Measurement and Modeling of Spontaneous Capillary Imbibition in Coal

Jiwei Yue, Zhaofeng Wang,* Yongxin Sun, Jinsheing Chen, Fenghua An, Hongqing Yu, and Xuechen Li

Abstract: Coal is a typical dual-porosity medium. The implementation process of water invasion technology in coal is actually a process of spontaneous imbibition of external water. To obtain a model of spontaneous capillary imbibition in coal, the spontaneous imbibition of water in coal samples with different production loads is conducted experimentally. Due to the coal particle deformation and the cohesive forces, the porosity and maximum diameter decrease gradually with increasing pressing loads. Due to the filling effects and occupying effects, the proper particle grading can reduce the porosity and tortuosity. The Comiti model can be used to describe the tortuosity. The tortuosity increases with decreasing porosity. The smaller the porosity, the smoother the surface of the coal sample. The contact angle is negatively correlated with the surface roughness. The fractal dimension decreases with increasing pressing load. The difference in the pore characteristics between particles is the main reason for the difference in the fractal dimension. The proposed model of spontaneous capillary imbibition in coal is consistent with the experimental data. The implications of this study are important for understanding the law of spontaneous imbibition in coal and the displacement of gas by spontaneous capillary imbibition in coal, which is important for optimizing the parameters of coal seam water injection.

1. Introduction

Imbibition is a process of spontaneous absorption of a wetting fluid in a porous medium. There are many examples of capillary imbibition in nature and engineering, such as oil recovery, ink-jet printing, water imbibition of plant stems and leaves, water adsorption of building foundation, manufacturing, chalk ink interactions, and microfluidic technology. This spontaneous imbibition phenomenon can be interpreted as wetting fluids penetrating porous materials under the action of capillary pressure. Coal is a typical dual-porosity medium that contains pores and fractures. When the wetting fluid penetrates the coal matrix, such as during hydraulic fracturing, coal seam injection, hydraulic cutting, and hydraulic slotting, the amount of water in the coal matrix increases due to the capillary pressure. After the water penetrates the coal matrix, the coal matrix becomes moist. This phenomenon is also called imbibition. Wang et al. used an imbibition device with the functions of isobaric imbibition and isobaric desorption to quantitatively study the amount of imbibition gas with different gas pressures and water contents, and the experimental results revealed that adding water is more conducive to imbibition. Wang and Jiang found that the imbibition process of static water could enhance coalbed methane recovery. High-energy sites can be easily occupied by water molecules under the...
action of oxygen-containing functional groups in the coal matrix and hydrogen bonds in water molecules. To summarize, the spontaneous imbibition of external water can lead to four effects, namely, accelerating methane desorption, decreasing the coal seam gas content, eliminating the danger of coal and gas outbursts, and increasing the production of coalbed methane (CBM). The gas content in the coal is closely related to the wetting range of the coal body. The wetting range of the coal body is also related to the layout parameters of the water injection hole. However, the wetting range of the coal body is actually the imbibition distance. Therefore, measuring the imbibition distance in coal and modeling the relationship between the imbibition distance and imbibition time in coal are imperative.

To describe the spontaneous imbibition process of porous media, since 1918, many experts and scholars have studied the imbibition characteristics and proposed theoretical models of porous media. The classic theoretical models of imbibition include the pure inertial force model, Bosanquet model, Lucas—Washburn model, gravity factor model, Terzaghi model, and Handy model. The pure inertial force model, Bosanquet model, Lucas—Washburn model, gravity factor model, and Terzaghi model consider the imbibition height and imbibition time. The handy model considers the model imbibition volume and imbibition time. Quere derived the pure inertial force model that ignores the gravity, viscous force, and external pressure. Bosanquet believed that the gravity of water was small and could be ignored during the initial stage of capillary imbibition and proposed an imbibition model that ignored the gravity factor. Lucas and Washburn proposed an imbibition model that ignored the gravity and inertia effects of the fluid itself, called the Lucas—Washburn model. Fries and Dreyer derived the imbibition model considering the gravity factor. Terzaghi applied Darcy’s law to unsaturated fluid flow and developed a cylindrical soil spontaneous imbibition model. Handy ignored the gas-phase pressure gradient at the leading edge of the water phase, assumed that the process of water self-adsorption is a piston displacement process, and established a spontaneous imbibition model for the core. Mason et al. proposed a piston-shaped spontaneous imbibition model and explained the behavior of the spontaneous imbibition of the Berea sandstone. Although great progress has been made in the modeling of spontaneous imbibition of porous materials, some problems still remain. Regarding the description of the spontaneous imbibition models proposed above, the pores of the considered porous media are usually assumed to have a uniform diameter and form a vertical capillary bundle. Porous media have anisotropic properties, such as permeability, elastic modulus, yield strength, thermal expansion coefficient, and thermal conductivity. A model assuming a uniform pore diameter and a vertical capillary tube bundle cannot properly describe the process of spontaneous capillary imbibition. A coal matrix has a dual-porosity structure. Some studies have reported that remolded coal also has a dual-porosity structure. The pore sizes in remolded coal are not the same, and some of them are several orders of magnitude different. In addition, the capillaries of remolded coal are actually tortuous. Clearly, assuming that pores have a uniform diameter and form a vertical capillary bundle is also inappropriate. As a consequence, the theoretical models proposed above may not be suitable for studying spontaneous imbibition in coal.

