An engineering approach for permeability assessment of virtual cement-based materials

Li, Kai; Stroeven, Piet; Stroeven, Martijn; Sluijs, Bert

Publication date
2016

Document Version
Final published version

Published in
2016 International Concrete Sustainability Conference, Washington, USA

An engineering approach for permeability assessment of virtual cement-based materials

Kai Li[1], Piet Stroeven[2], Martijn Stroeven[3], Bert Sluys[4]

[1]Delft University of Technology, Faculty of Civil Engineering and Geosciences, Stevinweg 1, 2628 CN Delft, the Netherlands; PH (+31) 15278-8266; FAX (+31) 15278-5767; email: K.Li-1@tudelft.nl
[2]Delft University of Technology, Faculty of Civil Engineering and Geosciences, Stevinweg 1, 2628 CN Delft, the Netherlands; PH (+31) 15278-2767; FAX (+31) 15278-5767; email: P.Stroeven@tudelft.nl
[3]Delft University of Technology, Faculty of Civil Engineering and Geosciences, Stevinweg 1, 2628 CN Delft, the Netherlands; PH (+31) 15278-2767; FAX (+31) 15278-5767; email: M.Stroeven@tudelft.nl
[4]Delft University of Technology, Faculty of Civil Engineering and Geosciences, Stevinweg 1, 2628 CN Delft, the Netherlands; PH (+31) 15278-2728; FAX (+31) 15278-5767; email: L.J.Sluys@tudelft.nl

Abstract

A quick approach to estimate water permeability of virtual cement paste is outlined in this paper. Although the computational models developed by the authors are able to provide satisfactory results in comparison with experimental data, it is still interesting and meaningful to further limit the efforts. This paper therefore presents a simple mathematical model for permeability assessment. Only one parameter, *i.e.* water-filled porosity, is required for the proposed model. The results from the presented mathematical model are compared with data obtained by other approaches. A good agreement can be observed, validating the presented method. Additionally, median pore throat size and connectivity of the capillary pores are plotted as a function of water-filled porosity to help understand the permeability changes in structural terms.

1. Introduction

Permeability of concrete plays an important role in determining its transport-based durability properties. It is defined as the movement of an agent through the porous medium under an applied pressure (Banthia *et al.*, 2005). Hence, the pore microstructure in the hardened paste is of prime relevance for permeability studies since aggregates can usually be assumed impermeable. Cement paste is therefore selected in this paper instead of concrete or mortar. Measurements of cement paste's permeability can be conducted by experiments, and estimated by empirical formulas or computer simulations. Experimental tests are widely performed in laboratories. Samples should be fully saturated such that Darcy's law can be applied to calculate permeability. However, this state is difficult (if not impossible) to establish and maintain. Additionally, physical experiments are typically time-consuming, laborious, tedious and thus expensive. Several empirical models (EI-Dieb *et al.*, 1994; Zamani *et al.*, 2014) have therefore been proposed to predict the permeability of cementitious materials based on some parameters, *i.e.* capillary pressure, porosity and pore throat size. These empirical formulas are mostly derived from experimental data that critically depend on the type of materials, since the reported value in the same model for concrete and cement paste varies a lot. Over the past decades, a series of computer modelling approaches have therefore become popular in concrete research (Bishnoi and Scrivener, 2009; van Breugel, 1995; Garboczi and Bentz, 2001; Le *et al.*, 2013; Li *et al.*, 2016). They require less efforts, are cheaper and are mostly able to provide reliable results.

A relationship between permeability and pore characteristic of cementitious materials has been revealed by the abovementioned studies. In the case of a fully saturated specimen, median pore throat size seems to be a critical parameter in determining permeability (Gao and Hu, 2013; Le, 2015). Due to environmental humidity changes, water evaporation takes place, inevitably resulting in a partially saturated specimen. Once the effective porosity for fluid transport reaches a low value, the influence of connectivity of the capillary pores on permeability assessment will become apparent and should thus be taken into consideration (Ye, 2005; Li *et al.*, 2016). The effective porosity herein is defined as the porosity available for water permeation. Ye (2005) reported that the water permeability of cement pastes depends on the pore connectivity and the pore size distribution regardless of water/cement ratio (*w/c*) and curing age. Li *et al.* (2016) further found by application of the discrete element method (DEM) that both median pore size and connectivity of the capillary pores are actually correlated to the effective porosity. It indicates that the permeability could be simply represented by the effective porosity. Note that the effective pores for water permeation are water-filled ones, as indicated earlier.

