 doi:10.20944/preprints201812.0073.v1

Peer-reviewed version available at *Magazine of Concrete Research* **2019**; doi:10.1680/jmacr.19.00135

Relationship between water-permeability coefficient and pore-structure indicators of cementitious materials

Yuya Sakai[a]

[a] Institute of Industrial Science, The University of Tokyo, 4-6-1 Komaba, Meguro, Tokyo 153-8505, Japan, ysakai@iis.u-tokyo.ac.jp

Abstract

To understand the relationship between water permeability and the pore structure of cementitious materials, literature that studied both water permeability and the pore structure are reviewed, and their correlation is studied. Although the majority of this data is for cement paste, mortar, concrete, and cemented soil are also included. Based on this study, it is established the determination coefficient between water permeability and the total pore volume is very low; however, those between water permeability and the critical, threshold, and median pore diameters, respectively, are moderate. On the other hand, the threshold pore diameter, derived based on the percolation theory, exhibits a very high determination coefficient. The calculated water permeability, using the Katz and Thompson equation, agrees with the reported water permeability quantitatively to a certain level, but is overestimated.

Key words: Pore structure, water permeability, cement paste, mortar, concrete, cemented soil

1. Introduction

Most of the deterioration phenomena in concrete structures, including freezing and thawing, rebar corrosion, alkali silica reactions, etc., are caused by water penetration into concrete. Hence, the evaluation of the resistance against water mobility in concrete is critical. Water permeability is one of the indicators of water mobility; however, its evaluation is not easy. For example, for very dense concrete, water permeability evaluation is time consuming, even requiring few weeks at times [1]. This is not preferable because the cement paste matrix may change during this period due to hydration or leaching. Applying high pressure to the permeating water can reduce the time required for this test, but it may increase the risk of water leakage [2]. If water permeability is correlated with another property, such as the pore structure, this difficult test is not required. Moreover, this can be beneficial for numerical simulation because researchers are trying to develop numerical simulations that simulate the pore structure based on the mix design [3, 4], and this relationship is important for estimating the water permeability based on the pore structure. Thus far, several researchers have studied the relationship between water permeability and the pore structure; the relationship between water

---

Mercury Intrusion Porosimetry (MIP)

permeability and the total pore volume or the pore volume of a certain pore-size range [5-9] or critical pore diameter [10-15], have been reported. Others have used the critical pore diameter [16], median pore diameter [17], or threshold pore diameter [18] as indicators of the pore structure. In these researches, most of the pore-structure indicators in each paper showed good correlation with the water permeability. This is because each research used only their data or few additional data from other researches. As a result, the pore-structure indicator that governs water permeability or the indicator that should be used to estimate water permeability, is not yet clear. It is also important to understand the water permeation phenomena in concrete. With numerical simulation, the water permeability can be accurately calculated based on the simulated pore structure, if their relationship is established. Furthermore, as mentioned earlier, it is difficult to perform the water permeability test appropriately. However, once the relationship between the pore structure and water permeability is understood, the water permeability can be estimated by pore-structure analysis, and the result can be used for validating the appropriateness of the test, by comparison.

In this research, by reviewing literature that studied both water permeability and the pore structure of cementitious material, the pore-structure indicator that exhibits the best correlation with water permeability was studied. In addition, the applicability of the Katz-Thompson model [19, 20] to the water permeability of cementitious material was studied because the applicability of this model to cementitious material is still being debated [11, 13, 15, 21].

2. Literature Reviewing

In this paper, literature on water-permeability measurement as well as pore-structure analysis using MIP were reviewed. There are several reports on the relationship between water permeability and the total pore volume [5-9] or critical pore diameter [10-15]; if they are included, the quantity of data on these indicators would become considerably more than that on the other indicators, rendering comparison unfair. Therefore, in this paper, only literature that reported the pore-size distribution and the various pore-structure indicators, which can be calculated, were reviewed. The literature used in this paper are listed in Table 1; along with the authors, the types of samples are listed. Although most of the data is for cement paste, the data by Tatekawa et al. [24] is for mortar, and those by Sakai et al. [18] is for concrete. The data by de Souza Rodrigues et al. [27] is for cement paste but it includes pulp, whereas that by Matsui et al. [25] is for cemented soil.

