used the minimization method of constructing objective function. Considering analytical solution given by Eq. (1), the calculation method of time varying diffusion coefficient of different absorption gases in coal particles was proposed. That is, the diffusion coefficient is obtained by calculating the minimum value of Eq. (3), measuring diffusion amount of test gases in coal particles at different time, and divided-period fitting.



(a) 0.2-0.25 mm (b) 0.25-0.5 mm (c) 0.5-1 mm (d) 1-3mm

Fig. 1. Coal samples of different particle sizes.