A dynamic DEM-based modelling approach (the so-called Complete Methodology, CM) has been developed in the authors group (Li *et al*., 2016; Stroeven *et al.*, 2015) for estimating the permeability of cement pastes on the basis of their pore structures. The CM consists of five stages (particle packing simulation - hydration simulation - pore delineation - pore measuring - tube network modelling) of which the relevant details can be found in (Li *et al.*, 2015; Stroeven *et al.*, 2015). CM is advanced and robust; the results obtained by CM show a satisfactory agreement compared to experimental data. Nevertheless, a complete solution still costs a few hours. Thus, it is interesting and meaningful to further limit the efforts in permeability estimation by defining a shorter way while at the same time maintaining reliability at an appropriate level. A mathematical model has been developed and is presented in this paper. The data used for developing the shortcut method are obtained from the CM. Since the CM has been described in detail in (Li *et al.*, 2015; Stroeven *et al.*, 2015), only a brief introduction of the CM will be given in Section 2. Next, the proposed model for permeability assessment of cement pastes will be presented in Section 3. Only one input parameter, *i.e.* water-filled porosity, is required to calculate the permeability. The total computation after porosimetry only takes a few minutes, which can be considered as an obvious improvement in contrast to the CM which takes a few hours. The confidence limit is set to 0.91, indicating an appropriate level of reliability. Moreover, a good agreement is observed between the results calculated by the proposed model and data from other studies. Afterwards, the various inter-relationships between the median pore throat size, pore connectivity, water-filled porosity and water permeability are studied. This information will serve as support for the presented mathematical approach.

2. The complete methodology (CM)

The CM consists of five stages, as illustrated in Fig. 1. Although the simulations performed are three-dimensional, a two dimensional overview is given for simplicity reasons. All relevant details can be found in (Li *et al.*, 2015; Stroeven *et al.*, 2015).

In stage 1, cement grains (the red circles in Fig. 1) are dynamically mixed in a cubic box (final size of 100 μm). The Rosin-Rammler function was chosen to represent the particle size distribution. The minimum size is kept as 1 μm, while the maximum one ranges from 30 μm via 40 μm to 45 μm. Six periodic boundaries were used to properly represent bulk phase. In contrast to the random sequential addition (RSA) system, the DEM-based package HADES is chosen for the particle packing simulation since the pore network characteristics are basically structure-sensitive. Therefore, a DEM approach is more approximate for this purpose. The packing simulation terminates once a certain packing density (that is, the required *w/c*) is reached. In this paper, three different water/cement ratios (*i.e.* 0.4, 0.45 and 0.5) were selected to represent ordinary cement paste.

In stage 2, particles are assumed to consist of the four major compounds of the Portland cement, tricalcium silicate, dicalcium silicate, tricalcium aluminate and tetracalcium aluminoferrite to simulate the cement hydration stage (Le *et al.*, 2013). The ratio of the successive compounds in the clinker were 61%, 20%, 8%, 11% by volume, respectively. The chemical reaction of cement grains and water results in a growth of the particles, thereby decreasing the pore space of the specimen. The pastes hydrated for 3 days, 7 days and 28 days were selected for permeability investigation. Although calcium silicate hydrate (C-S-H) has been proven porous and weakly-permeable (Jennings, 2000; Jennings, 2008), its contribution is only of importance for the total permeability at very low water-filled porosity. Therefore, the C-S-H permeability is neglected in this paper.

In stage 3, the Double Random Multiple Tree Structuring (DRaMuTS) method (Stroeven *et al.*, 2012) is used to assess the pore topology and relevant pore fractions of the produced virtual matured material. In brief, random points are distributed inside the specimen and only the points located in pore space are used for porosimetry. Since the isolated and dead-end paths do not contribute to the fluid transport, only the continuous pore channels should be known. To find the possible paths through the sample, points are connected to each other by non-obstructive lines. In this way, it is straightforward to establish continuous pore network through the specimen. However, the size of each individual pore is still unknown, which is a necessary parameter for constructing the tube network used for fluid transport simulation.