Fractal theory was given by Mandelbrot in the 1970s and can be used to describe the flow characteristics of porous media. The pore size and pore surface distribution of porous media can be described by the law of fractal scaling. The irregular degree is an important feature of fractal objects, and it is independent of scale. A part of a fractal object and the whole object have self-similarity. The dimension of fractal geometry is called the fractal dimension, and its value is not less than its topological dimension. Many studies have shown that porous materials have fractal characteristics. The fractal theory has been used in a number of applications, such as in petroleum engineering, fuel-cell research, and coal industries. Cai et al. investigated the capillary rise of water in a single curved capillary based on the fractal geometry and obtained a model of imbibition height and imbibition weight with respect to time. Tortuosity indicates the bending degree of a curved capillary. Cai and Yu also used the fractal geometry to analyze the effect of tortuosity on spontaneous capillary imbibition and found that the imbibition time exponent is less than 0.5. Fractal characteristics have a certain influence on the permeability of porous media. Remolded coal can be characterized as a fractal porous medium. The fractal theory can also be used in the study of spontaneous capillary imbibition of porous media. In view of the above work, this paper studied capillary spontaneous imbibition in remolded coal from the perspective of the pore fractal dimension. The main purpose of this study is twofold. The first purpose of this work is to experimentally measure the spontaneous capillary imbibition height of remolded coal samples with different pressing loads. The second purpose of this work is to establish a modified capillary imbibition model for describing the imbibition height of remolded coal samples.

The structure of this article is as follows. In Section 2, a model of the spontaneous capillary imbibition in remolded coal is established. In Section 3, the experimental processes of remolding coal samples and the corresponding experimental procedures are described. In Section 4, the imbibition height, porosity, tortuosity, contact angle, fractal dimension, and maximum capillary diameter are analyzed, and the accuracy of the model of spontaneous capillary imbibition is proven. In Section 5, the main conclusions are drawn.

2. MODELING OF SPONTANEOUS CAPILLARY IMBIBITION IN REMOLDED COAL

The cumulative number of pores with a pore diameter greater than or equal to the diameter $d$ in a porous medium can be described by eq 1.

$$N(\geq d) = \left( \frac{d_{\text{max}}}{d} \right)^{D_{p}}$$

where $D_p$ is the pore fractal dimension, $d_{\text{max}}$ is the maximum pore diameter, $d$ is the pore diameter, and $N$ is the number of pores.

The cross-sectional area of all of the pores and the whole cross-sectional area can be described by eqs 2 and 3, respectively.

$$S_p = \frac{\pi d_{\text{max}}^2}{4} \frac{D_p}{2 - D_p} (1 - \phi)$$

(2)

$$S_s = \frac{S_p}{\phi} = \frac{\pi d_{\text{max}}^2}{4} \frac{D_p}{2 - D_p} \frac{1 - \phi}{\phi}$$

(3)

where $S_p$ is the cross-sectional area of all of the pores, $\phi$ is the porosity of the remolded coal, and $S_s$ is the whole cross-sectional area.
The cumulative number of pores in a certain cross-sectional area can be calculated by eq 4.

\[ N(z, d) = \frac{S_{i}}{S_{t}} N = \frac{S_{i}}{S_{t}} \left( \frac{d_{\text{max}}^{2} - D_{p}}{d} \right)^{D_{p}} \]  

(4)

where \( S_{i} \) is the cross-sectional area of a certain area.

\[ N(z, d) = \frac{4S_{i}}{\pi d_{\text{max}}^{2} - D_{p}} \left( \frac{2 - D_{p}}{1 - \phi} \right) d^{D_{p}} \]  

(5)

The tortuosity can be described by eq 6. The tortuosity has a greater impact on the movement of liquids in porous materials. The capillary rise of a single curved capillary is shown in Figure 1.

\[ \tau = \frac{L_{c}}{L_{v}} \]  

(6)

where \( \tau \) is the tortuosity, \( L_{c} \) is the straight tube length, and \( L_{v} \) is the bend capillary length.

