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Experimental study of the radial multi-scale dynamic diffusion model for gas-bearing coal

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Abstract

Purpose. The purpose of this paper is to solve the scientific problem that the classical diffusion model in columnar coal cores cannot accurately describe the whole process of gas diffusion.

Methods. The diffusion-percolation experiments were carried out using the laboratory's homemade experimental equipment with standard ϕ 50mm×100 mm columnar raw coal cores under different air pressures.

Findings. The classical diffusion model was used to fit the experimental data. The experiment has found that the classical diffusion model of the columnar coal core can only partially describe the gas diffusion process. The longer the experimental time, the larger the error between the model and the experiment, and the analysis has found that the apparent diffusion coefficient shows decay changes with time. The dynamic diffusion coefficient concept is then proposed in order to con-struct a radial multi-scale dynamic prominent diffusion-percolation model for columnar coal cores. The theoretical curve of the new model nearly coincides with the experimental curve, and the new model can describe the gas diffusion-percolation process of columnar coal cores more accurately. The multi-scale dynamic diffusion-percolation model covers the classical diffusion model. It explains the mechanism of gas diffusion-percolation in multi-scale pores, i.e., at the beginning of the flow, gas flows out from the large external pores first, from the surface inwards. Over time, the pore size through which gas flows gradually becomes smaller, the diffusion resistance gradually increases, and the apparent diffusion coefficient slowly decreases.

Originality. This paper proposes a new multi-scale dynamic diffusion-percolation model to compare the old and new model analysis, as well as carefully studying the mechanism of gas flow in coal.

Practical implications. This research has important engineering significance for the accuracy of measuring the gas content of coal seams, as well as predicting coal and gas content.

Keywords: *apparent diffusion coefficient, columnar coal core, multiscale, diffusion-percolation model*

1. Introduction

Coal seam gas content is one of the factors causing coal and gas protrusion, so it is essential to accurately predict the gas content of coal seams and their storage capacity. However, when measuring the coal seam gas content at the surface of the field as part of the CBM project, the diffusion characteristics of coalbed methane from columnar coal cores extracted by drilling are very different from the gas diffusion characteristics of granular coal cores drilled in coal mines. The gas content measurement primarily uses a spherical diffusion model, and the relevant results are unsatisfactory. Therefore, it becomes necessary to study the diffusion characteristics and permeability properties of the original coal body.

Most experimental studies have been conducted using fine coal to study gas diffusion in coal. Barre [1] and Crank [2] have derived the exact solution of the spherical classical model to obtain the precise resolution and simplified equation for diffusivity. Ruckenstein [3] proposed a double pore diffusion model, but the diffusion model is difficult to calculate and promote, and it is not suitable for engineering field surveys. Yang [4] derived the exact solution and simplified formula of the classical diffusion model and used the approximation of the classical diffusion model to calculate the diffusion coefficient. After that, Nie [5]-[7] derived on this basis the trigonometric expression of the classical diffusion model, considering the surface diffusion resistance of the boundary layer and introducing the third type of boundary conditions. Zhang [8] proposed a new model of the dynamic diffusion coefficient, in which the diffusion coefficient is considered as a pore diameter. Li et al. [9], [10] discovered the dynamic decay characteristics of the gas diffusion coefficient in coal. They proposed a new multi-scale model of the dynamic diffusion coefficient, which can accurately describe the full-time diffusion process of gas in fine coal compared with the constant diffusion coefficient model.

The spherical model does not describe the gas diffusion characteristics of columnar coal cores due to the significant difference in size between granular and columnar cores. There are few studies on the columnar model. Pan [11] and Tan [12] conducted gas diffusion experiments using colum-

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nar coal cores. However, they used the spherical double pore model to analyze gas diffusion characteristics in columnar coal samples. The pressure function is still used in the spherical dual pore model to study lumpy coal for unidirectional diffusion. Through the spherical double porosity model, only effective diffusion coefficients were obtained but not diffusion coefficients. Liu [13] has studied a theoretical model when a single gas is adsorbed and expanded, and carried out column coal experiments to calibrate the model. However, the model treated the diffusion coefficients as constants independent of time and pressure, so many constant coefficients has no physical significance and could not be used in a theoretical model. Li [14] determined a radial diffusion equation for gas in columnar coal cores, carried out an experimental study of radial diffusion, derived an approximate solution of the columnar radial model, measured the diffusion coefficient of the gas in large-scale coal cores, and analyzed its flow characteristics. However, he does not considered the changes of different gas diffusion properties during the evolution of the micro-pore system to the fracture system.