In stage 4, the star volume method (SVM) is applied to calculate the pore size (Stroeven *et al.*, 2010). The smallest pore sections (*i.e.* pore throats) determine the available flow going through the transport channels, due to the ink bottle effect. Hence, median pore throat size is selected herein as pore characteristic for permeability calculation. It is obtained by calculating the smallest area among all cross-sections of each random point located in pore space.

Lastly, a tube network is constructed to represent the pore channels on the basis of the locations and sizes of pores in stage 5. By applying a pressure gradient between inlet and outlet nodes located at the top and bottom surfaces of the paste, the water flow,

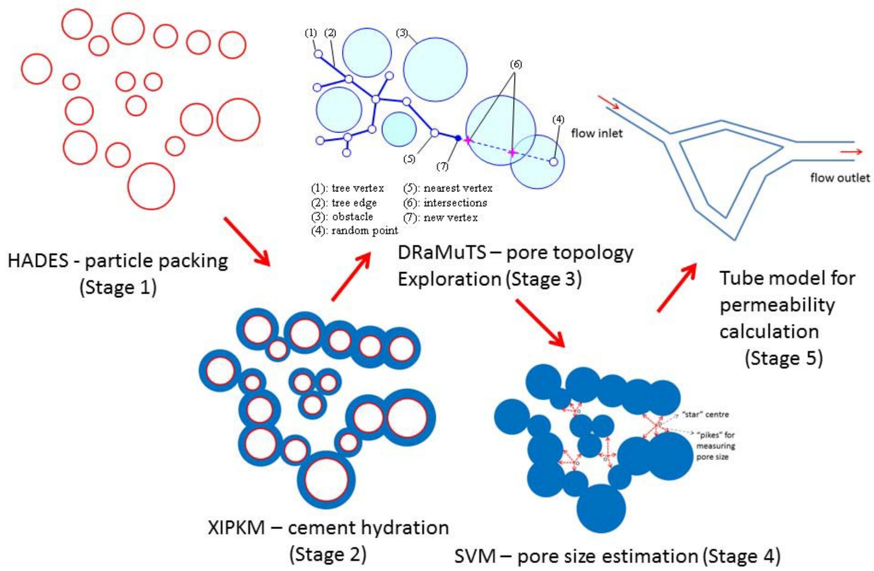

Figure 1. Two dimensional schematic diagram of the CM for water permeability estimation.

assumed to be slow, incompressible and laminar, penetrates the specimen. Then, Darcy's equation can be used to calculate permeability. However, it may only be applied to fully saturated samples, which is seldom the case in practice. The humidity changes in the environment inevitably result in water evaporation of the specimen. According to the Kelvin-Laplace equation, larger pores are easier to lose water than smaller ones. To obtain the pastes at various degrees of water saturation, the algorithm developed in (Li *et al.*, 2016) was implemented mimicking these empty pores. Details can be found in (Li *et al.*, 2015; Stroeven *et al.*, 2015).

3. Results and discussion

3.1 A quick approach for permeability assessment

The simulation results on water permeability versus water-filled porosity of cement pastes with variable water/cement ratio (0.4, 0.45, 0.5), curing age (3 days, 7 days, 28 days) and particle size range (1 µm - 30 µm, 1 µm - 40 µm, 1 µm - 45 µm) are presented in Fig. 2. A decline in permeability with decreasing water-filled porosity can be observed. All data in Fig. 2 are obtained from the CM. Note that the water permeability is plotted in a logarithmic way versus the water-filled porosity. A fitted equation can therefore be derived from the data using regression analyses:

$$\log_{10}(K) = -13.58\exp(-0.71P) - 16.59\exp(-0.007163P)$$

In this equation, *K* and *P* represent the water permeability and the water-filled porosity of the cement pastes, respectively. The curve is shown in Fig. 2 as a green line. The confidence limit of the developed model is set to 0.91, indicating a satisfactory accuracy for the chosen samples. In contrast to the computational efforts required by the CM (in term of hours), water permeability can be directly calculated

