

# Study on the Solution and Variation Law of Diffusion Coefficient Based on the Numerical Simulation Optimization Method

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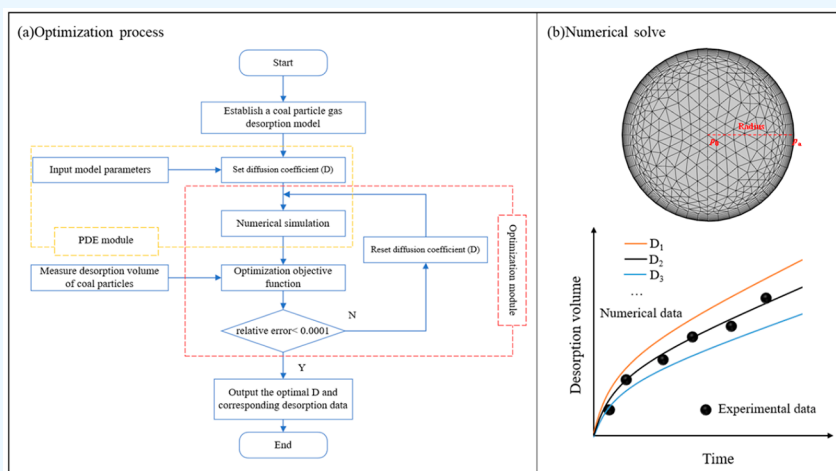
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**ABSTRACT:** Since the diffusion coefficient is a key parameter to characterize the diffusion rate of methane molecules, its measurement and solution have always been a research hotspot. The diffusion coefficient is normally solved through analytical solutions of theoretical models, which is complex and poorly applicable. In comparison, the numerical simulation optimization method can seek a solution easily and quickly, providing a clue for solving such problem. In this paper, first, gas desorption experiments were conducted on coal samples with different initial gas equilibrium pressures, coal particle sizes, and metamorphic degrees. Combined with existing theoretical models, the numerical simulation optimization method was adopted to solve the diffusion coefficient of the coal particle. Furthermore, the applicability and advantages of the numerical simulation optimization method were discussed. Finally, the variation law of the diffusion coefficients was analyzed. The results demonstrate that the numerical simulation optimization method can not only solve the diffusion coefficient easily and quickly but also reveal the law of diffusion concentration with time. The  $d$  values between the solution results and the experimental data under different conditions are all smaller than 0.2, which proves the effectiveness and accuracy of the simulation optimization method. The diffusion coefficient of gas from coal particles is unrelated to the initial gas equilibrium pressure, yet it has a Z-shaped relationship with the coal particle size and a V-shaped relationship with the metamorphic degree.

## 1. INTRODUCTION

Realizing efficient extraction and utilization of coalbed methane (CBM) can achieve triple benefits of “safety, energy, and environment”.<sup>1</sup> Coal is a kind of dual-porosity medium, in which the migration of gas can be divided into three processes: desorption, diffusion, and seepage. In matrix pores, gas flows mostly in the form of diffusion determined by the concentration gradient. In fractures, gas flows mainly in the form of seepage affected by the pressure gradient.<sup>2–4</sup> Diffusion can directly affect the equilibrium of gas pressure in the coal seam and has a certain influence on seepage. Therefore, in-depth exploration of diffusion law of gas from coal seam is to the fundamental task for revealing the migration mechanism of gas from coal seam,

and has a great significance for calculating the gas content of coal and the amount of gas emission from fallen coal and designing gas extraction.<sup>5,6</sup>

Diffusion mainly refers to the process in which molecules of a substance transfer from a high concentration to a low

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concentration under the action of the concentration gradient until the concentration is uniformly distributed.<sup>7,8</sup> The rate of diffusion is proportional to the concentration gradient of the substance. Since the diffusion coefficient is a key parameter to characterize the diffusion rate of methane molecules, its measurement and solution has always been research hotspot.<sup>9,10</sup>

The diffusion coefficient of gas from the coal seam generally lies in the range of  $10^{-15}$ – $10^{-7}$  m<sup>2</sup>/s.<sup>11,12</sup> It is normally calculated by the coal particle desorption method which is to solve the diffusion coefficient by fitting the analytical solution of the diffusion theoretical model on the basis of desorption data.<sup>13–17</sup>

According to different assumptions, diffusion theoretical models are divided into the single-pore diffusion model<sup>18,19</sup> and the double-pore diffusion model.<sup>20,21</sup> In accordance with different solution conditions, diffusion theoretical solution can be classified as constant pressure solution and constant volume solution.<sup>22–24</sup> Crank,<sup>10</sup> Yang,<sup>25</sup> and Nie et al.<sup>14</sup> have derived approximate analytical solutions under constant-pressure condition based on the Fick's law of diffusion. Analytical solutions are generally derived in the form of infinite series under specific assumptions and boundary conditions. Although scientific, its scope of application is limited. Encountered with complex theoretical models, such as constant volume gas desorption, gas desorption under the assumption of a cube coal matrix and diffusion coefficient is assumed to have time-related characteristics, etc., analytical solutions will be more difficult to solve. When the analytical solution cannot be obtained or is difficult to seek, it can be obtained by numerical analysis methods, including numerical approximation, the difference method, the interpolation method, etc.<sup>18,20,21,26</sup> But the calculation process is also cumbersome. According to the characteristic and law of experimental data of gas desorption from coal particle, there appeared simplified analytical empirical formulas such as the  $\sqrt{t}$  model,<sup>25,27</sup> Barrer's power function formula,<sup>8</sup> Sun Zhongxu's formula,<sup>11</sup> and Winter's formula.<sup>28</sup> Nevertheless, important parameters such as the diffusion coefficient cannot be obtained from these formulas for lack of strict theoretical derivation. Currently, no simple and efficient method can be adopted for calculating the diffusion coefficient.

The numerical simulation optimization method has certain advantages in solving partial differential equations and can provide a new clue for solving the diffusion coefficient. The method improves certain indicators by changing the model input data under certain constraints. The optimization module in COMSOL Multiphysics is a universal interface used to study the optimal solution. It can be used with any module in the product library, and applied to parameter, shape, topology optimization, and parameter estimation in practical problem.<sup>29,30</sup> According to the type of problem, the functions of the optimization module can be broadly classified into single objective design and parameter solving. Parameter solving is mainly to compare the simulation data with the experimental data and select the parameter corresponding to the most matching simulation data as the optimal parameter. It is suitable for parameter estimation and curve fitting of a partial differential equation. Compared to the experimental method, the numerical simulation optimization method for solving parameters can improve solving speed and reduce economic cost.

In this paper, gas desorption experiments were conducted on coal samples with different initial gas equilibrium pressures, particle sizes, and metamorphic degrees. The applicability and superiority of the numerical simulation optimization method for solving the diffusion coefficient were discussed. Furthermore,

the variation of the diffusion coefficient of gas from coal particles under different conditions were obtained. The study in this paper can provide a simpler and more accurate algorithm for solving the diffusion coefficient and is crucial for further exploring the law of gas migration in coal particles.

