1. Introduction

Concrete is a complex composite material, the cement paste matrix of which is formed by the hydration of cementitious materials including Portland cement and supplementary cementitious materials (SCMs). Owing to the porous nature, the aggressive species from the surrounding environment, e.g., water, chloride, and carbon dioxide, can transport into the interior of concrete via its porous matrix, leading to its performance degradation and durability reduction (Liu et al. 2020a; Nukushina et al. 2021). During the service process, concrete is inevitably subject to damage under external service loads. The loading damage is normally insufficient to result in significant degradation to the performance of concrete (Picandet et al. 2001). However, the presence of damage adversely modifies the transport properties of concrete since the damage-induced microcracks facilitate a path of aggressive species (Li et al. 2019). Transport properties including diffusivity and permeability are usually regarded as the important indicators that directly govern the ingress of aggressive species to evaluate the concrete durability (Sakai 2017; Nguyen et al. 2020; Liu and Zhang 2021). As a result, accurate understanding the influence of mechanical damage on the transport properties of cementitious materials is crucial and significant for their durability design.

Permeability is one of fundamental transport-related properties of cementitious materials, which is defined as the movement of fluid through the porous medium under an applied pressure (Li et al. 2017). Over the last few decades, both experimental and modelling attentions have been paid to the effect of mechanical damage on permeability of cementitious materials. Regarding experimental investigations, Breyssse et al. (1994) investigated the water permeability of concrete with tension-induced microcracks and declared that water permeability perpendicular to the direction of tensile loading increases with the increasing damage degree. Picandet et al. (2001) experimentally examined the effect of axial compressive loading-induced damage on the gas permeability of concrete, and suggested that the gas permeability increases with the load-induced strain due to the increasing connection of the microcrack network. Choinhska et al. (2007) investigated the effect of compressive loading on gas permeability of concrete subjected to various temperatures. The results indicated that the variation of permeability is less dependent on the stress levels lower than 80%, while a significant increase of permeability can be observed as the load exceeds 80% of the peak stress. Zhou et al. (2012b) quali-
fied the compressive load-induced damage of concrete with ultrasonic pulse velocity and declared that the air permeability is significantly influenced by the microcracking density. Tegguer et al. (2013) experimentally correlated the change in chloride diffusivity and gas permeability with the increasing uniaxial compressive load indicated as residual strain and the relative decrease in elastic modulus (i.e., so-called damage value) on concrete. They found that transport properties both increase with the increase of compressive damage and the gas permeability shows an exponential correlation with chloride diffusivity for damaged concrete. Based on the aforementioned experimental analysis, it is generally accepted that the damage-induced microcracks in the cement paste matrix should be responsible for the increasing permeability of concrete. Although some meaningful findings can be observed from the experimental results, it is still difficult to experimentally quantify the relationship between permeability and mechanical damage of concrete because of complex and uncontrollable prefabricated procedure for mechanical damage and limited characterisation methods for microcracking apart from the time-consuming experimental process.

Modelling is an alternative tool to investigate the permeability in mechanically damaged cementitious materials without the above-mentioned drawbacks. For example, Chatzigeorgiou et al. (2005) used a discrete lattice model to obtain the mechanical damage-permeability coupling in homogeneous concrete. They demonstrated that there is an intrinsic relationship between permeability and damage degree, independent on the type of concrete. To take the heterogeneity of concrete into account, Raghavan et al. (2016) used mesoscale hygro-mechanical modelling with a morphological 3D two-phase mortar-aggregate model to investigate the coupling between mechanical loading and the permeability of the concrete and concluded that the aggregate content has less effect on the increased rate of permeability as a function of the ratio of stain over that at the peak of the compression curve. According to the damage mechanics of concrete at mesoscale in conjunction with Monte Carlo simulation and finite element analysis, Wimalasiri et al. (2018) developed a 2D methodology to simulate the effects of aggregate attributes on the permeability of stressed concrete. They revealed that the aggregate content is the most influential factor for permeability degradation of concrete and the aggregates with more sharper edges tend to result in higher permeabilities at a given damaged state. However, these aforementioned investigations were all focused on the simulation at mesoscale or macroscale without considering the heterogeneity of the cement paste matrix. Microscopically, the failure of concrete under various types of stresses is regarded as the consequence of the crack initiation and propagation of cement paste matrix at highly localised regions under large tensile stress concentration (Bernard et al. 2008). As a result, the tensile behaviour of the cement paste matrix is very important for the failure mode of concrete. Nevertheless, no related study has been reported regarding the effect of mechanical damage on permeability of cement paste at microscale until now. Furthermore, in addition to the mechanical damage, the cementitious materials in practice are partially saturated, the transport properties of which are moisture-dependent. The coupling effects of mechanical damage and moisture conditions (water content and moisture distribution) on transport properties remain poorly understandable.