Preprints (www.preprints.org) | NOT PEER-REVIEWED | Posted: 6 December 2018 doi:10.20944/preprints201812.0073.v1

Peer-reviewed version available at *Magazine of Concrete Research* **2019**; doi:10.1680/jmacr.19.00135

Table 1 List of literature used in this study

Authors	Specimen	Measured parameter
Cui et al. [22]	Cement paste	Flow
Hughes [23]	Cement paste	Flow
Tatekawa et al. [24]	Mortar	W/C<75%: Depth, W/C>100%: Flow
Matsui et al. [25]	Cemented soil	Flow
Phung et al. [26]	Cement paste	Pressure
de Souza Rodrigues et al. [27]	Cement paste	Flow
Sakai et al. [18]	Concrete	Flow

Li et al. [6], and Reinhardt and Gaber [12] reported only the derivative curves, and pore-structure indicators, other than the critical pore diameter, could not be calculated; therefore, their data was not used. On the other hand, Tatekawa et al. [24] reported the pore-size distribution as the derivative curves, but in their case, the intrusion steps were clear and the data could be converted into cumulative curves for obtaining the pore-structure indicators. The data reported by Goto and Roy [28] could also be converted into cumulative curves, but the obtained maximum cumulative pore volume did not agree with the reported total pore volume and the partial pore volume could not be obtained; therefore, their data was not used in this paper. Research that measured the water permeability of a specimen with intentional damage [29] was not included because the effect of the damage on the water permeability may be predominant rather than the pore structure, and this is not within the scope of this paper. Some studies used SEM [30] and X-ray CT [7] to measure the pore-size distributions, and compared them with the water permeability; however, the scale of the pore size obtained by MIP is different and comparison becomes difficult; therefore, such data were not used in this study.

In Table 1, the parameters measured in the water permeability test are also shown. "Flow" indicates the amount of penetrated water from one side and was measured by applying pressure on the other side. "Pressure" indicates the pressure change, and was monitored, while by applying constant flow. "Depth" indicates the penetrated depth, and was measured, when water was forced into dry specimens. In this paper, the reported values are used as such. Sakai et al. [18] have not published the obtained pore size distribution, but it is as indicated in Figure 1.

Preprints (www.preprints.org) | NOT PEER-REVIEWED | Posted: 6 December 2018 doi:10.20944/preprints201812.0073.v1
Peer-reviewed version available at Magazine of Concrete Research 2019; doi:10.1680/jmacr.19.00135

