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# Constitutive model for methane desorption and diffusion based on pore structure differences between soft and hard coal

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ABSTRACT

This paper aims to improve the accuracy and applicability of gas diffusion mathematical models from coal particles. Firstly, a new constitutive model for gas diffusion from coal particles with tri-disperse pore structure is constructed by considering the difference in characteristics between soft coal and hard coal. The analytical solution is then derived, that is, the quantitative relationship between gas diffusion rate ($Q_t/Q_\infty$) and diffusion time ($t$). The pore structure parameters of soft coal and hard coal from Juji coal mine are determined. Gas diffusion rules are numerically calculated and investigated by physical simulation methods. Lastly, the applicability of this model is verified. The results show that the homogeneous model only applies to the gas diffusion process of hard coal during the initial 10 min. The calculation results from this model and the physical experimental results of soft coal and hard coal are nearly identical during the initial 30 min.

## 1. Introduction

Gas diffusion law and model from coal particles are the key theoretical bases for determining gas content in a coal seam [1–4], coal and gas outburst prediction parameters [5,6], gas emission from falling coal [7,8], and coal-bed methane (CBM) exploitation [9,10]. The dynamic law of gas adsorption and desorption from coal particles belongs to the research category of gas adsorption, desorption and migration theory in porous media [11]. Coal particles refer to the loose coal spalled from a coal seam (wall), which don't load ground stress.

Currently, there are only a few theoretical models to describe the law of gas diffusion from coal particles based on different perspectives. In terms of internal pore structure in coal particles, these models can be classified into two types, including unipore models [6,12–15] and bi-disperse diffusion models [16–18]. In addition, by using the analogy of relaxation phenomena in glassy polymers to study gas diffusion in coal, Staib et al. [19] has produced a stretched exponential model of $CO_2$ and $CH_4$ diffusion that assumes a distribution of characteristic diffusion times. Nevertheless, there are usually notable deviations that lead to not meeting the requirement of engineering when these models are applied to describe the gas diffusion laws of outburst-prone soft coal or tectonic coal [20,21]. The following are some specific problems that arise during engineering application.

Firstly, the indices of outburst prediction, including the gas content and gas desorption index by drill cuttings, cannot be determined accurately. Coal and gas outburst should occur only if the outburst indices are greater or equal to a threshold value. However, due to errors in the estimation of outburst prediction parameters, accidents caused by coal and gas outburst have occurred, and resulted in casualties. For example, the accident of "10.27" coal and gas outburst caused 18 deaths and 5 injuries at Jiulishan mine in Jiaozuo city, China. The estimation error or deviation was caused by the limitations of the theoretical assumptions and errors, especially the mathematical models that are inadequate to describe gas emission rules from soft coal layers where outburst is apt to take place [20,21]. Secondly, due to the deviation errors of gas content, there emerges an abnormal phenomenon where the recovery amount of gas resource is greater than that estimated by exploration in China [22,23]. Such deviation error also results in inaccurate forecast of gas emission from mined coal in underground coal mines. Therefore, it is essential to develop a more precise mathematical model that can reflect the difference in pore structure and gas diffusion law between soft coal and hard coal.

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The conventional model that is commonly used is a transient mathematical model of gas diffusion based on uniform porous coal particles and Fick's second law for spherically symmetric flow [13]. As shown in Eq. (1), the model is a classical homogeneous diffusion model from coal particles. The additional assumptions are summarized as follows:

(1) The intra-particle gas flow follows the law of conservation of mass and continuous theorem.
(2) Diffusion coefficient is independent of concentration, location and time.
(3) Isothermal system.

$$\frac{\partial C}{\partial t}=\frac{D}{r^2}\cdot\frac{\partial}{\partial r}\left(r^2\frac{\partial C}{\partial r}\right)=D\left(\frac{\partial^2 C}{\partial r^2}+\frac{2}{r}\cdot\frac{\partial C}{\partial r}\right) \tag{1}$$

where $C$ is the adsorption concentration, kg/cm$^3$; $r$ is the radius of the coal particle, m; $D$ is the diffusion coefficient of gas in coal particle, m$^2$/s; and $t$ is the time of gas diffusion, s.

Yang, Nie and Crosdale have conducted research regarding the homogeneous diffusion model, such as the analytical solution of the model, the related experiments, and the simplified numerical calculation [6,14,24]. The results show that the relationship between the amount of gas transported and time is a series solution, as shown in Eq. (2). They believe that it is adequate to describe the initial gas emission rules from hard coal.

$$\frac{Q_t}{Q_\infty}=1-\frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{n^2}e^{-n^{2Bt}} \tag{2}$$

where $Q_t$ is the total volume of the diffusing gas at time $t$, cm$^3$/g; $Q_\infty$ is the total diffusion volume, cm$^3$/g; $B=\pi^2D/a^2$, and the value of $B$ ranges from $6.5797\times10^{-6}$ to $6.5797\times10^{-3}$.