To fill this gap, the objective of this study is to investigate the gas permeability in partially saturated cement paste with mechanical damage. An integrated finite element-lattice Boltzmann modelling framework was proposed to investigate the coupling effects of mechanical damage and moisture conditions on gas permeability of heterogeneous cement paste. First, the sound 3D microstructure of hardened cement pastes with water-to-cement (w/c) ratios from 0.3 to 0.6 was simulated using a voxel-based hydration model. Subsequently, the fracture process of cement paste under the uniaxial tensile loading was simulated using a finite element model, based on which the cement paste at various damage states can be obtained. Afterwards, using the damaged cement paste with various damage degrees as input, a lattice Boltzmann (LB) modelling framework was proposed to simulate the equilibrium distribution of water and gas phase in pore structure (capillary pores and microcracks), and gas permeability through the damaged cement paste with various water saturation levels. The coupling effects of mechanical damage and moisture conditions on the gas permeability in cement paste were estimated quantitatively. The simulation results of gas permeability in partially saturated cement paste as a function of mechanical damage were compared with the experimental data from the literature.

2. Generation of sound cement paste

In this study, the heterogeneous microstructure of sound cement paste employed as the input was simulated using the voxel-based CEMHYD3D model (Bentz 1997), which consists of three main steps, i.e., particle packing, phase segmentation, and hydration. First, the cement particles were regarded as digital spheres and randomly packed in a cubic box according to the particle size distribution and w/c ratio. The spherical particles cannot overlap with each other, and the packing sequence is from the largest particles to the smallest ones. The periodic boundary conditions were used at faces and edges of cubic box in order to minimise finite size effects. Subsequently, each cement particle was then segmented into four mineral phases, i.e., tricalcium silicate (C\textsubscript{3}S), dicalcium silicate (C\textsubscript{2}S), tricalcium aluminate (C\textsubscript{3}A), and tetracalcium aluminoferrite (C\textsubscript{4}AF), based on the bulk phase composition and auto-correlation functions of the mineral phases. Afterwards, the cellular automa-
ton-like evolution rules with pre-defined dissolution and reaction probabilities were employed to manipulate the movement and phase transition of basic voxels accounting for the reactions in the system, which simulates the cement hydration process and leads to the microstructural development. More details about the modelling procedure can be found in a previous publication (Liu et al. 2020c). For the attributes of binder used in the simulations, Portland cement with Blaine fineness of 369 m²/kg consists of 53.72% C₃S, 24.09% C₂S, 7.61% C₃A, 8.98% C₄AF and 5.60% CaSO₄·2H₂O. In the simulations, the size of representative volume element (RVE) of cement paste was set as 100 × 100 × 100 μm³, which is sufficient to guarantee the representativeness of cement paste for simulating the microstructure of cement paste (Liu et al. 2019). A resolution of 0.5 μm/voxel was employed, which is identical to that of real paste microstructure obtained using X-ray computed tomography images by Promentilla et al. (2009) and Zhang (2017). It is worth noting that owing to the higher resolution compared to the widely used 1 μm/voxel (Bentz 1997), the 3 × 3 × 3 basic cubic dissolution element in the CEMHYD3D model is switched into a 5 × 5 × 5 one for increasing the ultimate hydration degree of cement (Garboczi and Bentz 2001). In this study, the cement pastes with four w/c ratios (0.3, 0.4, 0.5 and 0.6) were investigated. The typical microstructure of cement pastes with w/c = 0.3 and 0.5 at 180 d is displayed in Fig. 1.