![Figure 1. Schematic diagram of the imbibition liquid of a single curved capillary.](image)

The imbibition liquid of bottom water in a porous medium is similar to an incompressible Newtonian fluid flowing upward. The flow behavior can be described by Hagen–Poiseuille’s law. The flow in a capillary can be described by eq 7.

\[ q(d) = \frac{\pi}{128} \frac{d^{4}}{\mu L_{v}} \left( \frac{4\sigma \cos \theta}{d} - \rho g L_{v} \right) \]  

(7)

where \( q(d) \) is the flow rate in the capillary, \( g \) is the acceleration due to gravity, \( \mu \) is the liquid viscosity, \( \sigma \) is the surface tension of the liquid, and \( \rho \) is the density of water.

The total flow rate in a certain cross-sectional area can be calculated by eq 8.

\[ Q = -\int_{l_{\text{min}}}^{d_{\text{max}}} q(d) dN_{t} = \frac{\sigma \cos \theta}{8\mu L_{v}} \left( \frac{2 - D_{p}}{3 - D_{p}} \right) S_{i} \phi d_{\text{max}} \left( 1 - \xi^{3 - D_{p}} \right) \]  

\[ \left\{ \begin{array}{l} \frac{S_{i} \rho g}{32\pi r^{2}} \left( \frac{2 - D_{p}}{4 - D_{p}} \right) d_{\text{max}}^{2} \\ 3 - D_{p} 1 - \phi \end{array} \right\} \]  

\[ dN_{t} = \frac{4S_{i}}{\pi d_{\text{max}}^{2} - D_{p}} \left( \frac{2 - D_{p}}{1 - \phi} \right) d^{D_{p} + 1} dd \]  

(8)

where \( \xi = \frac{L_{c}}{L_{v}} \). Generally, \( \xi \) is less than 10\(^{-2} \). Therefore, eq 8 can be simplified to eq 9.

\[ Q = \frac{\sigma \cos \theta}{8\mu L_{v}} \left( \frac{2 - D_{p}}{3 - D_{p}} \right) S_{i} \phi d_{\text{max}} \left( 1 - \phi^{3 - D_{p}} \right) \]  

\[ \left\{ \begin{array}{l} \frac{S_{i} \rho g}{32\pi r^{2}} \left( \frac{2 - D_{p}}{4 - D_{p}} \right) d_{\text{max}}^{2} \\ 3 - D_{p} 1 - \phi \end{array} \right\} \]  

(9)

Thus, the imbibition rate of a curved capillary can be described by eq 10.

\[ v_{i} = \frac{Q}{S_{i}} = \frac{\sigma \cos \theta}{8\mu r^{2} L_{v}} \left( \frac{2 - D_{p}}{3 - D_{p}} \right) \phi d_{\text{max}} \left( 1 - \phi \right) \]  

(10)

The imbibition rate of a straight capillary can be calculated by eq 11.

\[ v_{i} = \frac{\phi L_{v}}{t} = \frac{a}{b} - b \]  

(11)

The function of the imbibition rate of a curved capillary and the imbibition rate of a straight capillary can be described by eq 12.

\[ v_{i} = \frac{\phi L_{v}}{t} = \frac{a}{\tau} \]  

(12)

Substitution of eq 10 into eq 12 leads to eq 13.

\[ v_{i} = \frac{\sigma \cos \theta}{8\mu r^{2} L_{v}} \left( \frac{2 - D_{p}}{3 - D_{p}} \right) \phi d_{\text{max}} \left( 1 - \phi \right) \]  

(13)

Combining eqs 11 and 13 and integrating eq 11, the function of between imbibition time and imbibition height is as follows.

Equation 14 is an implicit analytical solution.

\[ t = \frac{a}{b^{2}} \ln \left( 1 - \frac{b}{a} \right) - \frac{L_{v}}{b} \]  

\[ \left\{ \begin{array}{l} a = \frac{\sigma \cos \theta}{8\mu r^{2}} \left( \frac{2 - D_{p}}{3 - D_{p}} \right) \phi d_{\text{max}} \\ b = \frac{\rho g}{32\pi r^{2}} \left( \frac{2 - D_{p}}{4 - D_{p}} \right) d_{\text{max}}^{2} \\ 3 - D_{p} 1 - \phi \end{array} \right\} \]  

(14)

The expression of the exact analytical solution can be described by eq 15.

\[ h = \frac{a}{b} \left[ 1 + W \left( -e^{-1} \right) \right] \]  

\[ \left\{ \begin{array}{l} W(x) \approx \frac{2ex - 10.7036 + 7.56859\sqrt{x^{2} + 2ex}}{12.7036 + 5.13501\sqrt{2} + 2ex} \end{array} \right\} \]  

\[ \leq x \leq 0 \]  

(15)

where \( W(x) \) denotes the Lambert W function, which is shown in Figure 2.