Thus, the current research on the radial gas flow model of columnar coal cores is still inaccurate, which dramatically affects the accuracy of gas content measurement in columnar coal cores and determining the intrinsic permeability of raw coal. The experimental aspects are fragmented and their standardization, relevance, clarity, and scientificity should be improved. Therefore, further practical and theoretical research on columnar coal cores require conducting diffusion-percolation experiments on columnar coal cores and creating a radial multiscale dynamic apparent diffusion-percolation model, thereby providing a relatively accurate theoretical model for determining gas content and studying intrinsic permeability in deep drilling.

2. Column coal gas diffusion-percolation experiment

2.1. Experimental setup and method

The column coal core diffusion-percolation experimental system is shown in Figure 1. The practical system consists of a methane inlet unit, a vacuum pumping unit, a column coal core adsorption unit, and a gas flow measurement unit.

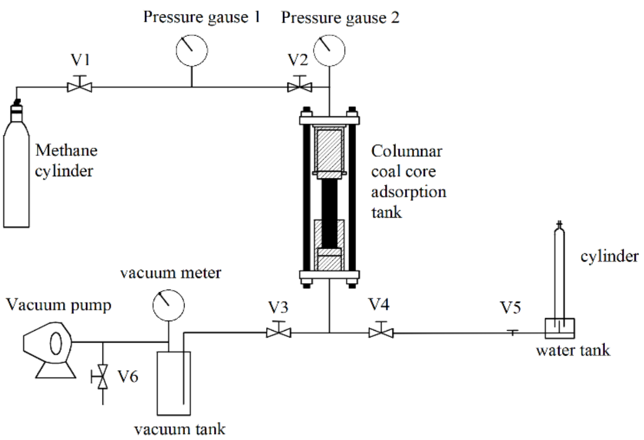

Figure 1. Coal gas diffusion-percolation experimental system

The methane gas cylinder, valve V1, and pressure gauge 1 form the inlet unit, the vacuum pump, vacuum tank, vacuum gauge set, and valve V6 condition the vacuum pumping unit, the column coal core adsorption balance tank, valves V2, V3, V4 and pressure gauge 2 forms the column coal core adsorption unit, and valve V5 and water tank form the gas flow monitoring unit.

2.2. Experimental method

1. Coal sample preparation

The coal samples were taken from the Shenmu coal mine, and ϕ 50×100 mm columnar raw coal cores were drilled axially along the seam. To exclude the effect of moisture, all coal samples were vacuumed and dried anaerobically in a muffle furnace at 100°C. The results of the basic parameters of the coal samples are shown in Table 1.

Table 1. Basic parameters of coal samples

No	Parameters	Values
1	Moisture	0.0108
2	Ash	0.1017
3	Volatile matter	0.089
4	Density	1.59
5	Porosity	0.0525
6	a_t (adsorption constant)	43.49
7	b_t (adsorption constant)	1.03