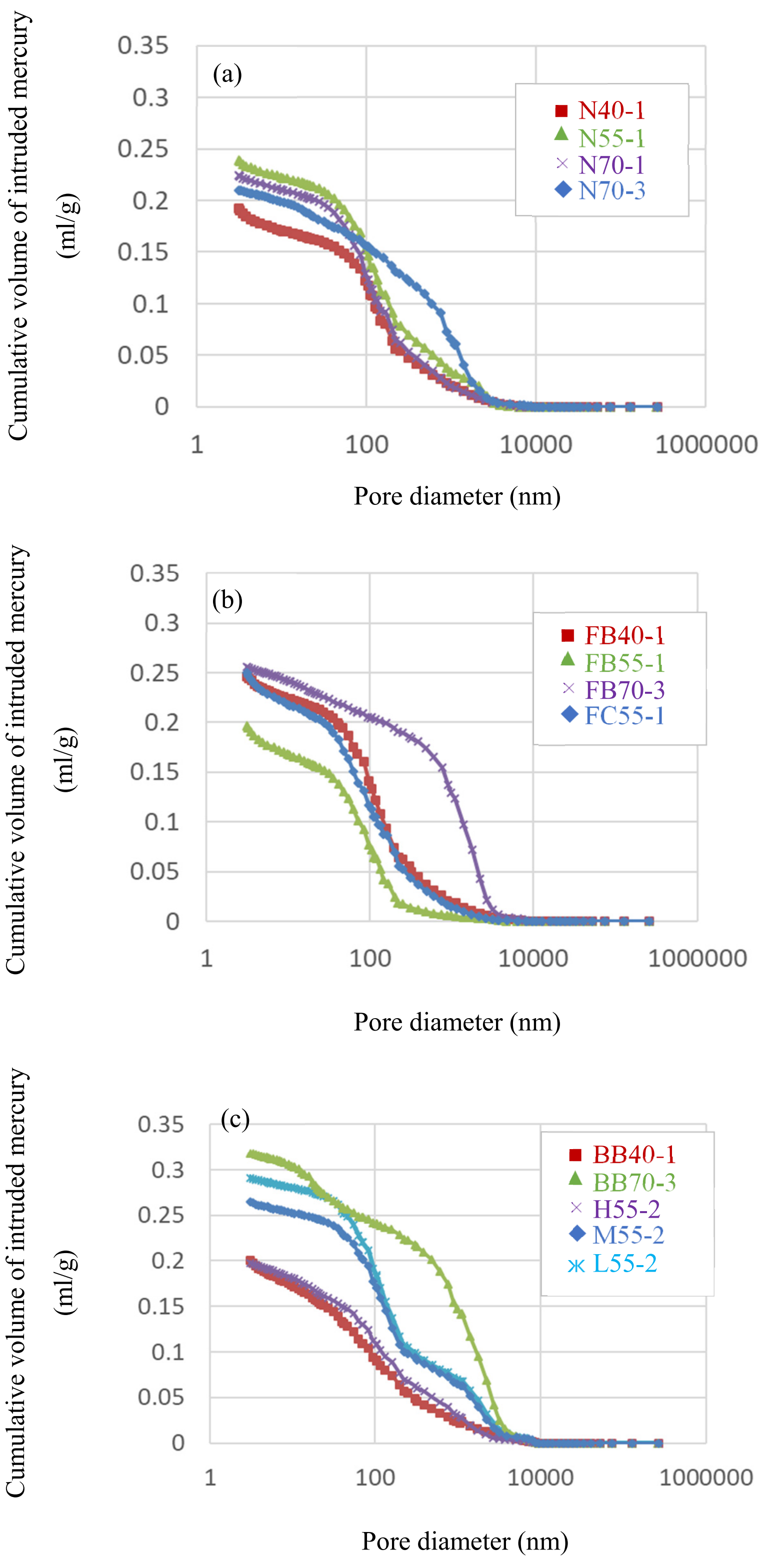

Fig. 1 Cumulative pore-size distribution measured by Sakai et al. [18]

Preprints (www.preprints.org) | NOT PEER-REVIEWED | Posted: 6 December 2018 doi:10.20944/preprints201812.0073.v1

Peer-reviewed version available at *Magazine of Concrete Research* **2019**; doi:10.1680/jmacr.19.00135

3. Methodology

The water-permeability coefficient and pore-structure indicators from the reviewed literature were read using the GSYS software (Japan Charged-Particle Nuclear Reaction Data Group, Japan), and their relationships were examined. As indicators, the total-pore volume, and the critical, threshold, and median pore diameters were considered. The total pore volume and median pore diameter were defined as the maximum cumulative intruded mercury volume, and the corresponding pore diameter, when half the total pore volume was intruded by mercury, respectively. The critical pore volume was defined as the corresponding pore diameter, when the derivative of the pore-size distribution reached a maximum. The definition of the threshold pore diameter has not yet been established [31]; however, in this research, it was defined as the intersection point between the tangential line of the mutational point of the curve slope and the pore-diameter abscissa [11]. Furthermore, we examined the threshold pore diameter defined by Sakai et al. [22] based on the percolation theory, assuming that a connected path was formed, when the volume of intruded mercury reached 16% of the cement paste volume [33, 34], wherein the corresponding pore size was the threshold pore diameter. Katz and Thompson [19, 20] and Thompson et al. [35] proposed the following equations for calculating the permeability coefficient of a porous rock sample using the critical pore diameter, based on the percolation theory:

$$k = \frac{1}{226} \cdot \frac{\sigma}{\sigma_0} \cdot d_c^2 \quad , \tag{1}$$

$$\frac{\sigma}{\sigma_0} = \frac{d_{max}^e}{d_c} \cdot \varphi \cdot S(d_{max}^e) \;, \tag{2}$$

where σ is the conductivity of rock saturated with brine solution having a conductivity σ_0, d_c is the critical pore diameter, d_{max}^e is the electrical conductivity characteristic dimension that produces the maximum conductance ($d_{max}^e = 0.34 \times d_c$ for a very broad pore-size distribution), and $S(d_{max}^e)$ is the fractional volume of the connected pore space involving pores > d_{max}^e. The permeability calculated using Eq. 1 was compared with the reported water permeability.

In some reports, darcy, m/s, or cm/s was used as the unit of water permeability, and these were converted to m^2, in this paper. m/s was converted to m^2 by multiplying by the water viscosity (0.001 Pa·s) and dividing by the water weight per unit volume (9800 N/m^3). Some reports used cc/g or ml/g for the pore volume, and these were converted into the pore volume per unit volume by multiplying with the bulk density, which was calculated using the following equations:

$$\varphi = \varphi_m \cdot \rho_a, \tag{3}$$

$$\rho_a = \frac{1}{\frac{1}{\rho_t} + \varphi_m} \;, \tag{4}$$

where ρ_a is the bulk density, ρ_t is the true density, and φ_m is the total pore volume per unit mass. ρ_t was assumed to be 2.7 g/cm^3 and 2.5 g/cm^3 for cement paste and mortar, respectively.