## 2. GAS DESORPTION EXPERIMENTS

The coal samples used in the experiments were taken from the Dongda Coal Mine in Shanxi Province, China. The fresh large coal samples selected on site were sealed and preserved well and transported to the laboratory for experiments. After the coal samples were crushed into coal particles by a crusher, four groups (0.074–0.2 mm, 0.2–0.5 mm, 0.5–1 mm, and 1–3 mm) of coal samples with different particle sizes were separated by a standard sample sieve and put into sealed glass bottles, and the mass of coal particles in each group was not less than 50 g. Then, the coal samples were dried in a vacuum drying oven, sealed, and labeled for later use. Certain basic parameters such as the proximate analysis data, adsorption constant and solidity coefficient of the samples were measured in the laboratory, as presented in Table 1.

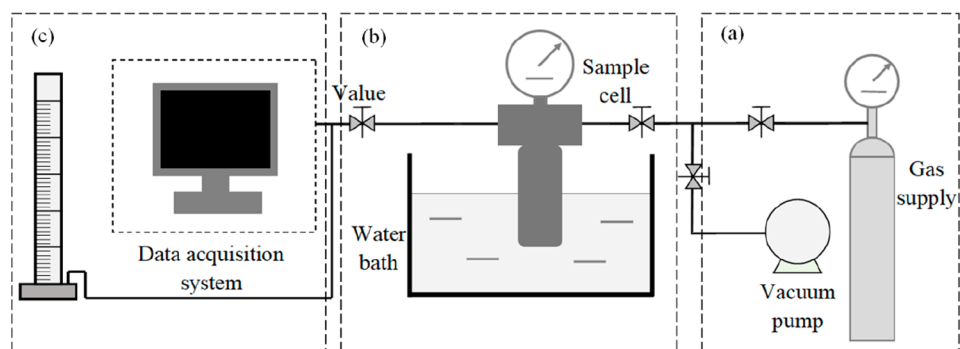
**Table 1. Basic Parameters of Coal Sample**

Test item	Mad	Ad	Vdaf	Fcad	<i>a</i>	<i>b</i>	<i>f</i>
Test result	2.95%	14.62%	5.79%	76.64%	40.4	1.3	0.96

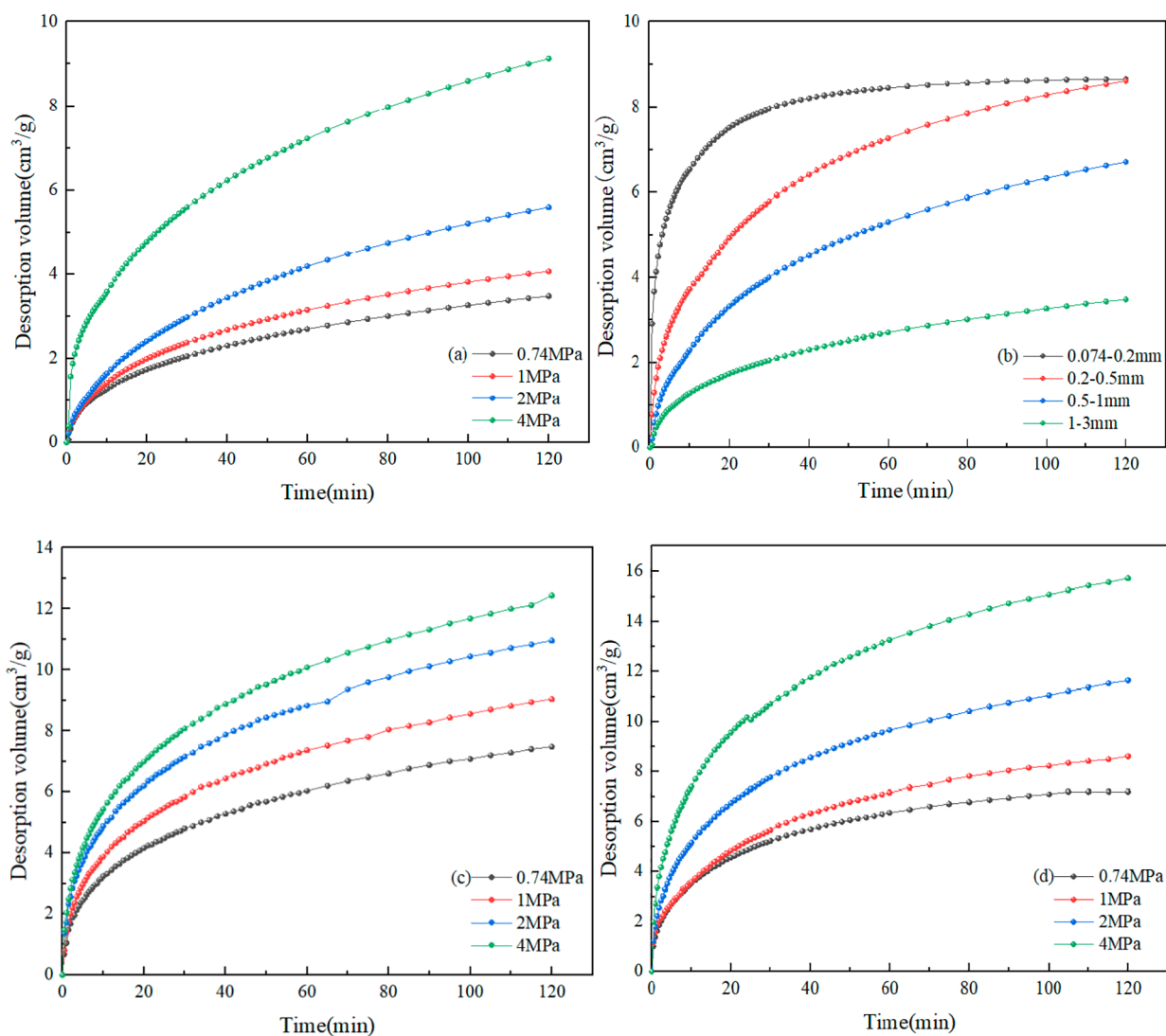
The experimental system includes a degassing and gas supply system, a adsorption desorption system, gas collection, and a data acquisition system and is depicted in Figure 1. First, the air tightness of the experimental system was checked, and the experiment was carried out after it met the requirements. A prepared coal sample was vacuumed in the water bath for 12 h, and then the water bath was filled with gas at a certain initial gas equilibrium pressures at 30 °C to maintain adsorption equilibrium for 12 h. Subsequently, the vent valve of the sample cell was closed, and the desorption valve of the sample cell was opened. Subsequently, the valve of the coal sample tank was quickly opened, and the free gas inside was released. Finally, the valve of the coal sample tank was connected to the measuring cylinder through a hose. The amount of coal sample gas desorption was recorded through the drainage and gas collection method. Under such condition, the gas desorption volume of the coal sample under different initial gas equilibrium pressures within 120 min was measured, and a gas desorption curve of the coal sample was drawn. By using the same method, the gas desorption volume of coal samples with different particle sizes and different metamorphic degrees were measured. The change trends of the cumulative desorption volume of coal samples over time are presented in Figure 2.