3. Modelling of mechanically damaged cement paste

According to the microstructure of cement paste obtained from the voxel-based CEMHYD3D model, the finite element model for fracture in cement paste can be established using the ABAQUS software. The modelling procedure is listed as follows. The microstructure of cement paste at each point was regarded as a solid unit, and the material behaviour was defined by brittle cracking in ABAQUS/Explicit material model. The elastic modulus and Poisson’s ratio of each phase as input were obtained from the measured values of nanoindentation in the literature (Monteiro and Chang 1995; Bernard et al. 2008), as summarised in Table 1. The tensile strength was set to 1/10000 of the elastic modulus (Liu et al. 2013) and the brittle failure was assumed as isotropic. In the simulations, the failure strain was set as 0.0001, while the shear retention factor was set as 1. To simulate the damage and failure under the uniaxial tension, the displacement load was applied on the 3D cement paste. As shown in Fig. 2(a), the reference point was established in the Y-direction and then coupled with the nodes of the upper surface of cement paste. Subsequently, a displacement load of 0.3 μm in the Y-direction was applied to the reference point and the degrees of freedom in other directions were limited. Finally, the six degrees of freedom of the nodes on the bottom of cement paste were limited to zero (Fig. 2(a)). Herein, all phases in the heterogeneous cement paste were assumed to be linear elastic materials. Rankine criterion (Kamali-Bernard and Bernard 2010) was used for the failure of the element, that is, when the stress of the element reaches the tensile strength of the phase, the failure occurs:

\[
\max (\sigma_1, \sigma_2, \sigma_3) \geq \sigma_f
\]  

where \(\sigma_1\), \(\sigma_2\) and \(\sigma_3\) are the principal stresses in three directions, and \(\sigma_f\) is the tensile strength. To avoid excessive distortion caused by the continuous increase of strain after the failure of the element, the element without bearing capacity was deleted in brittle failure.

Figure 2(b) displays the simulated stress-strain

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Table 1 Material parameters of each phase in cement paste obtained from (Monteiro and Chang 1995; Bernard et al. 2008).

| Phases   | E (GPa) | v   |
|----------|---------|-----|
| C₃S      | 117.6   | 0.314 |
| C₂S      | 117.6   | 0.314 |
| C₃A      | 117.6   | 0.314 |
| C₄AF     | 117.6   | 0.314 |
| Gypsum   | 45.7    | 0.330 |
| CH       | 38.0    | 0.305 |
| C-S-H    | 22.4    | 0.240 |
| AFt      | 22.4    | 0.250 |
| AFm      | 42.3    | 0.324 |
| FH₃      | 38.0    | 0.250 |
curves of cement pastes with various w/c ratios (0.3, 0.4, 0.5 and 0.6) under the uniaxial tension. Taking cement paste with w/c = 0.3 as an example, the stress distribution and crack evolution at the peak, sharp initial drop section and softening section are shown in Fig. 3, corresponding to Points A, B and C in Fig. 2(b). It can be seen that when the strength reaches the peak, a small part of the elements reaches the ultimate strength and the scattered microcracks appear. In the initial drop section after the peak, more elements are deleted and larger cracks are formed, resulting in a sharp decline in the bearing capacity of the cement paste. At the softening stage, the microcracks expand into a through microcrack perpendicular to the loading direction. The cement paste loses its bearing capacity, and the stress is released by the large crack. It is worth noting that since the microstructure of cement paste is voxel-based, the width of microcracks is thus a multiple of the resolution, i.e., 0.5 μm/voxel in this study.

To quantitatively describe the cracking state of cementitious materials, the damage degree \(d\) related with Young’s modulus is usually used (Choinski et al. 2007):

\[
d = 1 - \frac{E_d}{E_s}
\]

where \(E_d\) is the Young’s modulus of damaged cement paste, and \(E_s\) is the Young’s modulus of sound cement paste. In this study, a finite element code (named elas3d.f) developed by Garboczi and Day (1995) was employed to estimate the Young’s modulus of damaged cement paste. This program treats each cubic voxel as a tri-linear finite element, upon which the elastic equations are discretized and solved using a relaxation algorithm. The average stress for a given strain is used to determine the composite moduli, which are averaged over direction to minimize the effects of having a small, periodic model of a large microstructure. This algorithm has been widely used and its accuracy has also been well demonstrated in the field of cementitious materials and porous materials (Garboczi and Berryman 2001; Haecker et al. 2005). The detailed description can refer to our recent publication (Liu et al. 2018b).

Figure 4 displays the simulated Young’s modulus of sound cement paste in comparison with the other micromechanics models (Bernard et al. 2003; Sanahuja et al. 2007;
Pichler and Hellmich 2011; Zhang et al. 2018) together with experimental results (Haecker et al. 2005). As can be seen, the simulation results of Young’s moduli of cement pastes with different w/c ratios at 180 d agree well with the experimental data measured from elastic resonance measurements (Haecker et al. 2005) and the estimated results from other micromechanics models (Bernard et al. 2003; Sanahuja et al. 2007; Pichler and Hellmich 2011; Zhang et al. 2018).