In the method proposed by Sakai et al. [32], the volume fraction of the cement paste of the sample used in the MIP is required. For the mortar used by Tatekawa et al. [24], and the concrete used by Sakai et al. [18], based on the mix design, the volume fraction of cement paste was calculated by subtracting the volume of sand and gravel from unit volume (1 m^3). Matsui et al. [25] provide the

Peer-reviewed version available at Magazine of Concrete Research 2019; doi:10.1680/jmacr.19.00135

amount of cement and water per sand volume of 1 m^3. In this case, the weight of the cement paste was calculated by adding the weight of cement and water, and the volume of the cement paste was calculated, assuming a bulk density of 2 g/cm^3, for calculating the volume fraction of cement paste in cemented soil.

For comparing the correlations, the determination coefficients were calculated assuming an exponential approximation because all the indicators showed the highest correlation, when an exponential approximation was assumed. In the reviewed literature, the total pore volume was measured by performing MIP or measuring the saturated and oven-dried weights; however, as the latter was employed only in few studies, in this paper, the former was used for comparison.

4. Results

Figures 2–6 depict the comparison between the water permeability and pore-structure indicators. The broken lines in the figures are the regression lines, assuming an exponential approximation. Figure 2 shows the relationship between the total pore volume and water permeability; the determination coefficient was very low at 0.12. Here, data, other than the cement paste, include the effect of sand; hence, the comparison is not fair. However, omitting these data increased the determination coefficient only to 0.16. Therefore, the total pore diameter has low correlation with the water permeability.

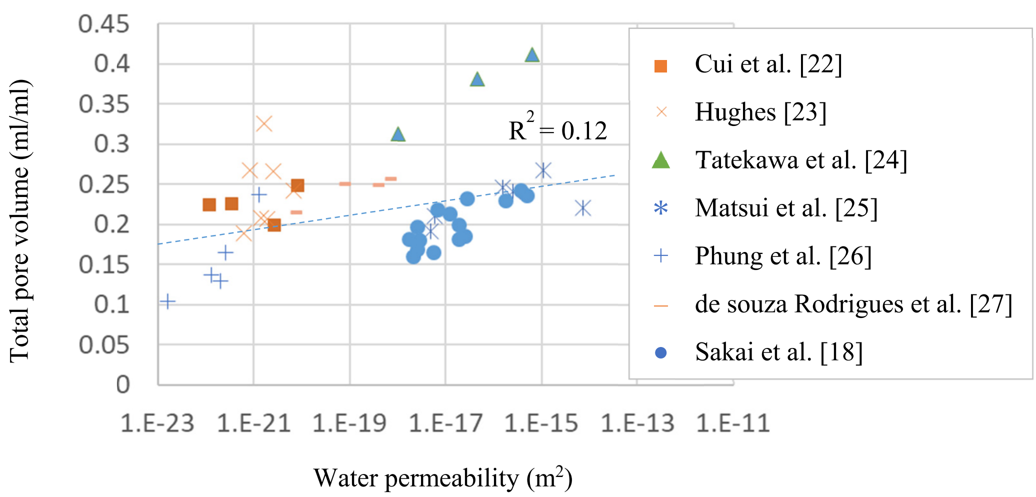

Fig. 2 Relationship between water permeability and the total pore volume

Figures 3–5 compare the water permeability and the critical, median, and threshold pore diameters, respectively. Among them, the critical pore diameter showed the highest determination coefficient at 0.73, whereas those of the median and threshold pore diameters were lower at 0.67 and 0.63, respectively. As mentioned earlier, many researchers have the used critical pore diameter, when discussing the relationship between the pore structure and water permeability, and this was proved to be appropriate. Figure 6 shows the relationship between the water permeability and threshold pore

Preprints (www.preprints.org) | NOT PEER-REVIEWED | Posted: 6 December 2018 doi:10.20944/preprints201812.0073.v1

Peer-reviewed version available at Magazine of Concrete Research 2019; doi:10.1680/jmacr.19.00135

diameter, obtained by a method based on the percolation theory; the determination coefficient was very high at 0.86, indicating that there is strong correlation between them.

