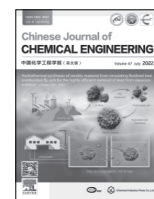




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Full Length Article

“Relay-mode” promoting permeation of water-based fire extinguishing agent in granular materials porous media stacks

Kang Wang¹, Wei Tan¹, Liyan Liu^{1,2,*}¹ School of Chemical Engineering and Technology, Tianjin University, Tianjin 300350, China² Tianjin Key Laboratory of Chemical Process Safety and Equipment Technology, Tianjin 300350, China

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ABSTRACT

Water-based fire extinguishing agent is the main means to deal with smoldering fires. However, due to the hydrophobic properties of the particle surface, the porous medium channel provide resistance and slow down the extinguishing agent flow during the downward permeation process. To promote the liquid permeation process in such porous media, this work studied liquid imbibition process and analyzed the oscillating and attenuating process of liquid level in capillary channel by theoretical, experimental, and numerical methods. An empirical mathematical equation was proposed to describe the oscillating process, and the effects of the capillary diameter and contact angle parameters on the transportation process were analyzed. Based on this, the “relay-mode” was proposed to promote the liquid transportation forward. Finally, the transient simulation results of liquid permeation in coal stacks showed when the liquid flowed through the channel with changed diameter from large to small ones, the transportation distance was several times longer than that through the unidiameter ones. The trend of liquid “relay-mode” in capillaries can be used to promote the permeation in granular materials porous media stacks. The relevant results also provide new thoughts to develop the water-based fire extinguishing agents and then improve the firefighting efficiency of deep-seated fire in porous media stacks.

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1. Introduction

In order to achieve sustainable development, biomass energy has become an important method and trend to alleviate the energy crisis, the fire safety problems attract more attention during the biomass materials storage process. Among the fire accidents in the storage of biomass materials reported in China and other countries, deep-seated fires occupy a large part [1–4]. The ignition point of such fires is often located deep in the stack, and the hydrophobicity of the biomass particles surface often prevents the water-based fire extinguishing agents from reaching the ignition point quickly, which usually consume much manpower and financial resources [5–9]. Researches have been carried out on promoting liquid permeation in the stacks in fire safety field, such as reducing the surface tension of the liquid phase by adding some surfactants [10,11]. Tan *et al.* [12] did much work to investigate the effect of various potassium halide additives on improving extinguishing

water mist. Yu *et al.* [13] investigated that the addition of Xanthan Gum (XG) and Fluorocarbon Surfactant 1157 (FC1157) synergistically improving the spreading performance of aqueous foam over ethanol. Above efforts are mainly based on the improvement of fire extinguishing agents, which focus on the statics properties improvements of extinguishants. While, it is lack on the research of the igniting materials stacks and the interactions between the liquid and solid phases, which is of great significance to investigate the water-based extinguishing agents displacing air in granular materials stacks. The permeation paths in granular materials stacks are all interlaced, and with many factors effecting the permeation process, investigating the liquid permeation process in stacks with complex structure and large span of pores size is still challenging. Considering that the essence of liquid permeation in pores is the process of liquid phase displacing gas phase, and paths with complex structures can be regarded as a collection of finite pipes, the scholars have studied the capillary phenomenon in a single capillary to investigate the dynamic process of liquid permeation in porous media stacks. Sachin *et al.* [14] and Tang *et al.* [15] both studied the dynamic evolution of meniscus in a capillary, Lunowa *et al.* [16] used to account for non-equilibrium effects like dynamic

* Corresponding author.

E-mail address: liuliyang@tju.edu.cn (L. Liu).

capillarity and hysteresis during discussing the mathematical models for two-phase flow in a porous medium. Studying the liquid movement in a single capillary is extremely important for the subsequent study of the liquid penetration process in granular material stacks. However, above researches assumed the liquid simply rising in the capillary and finally stabilized at the equilibrium level, without specific consideration of the dynamic process of the meniscus movement in the capillary in detail, which is one of the innovative contents of this research.

In related fields of lithofacies and oil reservoir storage, many scholars have studied the liquid permeation flow in porous media [17–20]. Although many formulas and theories in the microscopic field have also been used, such as capillary equations, Young-Laplace equations, etc., the permeation models [21–26] are established from a macro perspective. The essence of permeation model is to describe the liquid accumulation flowing through porous media versus time, illustrating the influences on which of liquid flowing parameters, solid structure parameters, and interface interactions. As mentioned by Richards [27], the gas-liquid interface is shaped as spherical fan in the equilibrium state according to Young-Laplace equation. But during the meniscus formation process, the gas-liquid interface will deviate from the spherical fan shape due to viscous, interfacial tension and inertial forces. Thus, the gas-liquid interface changing is of especially great importance for the subsequent permeation state changing during the permeation process. The gas-liquid interface shape reflects the resultant force of the liquid molecules near the gas-liquid interface in real time. This is a great complement to the traditional static meniscus research method. Mohammad *et al.* [28] regarding the liquid level in the capillary as a rigid body and studied the oscillation process of the surface by transforming the nonlinear Lucas-Washburn equation into a linear mass-spring-damping system in 2017. However, the dynamic gas-liquid interface is not mentioned, resulting in a certain deviation between the resultant force analysis research of the meniscus in the permeation process and the actual situation. It is still of great importance for revealing the liquid movement in capillary [17,29–33] and then in real granular materials porous media stacks. The research of meniscus movement in capillaries is a supplement to the statics work in the liquid permeation in porous media, making up for the defects in related research fields and providing much realistic guidance and help for follow-up research. It can be much helpful to realize that the constantly changing parameters during the liquid permeation can be used to achieve the expected goals.

This research studied the dynamic movement of meniscus in capillary and found the oscillating characteristics of liquid level in capillary with the combination of experiment and numerical simulation methods. A series of transient simulation with computational fluid dynamics (CFD) of liquid rising and falling in capillaries were carried out. The effect of capillary diameter, contact angle on the gas-liquid interface deforming, the extreme liquid level, and the equilibrium liquid level was discussed. An oscillation attenuation function was proposed to mathematically describe the three-phase line moving process, coming up with a concept of “relay-mode” to realize promoting liquid permeation by narrowing the pore throats in porous media. A transient simulation of liquid permeation in real granular materials porous media stacks was carried out and the influences of pore size changing on the liquid permeating process were discussed. Relevant results in this research are expected to deepen the understanding of liquid dynamic rising process in capillaries, and the conclusions can provide some theoretical guidance for promoting liquid permeating in granular material stacks and then improving the extinguishing efficiency of smoldering fire, which is of great significance for engineering safety accidents prevention and disposal.

2. Methodology

2.1. Experimental set up of liquid dynamic rising in capillary

The experimental device for liquid dynamic rising in capillary was composed of capillary lifting and clamping device, high-speed photography acquisition device (OLYMPUS, i-speed 3), control and display unit (CDU), light source, silica capillary channel, liquid storage tank, data acquisition platform and lifting platform, as shown in Fig. 1. The high-speed photography acquisition device assists to record the liquid rising process in a capillary channel at required recording frame rate. The CDU was helpful to adjust the graphic capturing performance. The capillary channels were made of quartz glass with good light transmission performance. The diameter of capillary was set as 1 mm, 2 mm, 3 mm, 4 mm, 5 mm, respectively, and deionized water was selected as the wetting liquid.

The capillary clamping and lifting device were completely self-designed to satisfy the experimental requirements and manufactured using 3D printing technology, as shown in Fig. 2. The clamping arm was fixed on the nut of the ball screw, and can be moved with the screw by twisting the rotary knob. Due to the limiting effect of the support frame, the screw with the clamping arm can only move in the vertical direction without rotating movement. At the same time, a certain margin was left in the clamping notch of the clamping arm, which can fasten capillary channels of different diameters and keep the capillaries in a vertical direction. In every single experiment, the high-speed photograph acquisition device was turned on before the capillary was immersed into the liquid. Then the resolution was then adjusted to ensure the frame on the CDU clear, and the capillary channel was moved by twisting the rotary knob to immerse the capillary end into the liquid in the water tank. This capillary clamping and lifting device can keep the capillary vertical during the whole experimental operation process.

2.2. Numerical simulation of liquid imbibition in capillary

The establishment of the physical and mathematical models for the dynamic rising of meniscus in capillary is based on the following assumptions and simplifications:

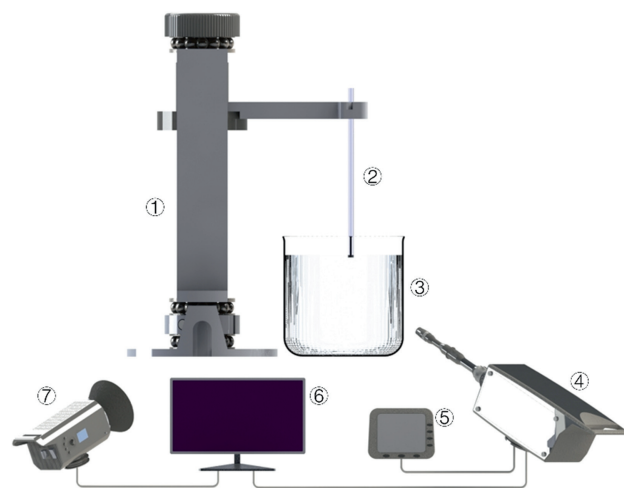


Fig. 1. The sketch of experimental set up of the liquid rising in capillary. ① Capillary clamping and lifting device; ② Capillary channel; ③ Liquid storage tank; ④ High-speed photography acquisition device; ⑤ CDU; ⑥ Data acquisition platform; ⑦ Light source.