4. Lattice Boltzmann simulations of gas permeation of partially saturated cement paste

Using the 3D damaged cement paste as an input, an LB modelling framework consisting of an LB model for permeation and a multiphase LB model was proposed to simulate the gas permeability of damaged cement paste with various water saturation levels. LB method originating from the kinetic Boltzmann equation is used to approximately describe the continuous Boltzmann equation by discretising the physical space into a set of uniformly spaced lattice nodes and the velocity space into a finite set of microscopic velocity vectors. Compared to the other mesoscopic numerical solver, the LB method has already achieved considerable success in simulating fluid flow and ionic transport in porous media as a result of its easy implementation of multiple interparticle interactions and complex geometry and boundary conditions (Liu et al. 2020b).

4.1 General lattice Boltzmann model

For single-relaxation-time (SRT) collision operator, the evolution of particle distribution functions is satisfied with the following discrete lattice Boltzmann equation (Martys 1999):

\[ f_i(x+e_i\delta t, t+\delta t) - f_i(x, t) = \frac{1}{\tau} \left[ f_{i_{eq}}(x, t) - f_{i_{eq}}^m(x, t) \right] \]  \hspace{1cm} (3)

where \( f_i \) is the non-equilibrium and equilibrium particle distribution function at location \( x \) and time \( t \); \( \tau \) is the relaxation time, \( e_i \) is the discrete lattice velocity, subscript \( i = 0, 1, \ldots (b-1) \) denotes the velocity direction where \( b \) represents the number of the discrete lattice velocity directions; \( \delta t \) is the lattice time step; \( f_{i_{eq}}^m \) is the equilibrium particle distribution function. To acquire high calculation accuracy, a cubic lattice model with 19 discrete velocity directions (indicated in Fig. 5) is usually applied to the 3D porous medium, i.e., D3Q19. The corresponding lattice velocity vector of the D3Q19 model is expressed as:

\[
e_{i} = \begin{bmatrix} 0 & 1 & -1 & 0 & 0 & 0 & 0 & 0 & 1 & 1 & 1 & 0 & 0 & -1 & -1 & -1 & -1 & 0 \ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & -1 & 0 & 0 & 1 & -1 & 1 & 0 \ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & -1 & 0 & 0 & -1 & 1 \ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 1 & 1 & 1 & 1 \ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -1 & -1 & -1 & -1 & -1 \ \end{bmatrix}
\]  \hspace{1cm} (4)

In Eq. (3), \( f_{i_{eq}}^m \) is expressed as:

\[
f_{i_{eq}}^m(x, t) = \omega_i \rho(x, t) \left[ 1 + \frac{u_{eq}^m e_i}{c_s^2} + \frac{(u_{eq}^m e_i)^2}{2c_s^4} - \frac{u_{eq}^m}{2c_s^2} \right]
\]  \hspace{1cm} (5)

where \( \rho(x, t) \) is the macroscopic density; \( u(x, t) \) is the macroscopic velocity; \( u_{eq}^m \) is the equilibrium velocity; \( c_s \) is the sound lattice speed, equal to \( \frac{\delta x}{\sqrt{3} \delta t} \); \( \omega_i \) is the weight factor dependent on \( i \)th direction that is expressed as:

\[
\omega_i = \begin{cases} \frac{1}{3} & \text{if } i = 0 \\ \frac{1}{18} & \text{if } i = 1, 2, \ldots, 6 \\ \frac{1}{36} & \text{if } i = 7, 8, \ldots, 18 \end{cases}
\]  \hspace{1cm} (6)

\( \rho(x, t) \) and \( u(x, t) \) can be further expressed as:

Fig. 4 Simulated Young’s modulus of sound cement paste in comparison with other studies (Bernard et al. 2003; Haecker et al. 2005; Sanahuja et al. 2007; Pichler and Hellmich 2011; Zhang et al. 2018).

Fig. 5 D3Q19 cubic lattice model.
\[ \rho(x,t) = \sum_{i=1}^{N} f_i(x,t) \]  
\[ \rho(x,t) u(x,t) = \sum_{i=1}^{N} f_i(x,t) e_i \]

where \( \rho(x,t) u(x,t) \) is the momenta of fluid elements.