## 3. MODEL AND SOLUTION OF DIFFUSION DRIVEN BY THE PRESSURE GRADIENT OF FREE GAS

**3.1. Model of Diffusion Driven by the Pressure Gradient of Free Gas.** Gas diffusion in coal particles conforms to the classical Fick's law of diffusion, based on which many scholars carried out in-depth research and proposed different diffusion models. The most widely used one is Crank's single-pore diffusion model (Figure 3) proposed by Oxford Crank in 1956.<sup>9,10</sup> The core idea of the single-pore diffusion model is to assume that coal particles are homogeneous isotropic media and



**Figure 1.** Schematic diagram of the experimental system: (a) degassing and gas supply system, (b) adsorption-desorption system, and (c) gas collection and data acquisition system.



**Figure 2.** Gas desorption volume of coal samples with different conditions: (a) initial gas equilibrium pressures, (b) particle sizes, (c) Zhaozhuang Coal Mine, and (d) Yongtai Coal Mine.

that matrix pores in coal are single-diameter pores. The polar coordinate form of the flow equation is

$$\frac{\partial c}{\partial t} = D \left( \frac{\partial^2 c}{\partial r^2} + \frac{2}{r} \frac{\partial c}{\partial r} \right) \quad (1)$$

where  $D$  is the diffusion coefficient,  $\text{m}^2/\text{s}$ ;  $r$  is the coal particle diameter,  $\text{m}$ ; and  $t$  is the time,  $\text{s}$ .

The initial and boundary conditions under constant pressure condition are

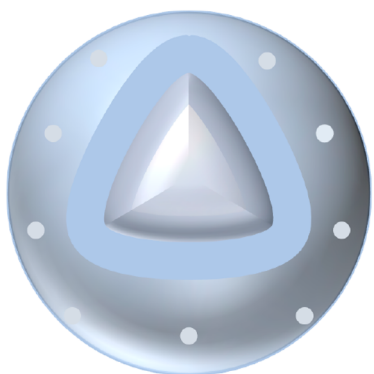


Figure 3. Single-pore diffusion model.

$$\begin{cases} c|_{t=0} = c_0 \\ c|_{r=0} = 0 \\ \left. \frac{\partial c}{\partial r} \right|_{r=0} = 0 \\ c|_{r=r_0} = c_1 \end{cases} \quad (2)$$

where  $c_0$  is the initial concentration of the coal particle, namely, the concentration at the center of the sphere, mol/m<sup>3</sup>;  $c_1$  is the concentration corresponding to the atmospheric pressure, namely, the concentration on the surface of the sphere, mol/m<sup>3</sup>.

Gas in coal particles mainly exists in the form of adsorbed gas and free gas. However, in the diffusion process, adsorbed gas is in a relatively static state and does not participate in the diffusion flow. The gas actually involved in the flow is free gas (including original free gas and free gas generated by desorption; Figure 4). The mass of gas in coal particles can be expressed by the Langmuir Equation as

$$m = \frac{ab\rho_s\rho_a p}{1 + bp} + \frac{\phi T_0 \rho_s}{p_a T} p \quad (3)$$

where  $a$  and  $b$  are Langmuir adsorption constants, cm<sup>3</sup>/g and 1/MPa, respectively,  $\rho_s$  is the apparent density of coal, g/cm<sup>3</sup>,  $\rho_a$  is the density of gas in the standard case, taken as  $7.17 \times 10^{-14}$  g/cm<sup>3</sup>,  $\phi$  is porosity,  $T_0$  is the temperature in the standard case, taken as 273.15 K,  $T$  is the experimental temperature, K,  $p_a$  refers to gas pressure in the standard case, taken as 0.101 MPa, and  $p$  represents the experimental pressure, K.

Diffusion of gas is essentially driven by the pressure gradient of free gas, and the flow velocity is proportional to the pressure gradient but not directly related to the gas content. Qin et al. derived the flow equation of diffusion driven by the density gradient of free gas:<sup>31,32</sup>

$$J = -D \text{grad } \rho_g \quad (4)$$

where  $J$  is the gas mass passing through unit coal particle per unit time, g/(cm<sup>2</sup>·s) and  $\rho_g$  refers to free gas density in coal particles, g/cm<sup>3</sup>.

According to the ideal gas equation, the calculation formula for the density of free gas  $\rho_g$  is expressed as

$$\rho_g = \frac{pM}{RT} \quad (5)$$

where  $M$  is the molecular mass of gas, taken as 16 g/mol;  $R$  represents the universal gas constant, taken as 8.314 J/mol·K.

Taking any control body, according to the conservation of mass, it can be obtained:

$$\frac{\partial m}{\partial t} \left( \frac{4}{3} \pi (r + dr)^3 - \frac{4}{3} \pi r^3 \right) = \frac{\partial (J 4 \pi r^2)}{\partial r} dr \quad (6)$$

where  $m$  refers to the gas content in the control body with thickness  $dr$ ; and  $r$  is the radius of control body, m.

The gas desorption diffusion equation driven by the free gas pressure gradient can be derived by associating eqs 3–6:

$$\frac{\partial}{\partial t} \left( \frac{ab\rho_a p}{1 + bp} + \frac{\phi T_0}{p_a T} p \right) = D \frac{T_0}{p_a T} \left( \frac{\partial^2 p}{\partial r^2} + \frac{2}{r} \frac{\partial p}{\partial r} \right) \quad (7)$$

The corresponding initial and boundary conditions of gas diffusion are

$$\begin{cases} p|_{r=0} = p_0 \\ \left. \frac{\partial p}{\partial r} \right|_{r=0} = 0 \\ p|_{r=r_0} = p_a \end{cases} \quad (8)$$

### 3.2. Numerical Simulation Optimization Method.

**3.2.1. Solving Principle.** Due to the complexity of eq 7, it is difficult to directly obtain an analytical solution, and the solving process is relatively complicated. COMSOL Multiphysics, which is simulation software for simulating real physical fields by solving partial differential equations, has been widely used for