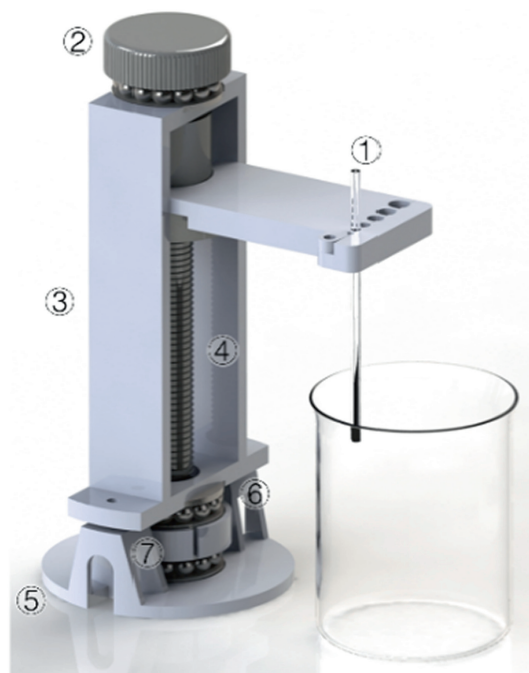


Fig. 2. Capillary clamping and lifting device. ① Clamping arm; ② Rotary knob; ③ Support frame; ④ Ball screw; ⑤ Support pedestal; ⑥ Thrust bearings; ⑦ Adapting piece.

- ① The fluid is incompressible and satisfies Boussinesq approximation;
- ② Except for surface tension, other physical parameters are constant;
- ③ The fluid velocity is low, and the flow is laminar;
- ④ All solid walls meet the non-slip condition;
- ⑤ The liquid in the capillary is isotropic.

Considering the rotational symmetry of the physical model shown in Fig. S1 (in Supplementary Material), the water moving scenario in capillary was simplified as two-dimensional. A series of liquid rising and falling simulation schedules in capillary of different diameters with different contact angles were established and solved by Fluent, the schematic diagram of the computational domain was shown in Fig. S2. The numerical models were all discretized into structured grids with ICEM (integrated computer-aided engineering and manufacturing) CFD using the least squares cell-based gradient evaluation method, where the minimum grid is 0.001 mm. The details of these meshing schedules were listed in Table S1, with the grid independence verification had been performed. The grid cells were all quadrilateral with good quality and low aspect ratio.

Volume of fluid (VOF) model was on to set the initial liquid level, where the gas-liquid interface was tracked by the geo-reconstruct scheme. Specified operating density was selected and set as the lightest density of the gas-liquid system to eliminate the influence of hydrostatic pressure accumulation. To improve the convergence of the calculations, the implicit body forces were considered in the VOF model to balance the pressure gradient and volume force in the momentum equation. The pressure implicit split operator (PISO) algorithm was adopted, where a two-step modification method was implemented for faster convergence by setting a larger time step. The pressure term was applied with the pressure staggering option method and momentum term with the second order upwind scheme. A double precision representation of real numbers was adopted to reduce the round off errors.

Table 1

Key variables in the liquid rising simulations

The item	Values
Water density $\rho_L/\text{kg}\cdot\text{m}^{-3}$	998.2
Water dynamic viscosity $\mu_L/\text{kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}$	1.003×10^{-3}
Air density $\rho_G/\text{kg}\cdot\text{m}^{-3}$	1.225
Air dynamic viscosity $\mu_G/\text{kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}$	1.7894×10^{-5}
Water surface tension $\sigma/\text{N}\cdot\text{m}^{-1}$	0.07275
Capillary diameter d/mm	1, 2, 3, 4, 5
Contact angle $\theta/(\circ)$	30, 60, 120, 150
System temperature/K	293.15

To clarify the gas-liquid interface movement in capillary, a series of transient simulation have been carried out. The key variables included in the liquid rising in capillary simulations were list in Table 1, where the time step was set as 10^{-6} s.

3. Results and Discussion

3.1. Experimental analysis and simulation verification

As shown in Fig. 3, the liquid level moving process from the quartz glass tube contact to the water surface until the liquid rises to the equilibrium level. From overall view, the liquid level underwent rising and falling rather than simply ascending to the equilibrium level. The phenomenon of oscillating was more obvious in the narrower tube with the diameter of 1 mm. With the diameter enlarging, the intensity of the oscillating gradually decreased and the duration became shortened.

The numerical simulation method was verified by conducting a series of liquid imbibition simulation to compare with the experiments, where the contact angle was measured as 77.13° . Fig. 4 showed the simulation results of liquid level moving in tubes with different diameters. The trajectory curves over time intuitively demonstrated the characteristic of oscillating rising of liquid level moving in capillary. According to capillary equation, the liquid level in a capillary can be calculated from the static force balance on the gas-liquid phase, which can be described as follows:

$$\Delta p = \sigma(1/r_1 + 1/r_2) \quad (1)$$

$$h_0 = 2\sigma\cos\theta/\Delta\rho gr \quad (2)$$

where Δp is the additional pressure, Pa; σ is the surface tension of liquid, $\text{N}\cdot\text{m}^{-1}$; r is the radius of curvature of the meniscus, mm; h_0 is the height of the equilibrium position, mm; θ is the contact angle, $(^\circ)$; $\Delta\rho$ is the density difference of liquid and gas, $\text{kg}\cdot\text{m}^{-3}$; and g is the gravity, $\text{m}\cdot\text{s}^{-2}$. For the liquid in a capillary, the gas-liquid interface will finally deform to the shape of meniscus, generating additional pressure, which is related to surface tension and curvature of liquid surface. From Fig. 4, the equilibrium levels under different diameter conditions were consistent with Eq. (2).

To illustrate the oscillating rising characteristics in detail, the equilibrium level, the extreme liquid level with its corresponding time of the capillary experiments and simulation were compared, as shown in Figs. S3 and S4. The errors between the experimental and simulation results of both the liquid levels and its corresponding time were very small about 9.0%, and with the maximum error being below 13.0%. The simulation value of the extreme levels during the capillary rising in capillary were close to the experimental data, indicating the numerical simulation method reliably representing the oscillating rising phenomenon in the capillaries.

Fig. 5 showed the deviation errors of the theoretical and the simulation results from the experimental data, where the theoretical values were calculated according to Eq. (2). The errors between the theory values and experimental data all exceeded 15.0% in

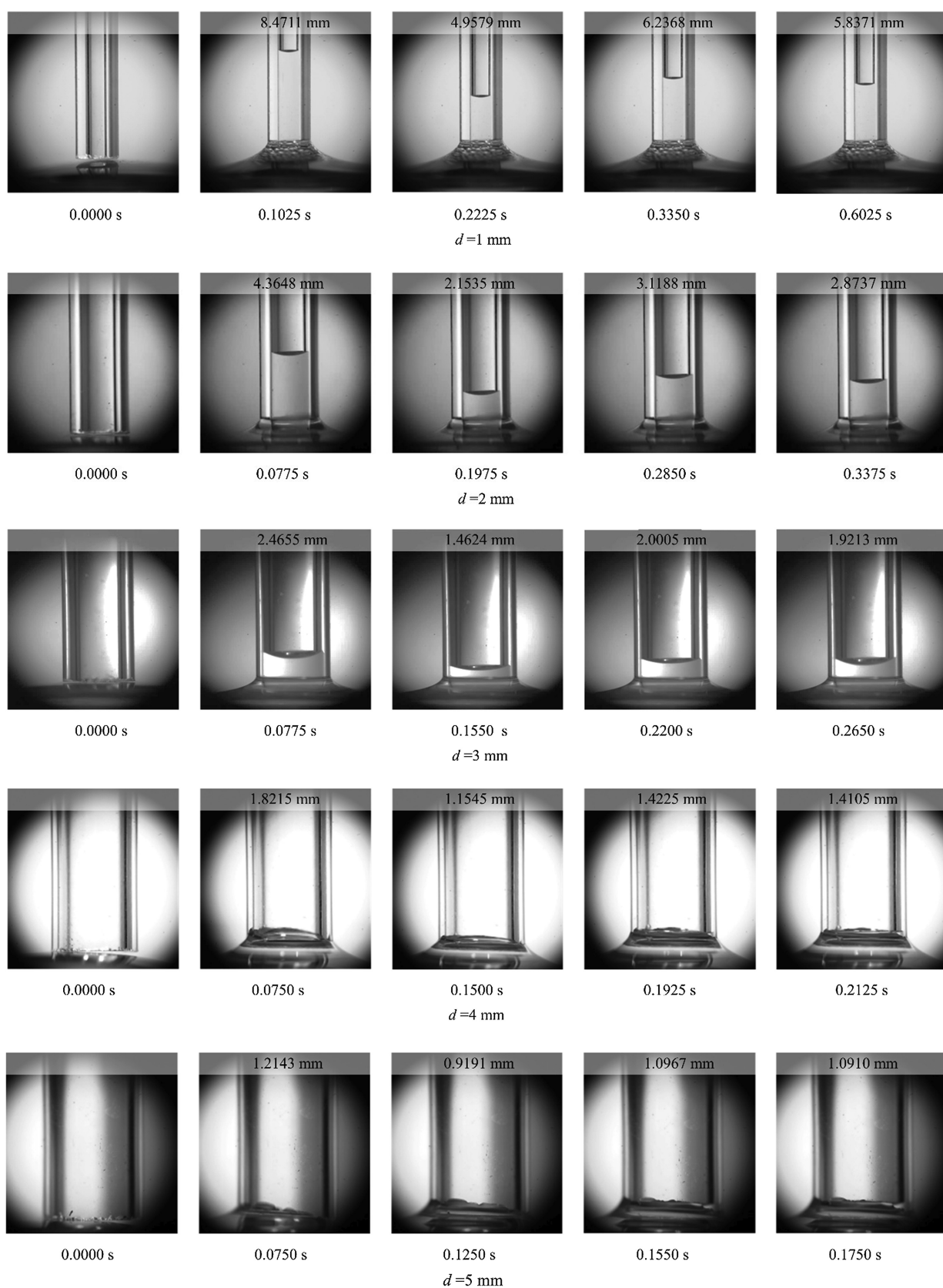


Fig. 3. The liquid level with time in different capillaries.

capillary channels with different channel diameters, and the maximum error even reached 22.511% with the capillary diameter of 5 mm. The deviation between the simulation results and the exper-

imental data was basically less than 10.0%, and the maximum error appears in the capillary with a diameter of 3 mm, which was 12.922%. The simulation value was closer to the experimental data

than the theoretical value, where the experimental data was larger than the simulation value and smaller than the theoretical value. In numerical simulation, the uniform inner wall was set for the boundary conditions in capillaries were controllable. During the liquid rising in capillary, the contact was only affected by the surface tension. However, the inner wall in experiments was not uniform and the roughness was large, which could promote liquid rising to a higher level. Combined above analysis with Fig. 4, the oscillating phenomenon weakened with the tube diameter enlarging, explaining the oscillating rising duration in experiments were longer than the simulation results.