The numerical calculation value of $B$ is $6.5797\times10^{-5}$, and the value of $n$ is 1, 10 and 10,000. The numerical solution to Eq. (2) verifies the adaptability of Eq. (1) by using the software of Maple which is a numerical calculation software according to a given model and related parameters. The results are shown in Fig. 1. With the increase of $n$ value, the calculation results using Maple software get close to the experimental results of gas diffusion in shape. When $n$ is equal to 1, 10, and 10,000, the theoretical diffusion rate is conducted correspondingly. The relationship between ln $[1-(Q_t/Q_\infty)^2]$ and $t$ is exhibited in Fig. 2, where no matter how what $n$ is equal to, the relationship of both is always linear. The phenomenon depends on the assumption that the diffusion coefficient is constant.

The experiment concerning the gas diffusion from soft coal and hard coal of Juji coal mine was performed. The experimental results show that the relationship between ln $[1-(Q_t/Q_\infty)^2]$ and $t$ is not linear, just as illustrated in Fig. 3. The results also reveal that gas diffusion coefficients of soft coal and hard coal both decrease with time, furthermore, gas diffusion coefficients of soft coal decrease more significantly with time.

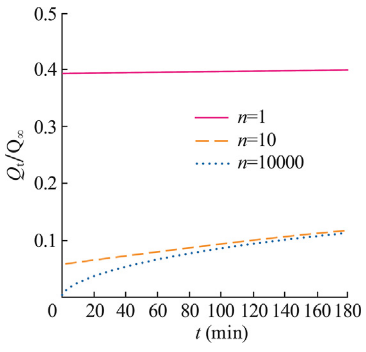

**Fig. 1.** Numerical solution of homogeneous diffusion from coal particles.

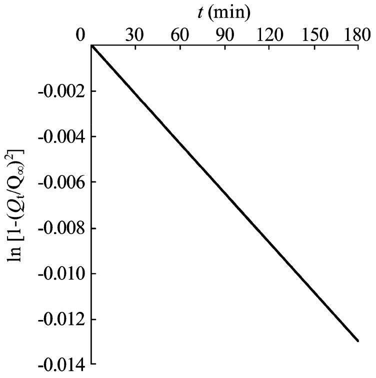

**Fig. 2.** Theoretical calculation based on Eq. (2) when $n$ = 1, 10, and 10,000.

Considering the mass transfer resistance of the surface of coal particles, Nie et al. adapted the unipore diffusion model [14]. The mathematical and physical model of gas diffusion through coal particles under the third type boundary condition was established and its analytical solution was using mathematical and physical methods. The simplification of the solution to the equation is similar to the empirical formula developed by Bolt and Innes [25].

Qin et al. believes that gas emission rate from coal particles is proportional to the gas pressure gradient in the inter-particles according to the experimental data [15]. Therefore, he developed a mathematical model based on Darcy's law and homogeneous coal particles. However, most coal is actually not homogenous. Rather, it can, and often is, quite variable in terms of maceral and mineral content. Pore content and size distribution can also be highly variable. Moreover, it is inconsistent with the opinion that the gas diffusion process from coal particles complies with Fick's law, which most scholars agree with. In addition, it is very difficult for gas migration in coal particle to form a pressure driving condition. Zhang [26] hypothesized that the diffusion coefficient increases with the emission time, however, gas concentration in the inter-particles areas is reduced. He also proposed a spherical diffusion model using time-varying diffusion coefficient. In the initial and terminal phases, the gas diffusion coefficient basically tends to be a constant, but the diffusion coefficient exhibits a sharp increase at a certain period in the middle of diffusion time. Zhang's [26] results are thus inconsistent with most scholars' experimental results including Yang, Nandi, and Liu [6,12,20,21].

Coal particles with complex porous structure are assumed to have a uniform pore system [6]. Therefore, the diffusion model is simple and easy to use, which is based on gas diffusion from homogeneous and spherical coal particles. However, the assumption also results in poor applicability of the models, and is inadequate to describe long-term diffusion rules of coal, especially soft coal. Many scholars demonstrated that there are remarkable errors in determining gas diffusion coefficients and lost gas volumes [1,6,21].

Bi-disperse diffusion models assume two pore sizes: micro-pore and macro-pore, which are classified into parallel-path models and continuum models according to diffusion pattern. Yi et al. [18] simplified the Pore plus Sorb-Phase Diffusion Model developed by Gray and Do [27], which belongs to the parallel-path diffusion model that assumes that gas molecules diffuse in parallel. Moreover, local equilibrium exists among the concentrations of gas in micro-pore and the macro-pore, as shown in Fig. 4 [28]. The mathematical model is expressed in Eq. (3). The resulting mathematics lends itself to analytical solutions, a convenience that has made the model quite popular. However, Helfferich believes that there are

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