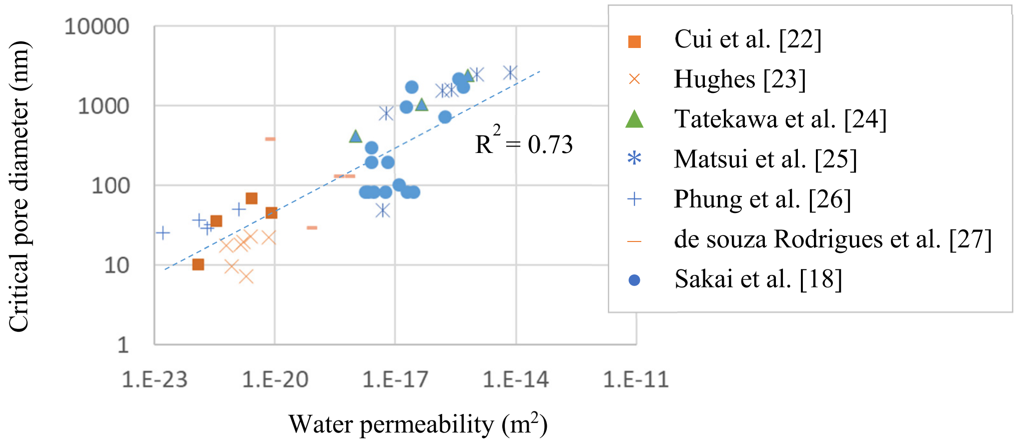

Fig. 3 Relationship between water permeability and the critical pore diameter

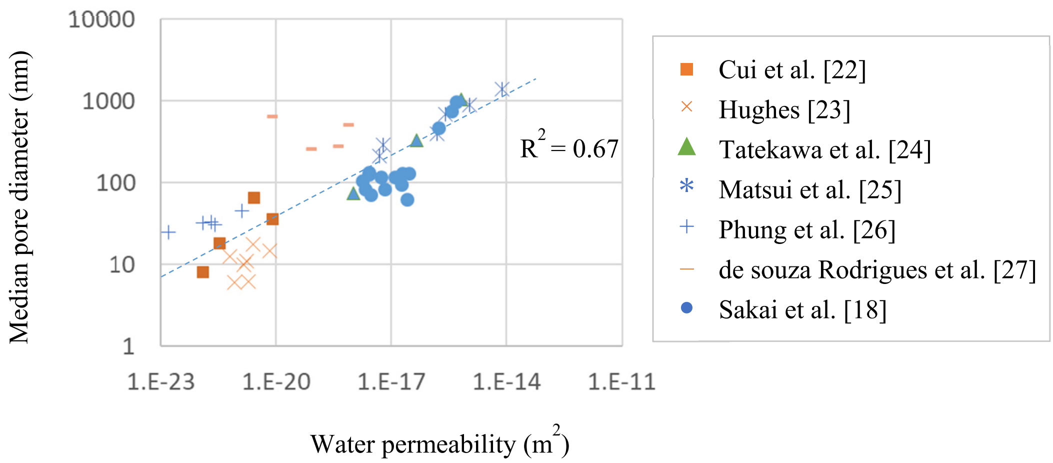

Fig. 4 Relationship between water permeability and the median pore diameter

Preprints (www.preprints.org) | NOT PEER-REVIEWED | Posted: 6 December 2018 doi:10.20944/preprints201812.0073.v1

Peer-reviewed version available at Magazine of Concrete Research 2019; doi:10.1680/jmacr.19.00135