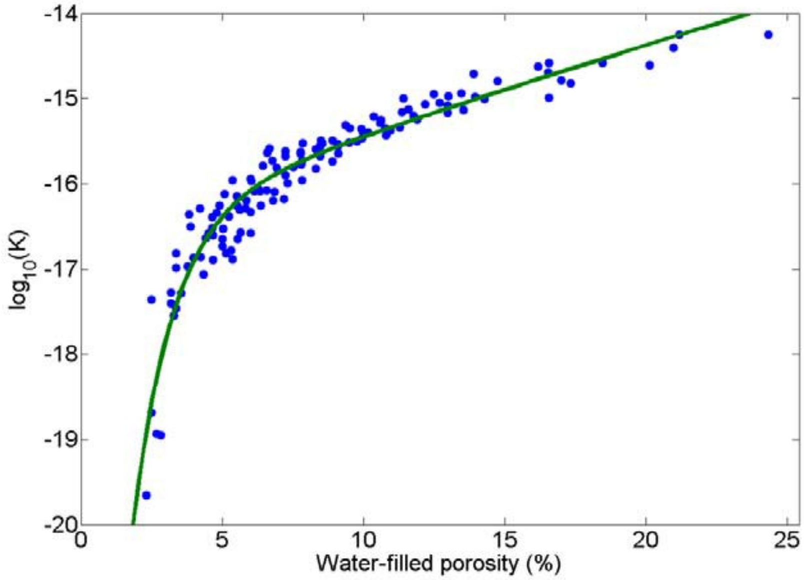

Figure 2. Log_{10} (K) versus water-filled porosity P of all samples; K: water permeability. The data obtained from the CM are plotted in blue points, while the green curve represents the linear regression equation.

using the abovementioned formula once the water-filled porosity is known. The water-filled porosity is measured by the points counting technique in this paper.

Points are randomly distributed inside the sample of which those located in water-filled pore space are counted. The water-filled porosity is calculated by dividing the number of points in water-filled pores to that in the whole specimen. The calculation just costs a few minutes. Therefore, it can be considered as a quick method for permeability assessment compared to the CM. The water-filled porosity is also possible to be measured by physical experiments, including the oven drying technique (Lafhaj *et al.*, 2006; Kameche *et al.*, 2014) and the nuclear magnetic resonance (NMR) method (Muller *et al.*, 2013; Zamani *et al.*, 2014). Hence, the proposed model can potentially be used for permeability estimation for engineering purposes once the water-filled porosity is experimentally measured.

3.2 Validation

The relationship between water permeability and porosity of cement paste has been studied by (Pignat *et al.*, 2005; Zalzale *et al.*, 2013). The obtained results in these papers were used to validate the proposed model, as illustrated in Fig. 3. When the water-filled porosity is larger than 7%, a good agreement is found between the results obtained by the proposed model and the data from (Pignat *et al.*, 2005; Zalzale *et al.*, 2013). However, the differences become apparent once the water-filled porosity is below 7%. This is attributed to the fact that the network model is used in this paper while the model in (Zalzale *et al.*, 2013) is voxel-based. The resolution limit in the latter approach usually leads to a higher depercolation threshold in contrast to that in the network model. It indicates that smaller pores ignored in the voxel-based model may still be considered to contribute to the fluid transport in the model presented here. Hence, a larger water permeability is obtained by the proposed model at low