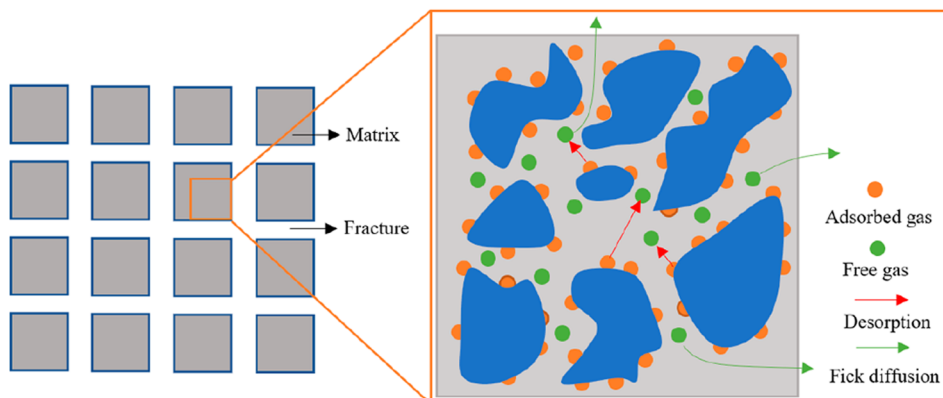
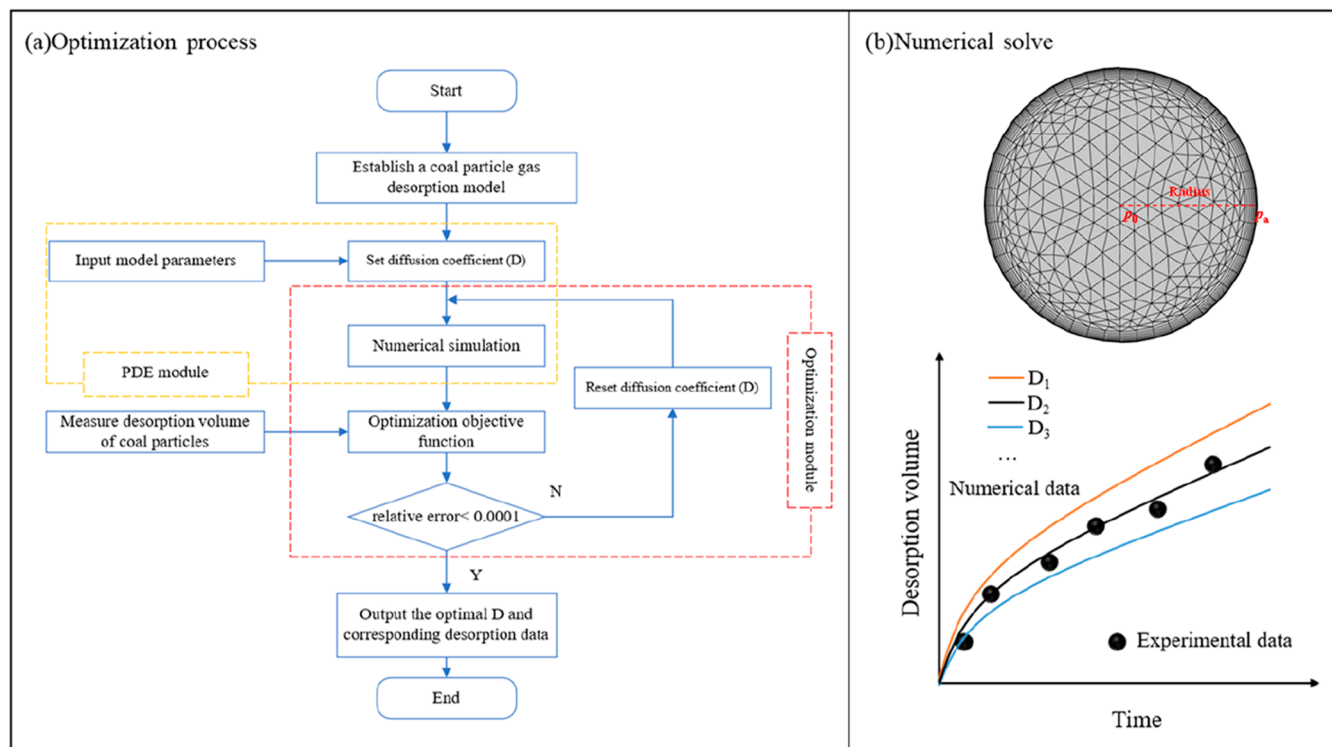


Figure 4. Theoretical model for gas flow in a coal matrix

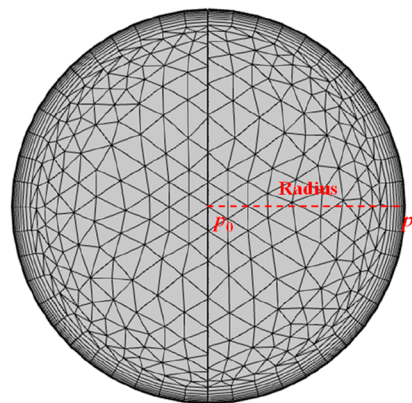


**Figure 5.** Schematic diagram of the optimization module in COMSOL Multiphysics for solving the diffusion coefficient: (a) optimization process and (b) numerical solve.

solving multiple physical field coupling problems. As a universal interface for analyzing optimal solution, its optimization module can be used in combination with any of its other modules and is widely applied to parameter calculation, shape optimization, and parameter estimation. The gradient analysis solvers in the optimization module mainly include the Levenberg–Marquardt solver, the interior point optimizer, the moving asymptote solver, and the sparse nonlinear optimizer, among which the Levenberg–Marquardt solver is the most extensively used.<sup>30</sup>

The optimization is to find the control variable that minimizes the predicted output or objective function under the given constraint conditions. During this process, numerical simulation is conducted mainly through the establishment of mathematical models and input of model parameters. These are continuously optimized and compared until the sum of squares of the difference between experimental data and model output data is minimized, and then the corresponding model parameters are output (Figure 5a). The process of solving the diffusion coefficient by numerical simulation can be considered as a process of constantly seeking the optimal solution of the model parameter value  $D$  under the constraints of the experimentally obtained desorption rate and time by taking  $\lambda = M_t/M_\infty$  as the predicted output and  $t$  as the dependent variable, until the sum of squares of the difference between the simulation results and the experimental data is minimized (Figure 5b).

**3.2.2. Solving Process.** To simplify the solution process, a three-dimensional axisymmetric model whose diameter is the average diameter of the coal particles was established (Figure 6). It is assumed that the gas pressure on the surface of the sphere is atmospheric pressure  $p_a$  and the internal initial gas equilibrium pressure is  $p_0$ . The general partial differential formula module and the Levenberg–Marquardt optimization solver module in COMSOL Multiphysics were selected for simulating the



**Figure 6.** Three-dimensional axisymmetric numerical model.

solution. The parameters required for the simulation process are listed in Table 2.<sup>19,30</sup>

Since the simulation process mainly predicts the relationship between the output desorption rate  $\lambda$  and the dependent variable time  $t$ , while the model solves the relationship between the pressure  $p$  and the time  $t$ , conversion is required as eq 9:

$$\lambda = \frac{M_t}{M_\infty} = \frac{\int_S c dv - \int_S c_a dv}{\int_S c_0 dv - \int_S c_a dv} \quad (9)$$

where  $c$  is the gas concentration under the pressure  $p$ , mol/m<sup>3</sup>,  $c = \frac{abp}{1 + bp} \frac{\rho_a}{V_M}$ ;  $c_0$  is the limit gas concentration under the pressure  $p$ , mol/m<sup>3</sup>;  $c_a$  is the gas concentration under atmospheric pressure  $p_a$ , mol/m<sup>3</sup>; and  $\rho_a$  is the apparent density of coal, kg/m<sup>3</sup>.