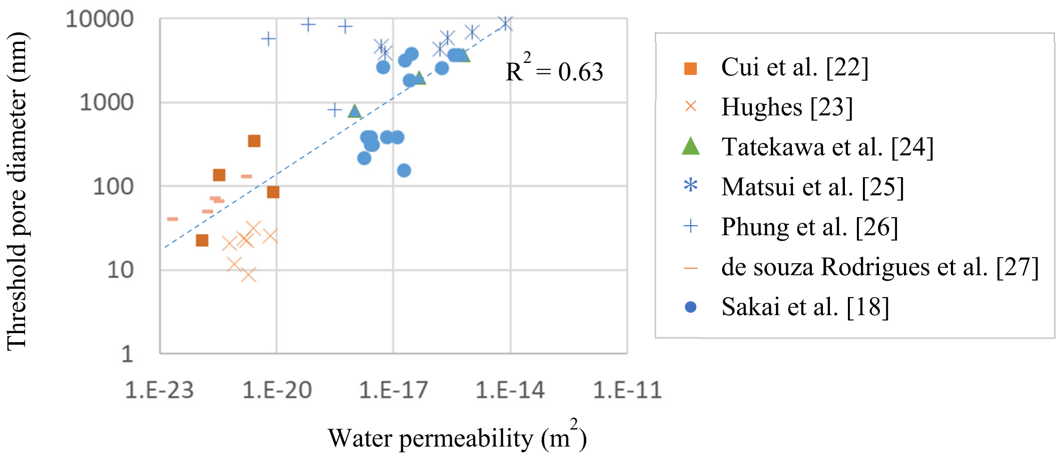

Fig. 5 Relationship between water permeability and the ordinary threshold pore

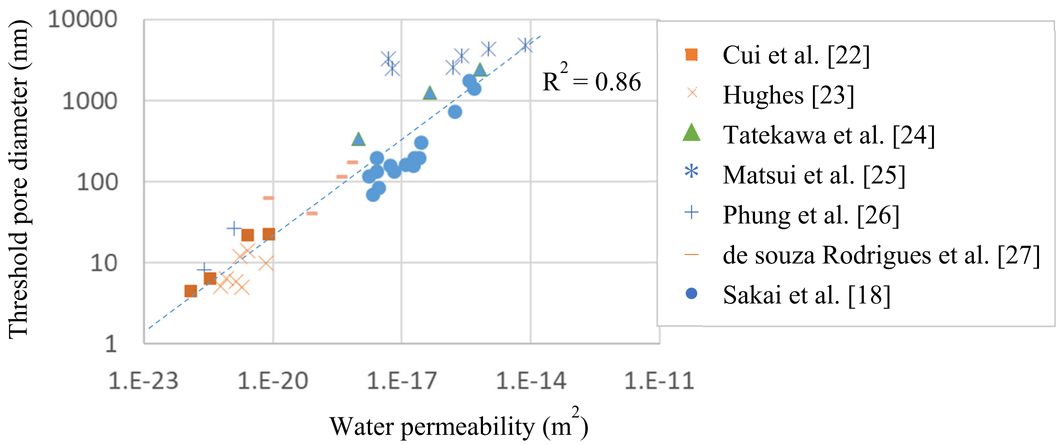

Fig. 6 Relationship between water permeability and the threshold pore diameter obtained based on the percolation theory

Figure 7 depicts the comparison between the water permeability calculated using the Katz-Thompson equation and the measured results. Although the equation overestimates, the values agree quantitatively to a certain degree, even when no adjustment factor is included. El-Dieb and Hooton [13] also reported that the calculation overestimates. Therefore, if some variation is permitted, the Katz-Thompson model can estimate the water permeability quantitatively without an adjustment factor.

Peer-reviewed version available at Magazine of Concrete Research 2019; doi:10.1680/jmacr.19.00135

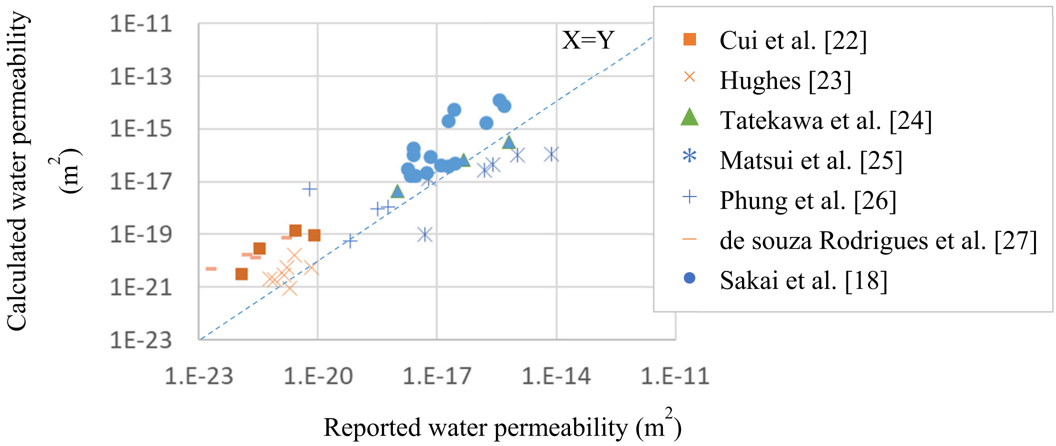

Fig. 7 Relationship between the reported and calculated water permeability using Eqs. 1 and 2

On the other hand, Figure 8 shows the relationship between the measured and calculated water permeability, using the following regression equation. This regression equation was obtained from Figure 6 and is used for calculating the water permeability, using the threshold pore diameter derived based on the percolation theory.

$$k = 2^{-23} \times d_{thp}^{2.2} \quad , \tag{5}$$

where d_{thp} is the threshold pore diameter derived based on the percolation theory.

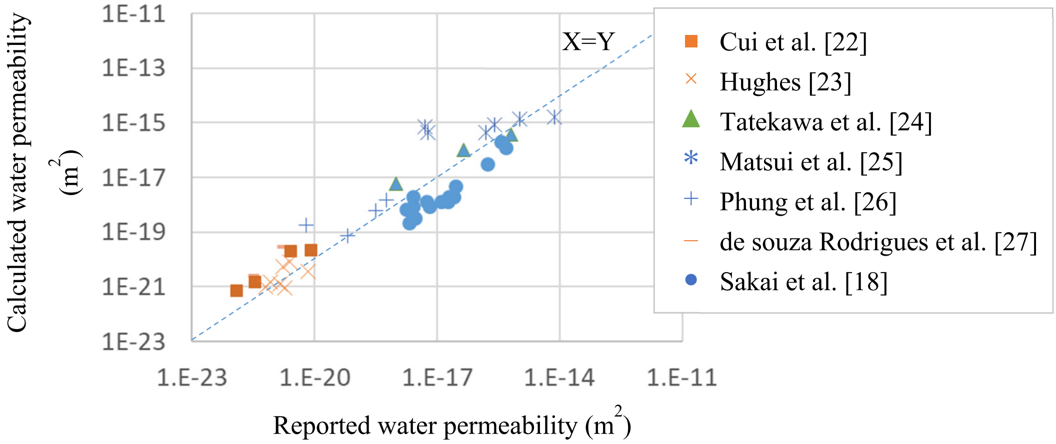

Fig. 8 Relationship between the reported and calculated water permeability using Eq. 5