Especially, there are fewer parameters in the capillary equation of Eq. (2), which have all been ideally simplified, such as the standard circular capillary cross section, the smooth inner wall of the capillary, and the uniform hydrodynamic properties of the liquid in the capillary. All the simplifications and assumptions will affect the real liquid rising process in the capillary, and then results in the deviation error. The bottom end and inner wall of the capillary channels were usually rough, and the liquid level in the water tank was not completely horizontal at the beginning. That explains why the theory value is larger than the experimental data.

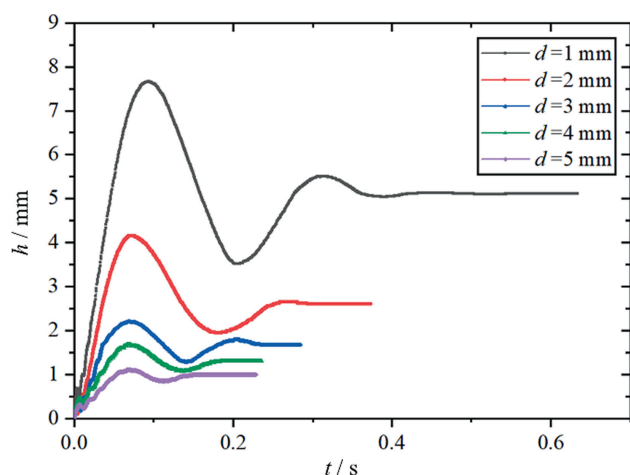
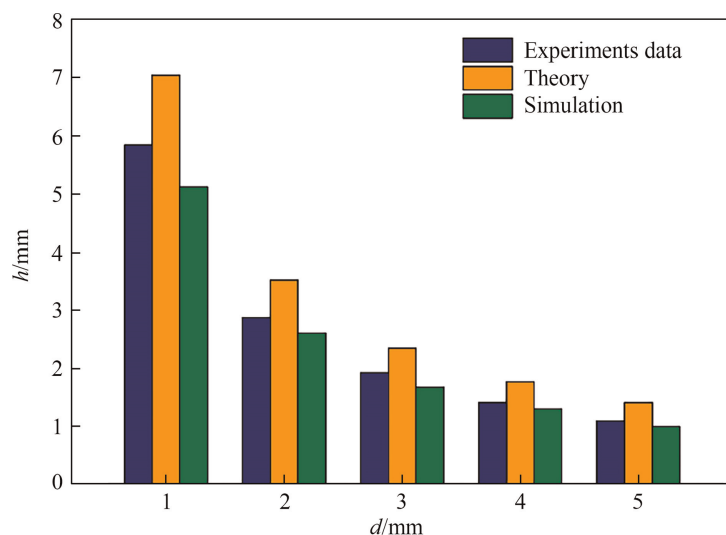
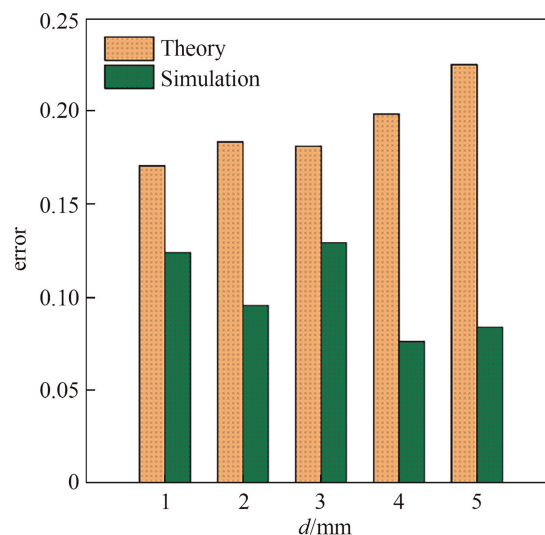


Fig. 4. The simulation results of liquid level moving in tubes of different diameters.



(a) Comparison of theory and simulation results with experimental data



(b) Error of theory and simulation with experimental data

Fig. 5. Comparison and error of theory and simulation results with experimental data.

It is also noteworthy that although the gas-liquid interface oscillation in the capillaries can be observed, the liquid level image was easily affected by the inner wall of the glass channel, as shown in Fig. 3. Besides, some liquid unexpectedly adhering to the glass channel wall interfered the experimental observation. Additionally, the dynamic monitoring of the gas-liquid interface in the capillary is originally faced with huge challenges and difficulties. Considering these factors, is not realistic to rely entirely on experimental method to carry out the research of liquid rising in the capillaries. From above comparison results, the simulated values of liquid level and its corresponding time were both reliable, with the most errors basically come from system errors, indicating the numerical simulation method was reliable. Moreover, the results obtained by numerical simulation methods are observable and maneuverable. To systematically study the dynamic movement process of liquid in single capillary and analyze in detail the influences of the capillary structural parameters, liquid physical parameters, and the gas-liquid interaction, the numerical simulation method was adopted to carry out the following research.

3.2. Dynamic meniscus moving process in capillary

The liquid in the capillary will move upwards and present a concave meniscus due to the hydrophilic capillary wall, or move downwards and present a convex meniscus due to the hydrophobic wall. Some researches [15,34–36] have studied the meniscus forming process in a semi-closed capillary, and find that the gas-liquid interface in the capillary changes dynamically rather than always maintain a circular fan shape, which has a very important reference for the understanding of the meniscus. Combining the dynamic change process of the meniscus with the liquid rising process in the capillary, this work simulated the dynamic liquid rising process in different capillary scenarios. The inner diameter was set as 1 mm, 2 mm, 3 mm, 4 mm, and 5 mm, respectively, and the contact angle of 30°, 60°, 120°, and 150°, respectively.

3.2.1. The gas-liquid interface oscillating process

Fig. 6 shows the phase line undergoes an oscillation process and finally stabilizes at the final equilibrium position. According to the phase line movement curves in Fig. 6, under the same contact angle condition, the smaller the inner diameter of the capillary,