2. Procedure of the diffusion-percolation experiment

As shown in Figure 1, the dried column coal is placed in the column coal core adsorption equilibrium tank, and therefore the valve is closed. Valves V2, V3, V4, and V5 are opened and the vacuum pump is turned on for pumping operation. When the absolute pressure of the vacuum gauge is 10 Pa and there are no more continuous changes, valve V3 is closed and valve V6 is opened to make the pressure in the vacuum pump vessel equal to zero. Then it is necessary to open the valves V1 and V2, close the valves V3 and V4, and control the pressure-reducing valve of the methane gas cylinder. The pressure is set at 3 MPa of the column coal for the inflation operation. To achieve equilibrium in coal sample adsorption for 72 h, close V1, V2 and pressure reducing valve switch, open the valve V4, release the gas in the coal sample tank. Then wait for 5s, the gas pressure in the coal sample tank drops to atmospheric pressure, immediately turn on valve V5 to desorb the gas in the coal within 180 min. The cumulative amount of gas desorbed is measured through the measuring cylinder in the water tank by using the drainage method. The cumulative desorption amount was recorded every 30 seconds until the cumulative desorption amount changed very little after 180 min, and the test was finished. The air pressure was adjusted to 3, 2, 1 and 0.5 MPa in turn, and the above experimental operation was repeated to complete the column coal core diffusion-percolation experiment.

2.3. Experimental data processing

After the diffusion-percolation experiment, the measured cumulative gas diffusion was converted into the diffusion quantity Q_t per unit mass of coal under standard conditions and compared with the ultimate diffusion quantity Q_∞ to obtain the diffusivity Q_t/Q_∞ versus the time curve. In the experiments, the drainage method was used to measure gas diffusion, and the limiting desorption quantity Q_∞ under this condition is the difference between the initial gas content Q and the final state gas content Q_a at atmospheric pressure. Thus, $Q_\infty = Q - Q_a$, where Q, Q_a under experimental conditions are calculated according to the following Equation:

$$Q = \frac{a_t b_t p}{1+bp}\left(1-A_{ad}\right)+\frac{10p\phi \cdot 273}{\rho\left(273+\theta_w\right)}, \tag{1}$$

where:

a_w, b_w – are the adsorption constants at corresponding temperatures;

p – the adsorption equilibrium pressure;

A_{ad} – the ash;

θ_w – the desorbed water temperature;

ρ – the apparent density;

$Ø$ – the porosity;

Q – the total gas content corresponding to the initial adsorption equilibrium pressure p;

Q_a – the gas content at atmospheric pressure.

When calculating the final state equilibrium gas content Q_a at atmospheric pressure, the pressure p in Equation 1 above is replaced by atmospheric pressure.

3. Comparison of the classical diffusion model of a columnar coal core with experiment

In the process of observing and analyzing experimental results, the normalized diffusivity versus time ($Q_t / Q_\infty \sim t$) curves are often used to reflect the changes in diffusion patterns. The classical model of columnar coal cores is commonly used to analyze the gas diffusion of columnar coal, and the analytical solution is shown in Equation 5 [2], [14].

$$\frac{Q_t}{Q_\infty} = 1 - 4\sum_{n=1}^{\infty}\frac{\exp\left(-a_n^2 Dt\right)}{a_n^2 R^2}, \quad (2)$$

where:

Q_t – the cumulative diffusion at moment t, cm^3/g;

Q_∞ – the limiting diffusion, cm^3/g;

Q_t / Q_∞ – the cumulative diffusion at moment t;

D – the apparent diffusion coefficient, cm^2/s;

a_n – the nth positive root of the first class zero-order Bessel function $J(a_n R) = 0$;

R – the radius of the columnar coal core, cm.

In Equation 2, when $n > 1$, it is more difficult to solve the apparent diffusion coefficient D, and $n = 1$ is taken to simplify the process.

$$\ln\left(1 - \frac{Q_t}{Q_\infty}\right) = -\lambda t + B; \qquad \lambda = a_1^2 D, \quad B = \ln\frac{4}{a_1^2 R_0^2}, \quad (3)$$

where:

a_1 – the first positive root of the first class zero-order Bessel function $J(a_n R) = 0$.

The experimental data of the column coal cores were processed according to Equation 3 and 2, and the experimental data of 0.5 and 3 MPa were selected to plot the experimental versus theoretical graphs and $\ln(1 - Q_t / Q_\infty) \sim t$ shown in Figure 2.