Figure 8 shows that using Eq. 5, we can evaluate the water permeability of various cementitious material, such as cement paste, mortar, concrete, and cemented soil, more accurately than the Katz-

Peer-reviewed version available at Magazine of Concrete Research 2019; doi:10.1680/jmacr.19.00135

Thompson equation. Therefore, even though the Katz-Thompson equation can estimate the water permeability with a certain accuracy, Eq. 5 provides higher accuracy, as per Figure 8. Therefore, the results indicate that for calculating the water permeability based on the pore structure, the threshold pore diameter can be derived based on the percolation theory and converted to water permeability using Eq. 5. However, in the method proposed by Sakai et al. [28] for obtaining the threshold pore diameter, when the porosity of the cement paste portion is less than 16%, the threshold pore diameter cannot be obtained. This is because the pore is judged to be discontinuous, according to the general percolation theory [29]. We can use the Katz-Thompson equation, but as per Figure 7, this equation overestimates when the water permeability is small; therefore, this equation is unsuitable for estimating the water permeability in this region. Thus, calculation using the critical diameter can be a possible option and the following is the regression equation:

$$\mathrm{k} = 2^{-24} \times d_{cr}^{2.5} \quad , \tag{6}$$

where d_{cr} is the critical pore diameter.

Another problem in the method proposed by Sakai et al. [28] for obtaining the threshold pore diameter is that although the percolation threshold was assumed to be 16%, it is not clear whether this value is optimum because the threshold can deviate from 16%, depending on the conditions [29]. Bentz et al. [32] performed numerically simulated cement hydration and reported that the percolation threshold was 18%. By optimizing the percolation threshold, the correlation between water permeability and the threshold pore diameter can be improved. In either case, the high correlation indicates that water permeability in cementitious materials is likely to be governed by the threshold pore diameter derived based on the percolation theory.

5. Conclusion

Various literature were reviewed, and the relationship between water permeability and the pore structure was studied; the threshold pore diameter derived based on the percolation theory showed the highest determination coefficient. The critical pore diameter showed a relatively high determination coefficient but that of the total pore volume was very low. The water permeability calculated using the regression equation of the threshold pore diameter, derived based on percolation theory, showed higher accuracy compared to that obtained using the Katz-Thompson equation. These results indicate that the water permeability in cementitious materials is likely to be governed by the threshold pore diameter derived based on the percolation theory. Using the obtained regression equation, when the pore structure is obtained by analysis or simulation, the water permeability of cementitious materials can be estimated from the threshold pore diameter derived based on the percolation theory. In this paper, the data includes that of cement paste, mortar, concrete, and even cemented soil. Further studies on the relationship between water permeability and the threshold pore diameter, derived based on the percolation theory, may reveal the limitations of this correlation, if any.

Acknowledgments

This study was supported by JSPS KAKENHI, grant number 18K13809.

References

[1] A.S. El-Dieb, R.D. Hooton, Water-permeability measurement of high performance concrete using a high-pressure triaxial cell, Cem Concr Res. 25(6) (1995) 1199–1208.

[2] G.W. Scherer, J.J. Valenza, G. Simmons, New methods to measure liquid permeability in porous materials, Cem Concr Res. 37(3) (2007) 386–397.

[3] K. Maekawa, T. Ishida, T. Kishi, Multi-Scale Modeling of Structural Concrete: Integrated Material and Structural Mechanics, Taylor & Francis 2008.

[4] C. Pignat, P. Navi, K. Scrivener, Simulation of cement paste microstructure hydration, pore space characterization and permeability determination, Mater. Struct. 38(4) (2005) 459–466.

[5] B. Miloud, Permeability and porosity characteristics of steel fiber reinforced concrete, AJCE 6(4) (2005) 317–330.

[6] K. Li, M. Stroeven, P. Stroeven, L.J. Sluys, Investigation of liquid water and gas permeability of partially saturated cement paste by DEM approach, Cem Concr Res. 83 (2016) 104–113.