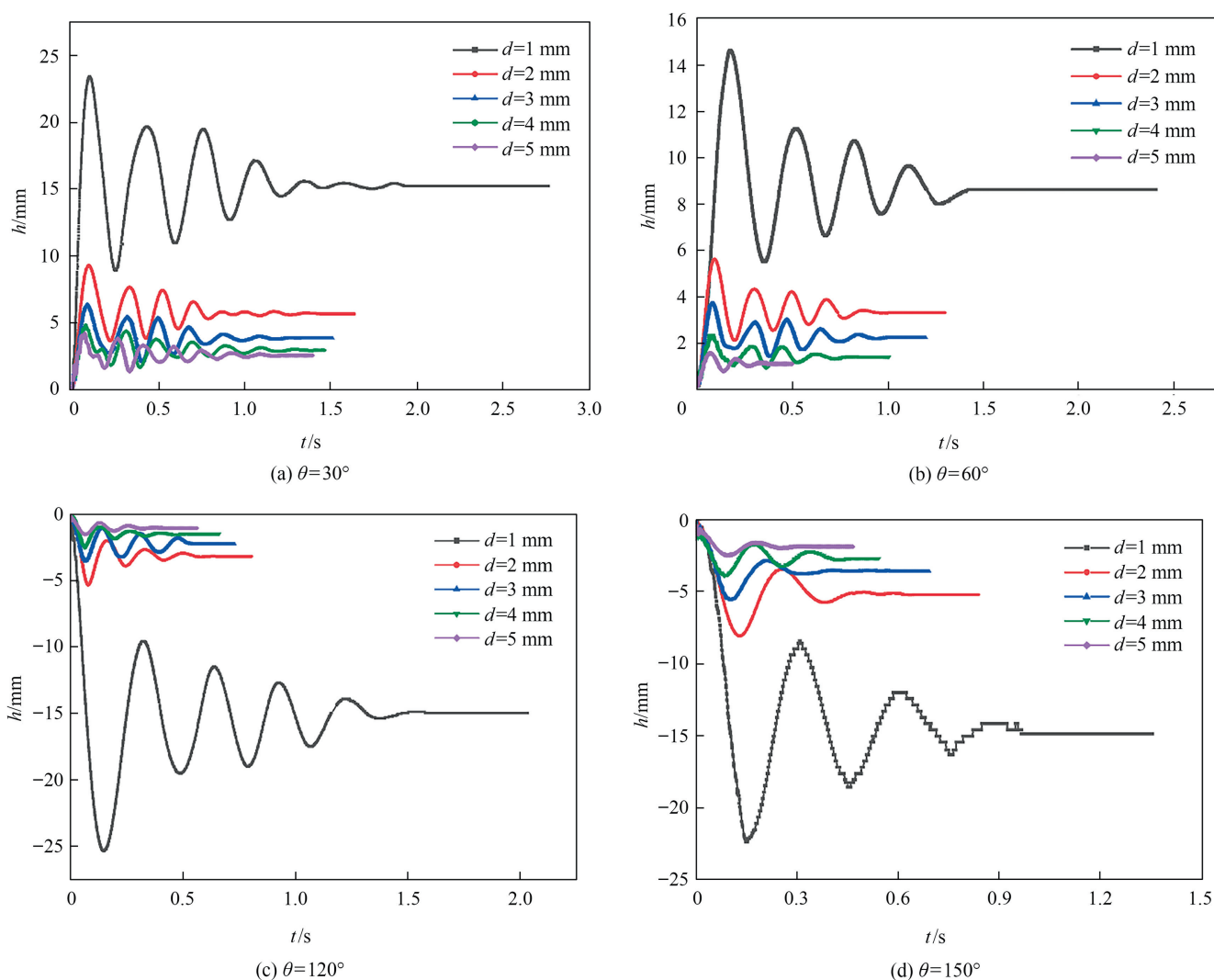


Fig. 6. Gas-liquid-solid phase line movement process in the capillary.

the longer the oscillation process. This phenomenon can be explained by the following.

The gas-liquid interface initially maintained a flat shape under the dynamic thermal equilibrium of the gas and liquid phases. When the capillary was immersed into the liquid phase, the original gas-liquid interface was destroyed by the channel cross section. The equilibrium state near the gas-liquid-solid phase line changed due to the difference of surface free energy between the gas and solid phases. The gas-liquid-solid phase had a toward the new final steady state, which has the minimum free energy. The hydrophilic wall had the tendency to form a concave surface, while the hydrophobic wall formed a convex surface. When the gas-liquid interface was no longer maintained horizontal, the gas-water interface deviated from the state of force balance, with their surface free energy constrained within the narrow capillary channel, forming the macroscopically said additional pressure. Meanwhile, as the capillary diameter became smaller, the role of gravity was gradually weakened, and the capillary pressure increased, providing larger initial energy for the liquid movement in the capillary. This is the reason why the amplitude is larger and the frequency is higher of the liquid oscillation process in the capillary channel with a smaller diameter.

The force analysis of the liquid sucked into the capillary channel was shown in Fig. 7. The additional pressure on the meniscus was

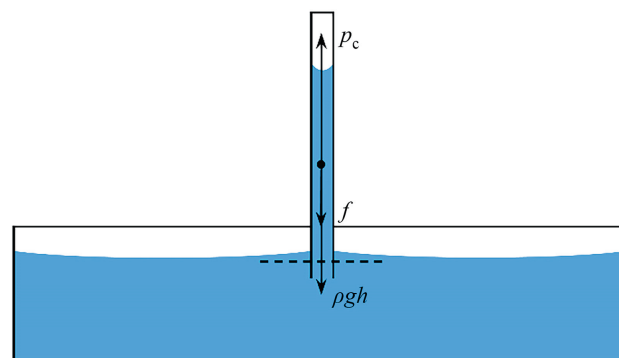


Fig. 7. Force analysis of the liquid level in the capillary.

the driving force term in the same direction as the upward movement of the liquid. The gravity force increased from zero, and with the viscous resistance together formed the resistance term in the opposite direction. Fig. 8 showed the schematic diagram of the liquid moving process during the capillary phenomena, Fig. 8(a)–(i) showed the moving characteristics of liquid level at different positions, where a represented the acceleration, v represented the moving velocity. The first solid arrow represented the direction,

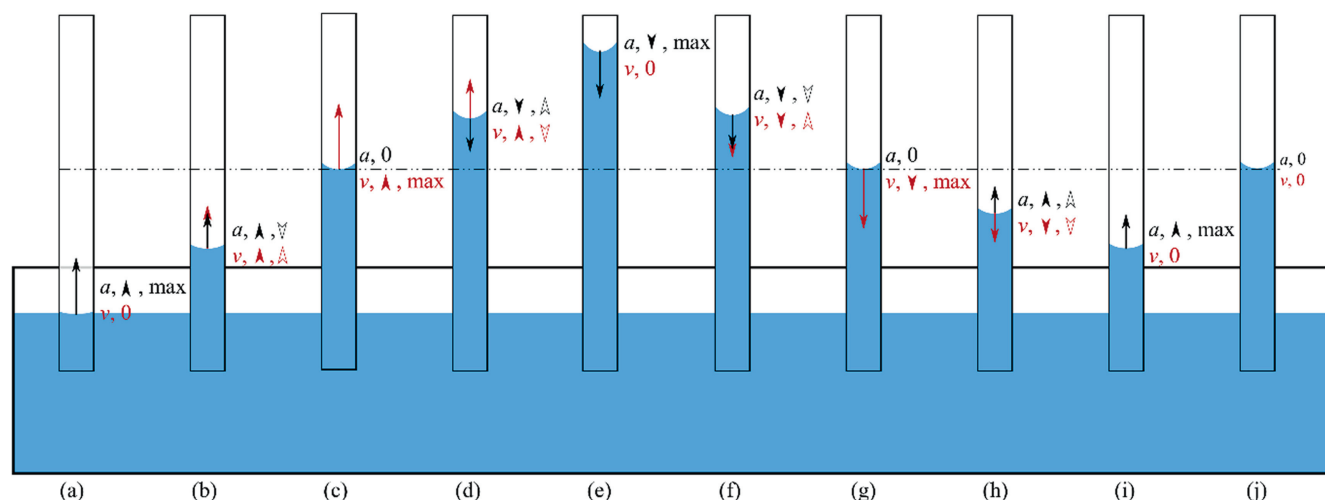


Fig. 8. Moving process analysis of liquid level in capillary.

and the second hollow one represented the changing trend. Before reaching the equilibrium level, the driving force of the additional pressure was greater than the resistance, resulting in the upward velocity and acceleration, as shown in Fig. 8(a). In Fig. 8(b), as the liquid level increased, the resistance increased, the acceleration decreased, and the velocity continually increased. When the liquid level reached to the equilibrium ones in Fig. 8(c), the resultant force on the liquid decreased to zero in the capillary, where the acceleration decreased to zero, the velocity was still upward and reached the maximum value. Therefore, the final steady state has not been reached at this time, and the liquid level continually rise, which also proves the rationality of the liquid oscillation process in capillary.

After the equilibrium level, the resistance on the liquid exceeded the driving force, the acceleration gradually increased downward and the velocity decreased, as shown in Fig. 8(d). When the velocity decreased to zero, the liquid reached the rising extreme position, that is, the first peak position in Fig. 8(e), and the acceleration reached the downward maximum. After that, the liquid fell with the downward acceleration decreasing and velocity increasing till the equilibrium level again, where the acceleration equaled to zero and the velocity reached the downward maximum value. Since the steady state has not been reached, the liquid continues to fall till the velocity reduces to zero, where was the first valley position and the acceleration is upward maximum in Fig. 8(i). Thus, the above process from Fig. 8(a)–(i) repeat until the liquid level stabilizes at the final equilibrium position as in Fig. 8(j), where both the acceleration and velocity decreases to zero. From above theoretical analysis, the process of liquid rising to form a stable meniscus in capillary is a dynamic oscillation process, rather than a one-way movement.

In the dynamic oscillation process, the peak position gradually decreasing each time can be contributed to the energy dissipation of the system. When the acceleration and velocity both reach to zero, the liquid level finally stabilizes at the equilibrium position. Fig. 6 showed the energy dissipation rate was related to the shape and structure parameters of the capillary channel, the physical properties of liquid and the gas-liquid interface interaction. In narrower capillaries, liquid level oscillates for a longer time with more oscillation peaks. Under the condition of the same capillary diameter, the more the contact angle deviates from 90° , the more violent the oscillation process is. From Fig. S5, the liquid surface deformed from the three-phase line, and then caused the deformation of the adjacent interface [31–33]. The similar phenomena take

place under the conditions of different capillary diameters and contact angles, is consistent with Tang Yicun's research [15].

Fig. 9 shows the movement of the three-phase line with time from the initial level to the first extreme value in different capillary channels with different contact angles. These curves are characterized as upward fluctuating in this initial period, which means the local oscillation process in these curves. According to Young-Equation, the gas and liquid molecules would move due to the unbalance state of surface free energy on the three-phase interface [37–39].

While, the essence of capillary phenomenon is also a process of the three-phase interface obtaining a new balance state, during which the liquid surface gradually formed a meniscus. In actual capillary phenomena, the flat gas-liquid-solid phase interface deformed due to the original equilibrium was destroyed, forcing the liquid surface to deviate from their original position and move up along the inner wall of the capillary. According to the assumption of fluid continuity, the water adjacent the three-phase line were deviated from their original position, effecting the movement of the whole gas-liquid interface. Considering the reverse effect, the water on the three-phase line will be constantly affected by the surrounding gas, liquid, and solid phases, changing their force state, and resulting in periodic acceleration and velocity changes. That was the reason of the fluctuation stage at the beginning of the three-phase line oscillation process. With the liquid level rising, the difference in the acceleration and velocity of the water phase decreased. When a stable equilibrium was finally reached, the movement curves gradually became smooth and the fluctuation periods disappeared.