3. EXPERIMENTS

3.1. Preparation of Remolded Coal Samples. The experimental coal samples were selected from the No. 3 mining seam of the Yonghong Coal Mine, which is located in Shanxi Province, China. The experimental coal samples are soft coals. The reasons for choosing remolded coal as the research object are as follows. Coal and gas outbursts occur easily in soft coal
seams. However, coal and gas outbursts do not occur easily in hard coal seams. In addition, the coal sample from this soft coal seam has a certain shape.

The anthracite samples are first crushed by a crusher. After crushing, the coal samples with diameters less than 60 mesh and between 60 mesh and 35 mesh are screened out. The remolded coal samples dimensions used in this experiment are 5 cm in diameter and 16 cm in height and 5 cm in diameter and 3 mm in height, and the remolded coal samples are made by a custom mold and a hydraulic universal testing machine (WES-1000B) (HUTM). The remolded coal samples with a diameter of 50 mm and a height of 160 mm are used to test the imbibition height, and the remolded coal samples with a diameter of 50 mm and a height of 3 mm are used to test the contact angle. In this paper, two particle size proportions of remolded coal samples are considered. One set of the remolded coal samples is made by coal particles with diameters less than 60 mesh, and the other is made by coal particles with diameters between 60 mesh and 35 mesh and less than 60 mesh in a proportion of 2:1.

The production technologies of the experimental coal samples, which are made by different pressing loads, are as follows.

1. To remove the interference of the particle coal sample moisture, the particle coal samples, which include those with diameters less than 60 mesh and those with diameters between 60 mesh and 35 mesh, are placed in a drying oven to dry. The drying temperature was 105 °C. To determine the end time of particle coal sample drying, the coal samples are weighed by a balance every half an hour. If the weights of coal samples do not change, the drying stage has ended. Then, the particle samples are stored in an evaporating dish for cooling and later use.

2. Coal samples with diameters less than 60 mesh are weighed. Then, some water is weighed. The quantity of pure water is equal to the quantity of coal samples multiplied by 20%. Then, the coal samples and pure water are combined and stirred thoroughly.

3. The experimental coal samples are made with a mold. The mold includes three parts, namely, a pressure head, a forming sleeve, and a retreat mold sleeve, which are shown in Figure 3.

4. A coal sample with a 20% water content is placed in the forming sleeve. To ensure that the coal samples are loaded enough, the pressure head is used for preloading. Then, the stable pressure function of the HUTM is used. Therefore, the HUTM is used to compress the coal samples with a load of 50 MPa for 30 min, and the schematic diagram of the coal samples compression is shown in Figure 4.

Figure 2. Lambert $W$ function.

Figure 3. Schematic diagram of the custom mold.

Figure 4. Schematic diagram of the compressing remolded coal.
(5) When the stabilization pressure time is over, the HUTM will automatically release pressure. Then, the retreat mold sleeve is used to drop out the remolded coal by HUTM. A sketch diagram of the retreating mold is shown in Figure 5. The resulting remolded coal sample is shown in Figure 6.

![Figure 5. Sketch diagram of the retreating mold.](image)

![Figure 6. Resulting remolded coal samples.](image)

(6) In this work, the remolded coal samples made of coal particles with diameters less than 60 mesh are studied by considering five pressing loads. To obtain the experimental coal samples, steps (2), (3), (4), and (5) are repeated with loads of 50, 100, 150, 200, and 250 MPa for these samples. These remolded coal samples with diameters between 60 mesh and 35 mesh and less than 60 mesh in a proportion of 2:1. Therefore, steps (1), (2), (3), (4), and (5) are repeated with loads of 50, 100, 150, 200, and 250 MPa for these samples. These remolded coal samples with diameters between 60 mesh and 35 mesh and less than 60 mesh in a proportion of 2:1 are referred to as $A_1$, $A_2$, $A_3$, $A_4$, and $A_5$, respectively.

3.2. Experimental Procedures. According to the experimental objectives in this paper, the experiments are divided into two groups, namely, for the determination of the imbibition height of the experimental coal samples during the spontaneous imbibition process (group I) and for the determination of the contact angle of the remolded coal with different pressing loads (group II).

For group I, the measurement steps of the imbibition height during the imbibition process are as follows.

(1) To exclude the effect of water, the remolded coal samples should be dried. The drying temperature is 378.15 K. If the weights of the remolded coal samples do not change, the drying stage has ended. Then, the remolded coal samples are placed in an evaporating dish for cooling. After cooling, the remolded coal samples can be used for testing.