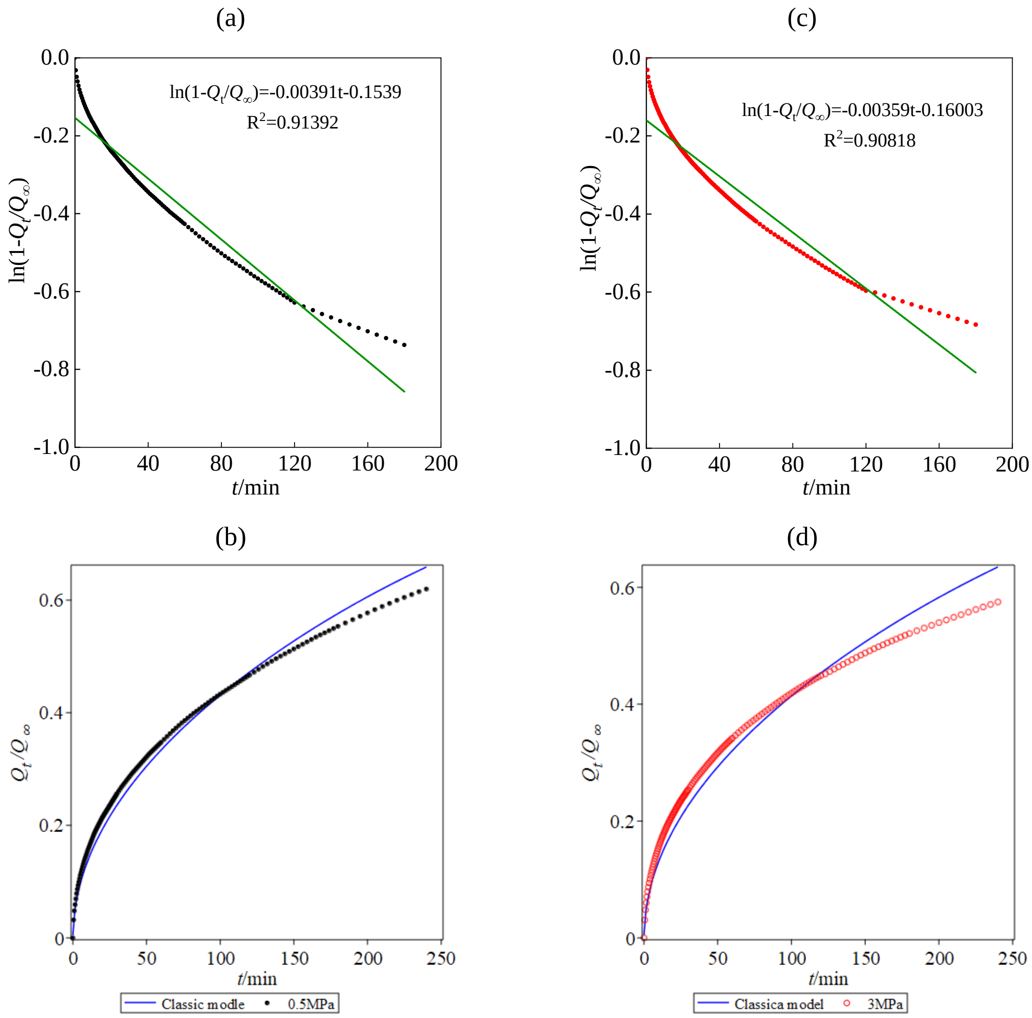

Figure 2. Fitted and classical model curves of columnar coal core diffusion parameters: (a) fitting of CH_4 diffusion parameters for 0.5 MPa; (b) comparison of classical model and experiment for 0.5 MPa; (c) fitting of CH_4 diffusion parameters for 3 MPa; (d) comparison of classical model and experiment for 3 MPa

Figure 2 shows that the theoretical value Q_t / Q_∞ of the classical diffusion model is larger than the experimental value at an early stage. The theoretical value Q_t / Q_∞ of the classical diffusion model is smaller than the practical value at the later stage.