Richard [27,40] used to mention the phenomenon of time relaxation, which is also consistent with the above analysis. The force on the liquid phase near the three-phase line varies dynamically, resulting in the acceleration and velocity changing constantly. Due to the continuity of the fluid, the motion state of the surrounding liquid keeps changing constantly also. From a macro point of view, the liquid on the three-phase line drive the entire gas-liquid interface being deformed, leading the contact angle also to change until the liquid forms a stable meniscus. The hysteresis time gradually decreased with the capillary diameter increasing in Fig. 9. From the analysis above, the effect of gravity force on the liquid weakens in narrower capillaries. In capillaries with large diameters, the additional pressure becomes smaller, the influence of gravity becomes more obvious, resulting in a smaller acceleration change of the liquid in capillaries. In such situation, the liquid

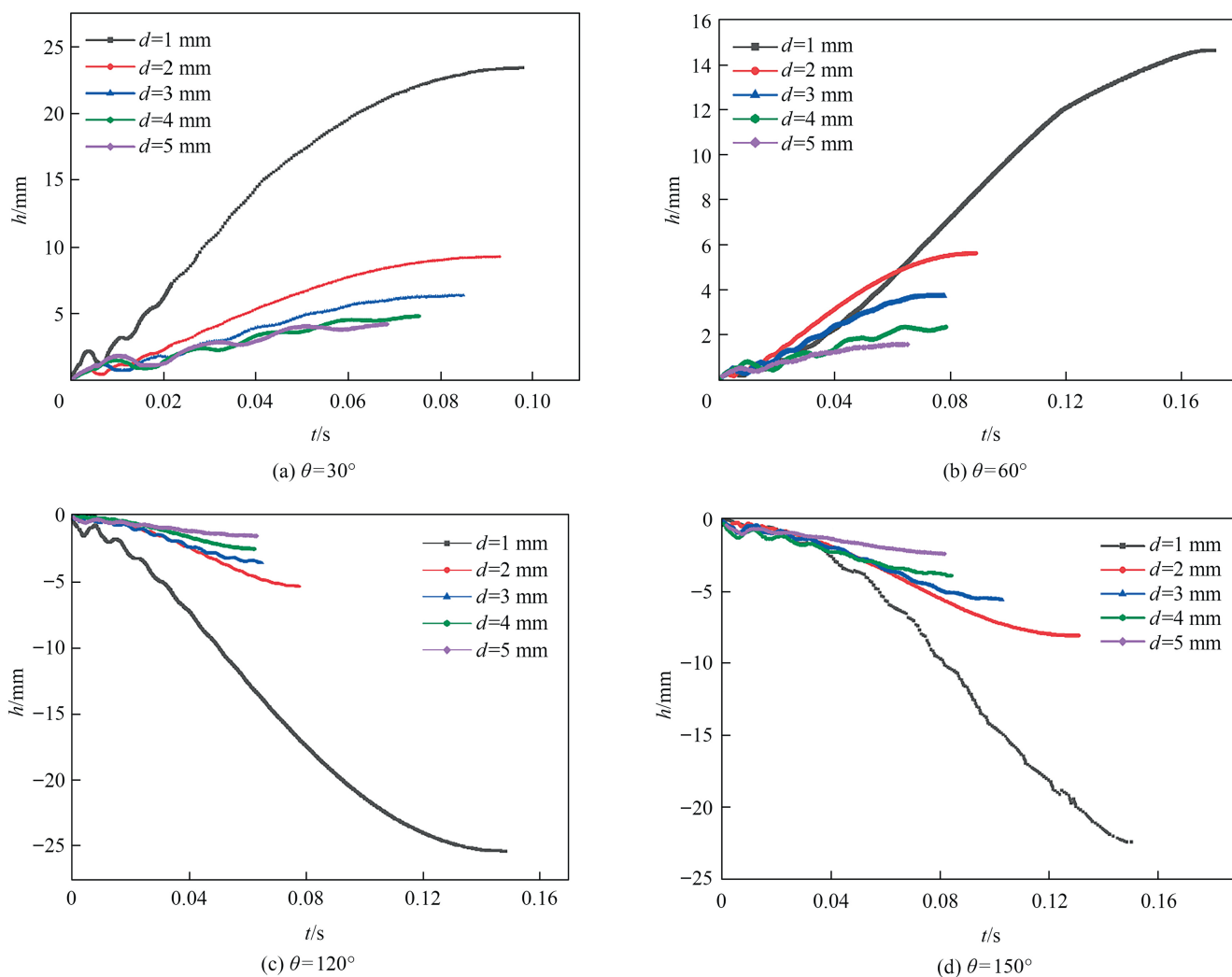


Fig. 9. The extreme position of the liquid oscillation process in capillary.

level reached the extreme positions with the liquid level changing slightly, easily reaching their equilibrium state.

3.2.2. Motion equation of gas-liquid-solid phase line

Different from the traditional permeation models, this research directly studied the liquid movement in a single capillary from the microscopic point of view. From the above analysis, the meniscus in the capillary undergoes several oscillations before reaching the equilibrium position, and so does the gas-liquid-solid phase line. It can be seen in Fig. 6, the peak amplitude, oscillating rate, and duration of the movement curve vary with pipe diameters and different contact angles. This research studied the oscillating pattern and attenuating characteristics of the three-phase line movement curves with mathematical method.

In Fig. 10, the black solid line represents the actual three-phase line movement in the capillary with the diameter of 2 mm and contact angle of 30°, the blue dashed line represents the equilibrium position, which is parallel to the abscissa and can be described as $h = h_0$, and the red dotted line is the attenuation curve, which is determined by the peak and trough positions of the three-phase line motion curve. Therefore, this research approximately described the three-phase line movement curve using a mathematical function of the oscillation attenuation function as follows:

$$h(t) = h' \exp(\alpha t) \sin(\omega t + \varphi) + h_0 \quad (3)$$

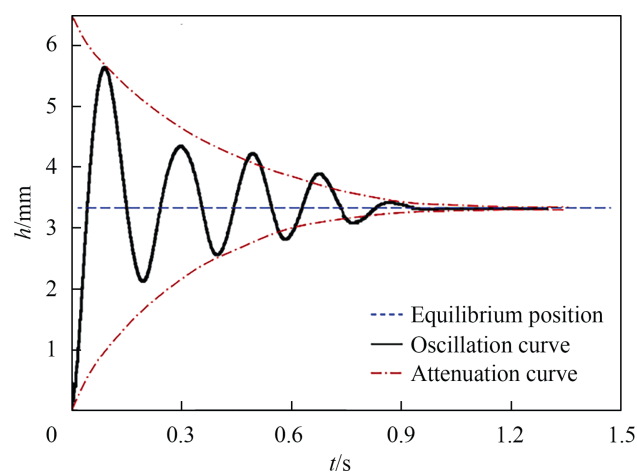


Fig. 10. Oscillation attenuation curve of gas-liquid-solid phase line.

Eq. (3) described the dynamic physical process of the three-phase line oscillating while rising in the capillary, where the first term described the oscillation process of the three-phase line, and the second term described the final static equilibrium position of the meniscus, expressed as h_0 . The physical meaning of h' , $\hat{t}_{\pm}$, ω , φ in Eq. (3) were illustrated as follows.

(1) Oscillating extreme height, h'

The envelope of the three-phase line movement curves can be shaped as an attenuation curves group, as shown in Fig. 10. According to the graphical characteristics of the curve of

$$y = \pm A \exp(Bx) \quad (4)$$

where the intercepts at $x = 0$ are coordinated at $(0, A)$ and $(0, -A)$, and the line of $y = 0$ is the asymptote where the curve eventually stabilizes. According to the positions of all the peaks and valleys in Fig. 10, the attenuation curves couple of

$$h = \pm h' \exp(\alpha t) \quad (5)$$

is plotted, and the asymptote is positioned at $h = h_0$. The upper attenuation curve intersects the y-axis at point $(0, h(0))$, and the lower attenuation curve at $(0, 0)$. When the attenuation curves couple is assumed to be moved by the distance of h_0 , the original asymptote reaches the position of $h = 0$, the intersection points are moved to $(0, h')$ and $(0, -h')$, respectively. Thus, the value of h' can be calculated numerically as follows:

$$0 - h_0 = -h' \quad (6)$$

$$h' = h_0 \quad (7)$$

Fig. 11 showed the value of h' under different capillary diameters and contact angles. According to Eq. (2), the inverse proportional function was used to fit the data points in Fig. 11. The value of adjusted R^2 exceeded 0.989 for every fitted curve, illustrating the inverse proportional function fitted the data points well.

According to the capillary equation, the liquid level rising distance for $\theta < 90^\circ$ was approximately equal to the falling distance for $\theta > 90^\circ$ when $\hat{I}_1$ deviated the same degree from 90° , where the contact angle only changed the direction without changing the distance, as shown in Fig. 12. The parabolas with the line of $x = 90^\circ$ as the axis of symmetry fitted these data points well with all the adjusted R^2 bigger than 0.989. And the further work would be carried out to study the influences of the capillary structure, liquid physical properties, and interface interaction on the parabola shape. Thus, the value of h' under the given contact angle condition can be calculated according to the parabola curve.