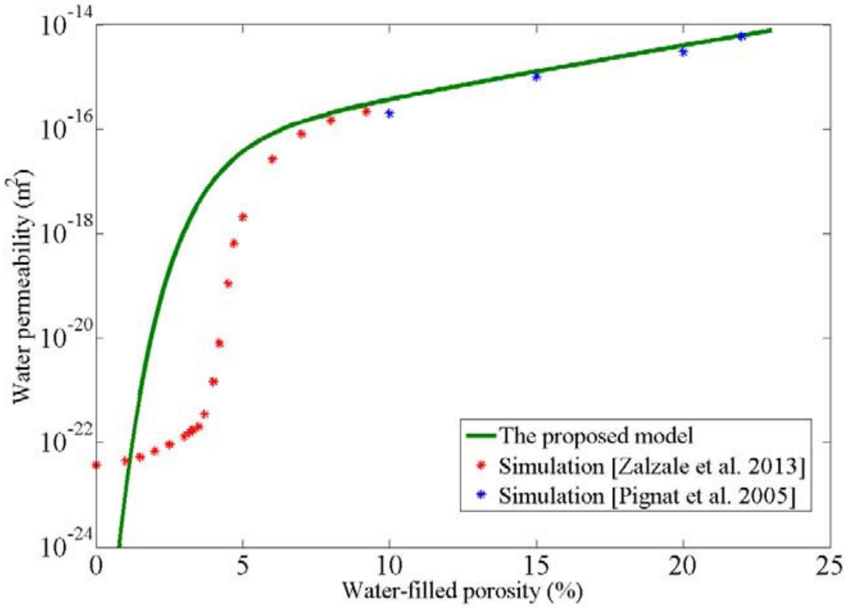

Figure 3. Comparison between the proposed model and other simulation data.

water-filled porosity. Another difference is also observed in water permeability at very low values of the water-filled porosity (below 3%). A sharp drop happens in the proposed model while the data reported in (Zalzale *et al.*, 2013) almost reach a stable value. This can be explained by the fact that C-S-H is treated as a medium with low permeability in (Zalzale *et al.*, 2013) while its permeability is neglected in this paper.

3.3 Relationship between median pore throat size and water-filled porosity

In this section, median pore throat size of cement pastes with variable water/cement ratio, curing age and particle size range are plotted as a function of water-filled porosity. Note that only one technological parameter of the samples is varied and the others are kept constant. For instance, water/cement ratio and particle size range are respectively maintained at 0.4 and 1 µm – 30 µm, while the curing age is varied from 3 days via 7 days to 28 days. In each case, the simulation starts from the fully saturated state and then water is gradually evaporated from the samples to obtain pastes with various water saturation degrees. The results are shown in Figs. 4-6. In all cases, median pore throat size declines almost linearly with the reduced water-filled porosity. This implies that the channels for water permeation become smaller, leading to a lower water permeability. Although a small difference in median pore throat size at the same water-filled porosity is observed, the resulting scatter in water permeability is negligible considering a confidence limit of 0.91. Unfortunately, the sharp drop in water permeability at low water-filled porosity (below 5%), as presented in Fig. 2, still seems to be inexplicable when only considering the linear decline in median pore throat size. Another important parameter (*i.e.* pore connectivity) should be taken into consideration, as will be discussed in the next section.

3.4 Relationship between pore connectivity and water-filled porosity

The relationship between connectivity of the capillary pores and water-filled porosity

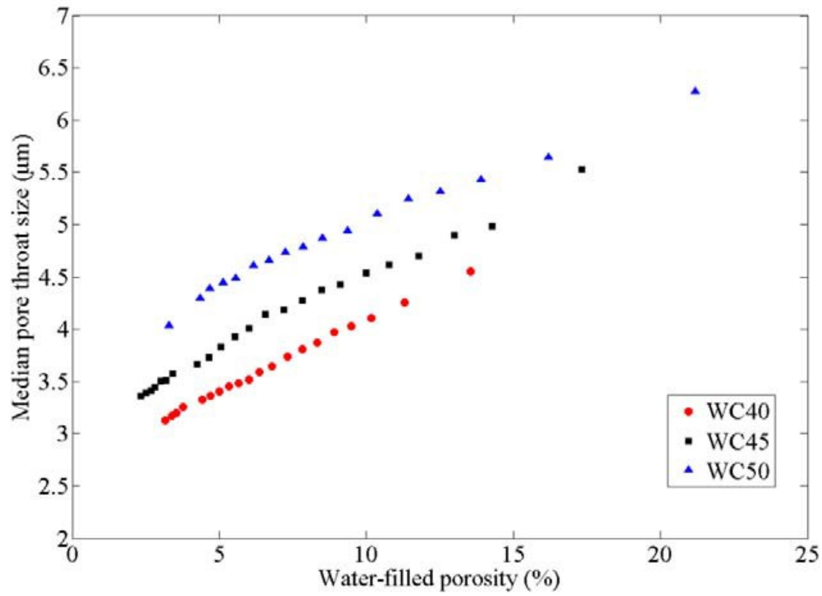

Figure 4. Random median pore throat size versus water-filled porosity of cement pastes with variable water/cement ratio. WC40: water/cement ratio = 0.4; WC45: water/cement ratio = 0.5; WC50: water/cement ratio = 0.5. Curing age: 28 days. Particle size range [1 µm – 30 µm].