The reason for the above phenomenon is that the apparent diffusion coefficient determined by the classical diffusion model is a constant that depends on the average diffusion capacity of gas in coal over time and contradicts the dynamic decay change of the apparent diffusion coefficient presented by the experimental results. In addition, this leads to the fact that the theoretical value of the classical diffusion model is more significant than the practical value at an early stage. The theoretical value of the classical diffusion model Q_t / Q_∞ is smaller than the experimental value at the later stage. Over time, the error of the curve between the classical diffusion model and the practical point becomes larger and larger. The classical diffusion model cannot describe the whole process of gas diffusion, and a dynamic diffusion-percolation model is needed that can describe the columnar coal core.

4. Multi-scale dynamic diffusion-percolation model

4.1. Development of a multi-scale dynamic diffusion-percolation model

The analysis of low-temperature liquid nitrogen and piezometric experiments shows that coal has pores of different sizes, ranging from several nanometers to several microns. These pore structures are continuously distributed and coal has multi-scale pore characteristics. The apparent diffusion coefficient indicates the ease of gas diffusion in coal, which is directly related to the pore structure of coal, and the ease of gas diffusion varies in different pore structures. Gas flow in coal is a continuous process of desorption-diffusion-percolation, so the dynamic apparent diffusion coefficient is used to describe the gas diffusion-percolation process in columnar coal cores.

To accurately describe the dynamic decay characteristics of the apparent diffusion coefficient, a negative exponential function is proposed to represent the variation law of the apparent diffusion coefficient [15]:

$$D(t) = D_0 \exp(-\beta t), \tag{4}$$

where:

$D(t)$ – the dynamic apparent diffusion coefficient that decays with time, cm^2/s;

D_0 – the initial apparent diffusion coefficient at $t = 0^+$, cm^2/s;

β – the decay coefficient of the dynamic apparent diffusion coefficient, s^{-1}, reflecting the degree of transition from the large outer pore to the small inner pore at different scales;

t – time, s.

The assumptions of the model are:

1) the radial dimension of the columnar coal core is smaller than the axial dimension, and the gas mainly flows out from the radial direction, ignoring the end effect;

2) the columnar coal core is assumed to be non-homogeneous, and the apparent diffusion coefficient of the columnar coal core varies dynamically with time, and the dynamic apparent diffusion coefficient contains both percolation and diffusion flow states;

3) the whole gas flow process is a continuous diffusion-percolation process and follows the law of mass conservation.

We get this by substituting Equation 5 into the apparent diffusion equation for columnar coal cores in columnar coordinates:

$$\begin{cases} \dfrac{\partial C}{\partial t} = D(t)\left(\dfrac{\partial C^2}{\partial r^2} + \dfrac{1}{r}\dfrac{\partial C}{\partial r}\right) \\ \dfrac{\partial C}{\partial r} = 0 \quad (r = 0, \quad t > 0) \\ C = C_0 \quad (t = 0, \quad 0 \le r \le R) \\ C = C_a \quad (r = R, \quad t > 0) \end{cases}, \tag{5}$$

where:

C – the methane diffusion mass concentration as a function of diffusion path r and time t, i.e., $C = C(r, t)$, g/cm^3;

r – the diffusion path, cm;

C_0 – the methane mass concentration of the initial adsorption equilibrium, the initial condition of the equation, g/cm^3;

C_a – the gas mass concentration on the surface of the coal core during diffusion, the boundary condition of the coal surface, g/cm^3;

R – the radius of the columnar coal core, cm.

By separating the variables and substituting Equation 4 into Equation 5 for the solution, a multiscale dynamic apparent diffusion model for columnar coal cores can be obtained:

$$\frac{M_t}{M_\infty} = \frac{Q_t}{Q_\infty} = 1 - \frac{4}{R^2}\sum_{n=1}^{\infty}\frac{\exp\left(a_n^2 \dfrac{D_0}{\beta}\left(\exp(-\beta t) - 1\right)\right)}{a_n^2}, \tag{5}$$

where:

Q_t and Q_∞ – the volumes of the gas in the standard condition by converting the masses M_t, M_∞ respectively, cm^3/g;

a_n – the n^{th} positive root of the first class zero-order Bessel function $J_0(a_n)$.