Under the condition of $\theta = 90^\circ$, the oscillating attenuating term on the right of the equal sign in Eq. (3) equals to 0, that is:

$$h' \exp(\alpha t) \sin(\omega t + \varphi) = 0 \quad (8)$$

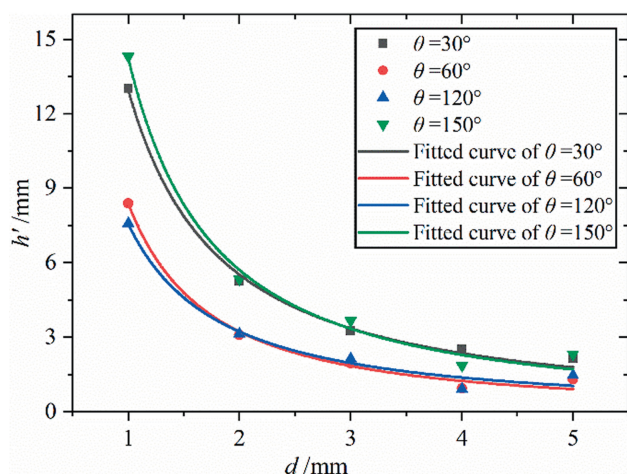


Fig. 11. The values of h' under different capillary diameters and contact angles conditions.

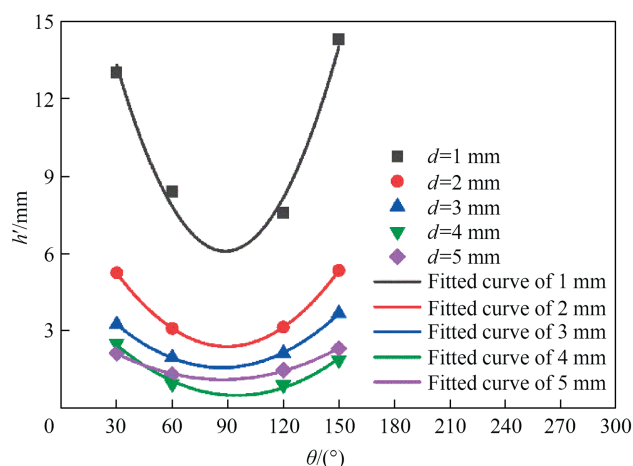


Fig. 12. The values of h' versus contact angles in every single capillary.

$$h(t) = h_0, \quad \theta = 90^\circ \quad (9)$$

The liquid level remains stationary, the oscillating and attenuating process disappears in the three-phase line movement curve, where $h' = 0$, $\alpha = 0$, $\omega = 0$, $\varphi = 0$. Thus, the fitted parabola curve in Fig. 12 is not suitable for the case of $\theta = 90^\circ$.

From the comparison of the simulation and theoretical calculation results, the value of h' is modified by the correction coefficient, β , which describes the deviation of the equilibrium liquid level:

$$h' = h_0 = 2\beta \sigma \cos \theta / \Delta \rho g r, \quad \theta \neq 90^\circ \quad (10a)$$

$$h' = 0, h_0 = 0, \theta = 90^\circ \quad (10b)$$

(2) Attenuation coefficient, α

The value of α mathematically represents the magnitude of the attenuating rate of the attenuation curves in Fig. 11. The larger α is, the steeper the curve is, thus making α as the attenuation rate during the liquid level oscillation process. The attenuation rate, α , is closely related to the energy dissipation. The faster the energy dissipation, the larger the attenuation rate is and the faster the attenuation curve decreases. From the overall trend, the attenuation rate increased with the contact angle increasing in Table 2. Due to the equation of

$$\cos(\pi - \theta) = -\cos(\theta) \quad (11)$$

the equilibrium positions shared the same distances but inverse directions under the contact angle condition of $\theta_1 + \theta_2 = \pi$. In the initial stage, the additional pressure was smaller, and gravity played a major role. With the naturally vertical downwards, gravity had a blocking effect on the oscillating process when liquid rising for $\theta < 90^\circ$, but a promoting effect when liquid falling for $\theta > 90^\circ$. Thus, the curve decayed faster to the stable state when $\theta > 90^\circ$.

Under the same contact angle condition, the attenuation rate increased first and then decreased with diameter increasing. In narrower capillaries, the capillary effect plays a major role rather than gravity. With capillary diameter increases, the gravity effect strengthens, increasing the attenuation rate. However, when the capillary was large enough, the capillary effect was very weak, weakening the volatility of the movement curves. Thus, the attenuation rate decreased with the diameter increasing. In summary, the attenuation rate is the result of the combined effect of the additional pressure and gravity.

Table 2The attenuation rate, α , under different diameter and contact angle conditions

Contact angle $\theta/(^\circ)$	Capillary diameter d/mm				
	1	2	3	4	5
30	−2.1866	−2.9698	−2.1852	−2.2460	−2.3780
60	−2.1561	−3.438	−3.0873	−8.7995	−3.7605
120	−2.1714	−5.7149	−2.2252	−5.2226	−7.1451
150	−3.0224	−5.2100	−9.1359	−11.8308	−5.7989

(3) Oscillating rate, ω

The parameter of ω reflects the oscillating characteristic of the curve in Fig. 6. During the liquid level rising, the three-phase line still constantly oscillates around the inner capillary wall, resulting the gas-liquid interface fluctuating. From the oscillation attenuation curve in Fig. 6, the three-phase line both undergoes a quarter of the oscillation period, $T/4$, from the initial position to the first peak position and from the first peak to the first valley position. The oscillation period has not remained constant, but continuously decreases during the whole liquid rising process. According to the equation of

$$\omega = 2\pi f = 2\pi/T \quad (12)$$

the oscillating frequency becomes larger, and then the oscillating rate, ω , also becomes larger. The energy dissipation occurs during the whole oscillation process. Thus, the amplitudes of the peaks and valleys continue to decrease, making it easier for the gas-liquid interface to reach the equilibrium position, then the period becomes shorter. At the extreme positions of peaks and valleys, the moving velocity of the three-phase line reduces to zero, and the acceleration obtains its maximum value. The duration between two adjacent peaks or valleys is about the half of the oscillation period, $T/2$. Besides, setting the equilibrium position as zero potential, the absolute value of the height reduction between the adjacent peak and valley can be regarded as the energy dissipation.

(4) Initial time phase, φ

In the sine function of

$$y(t) = A \sin(\omega t + \varphi) \quad (13)$$

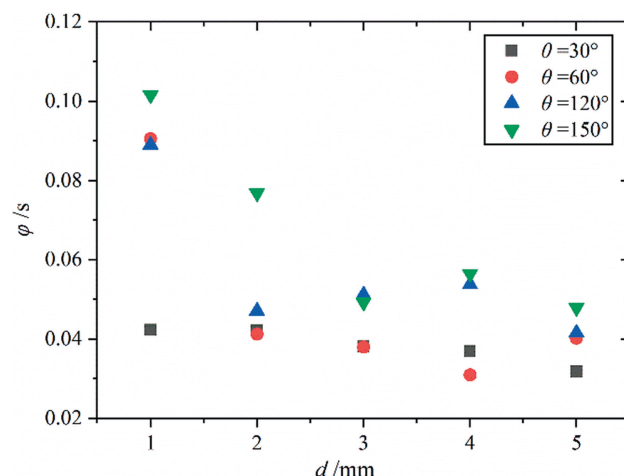
where φ is the initial phase of the sine wave. From the oscillation and attenuation curve in Fig. 6, φ represents the initial phase of the oscillation attenuation curve, which means the time when the liquid level reaches the equilibrium position for the first time:

$$\varphi = T_1/4 \quad (14)$$

where T_1 is the first oscillation period of the three-phase line movement curve. When the capillary channel was inserted into the water surface, potential energy was generated at the gas-liquid interface in the capillary. The initial potential together with the capillary structure not only determined the equilibrium position of the liquid level, but also determined its initial motion state. In narrower capillaries, the gravity effect is weaker, and the initial potential energy is greater. Then, the liquid rises faster in the initial stage, lasting a longer time. The initial time phase φ in Eq. (3) becomes larger with diameter decreasing, as shown in Fig. 13.

Based above analysis, the oscillation attenuation curve can well describe the three-phase line movement versus time in capillary. According to the illustration of all the parameters of the oscillation attenuation function, the three-phase line movement versus of liquid in capillary was expressed as follows:

$$h(t) = 2\beta\sigma\cos\theta/\Delta\rho g r \cdot \exp(\alpha t) \sin(\omega t - T_1/4) + 2\beta\sigma\cos\theta/\Delta\rho g r, \quad \theta \neq 90^\circ \quad (15a)$$

**Fig. 13.** The initial phase under different diameter and angle conditions.

$$h(t) = 0, \quad \theta = 90^\circ \quad (15b)$$

where h is the liquid level, mm; t is the time, s; β is the correction coefficient of the equilibrium position height; σ is the surface tension of liquid, $\text{N}\cdot\text{m}^{-1}$; θ is the contact angle, degree; $\Delta\rho$ is the density difference between the liquid and gas, $\text{kg}\cdot\text{m}^{-3}$; and g is the gravity, $\text{m}\cdot\text{s}^{-2}$; r is the radius of curvature of the meniscus, m; α is the attenuation rate; ω is the oscillating rate, $\text{rad}\cdot\text{s}^{-1}$; T_1 is the first oscillation period, s. The mathematical function included two terms, where the first term represented the oscillating and attenuating characteristics of three-phase line, and the second term was the equilibrium. Although there were still some fluid dynamics parameters not considered in Eq. (3), like the density, viscosity, the influence of these parameters was reflected in the coefficients of α , ω , φ , β . As a preliminary attempt, this equation well reflected the oscillation attenuation characteristics of liquid rising in the capillary with a certain degree of accuracy and reliability based on the verified numerical simulation method. It can provide new research ideas and reference for subsequent related research to investigating the dynamic moving process of meniscus in capillary. Subsequent work would continue to discuss in detail the influence of other fluid mechanic's parameters and capillary structure parameters to further modify and improve this equation, providing a supplement to the macroscopic permeation equations. Furthermore, from the oscillation attenuation function, there were several extreme positions above or below the final stable equilibrium positions, which provided some new thoughts for the liquid permeation strengthen in real porous media. The extreme position exceeded the equilibrium position to indicate the longest permeation distance of liquid flow along the real capillaries.