(2) In this paper, a water-sensitive indicator method is used to measure the imbibition height. The components of the water-sensitive indicator are carboxymethylcellulose, calcium carbonate, titanium dioxide, cobalt sulfate, cobalt thiocyanate, and water. The preparation ratio is 1:2:5:6:13:32. Compared with the drying method, the water-sensitive indicator method can be used to examine the imbibition height of porous materials. As shown in Figure 7a, the prepared water-sensitive indicator is painted on the wall of the experimental coal samples along a straight line. As shown in Figure 7b, the remolded coal is dried in a drier until the water-sensitive indicator turns gray.

(3) The remolded coal is sealed with plastic wrap. Then, the bottom of the remolded coal is sealed with fabric. As shown in Figure 8, a viscosity scale is pasted on the plastic wrap.

(4) The treated experimental coal samples created with different pressing loads are placed in the spontaneous imbibition experimental system. The experimental system of imbibition height shown in Figure 9 includes three sinks (a, c, and d), a piece of sponge (b), a miniature pump (e), some connecting pipelines, and some water. The sponge is placed in sink (a). The left side of sink (a) has an outlet. Sink (c) is placed below the outlet. The
water in the sinks (a, c, and d) and the pump form a circulating water stream. Therefore, the experimental device can keep the water level constant in sink (a).

(5) A remolded coal sample is placed in the experimental device. The height of change in the color of the indicator strip along the remolded coal sample is measured after a certain period of time. The height of change in the color of the indicator strip along the remolded coal sample is measured again after another period of time. The height of change in the color of the remolded coal is shown in Figure 10. According to the above method, the height of change in the color during the imbibition process is measured several times.

For group II, the measurement steps of the remolded coal contact angle are as follows.

The measurement methods of contact angle include the angle measurement method, force measurement method, penetration measurement method, and height measurement method. The height measurement method is used in this work. The height measurement method is based on the assumption that the droplet section is round. This assumption assumes that the droplet is small enough (less than 6 μL in volume) that the effect of gravity on the droplet shape can be ignored.\(^{52}\) The positive section of the droplet can be considered a part of the standard circle. The principle of calculating the contact angle by the height measurement method is shown in Figure 11.

Equations 16–18 can be obtained from Figure 11.

\[
\gamma + \alpha = 90^\circ \tag{16}
\]

\[
\gamma = \beta \tag{17}
\]

\[
\gamma = 2\alpha \tag{18}
\]

The substitution of eqs 16 and 18 into eq 17 leads to eq 19.

\[
\theta = 2\alpha \tag{19}
\]

For triangle OEG

\[
\tan \alpha = \frac{OE}{OG} \tag{20}
\]

Equation 20 can be changed to eq 21.

\[
\alpha = \arctan \left( \frac{OE}{OG} \right) \tag{21}
\]

The substitution of eq 19 into eq 21 leads to eq 22.

\[
\theta = 2\arctan \left( \frac{OE}{OG} \right) \tag{22}
\]

where \(\theta\) is the contact angle and OE is the height from the solid–liquid interface to the top of the droplet.
The measurement steps of the contact angle are as follows.

(1) The remolded coal slices shown in Figure 12 were prepared for the determination of the contact angle.

Figure 12. Remolded coal slices for determination of the contact angle.

(2) The contact angle shown in Figure 13 was measured by a JC2000D1 contact angle tester. The instrument was calibrated to ensure the sample feeder was in the central area of the screen and that the lofting table, camera, and lens were in the same horizontal plane.

Figure 13. JC2000D1 contact angle tester.

(3) The syringe with water was installed, and the position of the needle was controlled by a PC. The syringe was pushed slowly so that the drop fell freely onto the remolded coal slice. After the baseline was determined by the system software, the contact angle was determined according to the baseline, as shown in Figure 14.

Figure 14. Diagram of contact angle measurement.

The measurement steps of the contact angle are as follows.

(1) The remolded coal slices shown in Figure 12 were prepared for the determination of the contact angle.

(2) The contact angle shown in Figure 13 was measured by a JC2000D1 contact angle tester. The instrument was calibrated to ensure the sample feeder was in the central area of the screen and that the lofting table, camera, and lens were in the same horizontal plane.

(3) The syringe with water was installed, and the position of the needle was controlled by a PC. The syringe was pushed slowly so that the drop fell freely onto the remolded coal slice. After the baseline was determined by the system software, the contact angle was determined according to the baseline, as shown in Figure 14.