In gas diffusion-percolation experiments, gas flow in micro-nano pores is slow. The microscopic mechanism is more complex and difficult to observe, so it is generally converted into apparent permeability to describe and analyze the gas flow pattern in micro-nano pores from a macroscopic perspective.

According to the ideal gas equation and Darcy's theorem, the mass flux per unit time and per unit area is obtained:

$$J = -\frac{k(t)}{v} K_B T \nabla n_\rho, \tag{7}$$

where:

$k(t)$ – the apparent dynamic permeability;

v – the kinematic viscosity coefficient;

K_B – the Boltzmann constant;

T – the Kelvin temperature;

n_ρ – the number of molecular gas densities.

Equation 7 and Fick's law are combined to obtain:

$$k(t) = \frac{Mv}{R'T} D(t). \tag{8}$$

Further, the expression of the dynamic apparent diffusion coefficient and apparent dynamic permeability can be obtained as Equation 9:

$$k(t) = \frac{\mu}{p} D(t), \tag{9}$$

where:

μ – the dynamic viscosity coefficient, Pa·s;

p – the gas pressure, MPa.

The simple permeability conversion equation can convert a multi-scale dynamic diffusion-percolation model into a multi-scale dynamic apparent permeability model.

4.2. Dynamic apparent diffusion-percolation model testing and analysis

The experimental data were processed according to the multiscale dynamic diffusion-percolation model of columnar coal cores of Equation 6, and then further processed according to the apparent permeability conversion equation of Equation 9 to obtain the apparent permeability ratio k_t/k_∞ versus time curves and plot the comparison between the classical transparent diffusion model and the multiscale dynamic apparent-percolation model in Figure 3.

As shown in Figure 3, the experimental data at 0.5, 1, 2 and 3 MPa were processed. By comparing the classical apparent diffusion model and the multi-scale dynamic apparent diffusion-percolation model, it has been found that compared to the classical prominent diffusion model, the multi-scale dynamic transparent diffusion-percolation model for columnar coal cores can describe the whole process of gas diffusion in coal. The theoretical curve overlaps with the experimental curve and fits with higher accuracy. The problem is solved that the hypothetical value of the classical diffusion model is larger than the practical value at an early stage and smaller than the experimental value at the later stage. Using the apparent permeability conversion formula, the gas flow law in the pore space of a coal body can be analyzed more intuitively.