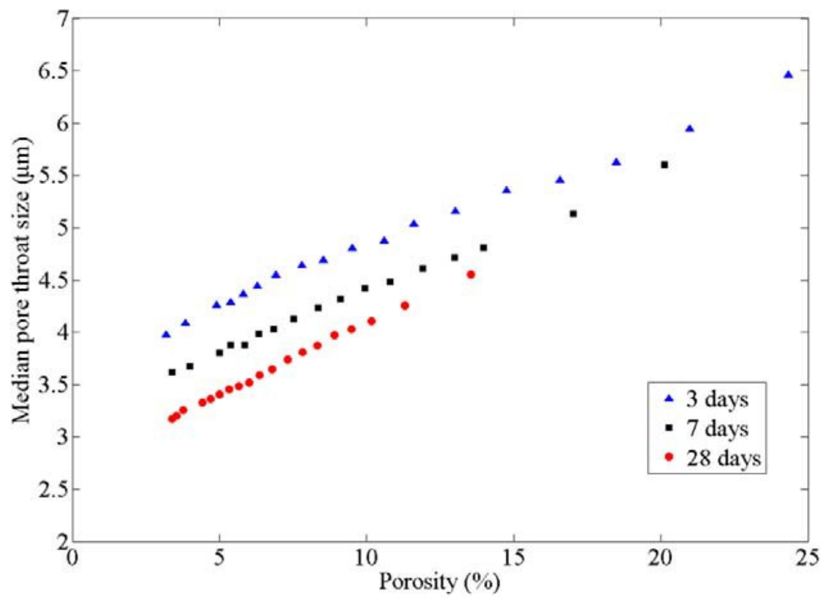

Figure 5. Random median pore throat size versus water-filled porosity of cement pastes with variable curing age (3 days, 7 days and 28 days). Water/cement ratio: 0.4. Particle size range [1 µm - 30 µm].

is studied and presented in this section. This is because water transport can only take place in the continuous phase. The same samples in accordance with Section 3.1 and 3.3 were used herein. Pore connectivity is defined as the volume ratio of the percolated pores to the total pores. The results obtained from all samples are shown

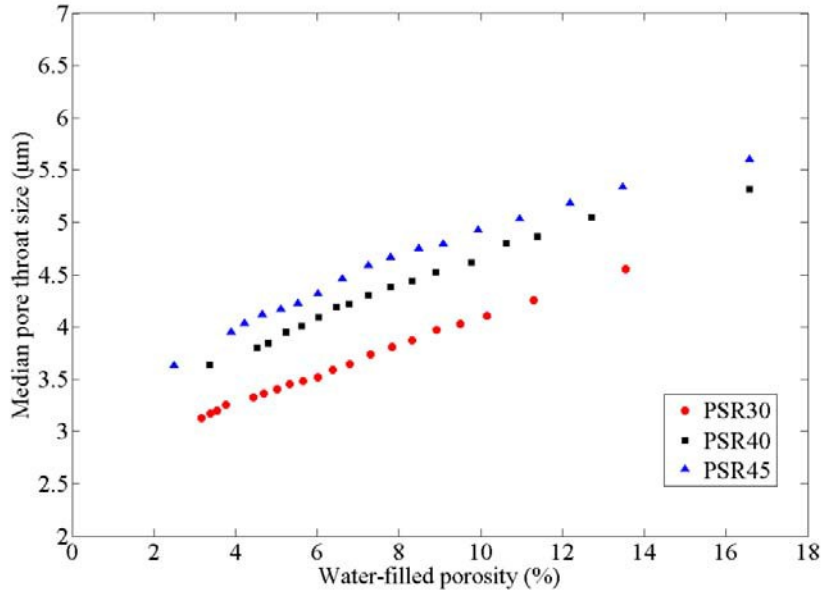

Figure 6. Random median pore throat size versus water-filled porosity of cement pastes with variable particle size range. PSR30: particle size range [1 µm - 30 µm]; PSR40: particle size range [1 µm - 40 µm]; PSR45: particle size range [1 µm - 45 µm]. Water/cement ratio: 0.4. Curing age: 28 days.