3.3. Transient simulation of liquid permeation in granular materials stacks

3.3.1. Peak level of capillary rising

Considering the actual solid fire accidents and the corresponding disposal for smoldering fire, it is expected to promote the liquid

penetrating into the deeper position of the porous media quickly. From Eqs. (1) and (2), changing the liquid density, viscosity, and surface tension is the main method to improve the permeation of fire extinguishing agent. Many researches pay more attentions on how to change the liquid properties, such as through adding surfactants to reduce the surface tension and then to promote the liquid permeation in the porous medium stacks. While, there are few studies focus on the structural properties of channels in the porous medium stacks.

In actual situations, the extremum positions the liquid level can reach have more practical application value, such as in fields of coating film, pharmacy, liquid transportation *etc.* From the comparison results in Fig. S6, the extreme positions were about 1.6 times higher than the corresponding equilibrium position in most simulations, regardless of whether the liquid level rises or falls. The maximum multiple reached up to about 1.85 times, with the capillary diameter of 2 mm and contact angle of 60° . Therefore, the farthest distance of the meniscus rising in the capillary is much higher than the final stable equilibrium position, which is of great reference value for research in related fields.

Meanwhile, regardless there still existed error between the simulation result and the theoretical value of the capillary equation, the simulated equilibrium liquid level was inversely proportional to the capillary diameter, and proportional to the cosine value of the contact angle. Interestingly, the extreme liquid level also followed the similar relationship. This phenomenon was consistent with Eq. (2), which meaning the theoretical calculation results needed to be corrected by certain parameters to approach the real situation. Also, as the capillary pore size became smaller, the liquid penetrated to farther positions.

3.3.2. The “relay-mode” in real granular coal stacks

From the oscillation attenuation function of Eq. (15), the liquid level can move to higher than the equilibrium level, which is closely related to the capillary diameter and liquid properties. Considering the essence of liquid transport in granular materials porous media is the displacement process of the gas phase in the pores by the liquid phase, the process of liquid transporting in porous media can be significantly affected by the size of pore throats. In actual granular materials stacks, the pore structure is very complicated, and the change of the internal pore throats will also affect the liquid transport. In capillary calculations, the capillary equilibrium level is usually obtained according to the Young-Laplace equation or the capillary equation. However, according to the above analysis, the extreme position of the wetting liquid rising in the capillary exceeds its equilibrium position. According to Fig. S6, the heights of equilibrium positions and extreme positions of the liquid rising were inversely proportional to the inner diameter of the capillaries. When the liquid in capillary reached the extreme position, the velocity dropped to zero, the acceleration was opposite to the velocity, and the liquid stopped rising. If the capillary is narrowed before the liquid velocity drops to zero, the resultant force state of the liquid will be changed, increasing the driving force item so that the liquid can maintain the current moving state and continue to rise. This process is named “relay-mode” process, which describes the process of keeping liquid in continuous motion by reducing the capillary diameter when the liquid level moves to an extreme position.

In order to illustrate the role of the “relay-mode” process in actual liquid permeation process, a real granular materials porous medium model was established based on the actual 200 mesh pulverized coal stack to simulate the internal liquid permeation process [41], as shown in Fig. S7, and the porosity was measured as 49.09%. With image processing technologies, the effective flow field pores of the coal sample were extracted, as shown in Fig. S8.

The solid framework of the granular materials porous media mainly formed by the mutual bonding between particles, and the spaces not filled by porous media particles constitute the pore throats, which are full of gas phase. Considering the small size capillary pores followed by tiny grids, which can be 0.0001 mm, it costs much more calculation resource and simulation time to carry out the full-model transient simulation. The framed region in the full-scale flow field model in Fig. S8 was selected as the simulation flow field, as shown in Fig. 14. The local pores region shares the same typical spacial structure characteristics of granular materials porous media, the pore size distribution in the simulation area is almost the same as the pore size distribution in the full-model flow field. Thus, it is reasonable to study the liquid permeation process in the local simulation area in Fig. 14 for the research of liquid displacing air phase in real granular materials porous media.

In the transient simulation, VOF model was adjusted to set the initial liquid level to simulate the state when the water just touches the upper surface of the granular porous media stacks. The water phase was set as the primary phase, and the air was the secondary phase. In surface tension force modeling in the phase interaction, the wall adhesion was selected as the wall adhesion option, where the surface tension on the liquid–air interface was set as $0.07275 \text{ N}\cdot\text{m}^{-1}$. The upper line was set as Velocity Inlet as the water injection. For transient simulation with small time step coming with large flowing velocity, the calculation amount was usually large with a slow calculating speed. This research set the initial water inlet velocity as $0.2 \text{ mm}\cdot\text{s}^{-1}$ and time step as $1.0 \times 10^{-5} \text{ ms}$ to ensure the transient simulation convergence. After the residuals of the continuity equation and velocity equation in the *x* and *y* directions stabilized, the time step was adjusted to $1.0 \times 10^{-4} \text{ ms}$ and the inlet velocity was improved to $1.2 \text{ mm}\cdot\text{s}^{-1}$ and then $2.0 \text{ mm}\cdot\text{s}^{-1}$.

The permeation process of liquid front end in the real granular materials porous media pores is shown in Fig. 15, the figures (a)–(p) show the permeation state at different time points from the initial liquid injection to 0.02 s. The liquid front end undergoes varying movement states in the completed capillaries. Even the fluctuation of gas–liquid interface can be still observed, it is not that obvious like in the single capillary. From the overall permeation process in Fig. 15, it is obvious that more liquid flowed in the right part of the pores, while less liquid existed in the left and middle parts in the flow field. On one hand, the simulation time is too short as about 0.02 s due to the tiny time step of

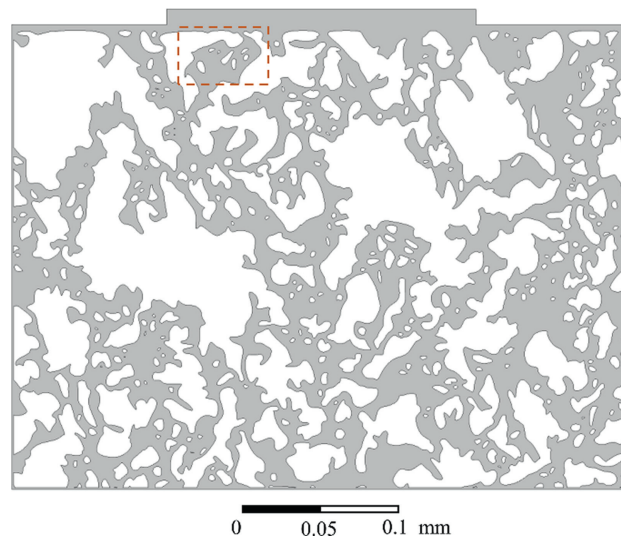


Fig. 14. The local simulation flow field in real pulverized coal stacks.