**Table 2. Parameters Required for the Simulation Process**

Parameter	Description	Value
$\phi$	Porosity	0.02
$p_0$	Initial gas pressure	0.74 MPa
$p_a$	Atmosphere	0.1 MPa
$r_0$	Average size of coal particles	2 mm
$a$	Langmuir adsorption constant	40.4 m <sup>3</sup> /t
$b$	Langmuir adsorption constant	1.3 MPa <sup>-1</sup>
$M$	Molecular mass of gas	16 g/mol
$R$	Universal gas constant	8.314 J/mol/K
$T$	Experimental temperature	303.15 K
$\rho_a$	Density of coal	1.25 kg/m <sup>3</sup>
$D$	Initial diffusion coefficient	$1.0 \times 10^{-11}$ m <sup>2</sup> /s

During simulation, the geometric distribution of the gas concentration  $c$  along any reference line from the center to the surface of the sphere was mainly observed. The gas emitted from the surface boundary of the sphere was monitored with the aid of the surface pointer tool, based on which the variation law of the desorption rate  $\lambda$  with the time  $t$  was obtained. With the diffusion coefficient  $D$  set as the global variable, the initial value was selected according to the value solved by the experimental method (for example,  $1.0 \times 10^{-11}$  was selected as the initial value under 0.74 MPa), and  $1.0 \times 10^{-9}$  and  $1.0 \times 10^{-11}$  were set as the upper and lower bounds of the parameter. To ensure accuracy of the solution, the optimization tolerance was set as  $1.0 \times 10^{-12}$ , and the maximum number of calculations for the model was set as 1000 times. Finally, the values of diffusion coefficient  $D$  under different pressures were calculated with the aid of the parametric scanning tool. The diffusion coefficients determined by the numerical simulation optimization method under different initial gas pressures are given in Table 3.

Under the initial equilibrium gas pressure of 0.74 MPa, the variation of the gas concentration with position and time is displayed in Figure 7. The cloud images can directly reveal the variations of concentrations at different internal locations over time, providing certain support for theoretical analysis and experimental research. Since the law of gas desorption and diffusion is not the focus of this study and many explanations about it have been put forward, it is only presented here without detailed explanation.

**3.3. Results.** The Fréchet distance, proposed by French mathematician Maurice René Fréchet in 1906, is a description method based on spatial path similarity (Figure 8). Its main advantage lies in considering the spatial distance factor of discrete sampling point, which is suitable for spatial path similarity evaluation.<sup>33,34</sup>

The Fréchet distance is suitable for comparative evaluation calculations between the numerical simulation optimization method and the experimental data. Since the Fréchet distance algorithm is mainly based on the spatial distance between curves, the range of the  $d$  value is positive. Meanwhile, according to the algorithm principle, the smaller the  $d$  value, the higher the similarity degree is. When the value is 0, it means that the two curves are the same.

**Table 3. Diffusion Coefficients Solved by the Numerical Simulation Optimization Method under Different Initial Gas Equilibrium Pressures**

Initial gas equilibrium pressure $p_0$ (MPa)	0.74	1	2	4
Diffusion coefficient $D$ (m <sup>2</sup> /s)	$1.077 \times 10^{-10}$	$1.043 \times 10^{-10}$	$1.062 \times 10^{-10}$	$1.050 \times 10^{-10}$

**3.3.1. Analysis of the Similarity with Different Initial Gas Equilibrium Pressures.** According to the preceding steps, the similarity  $d$  between the curves solved by the numerical simulation optimization method and the experimental curves was obtained by Python and is presented in Table 4, and the comparison results are shown in Figure 9. Under different gas equilibrium pressures, the  $d$  values are all smaller than 0.1 and basically stable at 0.0835, indicating that the simulation curves are highly similar to the experimental curves. The diffusion coefficient obtained boasts high accuracy. Despite a slight difference between the numerical simulation curves and the experimental curves, which is caused by the assumption of constant diffusion coefficient and objective conditions, such deviation can be ignored on the whole.

**3.3.2. Analysis of the Similarity with Different Coal Particle Sizes.** The solution results and comparison effects under different particle sizes are exhibited in Table 5 and Figure 10. The  $d$  values between the numerical simulation optimization curves and the experimental curves are all smaller than 0.2, demonstrating a high similarity between the two curves and remarkable accuracy of the diffusion coefficient solution. The  $d$  value varies under different coal particle sizes. The smaller the particle size is, the larger the  $d$  value and the lower the fitting accuracy are. This phenomenon can be explained as follows: the classical diffusion model has a good fitting effect on the initial stage of diffusion, and the fitting deviation grows with time; however, under a smaller coal particle size, the gas diffusion rate is higher; resultantly, the initial stage (0–10 min) in which the fitting effect is good accounts for a smaller proportion, and the overall fitting deviation is larger. Nevertheless, such a deviation can be ignored from the perspective of research.

**3.3.3. Analysis of the Similarity with Different Metamorphic Degrees of Coal Samples.** The solution results and comparison effects of Yongtai Coal Mine (high metamorphic degree), Dongda Coal Mine (medium metamorphic degree), and Zhaozhuang Coal Mine (low metamorphic degree) under 0.74 MPa are displayed in Table 6 and Figure 11. The  $d$  values between the numerical simulation optimization curves and the original experimental curves are all smaller 0.1, and the variation law of the other two are basically similar to that of Dongda Coal Mine, suggesting that the numerical simulation optimization curves are highly similar to the original experimental curves and the diffusion coefficient solved is pretty accurate. The  $d$  values of coal samples with different metamorphic degrees are different, and the coal samples with high and low metamorphic degrees have smaller  $d$  values than the one with a medium metamorphic degree, which is a mark of their higher fitting accuracy. The reason is that the metamorphic degree has a V-shaped variation law with the volume of large pores and the specific surface area of small pores,<sup>35</sup> resulting in different proportions of the initial stage of diffusion, which affects the fitting effect.

## 4. DISCUSSION AND PROSPECTS

**4.1. Discussion.** **4.1.1. Influence of Different Initial Gas Equilibrium Pressures on the Diffusion Coefficient.** As the law of gas flow in coal particles is not completely clear yet,

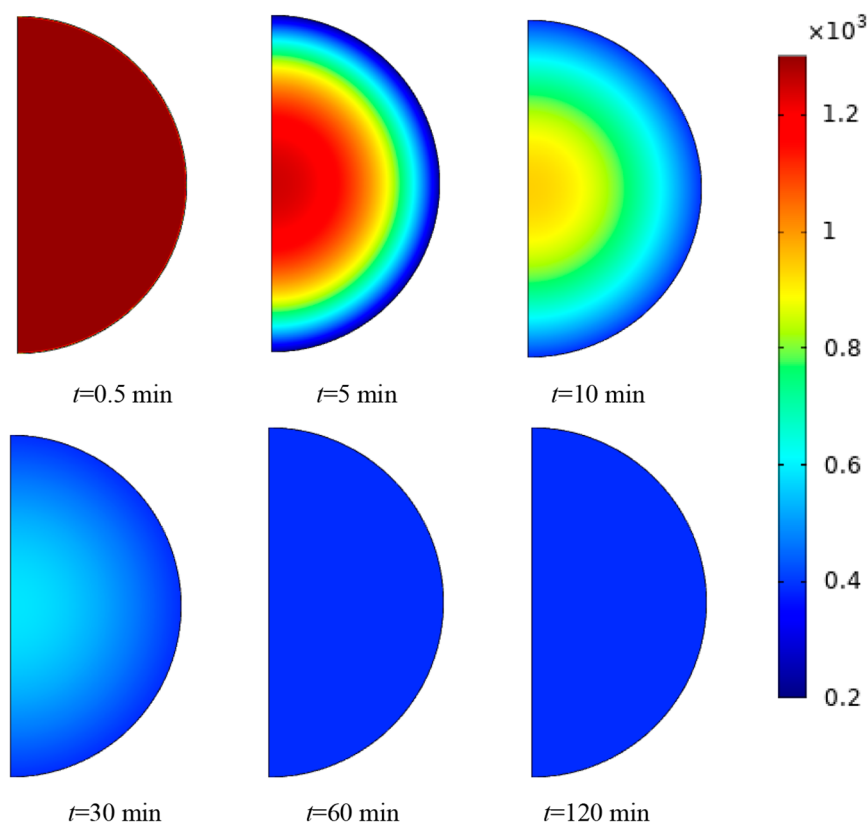


Figure 7. Cloud images of concentration variation in the model at different times.