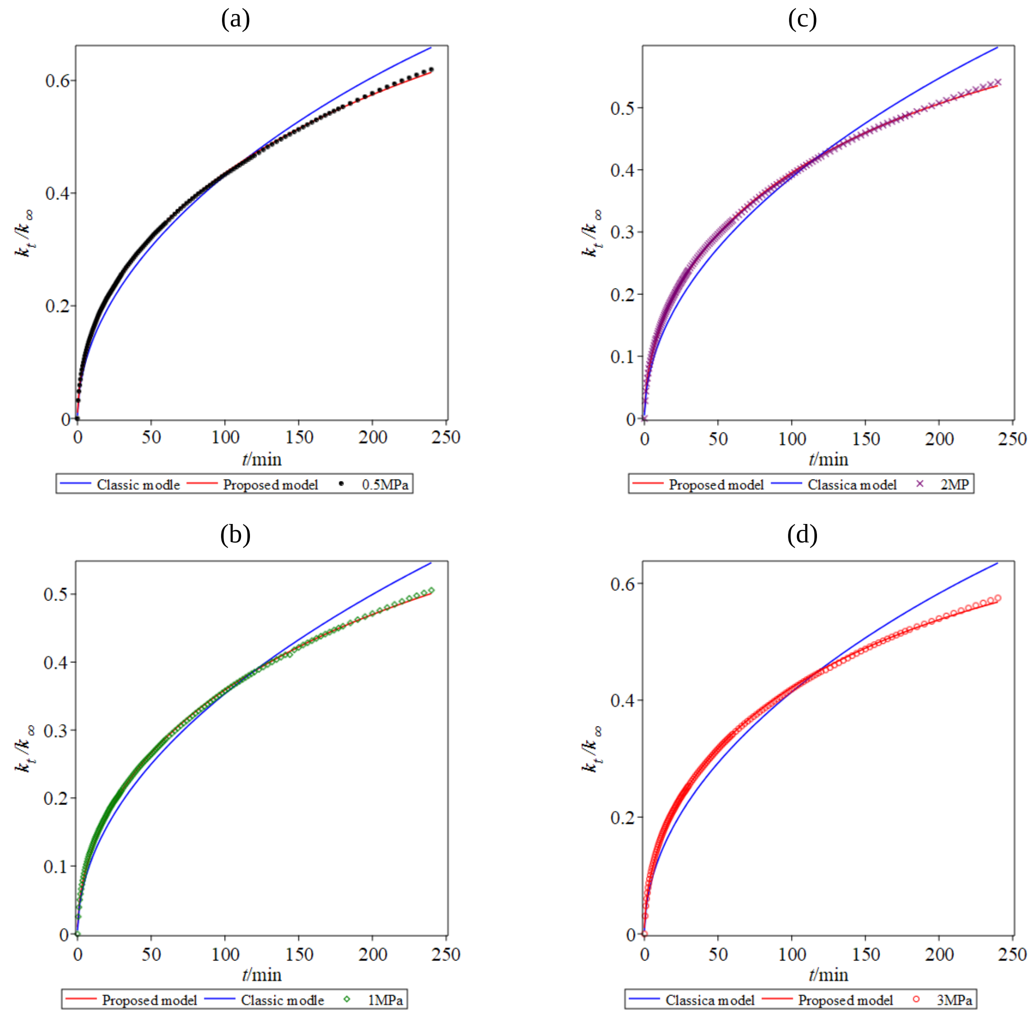

Figure 3. Multi-scale dynamic diffusion-percolation model of columnar coal cores compared with the classical model: (a) methane comparison chart for 0.5 MPa; (b) methane comparison chart for 1 MPa; (c) methane comparison chart for 2 MPa; (d) methane comparison chart for 3 MPa

4.3. Multi-scale dynamic apparent diffusion-percolation model with classical model relationships

Comparing Equation 2 and 6, it can be seen that:

$$\bar{D}(t)=\int_0^t D_0\exp(-\beta t)\,dt$$
$$\bar{D}=\frac{D_0}{\beta t}\left(1-\exp(-\beta t)\right) \quad , \qquad (10)$$
$$\bar{D}=D \qquad \left(Eq(2)=Eq(6)\right)$$

where:

$\bar{D}$ – the average value of the dynamic diffusion coefficient in the period from 0 to t, cm^2/s.

It can be seen from Equation 10 that the average value of the apparent dynamic diffusion coefficient $\bar{D}$ is equal to the classical model diffusion coefficient D, when Equation 2 and 6 are equal at a specific moment of time, which is the intersection of the multi-scale dynamic apparent diffusion-percolation model and the classical model curve in Figure 3.

4.4. Mechanistic study of multi-scale dynamic apparent diffusion-percolation model

The dynamic decay change of the apparent diffusion coefficient reflects the narrowing of the pore structure of gas flowing in coal, since coal has a multi-scale pore structure. The pores are connected with self-similar morphology, as shown in Figure 4. The pores in coal can be viewed as circular tubes of unequal diameters connected in series.

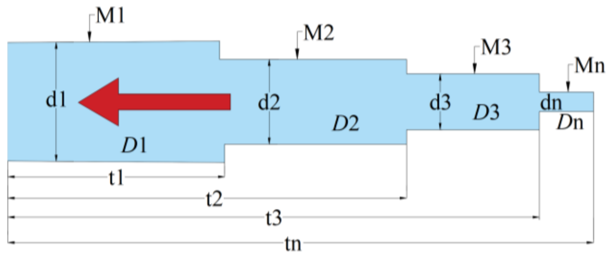

Figure 4. Multi-scale pore structure of coal sample

At the initial stage of gas diffusion t1, gas in coal first flows out from the large pores and fissures of M1, when the gas flow is subject to minor diffusion resistance and the most significant diffusion coefficient. At stage t2, the gas starts to flow out of the pores M2, the diffusion resistance starts to become more extensive, and the diffusion coefficient D2 decreases. As the diffusion time from the surface inwards increases, the apparent diffusion coefficient of each level gradually decreases. The dynamic apparent diffusion coefficient is composed of apparent diffusion coefficient of each successive pore level.