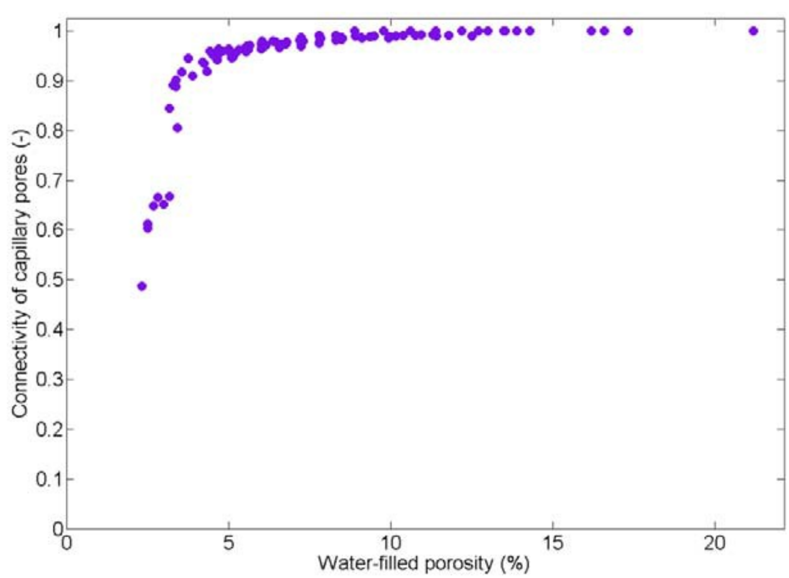

Figure 7. Pore connectivity versus water-filled porosity of all the samples.

in Fig. 7. The so-called depercolation phenomenon of capillary pores is observed, as already reported by (Chen *et al.*, 2006; Garboczi and Bentz, 2001; Ye, 2005). The pore connectivity slightly decreases first and then dramatically drops to a low value when the water-filled porosity reaches values below 5%.

This allows interpreting water permeability of cement pastes in the following structural terms. When water-filled porosity is large, availabe channels for water transport are highly connected, so that the reduction in water permeability is predominatly attributed to the decrease in the effective porosity and pore throat size. Once the water-filled porosity passes to the depercolation threshold, pore connectivty starts to play a dominant role in determing water permeability compared to the abovementioned two factors. After the system becomes fully depercolated, there are no continuous paths left for fluid transport, leading to a negligible permeability.

4. Conclusions

The permeability of normal cement paste is governed by water-filled porosity. This is an important indicator for durability assessment. A mathematical permeability model is therefore presented in this paper based on the water-filled porosity. In contrast to the CM (in term of hours for pore characteristics assessment and the network modelling stage), this shorter but reliable method for permeability estimation just costs a few minutes in total. This can be considered as a significant improvement in computational efforts. At structural level, pore throat size and connectivity of the capillary pores are studied to better understand the phenomenon driving the permeability. A gradual decrease in the water-filled porosity is observed to a linear decline in median pore throat size. After passing the de-percolation point, pore connectivity seems to mainly influence permeability.

Acknowledgment

The first author would like to thank the China Scholarship Council (CSC) for the financial support to this work.

References

Banthia, N., Biparva, A., Mindess, S. (2005). “Permeability of concrete under stress.” *Cememt and Concrete Research*, 35(9), 1651-1655.

Bishnoi, S., and Scrivener, K. L. (2009). “μic: A new platform for modelling the hydration of cements.” *Cememt and Concrete Research,* 39(4), 266-274.

Breugel, K van (1995). “Numerical simulation of hydration and microstructural development in hardening cement-based materials (II) applications.” *Cememt and Concrete Research,* 25(3), 522-530.

Chen, H. S., Stroeven, P., Ye, G., Stroeven, M. (2006). “Influence of boundary conditions on pore percolation in model cement paste.” *Key Engineering Materials,* 302-303, 486-492.