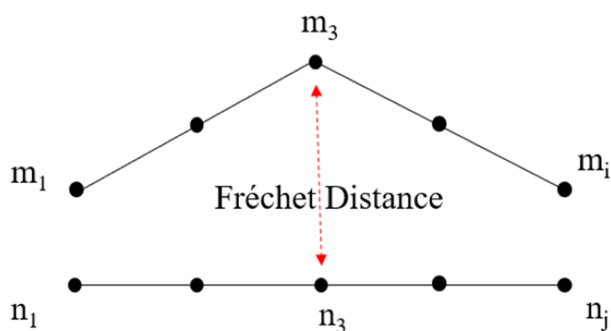


Figure 8. Schematic diagram of the Fréchet distance.

controversy still exists regarding the influence law of gas pressure on diffusion coefficient. Different conclusions were drawn, including positive, negative and irrelevant correlations between the diffusion coefficient and the gas pressure.<sup>36–40</sup> Yang et al. believed that the diffusion coefficient is positively correlated with the gas pressure.<sup>37</sup> Yi et al. held that the diffusion coefficient declines with the decrease of the equilibrium pressure.<sup>19</sup> Wu et al. consider that the diffusion coefficient is unrelated to the gas pressure.<sup>18</sup> In Figure 12 (a), the diffusion coefficient substantially lies near  $1.062 \times 10^{-10}$  m<sup>2</sup>/s. The initial equilibrium gas pressure has an insignificant influence on the diffusion coefficient, demonstrating that the law of gas diffusion

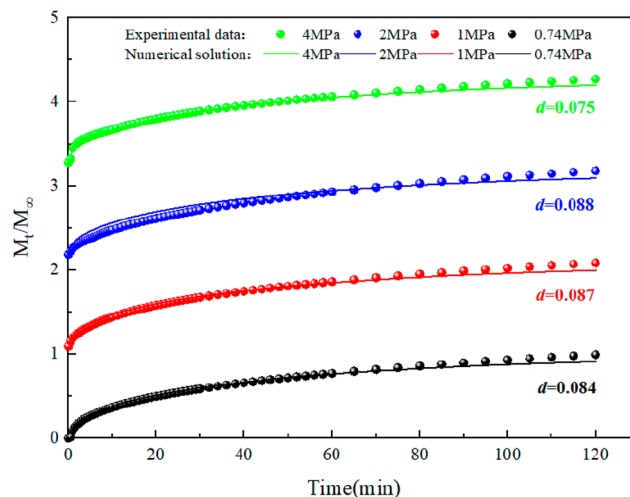


Figure 9. Comparison between numerical simulation curves and experimental curves under different initial gas pressures.

in the micropore and small-pore system accords with Darcy's law rather than the diffusion law. Actually, the model of diffusion driven by the pressure gradient is consistent with the Darcy's model of seepage derived by Hao,<sup>22</sup> and it is equivalent to a form of Darcy's law. Although the physical significances of diffusion

Table 4. Solution Results and Comparison Effects under Different Initial Gas Equilibrium Pressures

Initial gas pressure $p_0$ (MPa)	0.74	1	2	4
Diffusion coefficient $D$ (m <sup>2</sup> /s)	$1.077 \times 10^{-10}$	$1.043 \times 10^{-10}$	$1.062 \times 10^{-10}$	$1.130 \times 10^{-10}$
Similarity $d$	0.084	0.087	0.088	0.075

Table 5. Solution Results and Comparison Effects under Different Coal Particle Sizes

Particle size	0.074–0.2 mm	0.2–0.5 mm	0.5–1 mm	1–3 mm
Diffusion coefficient $D$ ( $\text{m}^2/\text{s}$ )	$3.024 \times 10^{-12}$	$4.456 \times 10^{-12}$	$1.502 \times 10^{-11}$	$1.077 \times 10^{-10}$
Similarity $d$	0.115	0.103	0.085	0.084

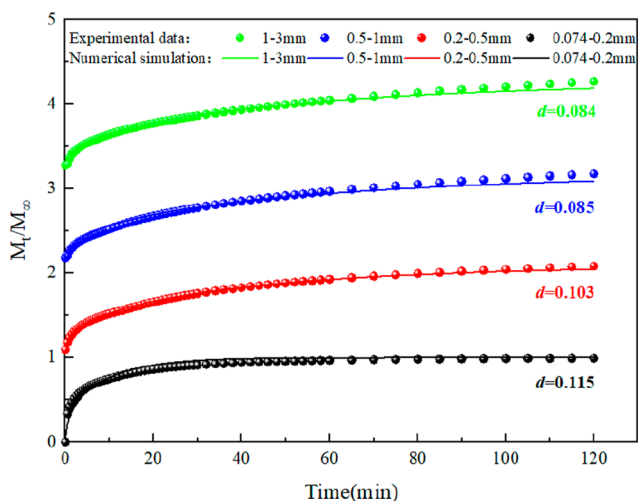


Figure 10. Comparison between numerical simulation curves and experimental curves under different coal particle sizes.

Table 6. Solution Results and Comparison Effects under Different Metamorphic Degrees

Metamorphic degree	Zhaozhuang Coal Mine	Dongda Coal Mine	Yongtai Coal Mine
Diffusion coefficient $D$ ( $\text{m}^2/\text{s}$ )	$1.350 \times 10^{-10}$	$1.077 \times 10^{-10}$	$1.907 \times 10^{-10}$
Similarity $d$	0.053	0.084	0.039

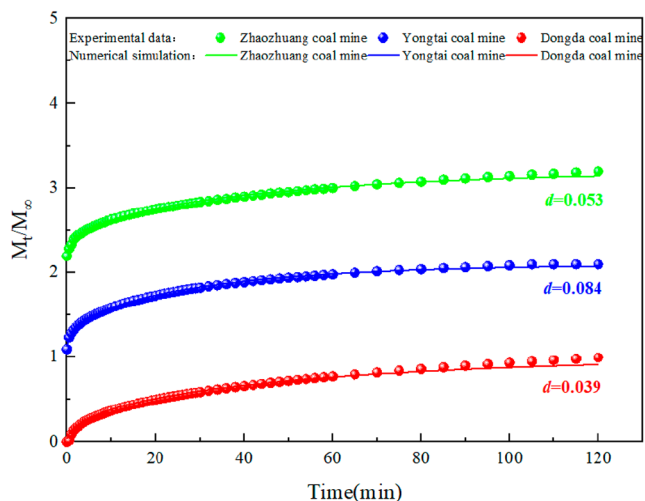


Figure 11. Comparison between numerical simulation curves and experimental curves under different metamorphic degrees.

and seepage behaviors are different, the concentration and pressure of a continuous gas medium can be transformed through the ideal gas state equation, which provides a bridge for the conversion of the diffusion behavior and seepage behavior. As revealed in eq 10, the diffusion coefficient and the permeability can be converted through a certain mathematical relationship, and the mathematical expression of conversion between the diffusion coefficient and the permeability has also