As shown in Figure 4, the apparent diffusion coefficient consists of D1 and D2 in series when the gas diffuses into the M2 matrix, and D1, D2 and D3 in series when the gas diffuses into the M3 level pores, and so on until the end of diffusion. More and more pores are connected in series, the pore size gradually decreases, the resistance to diffusion in series increases, and the apparent diffusion coefficient of series dynamics decreases slowly with time.

5. Conclusions

The classical diffusion model cannot describe the whole process of gas diffusion in columnar coal cores. The experiments have shown that the apparent diffusion coefficient is not a constant, but shows a changing trend of dynamic decay with time, so a multi-scale dynamic transparent diffusion-percolation model has been developed. The conversion relationship between the dynamic apparent diffusion coefficient and the apparent dynamic permeability is obtained according to the principle of mass conservation, which is useful for studying the intrinsic permeability of coal.

The mechanism of the apparent diffusion coefficient decaying with time is elaborated. Coal has multi-scale pore structure characteristics, and multi-level pores in coal are connected in series and permeable. The apparent diffusion coefficient gradually decreases as the gas in coal diffuses from the large to tiny diffusion pores in the process of diffusion, from the surface inwards.

Comparing the classical diffusion model and the multi-scale dynamic apparent diffusion-percolation model, it has been found that the constant diffusion coefficient in the classical model is equivalent to the average dynamic apparent diffusion coefficient.

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Експериментальне дослідження багатомасштабної радіальної динамічної дифузійної моделі газомісткого вугілля

Я. Сю, С. Чен, Ц. Ю

Мета. Вирішення наукової проблеми більш точного описання процесу дифузії газу у стовпчастих вугільних кернах на основі експериментальних досліджень.

Методика. Дифузійно-перколяційні експерименти проводилися із використанням саморобного експериментального обладнання лабораторії зі стандартними стовпчастими кернами з необробленого вугілля розміром 50×100 мм під різним тиском повітря. Для узгодження експериментальних даних використовувалась класична модель дифузії.

Результати. Експериментально встановлено, що класична дифузійна модель стовпчастого вугільного керна може лише частково описувати процес дифузії газу, при чому чим довший час експерименту, тим більша похибка між моделлю та експериментом. В результаті аналізу виявлено, що уявний коефіцієнт дифузії показує зміни затухання з часом. Запропоновано концепцію коефіцієнта динамічної дифузії для побудови багатомасштабної радіальної динамічної моделі помітної дифузійно-перколяційної фільтрації для стовпчастих вугільних кернів. Визначено, що теоретична крива нової моделі майже збігається з експериментальною кривою, і нова модель дозволяє більш точно описати процес дифузії-перколяції газу у стовпчастих кернах вугілля. Встановлено, що багатомасштабна динамічна модель дає пояснення механізму дифузії-перколяції газу в багатомасштабних порах, тобто на початку потоку газ витікає з великих зовнішніх пор, з поверхні всередину, при цьому з часом розмір пор, через які проходить газ, поступово зменшується, опір дифузії поступово збільшується, а уявний коефіцієнт дифузії повільно зменшується.

Наукова новизна. Вперше запропоновано нову багатомасштабну динамічну модель дифузії-перколяції для порівняння аналізу старої та нової моделей, а також ретельно вивчається механізм потоку газу у вугіллі.

Практична значимість. Це дослідження має важливе інженерне значення для точності вимірювання газоносності вугільних пластів, а також прогнозування вмісту вугілля та газу.

Ключові слова: *уявний коефіцієнт дифузії, стовпчастий вугільний керн, багатомасштабний, модель дифузії-перколяції*