4. RESULTS AND DISCUSSION

4.1. Analysis of the Results of the Imbibition Height of the Experimental Coal Samples. According to the experimental procedures (group I) in Section 3.2, these experiments test the imbibition height of the experimental coal samples ($A_1$–$A_4$, $B_1$–$B_4$). The test results of the imbibition height of the experimental coal samples are shown in Figure 15. Figure 15 shows that the imbibition height increases as time increases. The imbibition height increases as the pressing load increases. The reason for this phenomenon is that the greater the pressing load, the smaller the pore radius. The smaller the pore radius, the greater the capillary force. Capillary force drives spontaneous imbibition. Therefore, the imbibition height increases with increasing pressing load. The imbibition height of the $B$ remolded coal samples is greater than that of the $A$ remolded coal samples under the same pressing load. The reasons for this finding are as follows. The $B$ remolded coal samples can form a large capillary force. An experimental coal sample is a porous material that can be wetted by water. The $B$ remolded coal samples are made of coal with two particle size ranges, which increases the wettability of the remolded coal. The force between the liquid and solid molecules is greater than that between liquid molecules. To reveal the evolution law of the imbibition height, eq 14, which is an implicit analytical solution, can be used to fit the data presented in the scatterplots in Figure 15. The fitting parameters based on eq 14 are listed in Table 1. Therefore, the relationship between the imbibition time and imbibition height of the remolded coal can be obtained from Table 1.
As shown in eq 14, parameter $a$ is expressed in terms of surface tension, contact angle, viscosity, tortuosity, fractal dimension, maximum pore diameter, and porosity; and parameter $b$ is expressed in terms of water density, gravitational acceleration, viscosity, tortuosity, fractal dimension, maximum pore diameter, and porosity. The parameters $a$ and $b$ are known from Table 1. The viscosity, contact angle, porosity, and surface tension can be measured. The tortuosity can be calculated by the spherical particle filling model. Therefore, the maximum pore diameter and fractal dimension can be calculated by Maple software by solving equations simultaneously.

### 4.2. Analysis of the Porosity and Tortuosity of Remolded Coal

The coal industry standard of the People's Republic of China (MT/T 918-2002 and GB/T 217) can be used to measure the porosities of remolded coal. The solution of porosity is shown in eq 23, and the method of measuring the apparent relative density (ARD) is given in the coal industry standard of the People’s Republic of China (MT/T 918-2002). The true relative density (TRD) can be obtained using the coal industry standard of the People’s Republic of China (GB/T 217). The calculation results of the experimental coal sample porosities are shown in Figure 16.

$$\phi = \frac{\text{TRD} - \text{ARD}}{\text{TRD}}$$

(23)

where $\phi$ denotes the porosity of the remolded coal, TRD denotes the true relative density of the remolded coal, and ARD denotes the apparent relative density of the remolded coal.

Figure 16 shows that the porosities of the experimental coal samples decrease as the pressing load increases. The cause of the phenomenon is that a greater pressing load can increase coal particle deformation and bring the coal particles more close to other coal particles, which reduces the number and diameter of the pores. Therefore, the porosity decreases gradually with increasing pressing load. The porosities of the experimental specimens $B$ are smaller than those of the experimental specimens $A$. The reasons are as follows. Small particles will fill the central parts of the pores around large particles (filling effect), reducing the pore diameter and porosity, while large particles will occupy more space, reducing the number of particle accumulation pores (occupying effect). Both remolded coal samples $A$ and $B$ are formed by the adhesion of compressed particles. Compared with the $A$ remolded coal sample, $B$ remolded coal includes particle grading. It can be seen that the porosity can be reduced by a proper particle grading. As shown in Figure 16, the porosity and pressing load can be described by a power function, given in eq 24. The fitting parameters based on eq 24 are listed in Table 2.

$$\phi = \sigma^d$$

(24)

where $\phi$ is the porosity, $\sigma$ is the pressing load, and $c$ and $d$ are the fitting parameters.

| remolded coal | $c$   | $d$   | $R^2$ |
|---------------|-------|-------|-------|
| $A$           | 0.40644 | -0.07625 | 0.99709 |
| $B$           | 0.40644 | -0.07622 | 0.99659 |

Table 2. Fitting Parameters $c$ and $d$ Based on Equation 24

The expression of tortuosity is given in eq 6. The relationship between the tortuosity and porosity is shown in eq 25, which was established by the spherical particle packing model and derived from the fluid pressure drop equation. Since the remolded coal $A$ and remolded coal $B$ are made via particle packing, eq 25 can be used to calculate the tortuosity of the remolded coal samples. Figure 17 shows the calculation results of the tortuosity. As shown in Figure 17, the tortuosity increases with decreasing porosity. By substituting eq 24 into eq 25, the relationship between the tortuosity and pressing load can be obtained, which is shown in eq 26. Equation 25 implies that the greater the porosity, the less the tortuosity. As shown in Figure 17, the tortuosity increases with decreasing porosity. In other words, the tortuosity increases with increasing pressing load. The tortuosity of remolded coal $B$ is greater than that of remolded coal $A$. Due