### 4.2 Modelling of partially saturated cement paste

To obtain the partially saturated cement paste, it is necessary to mimic the distribution of water and gas in the capillary pore structure (plus microcracks). In this study, a multiphase LB model named pseudopotential model (Martys and Hagedorn 2002) was used to simulate the solid-fluid (water and gas) interaction in cement paste. In the pseudopotential multiphase model, the macroscopic fluid equilibrium velocity used in Eq. (3) is given by:

\[ u^{eq} = u + \frac{\tau}{\rho} \left( F_{int} + F_{ads} + F_g \right) \]

where \( F_{int} \) is the fluid-fluid interaction force, \( F_{ads} \) is the fluid-solid adhesion force, and \( F_g \) is the body force that is generally neglected.

The phase separation results from the cohesive force between liquid particles, i.e., fluid-fluid interaction force \( F_{int} \), which can be described as:

\[ F_{int} = -G \psi(x,t) \sum_{i=0}^{N} \omega \psi(x+\mathbf{e}_i,t) \mathbf{e}_i \]

where \( G \) is the coefficient of attractive forces between fluid particles representing the intensity of interparticle interaction, and \( \psi \) is the effective mass that is a function of local fluid density \( \rho \). \( \psi \) is negative for the attraction and positive for the repulsion between particles. The increasing \( G \) within the critical value can lead to a tendency to phase separation and a sharper interface.

The effective mass function \( \psi \) directly relates to the interaction force. The form of \( \psi \) controls the detailed nature of the interaction potential and determines the equation of state (EOS) of the fluid system. The effective mass \( \psi \) can be expressed as:

\[ \psi(\rho) = \sqrt{\frac{2(\rho-c \rho)}{c \rho g}} \]

To simulate the multiphase flow with a large density ratio, the following Carnahan-Starling EOS (Yuan and Schaefer 2006) can be introduced and incorporated into Eq. (11):

\[ p = \rho R_e T \left( \frac{b \rho}{4} + \left( \frac{b \rho}{4} \right)^2 - \left( \frac{b \rho}{4} \right)^3 \right) \]

\[ \left( 1 - \frac{b \rho}{4} \right)^3 - a \rho^2 \]

\[ a = 0.4963 R^2 T_e^2 / P_c \]

\[ b = 0.18727 R T_e / P_c \]

where \( a \), \( b \), and \( R \) are set as 1, 4 and 1 in the simulation, \( T_e \) is the critical temperature, and \( P_c \) is the critical pressure.

Then fluid-solid adhesion force \( F_{ads} \) can be expressed as:

\[ F_{ads} = -G \psi(x,t) \sum_{i=0}^{N} \omega \psi(x+\mathbf{e}_i,s) \mathbf{e}_i \]

where \( \rho_{w} \) is the density of solid phase that can be freely adjusted for achieving different contact angles at the interface between fluid and solid phases, and \( s \) is an indicator function for checking the neighbouring solid-phase site \( (x+\mathbf{e}_i,s) \) (1 for solid phase and 0 for liquid or gas phase).

To simulate the equilibrium distribution of water and gas phases in cement paste with various water saturation levels, the pore voxels are randomly transferred into liquid and gas voxels according to the pre-set degree of water saturation. Subsequently, the parameters, e.g., \( T \) and \( \rho_{w} \), need to be qualified for achieving the attributes of real water-gas distribution including water/gas density ratio and contact angle of water on the surface of solid phases composed of hydration products and unhydrated cement particles in cement paste. Based on the bubble benchmark test demonstrated in (Zhang et al. 2012), \( T \) that determines the liquid/gas density ratio was set as 0.585, corresponding to a liquid/gas density ratio of 0.4152/0.00053 = 783 that is close to a real water/gas density ratio, i.e., 1000/1.29 = 775. Since the contact angle of water on the surface of cementitious materials is usually regarded as 0° (Lura et al. 2003), the virtual density of solid phase \( \rho_{w} \) was thus set to be close to the liquid density in lattice units.

### 4.3 Modelling of gas permeability in cement paste

Treating gas-filled pores and microcracks as permeable phases, an LB model for permeation was proposed to simulate gas permeability of damaged cement paste with various water saturation levels. In the simulations, three kinds of boundary conditions are included for LB simulation of permeation, i.e., inlet/out boundary condition at inlet and outlet of the computational domain (i.e., X-direction in this study), periodic boundary conditions for the other four boundaries (i.e., Y- and Z-direction) and half-way bounce back condition at the interface of transport and non-transport phases inside the domain. The detailed boundary conditions are similar to those reported in Ref. (He et al. 1997). After achieving the steady-state gas velocity distribution in the domain, the permeability of the porous medium can be calculated according to Darcy’s law described as follows (Liu et al. 2021):
where $\kappa$ is the lattice permeability of porous medium; $\zeta$ is the kinematic viscosity of the fluid as $\zeta = c_s^2(\tau - 0.5)$; $\Delta \rho$ is the density difference of fluid between the inlet and outlet; $N_x$, $N_y$ and $N_z$ are the total number of nodes in X-, Y- and Z-direction; $u_x$ is the average velocity of the fluid in the X-direction. The dimensionless permeability can be converted into physical permeability ($\kappa$) using:

$$\kappa = \kappa R_s^3$$

where $R_s$ is the spatial resolution of the medium. In this study, the spatial resolution in the microstructure of cement paste is 0.5 $\mu$m/voxel, i.e., $R_s = 5 \times 10^{-7}$ m.

It is worth mentioning that cement paste is usually regarded as a two-phase transport medium, i.e., capillary pore and porous C-S-H. However, for fluid permeation in cement paste the permeable channel can be only regarded as a capillary pore (plus microcrack) path because the permeability of cement paste is less dependent on the weakly permeable C-S-H (Zalzale et al. 2013). Meanwhile, this operation was adopted in the simulation of permeability of sound Portland cement paste (Zhang et al. 2013; Li et al. 2017). As such, to improve the numerical efficiency, C-S-H is regarded to be non-permeable in cement paste.

5. Results and discussion

To evaluate the coupling effects of mechanical damage and moisture conditions on gas permeability of cementitious materials, the relative gas permeability that is equal to permeability of damaged cementitious materials over that of sound ones is usually used. In the following section, the gas permeability of sound cement paste at a dry state, the relative gas permeability of damaged cement paste at a dry state, and relative gas permeability of damaged cement paste with various water saturation levels are simulated, respectively.

5.1 Permeability of sound cement paste

Based on the sound cement paste, the gas permeability was simulated using the LB model for permeation. Taking cement pastes with w/c ratios of 0.3 and 0.5 as examples (see Fig. 1), the steady-state gas velocity in microstructure is shown in Fig. 6(a). It can be found that with the increase of w/c ratio, the transport paths and gas velocity are both increased. It can be attributed to the increase of effective capillary porosity as a result of increasing capillary porosity and connectivity of capillary pores. Figure 7 shows the simulation results of gas permeability of sound cement paste as a function of capillary porosity (hollow points). It can be observed that the gas permeability is increased with the increase of capillary porosity as a result of the increasing transport channels for gas. The evolution of gas permeability can be divided into two stages: above and below capillary porosity of 0.15. As the capillary porosity is greater than 0.15, the simulation results of gas permeability for all cement pastes with various w/c ratios display a similar increasing trend with the increase of capillary porosity. A perfect linear relationship between the logarithmic value of gas permeability and capillary porosity of cement paste regardless of w/c ratios can be observed. When the capillary porosity decreases to below 0.15, there is a sharp drop in gas permeability. This finding is consistent with that reported by Zalzale and McDonald (2012), who used the virtual cement paste generated by μic (Bishnoi and Scrivener 2009) to predict permeability. The presence of critical capillary porosity is attributed to the depercolation of the capillary pore network. Liu et al. (2018a) indicated that the depercolation of capillary pore network in cement paste with a resolution of 0.5 $\mu$m/voxel occurs at capillary porosity of around 0.15.

To verify the simulations, the experimental data of gas permeability in dry cement paste as a function of porosity are shown in Fig. 8 (Wong et al. 2009; Hamami et al. 2012; Tracz 2016). As can be seen, the simulation results as a function of capillary porosity are
in good agreement with the experimental data reported by Tracz (2016), who used the mercury intrusion porosimetry technique to measure the porosity. However, the remaining experimental data (Wong et al. 2009; Hamami et al. 2012; Tracz 2016) are far smaller than the simulation results. One of the reasons is that the experimental porosity was measured using the water displacement method, which can be regarded as the total porosity including gel pores and capillary pores (Liu et al. 2020c). Treating the average gel porosity of C-S-H in cement paste as 0.28 (Powers 1958), the gas permeability can be expressed as a function of total porosity (Fig. 7, solid points). It can be observed that the simulation results are much closer to the experimental data; however, the difference of gas permeability between simulations and experiments still has one order of magnitude. It can be attributed to the fact that (i) the simulated microstructure of cement paste cannot represent the real one. For example, the resolution of the microstructure of cement paste, i.e. 0.5 μm/voxel is larger than the majority of capillary pores, which can overestimate the permeability at a given capillary porosity (Zalzale and McDonald 2012); (ii) the cementitious samples used for measuring gas permeability cannot reach a completely dried state during the experimental procedure. Since the available air-filled porosity for gas permeation is lower than the total porosity in cement paste, the measured gas permeability is thus decreased; (iii) the attributes of raw materials, e.g., particle size distribution, between experiments and simulations are different. For example, Ye et al. (2006) demonstrated that the minimum size of cement particles reduces from 2 μm to 1 μm, the permeability of cement paste at 30 d can decrease by approximately 2-3 orders of magnitude.