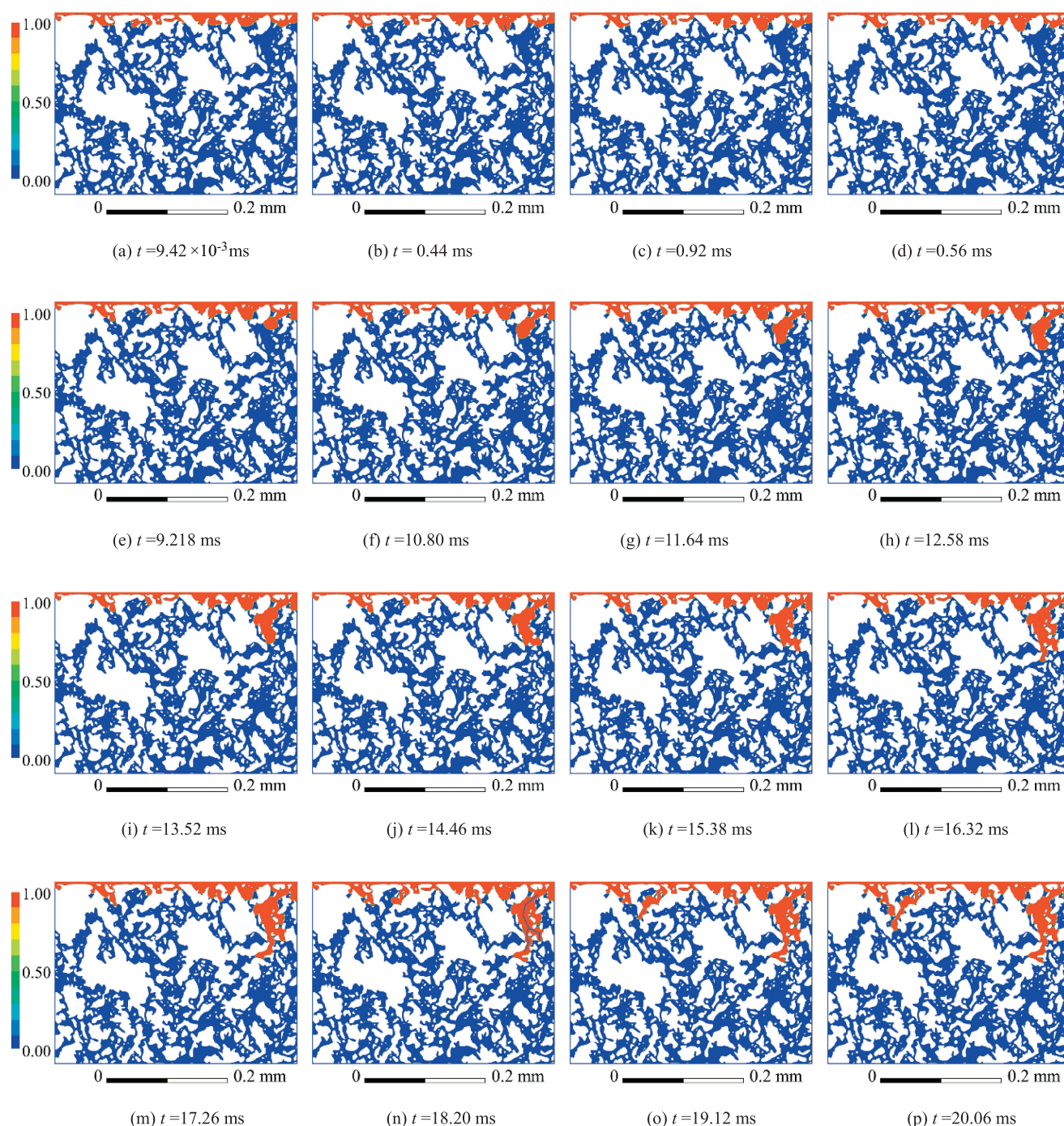


Fig. 15. Movement states of permeation liquid front end in pores of granular materials porous media.

1.0×10^{-5} ms or 0.01 ms, so the distance of liquid permeation is also short. On the other hand, the pore size played a significant role in the liquid permeation process.

The four dotted lines in Fig. 16 to represents the permeation paths namely A, B, C, D at different pores at the time of 0.02 s. The pore size of the permeation paths A, B, C on the left and the middle part are smaller, and the pore changes along the permeation paths are also smaller. The pore size along permeation path A is approximately from 8.45 μm to 5.00 μm and then to 11.71 μm , and permeation path B is from 6.1 μm to 13.6 μm to 9.4 μm and then to 6.0 μm , while, path C is from 5.5 μm to 5.1 μm and then to 6.1 μm . According to the gas-liquid interface

movement process in capillaries with different diameters and the analysis of “relay-mode” process, the additional pressure due to the capillary was the main driving force in such size pores. However, the size of the pores does not change a lot, so the liquid in these pores in this area cannot permeation downward quickly. It is necessary to increase the upward liquid replenishment above to promote the liquid permeation. At the time of 0.02 s, the lengths of permeation paths A, B, C are approximately 45.54 μm , 88.64 μm and 20.58 μm , and the average permeation flow rates of 2.28 $\text{mm}\cdot\text{s}^{-1}$, 4.44 $\text{mm}\cdot\text{s}^{-1}$ and 1.03 $\text{mm}\cdot\text{s}^{-1}$, respectively.

While, along the permeation path D in the right area, the pore where the liquid front end initially occupied was relatively larger,

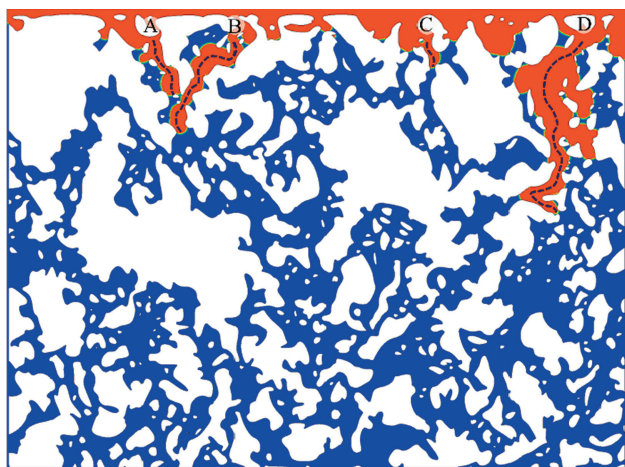


Fig. 16. Liquid permeation paths in granular coal stacks at $t = 0.02$ s.

which was about $41.17\ \mu\text{m}$. thus, the effect of gravity to promote liquid permeation downward was stronger. Besides, the pore size along path D decreased quickly, and the additional pressure was

also strengthened. The pore size decreased to $17.56\text{--}8.43\ \mu\text{m}$, and finally to $3.91\ \mu\text{m}$, and the distance was about $149.26\ \mu\text{m}$ with the average flowing velocity of $7.46\ \text{mm}\cdot\text{s}^{-1}$, as shown in Fig. 16. It is worth noting that more liquid flowed along the permeation path D, the flowing velocity can be about 1.68, 3.28, and 7.25 times of that in paths A, B, C, respectively. That means changing pore size can be used to adjust the movement state of the permeation front end in porous media stacks. And in narrower pores or when the pore size changes greatly, this trend might be more obvious.

Additionally, in the pore throats which enlarged along the permeation paths, the liquid front end was obstructed due to the “relay-mode” process. The flow field region in Fig. 17 was framed in Fig. 14, and the Fig. 17 (a)–(p) showed the dynamic movement of the liquid front end in the pore throats in the real granular porous media stacks, where the upward pore size was smaller than the downward pore. The dot line with double arrows in every figure represented the pore size of the position where the liquid front end flowed. In the initial stage, the small water supply came from the low flow rate. From $0.009\ \text{ms}$ to $0.180\ \text{ms}$, the liquid front end retreated from the larger pores to the smaller pores. As analyzed above, the effect of additional pressure was more obvious than the effect of gravity in smaller pores. The capillary force was the main driving force then. Thus, the phenomenon also proves that



Fig. 17. The dynamic changing of the liquid front end in the pores.

in the real capillary pore throats, the liquid has the tendency of moving from larger size pores to smaller size pores, which is also a manifestation of the “relay-mode” process in the liquid permeation in granular materials porous media stacks.

From Fig. 17(i)–(p), in the same time interval, liquid front ends moved faster and to longer distance in smaller size pores. From Fig. 17(i)–(l), the pore size less changed from 10.079 μm to 9.139 μm . The liquid front end slowly moved about 2.756 μm in 0.030 ms, and the average flowing velocity was about 0.092 $\text{m}\cdot\text{s}^{-1}$. While, from Fig. 17(m)–(p), the liquid front end moved significantly faster. The pore size changed from 6.414×10^{-3} mm in Fig. 17(m) to 1.874 μm in Fig. 17(n), and the liquid front end flowed at the average velocity of about 0.562 $\text{m}\cdot\text{s}^{-1}$. From Fig. 17(n)–(o), the pore size changed from 4.936 μm to 3.957 μm , the liquid front end moved about 5.654 μm at the average velocity of about 0.565 $\text{m}\cdot\text{s}^{-1}$. As analyzed about the “relay-mode” process, the liquid trends to move from large pores to small pores in granular materials porous media. From the comparison of the liquid front end positions in every figure in Fig. 17, the change of pore size significantly affected the movement state of the liquid front end. Under the action of the surface tension adjacent the three-phase line, the shape of the liquid front end deformed accordingly.

For granular materials porous media stacks in actual production and life, the “relay-mode” process can be used as a reference according to different purposes, which is accelerating the movement of liquid front end by reducing the pore size, and slow down the liquid permeation by enlarging the pore size. The conclusion has certain reference value and guiding significance for the pretreatment and stacking methods of the granular porous media stacks. Besides, this research provides a new thought of developing water-based fire extinguishing agents for solid deep-seated fire, which can be combined with the stacking methods improvement to promote the liquid permeation in such porous media stacks.

4. Conclusions

With the combination of capillary experiments and a series of transient capillary simulation with different diameters and contact angles, this research studied the movement characteristics of gas-liquid-solid phase line in capillary, the influences of which were analyzed in details. Based on the concept of “relay-mode”, the transient simulation of water permeation in real coal porous media stacks was performed to investigate the pore size significantly influence the liquid permeation process in actual granular materials stacks. The relevant results in this research are expected to provide new thoughts for the development of water-based fire extinguishing agents for deep-seated fires and then improve the firefighting efficiency of smoldering fire in porous media stacks. The further related research is also in preparation. And the following conclusions can be drawn:

During the dynamic rising or falling process of liquid in capillaries, the initial horizontal solid-liquid-gas interface underwent several oscillations before forming a stable meniscus at the eventually equilibrium position. The extreme position from the initial liquid level was about 1.6 times even 1.8 times of the equilibrium position in different simulations. In regard to the relationship with capillary diameter and contacting angle, the extreme position shared a similar law with the equilibrium position.

An oscillation and attenuation function was proposed to mathematically describe the dynamic moving process of the three-phase line in capillaries. The physical meaning of these parameters in the oscillation attenuation function were illustrated in detail with the simulation analysis, which lays the foundation for future work in related fields.

The liquid had an obvious tendency to move to the smaller capillaries during the permeation process, mainly being drove by the additional pressure in such pores. Meanwhile, the greater the pore size decreased, the more obvious the above tendency, and the faster the liquid permeation. The flowing distance and velocity in the narrowing path can be several times larger than that in such paths, where the pore size changes little, based on which the concept of “relay mode” was proposed.

The simulation of “relay-mode” was the preliminary attempt to promote liquid permeating in granular material stacks based on adjusting the shape and structure parameters of the pores in stacks. This is different from the traditional adding surfactants to change the liquid parameters. By adjusting the pre-processing and stacking method of materials, the spacial structure of the porous media can be more reasonable to promote the water-based extinguishing agent to flow through such self-heating substances stacks.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary Material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.cjche.2021.05.027>.

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