### Table 1. Fitting Parameters Based on Equation 14

| remolded coal | $a$ | $b$ | $R^2$ |
|---------------|-----|-----|-------|
| $A$           | 1.04121 $\times$ 10$^{-7}$ | 2.06784 $\times$ 10$^{-7}$ | 0.9999 |
to the occupying effect, the number of particles blocking the flow of fluid in remolded coal B is greater than that in remolded coal A.

\[ \tau = 1 + 0.41 \ln(1/\phi) \]  

(25)

where \( \tau \) is the tortuosity and \( \phi \) is the porosity.

\[ \tau = 1 + 0.41 \ln(1/(\sigma^4)) \]  

(26)

4.3. Test Results of the Contact Angle of the Remolded Coal Samples. The contact angle is the angle between the line tangent to the intersection of a solid, liquid, and gas and the solid–liquid boundary. Generally, the symbol \( \theta \) is used to represent the contact angle. If the contact angle is less than 90°, the surface of the solid is hydrophilic, meaning that the solid is easily wetted by the liquid. If the contact angle is greater than 90°, the surface of the solid is hydrophobic, meaning that the solid is not easily wetted by the liquid and the liquid easily moves on the surface. In other words, the wettability increases as the contact angle increases. If the contact angle is 90°, the solid (in this case, a porous material) exhibits neutral wetting.

According to the experimental procedures (group II) in Section 3.2, these experiments test the contact angle of the remolded coal samples (A1–A4, B1–B4). The test results of the contact angle of the experimental coal samples are shown in Figure 18.

As shown in Figure 18, the contact angle increases as the pressing load increases under the same particle ratio. This is because the proportion of solid particles per unit area decreases with decreasing pressing load. The lower the porosity, the smoother the surface, and the higher the porosity, the rougher the surface. The coarser the surface, the easier the wetting. A water droplet easily spreads on a rough surface, which leads to a decrease in the contact angle. For the remolded coal samples with the same pressing load, the contact angle of remolded coal A is greater than that of the remolded coal B. This is because the surface roughness of remolded coal A is greater than that of the remolded coal B, which results in different surface energies. The relationship between the pressing load and angle can be described by eq 27.

\[ \theta = \sigma^{1/4} \]  

(27)

4.4. Inversion of the Fractal Dimension and Maximum Diameter of a Capillary. As shown in eq 14, parameter \( a \) is expressed in terms of surface tension, contact angle, viscosity, tortuosity, fractal dimension, maximum pore diameter, and porosity; and parameter \( b \) is expressed in terms of the water density, gravitational acceleration, viscosity, tortuosity, fractal dimension, maximum pore diameter, and porosity. The parameters, including surface tension, contact angle, viscosity, tortuosity, density, and gravitational acceleration, are given in the previous section. Substituting these parameters (surface tension, contact angle, viscosity, tortuosity, density, and acceleration due to gravity) into parameters \( a \) and \( b \), a binary system of equations is created. Therefore, the fractal dimension and maximum diameter of the capillary can be inverted. The inversion results are shown in Figure 19.

As shown in Figure 19, the fractal dimension and maximum diameter decrease with increasing pressing load. In other words, different pressing loads will cause different pore structure shapes for the same particle size. The fractal dimension increases with increasing complexity of the pore structure shape. With increasing pressing load, the pore structure shape tends to become homogeneous. The fractal dimension of remolded coal A is greater than that of remolded coal B. The larger the fractal dimension, the stronger the heterogeneity of the pore distribution. Remolded coal B is composed of two particle size ranges, which produce some filling effects and occupying effects.
The difference in the pore characteristics between particles is the main reason for the difference in the fractal dimension. Due to the filling effects and occupying effects, the heterogeneity of pores between particles is reduced. Therefore, the pore structure shape of remolded coal \( A \) is less complex than that of remolded coal \( B \). The maximum diameter decreases with increasing pressing loads. For the experimental coal samples with the same pressing load, the maximum diameter of remolded coal \( A \) is greater than that of remolded coal \( B \). The decreasing rate of remolded coal \( B \) is greater than that of remolded coal \( A \) (Table 3).

| remolded coal | \( \epsilon \) | \( f \) | \( R^2 \) |
|--------------|------------|--------|--------|
| \( A \)      | 32.765     | 0.0638 | 0.9985 |
| \( B \)      | 33.196     | 0.0553 | 0.9988 |

The relationship between the fractal dimension and pressing load can be described by eq 28, and the relationship between the maximum diameter and pressing load can be expressed by eq 29. The parameters of eqs 28 and 29 are shown in Table 4.

\[
D_p = m + m_1 e^{m_2 \tau}
\]

(28)

\[
d_{\text{max}} = n + n_1 e^{n_2 \tau}
\]

(29)

### 4.5. Analysis of the Accuracy of the Spontaneous Capillary Imbibition Model of Remolded Coal

Equations 14, 24, 25, and 27–29 are combined, and a model of capillary imbibition in remolded coal is obtained, which is shown in eq 30. To verify the accuracy of the spontaneous capillary imbibition model, the remolded coal samples with a pressing load of 250 MPa were tested. The physical parameters of the remolded coal with pressing loads of 125 and 250 MPa can be calculated by eqs 14, 24, 25, and 27–29, and the results are presented in Table 5.