5.2 Permeability of damaged cement paste at a dry state

Using the cement paste with various cracking states as an input, the damage degree can be calculated using the finite element method, as indicated in Section 3. Figure 8 displays the evolution of the fracture pattern of cracking in cement paste with w/c ratios of 0.3 and 0.5, and damage degrees of 0.03, 0.07 and 0.12. It can be seen that with the increase of damage degree, the microcrack volume fraction of cement paste is increased. Additionally, for the cement paste with a lower w/c ratio, the microcrack volume fraction of the cement paste is higher. This is expected since the cement paste with a lower w/c ratio has a higher Young’s modulus, which leads to a more significant decreasing amplitude of Young’s modulus at a given damage degree. To simulate the gas permeability of damaged cement paste, the LB model for permeation was applied to the damaged mi-
crostructure, where the permeation direction is perpendicular to the direction of tensile loading. Figure 6 displays the steady-state distribution of gas velocity in damaged cement pastes with w/c ratios of 0.3 and 0.5, and damage degrees of 0.07 and 0.12 along X-direction. It can be observed that with the increase of damage degree (from 0 to 0.12), the permeation paths for gas is increased as a result of the increasing microcrack volume fraction. Meanwhile, the gas velocity in microcracks is higher than that in capillary pores, which can be attributed to the greater size.

Figure 9 shows the relative gas permeability of damaged cement paste as a function of damage degree. It can be seen that with the increase of damage degree, the relative gas permeability is increased. For example, for the cement paste with w/c ratio of 0.4, the relative permeability increases from 1 to 4.8 when the damage degree increases from 0 to 0.16. Regarding the effect of the w/c ratio, the permeability grows significantly for the damaged cement paste with a lower w/c ratio. For instance, the relative gas permeability of cement paste with damage degree of 0.16 is increased from 1.4 to 17.3 with the reduction of the w/c ratio from 0.6 to 0.3. It suggests that the relative gas permeability of cement paste with a lower w/c ratio is more sensitive to the damage, which can be attributed to the increasing brittle characteristic of cement paste with a lower w/c ratio (Picandet et al. 2001).

To validate the simulations, the simulation results of gas permeability of cement paste as a function of damage degree were compared with the experimental data of concrete with various mechanical damages (Fig. 9) (Picandet et al. 2001; Choinska et al. 2007; Zhou et al. 2012a; Tegguer et al. 2013). The detailed information on testing samples, sample preconditioning, and damage types are summarised in Table 2. As seen in Fig. 9, the experimental data are located in the range of the simulation results in general. However, some issues still need to be discussed. As seen in Table 2, the concrete samples, instead of cement paste, were used to investigate the relationship between gas permeability and damage degree. Picandet et al. (2001) demonstrated that the relationship between gas permeability and damage degree is dependent on the types of cementitious materials. It can be attributed to the difference in mechanical properties for different cementitious materials. Furthermore, the gas permeability in damaged cementitious materials is associated with the microcracking patterns as a result of different damage types (Zhou et al. 2012a). Herein, to approximate the cracking patterns with strong orientation induced by uniaxial tensile damage (Hearn 1999), the experimental data of damaged concrete under uniaxial compression was employed. Nevertheless, the cracking patterns as a result of various external loading are different. In terms of the simulated cracking pattern, since the microcracks with the width less than the resolution are not covered, it cannot completely represent...
the real one. The volume fraction and connectivity of capillary pores and microcracks are both underestimated to some extent, which may lead to the slight underestimation of damage degree and gas permeability.