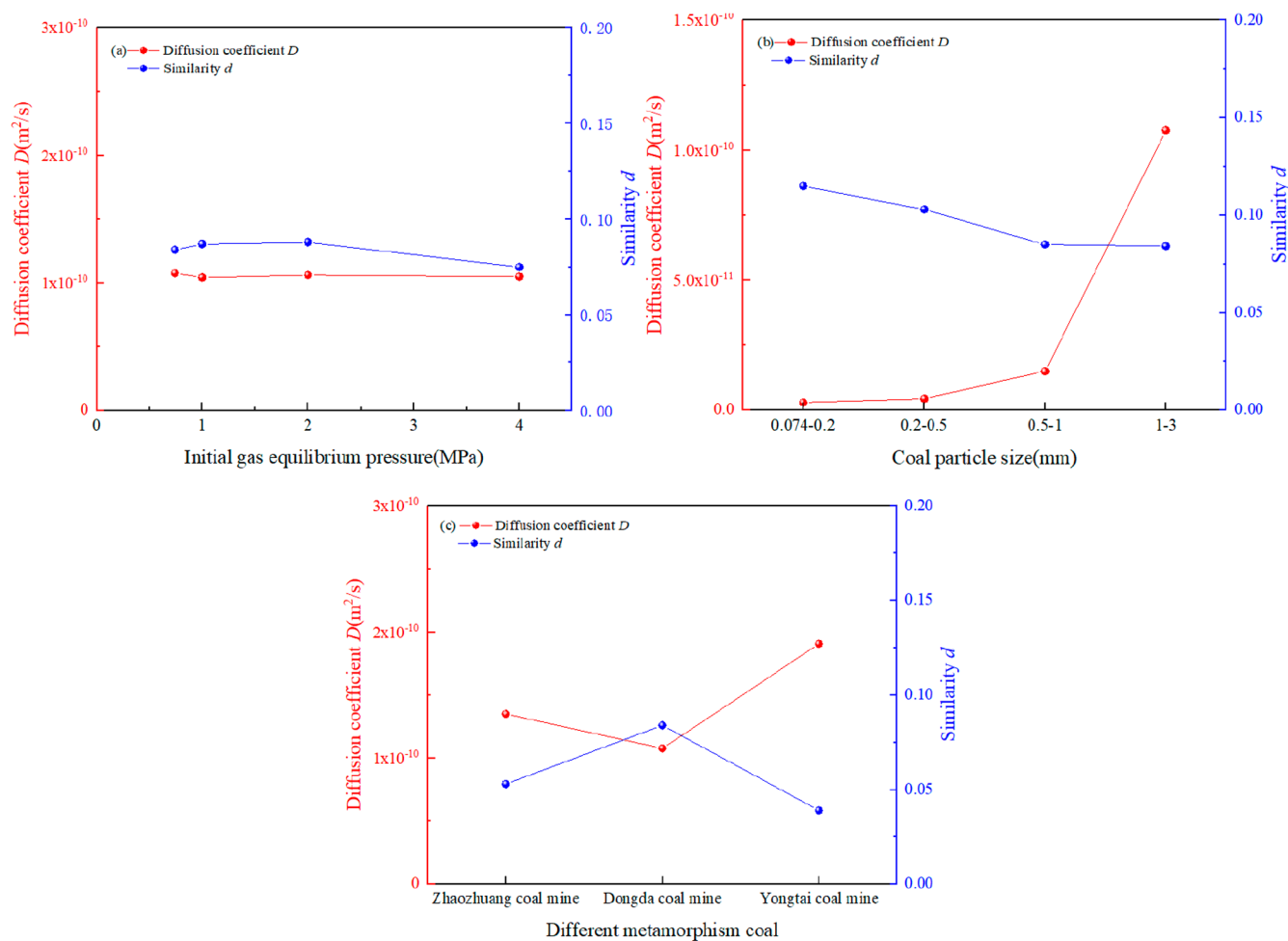
been derived in literature.<sup>41</sup> Therefore, the desorption and diffusion of gas from coal particles can be described by both the diffusion equation and the seepage equation. The difference lies in which is more suitable for experimental conditions and easier to reach conclusions.

$$\begin{cases} \frac{\partial}{\partial t} \left( \frac{ab\rho_a p}{1+bp} + \frac{\phi T_0 \rho_a}{p_0 T} p \right) = D \frac{T_0}{p_0 T} \left( \frac{\partial^2 p}{\partial r^2} + \frac{2}{r} \frac{\partial p}{\partial r} \right) \\ \frac{\partial}{\partial t} \left( \frac{ab\rho_a p}{1+bp} + B'm\rho_a p \right) = \lambda \left( \frac{\partial^2 p}{\partial r^2} + \frac{2}{r} \frac{\partial p}{\partial r} \right) \end{cases} \Rightarrow \begin{cases} B' = \frac{T_0}{p_0 T} \\ \lambda = D \frac{T_0}{p_0 T} = DB' \end{cases} \quad (10)$$

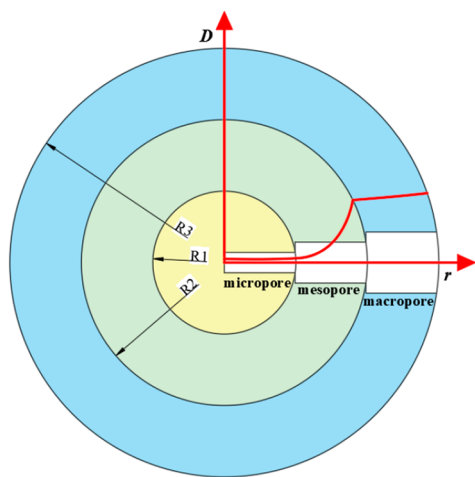
4.1.2. Influence of Different Coal Particle Sizes on the Diffusion Coefficient. Scholars all over the world have conducted plenty of research on the relationship between the coal particle size and the variation law of gas diffusion.<sup>41–48</sup> Through experimental research, Yuan proved that the diffusion coefficient is negatively correlated with the coal sample size.<sup>47</sup> The studies of Yang,<sup>25</sup> Li et al.,<sup>35</sup> and Yang et al.<sup>48</sup> showed that initial diffusion coefficient is positively correlated with particle size. In Figure 12b, the diffusion coefficient declines with the decrease in particle size. The diffusion coefficient of the sample with a particle size of 1–3 mm is 33 times that of 0.074–0.2 mm, showing an obvious scale effect. For small coal particles (in Figure 13, the radius is smaller than R1), large pores and cracks are damaged in the grinding and screening process, and the internal pores left are mostly large pores, manifesting a small diffusion coefficient. In contrast, for large-sized coal particles (in Figure 13, the radius is between R1 and R2), the pore structure is relatively complete; a large number of large cracks and pores contribute to a large diffusion coefficient. When the particle size is larger than the limit particle size (in Figure 13, the radius is larger than R2), the coal particle can be regarded as a whole composed of many particles of the limit size. In this case, the gas diffusion rate of the coal particle remains constant, independent of the particle size. In addition, when the particle size is smaller than 0.2–0.5 mm, the adsorption capacity does not change significantly with the particle size. This is attributed to the fact that when the particle is too small, the expansion in the external specific surface is approximately equivalent to the shrinkage in the pore surface. Macroscopically, the adsorption capacity does not change obviously and the corresponding diffusion coefficient barely varies.