EI-Dieb, A. S., and Hooton, R. D. (1994). “Evaluation of the Katz-Thompson model for estimating the water permeability of cement-based materials from mercury intrusion porosimetry data.” *Cememt and Concrete Research,* 24(3), 443-455.

Gao, Z. Y., and Hu, Q. H. (2013). “Estimating permeability using median pore-throat radius obtained from mercury intrusion porosimetry.” *Journal of Geophysics and Engineering,* 10(2), 025014.

Garboczi, E. J., and Bentz, D. P. (2001). “The effect of statistical fluctuation, finite size error, and digital resolution on the phase percolation and transport properties of the NIST cement hydration model.” *Cememt and Concrete Research,* 31(10), 1501-1514.

Jennings, H. M. (2000). “A model for the microstructure of calcium silicate hydrate in cement paste.” *Cememt and Concrete Research,* 30(1), 101-116.

Jennings, H. M. (2008). “Refinements to colloid model of C-S-H in cement: CM-II.” *Cememt and Concrete Research,* 38(3), 275-289.

Kameche, Z.A., Ghomari, F., Choinska, M., Khelidj, A. “Assessment of liquid water and gas permeability of partially saturated ordinary concrete.” *Construction and Building Materials,* 65, 551-565.

Lafhaj, Z., Goueygou, M., Djerbi, A., Kaczmarek, M. (2006). “Correlation between porosity, permeability and ultrasonic parameters of mortar with variable water/cement ratio and water content.” *Cememt and Concrete Research,* 36(4), 625-633.

Le, L. B. N., Stroeven, M., Sluys, L. J., Stroeven, P. (2013). “A novel numerical multi-component model for simulating hydration of cement.” *Computational Materials Science,* 78, 12-21.

Le, L. B. N. (2015). Micro-level porosimetry of virtual cementitious materials – Structural impact on mechanical and durability evolution, Ph.D. Thesis, Delft University of Technology, Delft, The Netherlands.

Li, K., Stroeven, P., Le, L. B. N. (2015). “Methodology for porosimetry in virtual cementitious composites to economically and reliably estimate permeability.” *Image Analysis and Stereology,* 34(2), 73-86.

Li, K., Stroeven, M., Stroeven, P., Sluys, L. J. (2016). “Investigation of liquid water and gas permeability of partially saturated cement paste by DEM approach.” *Cememt and Concrete Research,* 83, 104-113.

Muller, A. C. A., Scrivener, K. L., Gajewicz, A. M., McDonnald, P. J. (2013). “Densification of C-S-H measured by 1H NMR relaxometry.” *The Journal of Physical Chemistry C,* 117 (1), 403-412.

Pignat, C., Navi, P., Scrivener, K. (2005). “Simulation of cement paste microstructure hydration, pore space characterization and permeability determination.” *Materials and Structures,* 38(4), 459-466.

Stroeven, P., Hu, J., Koleva, D. A. (2010). “Concrete porosimetry: Aspects of feasibility, reliability and economy.” *Cememt and Concrete Composites*, 32(4), 291-299.

Stroeven, P., Le, L. B. N, Sluys, L. J., He, H. (2012). “Porosimetry by double-random multiple tree structuring in virtual concrete.” *Image Analysis and Stereology*, 31(1), 55-63.

Stroeven, P., Li, K., Le, L. B. N., He, H., Stroeven, M. (2015). “Capabilities for property assessment on different levels of the micro-structure of DEM-simulated cementitious materials. ” *Construction and Building Materials*, 88, 105-117.

Ye, G. (2005). “Percolation of capillary pores in hardening cement pastes.” *Cememt and Concrete Research,* 35(1), 167-176.

Zalzale, M., McDonnald, P. J., Scrivener, K. L. (2013). “A 3D lattice Boltzmann effective media study: understanding the role of C-S-H and water saturation on the permeability of cement paste.” *Modelling and Simulation in Materials Science and Engineering*, 21(8) 085016.

Zamani, S., Kowalczyk, R. M., McDonald, P. J. (2014). “The relative humidity dependence of the permeability of cement paste measured using GARField NMR profiling.” *Cememt and Concrete Research*, 57, 88-94.