5.3 Permeability of damaged cement paste with various water saturation degrees

To investigate the effect of water saturation degree on gas permeability in damaged cement paste, the equilibrium distribution of water and gas phases in pore spaces including capillary pores and microcracks was simulated using the multiphase LB model. Note that to keep consistent with the experiments (Sogbossi et al. 2020), the water saturation degree is defined as the ratio of water-filled capillary pores to the total capillary porosity in the simulation. Taking the cement paste with w/c ratio of 0.5 and damage degree of 0.12 as an example, Fig. 10 shows the equilibrium distribution of water and gas in capillary pore network and microcracks at water saturation levels of 25%, 50%, and 70%. It can be found that the water phase tends to fill smaller pore spaces, while the gas phase is subject to form air clusters in the larger pore spaces. This phenomenon conforms to the Kelvin-Laplace equation (Grasley and Lange 2007) that the air-filled pores in the porous medium are progressively occupied with water following the sequence from small pores to large pores with the increase of water saturation level. Meanwhile, compared to the capillary pores, the microcracks are liable to be occupied by the gas phase due to the larger space size and coarser pore space cluster. Treating air-filled pore spaces as permeable phases, the LB model for permeation was used to simulate gas permeability in partially saturated damaged cement paste. Figure 11 displays the steady-state distribution of gas velocity in damaged cement pastes with degrees of water saturation of 25%, 50% and 70% that corresponds to those in Fig. 10. It can be observed that with the increase of water saturation level, the permeation paths for gas are reduced. Additionally, the air-filled microcracks as permeation paths for gas are not significantly influenced by the increasing water saturation level from 0% to 70%. It suggests that the presence of microcracks can significantly enhance the gas permeation in partially saturated cement paste. Through calculation using Eqs. (16) and (17), the gas permeability of cement paste with w/c ratio of 0.5, damage degree of 0.12 and water saturation levels of 0%, 25%, 50% and 70% is $3.10 \times 10^{-15}$ m$^2$, $1.18 \times 10^{-15}$ m$^2$, $3.95 \times 10^{-16}$ m$^2$, and $1.30 \times 10^{-16}$ m$^2$, respectively. It should be noted that since microcrack volume fraction is underestimated due to the limitation of resolution in the simulations, partial gas-filled capillary pores and microcracks with a smaller size are converted into water-filled ones at a given water saturation level. The gas permeability in non-saturated damaged cement paste is thus underestimated. Nevertheless, this underestimation is insignificant because the gas permeability is less dependent on the smaller pores (Sakai 2020).

**Figure 11** shows the relative gas permeability of cement paste with w/c ratio of 0.5 and water saturation levels between 0% and 70% as a function of damage degree. It can be seen that the relative gas permeability of non-saturated damaged cement paste exhibits a similar evolution with the increase of damage degree in general. However, at a given damage degree the relative gas permeability is increased with the increasing water satu-

![Fig. 10](image1.png)

**Fig. 10** Equilibrium distribution of water and gas in capillary pore network and microcracks of cement paste with w/c ratio of 0.5, damage degree of 0.12, and degrees of water saturation of 25% (a), 50% (b), and 70% (c), at 180 d (Blue and grey colours represent water and gas, respectively).

![Fig. 11](image2.png)

**Fig. 11** Steady-state distribution of gas velocity in cement paste with w/c = 0.5, damage degree of 0.12 and water saturation levels of 25% (a), 50% (b), and 70% (c), along X-direction.
The experimental data are obtained from Ref. (Sogbossi et al. 2020).

For the sound cement paste, the evolution of gas permeability with capillary porosity can be divided into two stages by a critical capillary porosity, i.e., 0.15. When the capillary porosity is greater than 0.15, a perfect linear relationship between the logarithmic value of gas permeability and capillary porosity of cement paste regardless of w/c ratios is displayed. However, as the capillary porosity is below 0.15, there is a sharp drop in gas permeability due to the depercolation of the capillary pore network.

The microcrack volume fraction of the dry damaged cement paste is elevated with the increasing damage degree (from 0 to 0.12), which improves the permeation paths and thus results in the higher gas velocity due to the greater path size. The gas permeability of dry cement paste is increased with the increase of damage degree. The gas permeability of the dry cement paste with a lower water-to-cement ratio is more sensitive to the damage.

In the partially saturated damaged cement paste, water phase tends to fill smaller pore spaces, while the gas phase is subject to form air clusters in the larger pore spaces. Meanwhile, compared to the capillary pores, the microcracks are liable to be occupied by the gas phase due to the larger space size and coarser pore space cluster.

With the increase of water saturation level from 0% to 70%, gas permeation paths of damaged cement paste are reduced. However, due to larger size of microcracks in comparison with capillary pores, the microcracks as permeation paths for gas are not significantly influenced by the increase of water saturation. At a given damage degree, the relative gas permeability is increased with the increasing water saturation level. The gas permeability of cement paste with a higher water saturation level is more sensitive to the presence of microcracks.

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