From the above analysis, it can be concluded that the diffusion coefficient is influenced by the coal particle size with two limits, namely, “the upper limit particle size” and “the lower limit particle size”. As illustrated in Figure 14, the diffusion coefficient has a Z-shaped relationship with the coal particle size, that is, when the particle size is larger than the upper limit particle size or smaller than the lower limit particle size, the diffusion coefficient remains unvaried; when it is between the two limits, the diffusion coefficient grows with the increase of the coal



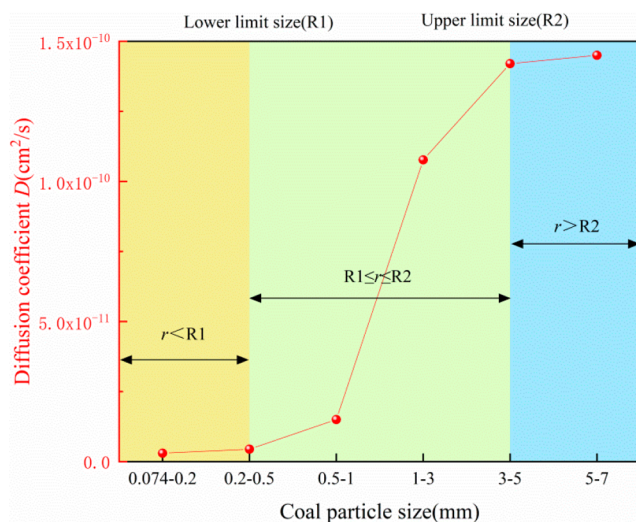


**Figure 12.** Variation law of the diffusion coefficient under different conditions: (a) different initial gas equilibrium pressures, (b) different coal particle sizes, and (c) different metamorphic degrees.



**Figure 13.** Model of pore distribution of coal particles with different sizes.

particle size. Under the pressure of 0.74 MPa, the lower and upper limit particle sizes of samples from the Dongda Coal Mine are 0.2–0.5 mm and 3–5 mm, respectively. Compared with the upper limit particle size, which has been extensively studied, research on the lower limit particle size is still insufficient.



**Figure 14.** Relationship between the diffusion coefficient and the coal particle size of samples from the Dongda Coal Mine.

Hence, a desorption experiment of smaller particle size is required to further improve the system of influence of coal particle size on diffusion coefficient.

**4.1.3. Influence of Coal Samples with Different Metamorphic Degrees on the Diffusion Coefficient.** The diffusion coefficient differs notably with different degrees of metamorphic. The main reason lies in the diversity of coal particle properties.<sup>47,49–51</sup> Yuan<sup>47</sup> and Zhang et al.<sup>50</sup> proved through experimental studies that the diffusion coefficient and the metamorphic degree presented a V-shaped relationship, that is, the diffusion coefficient of coal samples with high and low metamorphic degrees were higher than those of coal samples with a medium metamorphic degree. In Figure 12c, the diffusion coefficient and the metamorphic degree have an approximately V-shaped relationship, which is consistent with previous research results. From the microscopic point of view, the metamorphic degree largely affects the size, specific surface area, volume and other parameters of coal pores, especially the number of small and micropores, which directly determines the diffusion coefficient and diffusion law of coal particle. Although coal with a low metamorphic degree has more mesopores than that with a medium metamorphic degree, its open pores account for a relatively larger proportion, which leads to a certain increase in its diffusion coefficient.

**4.2. Prospects.** (1) During the whole experiment, the numerical simulation solution curves of coal samples with different initial gas equilibrium pressures, particle sizes and metamorphic degrees are basically consistent with the experimental data. In spite of some deviations, they can be ignored from the overall time scale. The reason for the deviations is that the diffusion coefficient is variable under certain experimental conditions, but the constant diffusion coefficient under allowable accuracy is recognized by the majority of scholars. Consequently, diffusion models can be highly consistent with experimental data only under specific experimental conditions, yet the deviations can hardly be eliminated. Meanwhile, due to the assumption of a constant diffusion coefficient, the single-pore diffusion model has a large deviation in describing desorption curve over a long period of time. For this reason, the time-dependent diffusion coefficient model and the fractal diffusion coefficient model have been proposed,<sup>52–55</sup> yet the principle and physical implication of diffusion coefficient variation have not been explained scientifically. Therefore, the diffusion coefficient solution and the diffusion law need to be further studied.

(2) The most direct way of seeking a solution is to obtain an analytical solution through equations. However, the analytical solution requires making plenty of assumptions and dealing with complicated situations, and has a relatively limited scope of application. The numerical simulation solution have advantages of high solving speed and simple operation, and its operation results have been proven to better reflect diffusion behavior of gas throughout the entire experimental process. Therefore, compared to the analytical solution method, the numerical simulation solution method is more applicable, faster, and more convenient, providing ideas for similar solution.

(3) Gas migration is a complex continuous process. Limited by experimental equipment, it cannot be subdivided by experiments and traditional theoretical methods. For instance, the adsorption state and free state of gas cannot be distinguished. In comparison, the numerical simulation method can reflect different stages of gas migration, realize the decoupling of gas migration process, and obtain diffusion coefficient or effective diffusion coefficient, which will be the focus of future research.

## 5. CONCLUSIONS

The diffusion coefficient is one of the key parameters for analyzing the diffusion law and is normally derived from desorption curves. The speed of solving the diffusion coefficient can be improved, and the economic cost can be reduced by applying the numerical simulation optimization method. In this paper, the numerical simulation optimization method was employed to solve the diffusion coefficient of coal samples with different initial gas equilibrium pressures, particle sizes, and metamorphic degrees.

The Fréchet distance similarity evaluation theory was used to compare and analyze the numerical simulation optimization results and the original experimental curves, and the variation law of the diffusion coefficient was explored. The main conclusions are drawn as follows.

(1) The numerical simulation optimization method can not only solve the diffusion coefficient, but also reveal the law of diffusion concentration with time.

(2) The similarity between the numerical simulation optimization result and the experimental data under different conditions was compared and analyzed based on the Fréchet distance similarity evaluation theory. The result show that the  $d$  values between the solution results and the experimental data under different conditions are all smaller than 0.2, which proves the effectiveness and accuracy of the numerical simulation optimization method.

(3) The diffusion coefficient of gas from coal particle is unrelated to the initial gas equilibrium pressure. It has a Z-shaped relationship with the coal particle size, that is, when the particle size is larger than the upper limit particle size or smaller than the lower limit particle size, the diffusion coefficient remains unvaried; when the particle size is between the two limits, it grows with the increase of the coal particle size. It presents a V-shaped relationship with the metamorphic degree, that is, the diffusion coefficient of coal samples with high and low metamorphic degrees are higher than those of coal samples with a medium metamorphic degree.

(4) The numerical simulation optimization method can also provide solution for solving the dynamic diffusion coefficient and decoupling the gas migration process.

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## Notes

The authors declare no competing financial interest.

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