As shown in Figure 20, both the calculated and the experimental imbibition heights of the remolded coal with a pressing load of 250 MPa are greater than the imbibition heights of the experimental coal samples with a pressing load of 200 MPa. The experimental data and theoretical values have little difference. In other words, the experimental data are consistent with the derived value. The calculated value of height for the remolded coal with a pressing load of 125 MPa is higher and lower than the imbibition heights of the experimental coal samples with pressing loads of 100 and 150 MPa, respectively. In general, the greater the pressing load, the higher the imbibition height. Therefore, eq 30 can be used to predict the imbibition height for the imbibition process in remolded coal with different pressing loads.

### 5. CONCLUSIONS

The implementation process of water invasion technology in coal is actually a process of the spontaneous imbibition of external water. To obtain the model of spontaneous capillary imbibition in remolded coal, the imbibition heights of the remolded coal made with different pressing loads are tested using a spontaneous imbibition experimental system. The major conclusions are as follows.

1. An implicit analytical solution and an exact analytical solution for the spontaneous imbibition water in remolded coal, which takes into consideration the gravity force, were obtained based on the fractal theory.

2. Due to the coal particle deformation and the cohesive force, the porosity and maximum diameter decrease gradually with increasing pressing load. Due to the included filling effects and occupying effects, the proper particle grading can reduce the porosity and tortuosity of remolded coal samples. The Comiti model can be used to describe the tortuosity. The tortuosity increases with decreasing porosity. The lower the porosity, the smoother the surface of the sample. The contact angle is negatively correlated with the surface roughness.

3. The fractal dimension decreases with increasing pressing load. The fractal dimension increases with increasing pore structure complexity. The difference in the pore characteristics between particles is the main reason for the difference in the fractal dimension. Due to the filling effects and occupying effects, the heterogeneity of the pores between the particles is reduced.

4. The pore radius decreases with increasing pressing load. The smaller the pore radius, the greater the capillary force. The imbibition height increases with increasing pressing load, and the experimental value is consistent with the theoretical value calculated by the model. The model not only reflects the influencing factors of imbibition but also can be used to predict the imbibition height at different pressing loads.

### Table 3. Fitting Parameters Based on Equation 27

| remolded coal | \( \epsilon \) | \( f \) | \( R^2 \) |
|--------------|------------|--------|--------|
| \( A \)      | 32.765     | 0.0638 | 0.9985 |
| \( B \)      | 33.196     | 0.0553 | 0.9988 |

### Table 4. Parameters of Equations 28 and 29

| remolded coal | \( m \) | \( m_1 \) | \( m_2 \) | \( n \) | \( n_1 \) | \( n_1 \) |
|--------------|-------|--------|--------|------|-------|-------|
| \( A \)      | 1.99699 | 0.00309 | -0.00202 | 5.125744 \( \times 10^{-5} \) | 5.87282 \( \times 10^{-5} \) | -0.0116 |
| \( B \)      | 1.99681 | 0.00328 | -0.00298 | 2.55032 \( \times 10^{-5} \) | 6.3786 \( \times 10^{-5} \) | -0.00479 |
Table 5. Physical Parameters of the Remolded Coal with a Pressing load of 250 MPa

| Remolded Coal | Pressing Load (MPa) | \( \phi \) | \( \tau \) | \( \theta \) | \( D_p \) | \( a \) | \( b \) |
|---------------|---------------------|------|------|------|---------|------|------|
| A             | 250                 | 0.26806 | 1.53978 | 46.6  | 1.99885 | 5.44808 \( \times 10^{-3} \) | 2.74 \( \times 10^{-2} \) | 3.82 \( \times 10^{-2} \) |
| B             | 125                 | 1.51811 | 44.6  | 1.9939 | 6.50333 | 1.89729 \( \times 10^{-2} \) | 3.04409 \( \times 10^{-2} \) |
|               | 250                 | 0.26682 | 1.54168 | 45.0  | 1.9937  | 4.47633 \( \times 10^{-3} \) | 3.28758 \( \times 10^{-2} \) | 3.66097 \( \times 10^{-2} \) |
|               | 125                 | 1.52002 | 43.4  | 1.99907 | 6.05355 | 2.73559 \( \times 10^{-2} \) | 4.00356 \( \times 10^{-2} \) |

Figure 20. Calculated values and experimental values of the imbibition height.

(a) Remolded coal A

(b) Remolded coal B

Notes

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