[7] M.A.B. Promentilla, S.M. Cortez, R.A.D. Papel, B.M. Tablada, T. Sugiyama, Evaluation of Microstructure and Transport Properties of Deteriorated Cementitious Materials from their X-ray Computed Tomography (CT) Images, Materials (Basel, Switzerland) 9(5) (2016) 388.

[8] Z. Yu, C. Ni, M. Tang, X. Shen, Relationship between water permeability and pore structure of Portland cement paste blended with fly ash, Constr Build Mater. 175 (2018) 458–466.

[9] T.C. Powers, Structure and Physical Properties of Hardened Portland Cement Paste, J Am Ceram Soc. 41(1) (1958) 1–6.

[10] B.K. Nyame, J.M. Illston, Relationships between permeability and pore structure of hardened cement paste, Mag Concrete Res. 33(116) (1981) 139–146.

[11] M.R. Nokken, R.D. Hooton, Using pore parameters to estimate permeability or conductivity of concrete, Mater Struct. 41(1) (2007) 1.

[12] H.W. Reinhardt, K. Gaber, From pore size distribution to an equivalent pore size of cement mortar, Mater Struct. 23(1) (1990) 3.

[13] A.S. El-Dieb, R.D. Hooton, Evaluation of the Katz-Thompson model for estimating the water permeability of cement-based materials from mercury intrusion porosimetry data, Cem Concr Res. 24(3) (1994) 443–455.

[14] P. Halamickova, R.J. Detwiler, D.P. Bentz, E.J. Garboczi, Water permeability and chloride ion diffusion in Portland cement mortars: Relationship to sand content and critical pore diameter, Cem Concr Res. 25(4) (1995) 790–802.

[15] B.J. Christensen, T.O. Mason, H.M. Jennings, Comparison of measured and calculated permeabilities for hardened cement pastes, Cem Concr Res. 26(9) (1996) 1325–1334.

[16] L. Bágel, V. Živica, Relationship between pore structure and permeability of hardened cement mortars: On the choice of effective pore structure parameter, Cem Concr Res. 27(8) (1997) 1225–1235.

[17] D.M. Roy, Relationships Between Permeability, Porosity, Diffusion and Microstructure of Cement Pastes, Mortar, And Concrete At Different Temperatures, MRS Proceedings 137 (1988) 179.

[18] Y. Sakai, C. Nakamura, T. Kishi, Evaluation of mass transfer resistance of concrete based on representative pore size of permeation resistance, Constr Build Mater. 51 (2014) 40–46.

[19] A.J. Katz, A.H. Thompson, Quantitative prediction of permeability in porous rock, Phys Rev B 34(11) (1986) 8179–8181.

[20] A.J. Katz, A.H. Thompson, Prediction of rock electrical conductivity from mercury injection measurements, Journal of Geophysical Research: Solid Earth 92(B1) (1987) 599–607.

[21] E.J. Garboczi, Permeability, diffusivity, and microstructural parameters: A critical review, Cem Concr Res. 20(4) (1990) 591–601.

[22] L. Cui, J.H. Cahyadi, Permeability and pore structure of OPC paste, Cem Concr Res. 31(2) (2001) 277-282.

[23] D.C. Hughes, Pore structure and permeability of hardened cement paste, Magazine of Concrete Research 37(133) (1985) 227-233.

[24] T. Tatekawa, K. Ueno, R. Chikamatsu, Water tightness of low-strength anti-washout under water mortar, Annual Conference of Japan Society of Civil Engineers, 2005.

[25] H. Matsui, K. Horikoshi, K. Higaki, H. Ishii, Permeability of Cement-treated Soil used for a Cutoff Wall, Report of Taisei Technology Center (2009) 1-4.

[26] Q.T. Phung, N. Maes, D. Jacques, G. De Schutter, G. Ye, Effects of W/P ratio and limestone filler on permeability of cement pastes, International RILEM Conference on Materials, Systems and Structures in Civil Engineering, 2016.

[27] C. de Souza Rodrigues, K. Ghavami, P. Stroeven, Porosity and water permeability of rice husk ash-blended cement composites reinforced with bamboo pulp, J Mater Sci. 41(21) (2006) 6925–6937.

[28] S. Goto, D.M. Roy, The effect of w/c ratio and curing temperature on the permeability of hardened cement paste, Cem Concr Res. 11(4) (1981) 575–579.

[29] G. Pijaudier-Cabot, L. Ecay, D. Grégoire, F. Khaddour, Permeability and relative permeability of mortar undergoing damage: a hierarchical capillary bundle approach, 9th International Conference on Fracture Mechanics of Concrete and Concrete Structures (Framcos-9), Berkeley, United States, 2016, p. online.

[30] J. Hu, P. Stroeven, Application of image analysis to assessing critical pore size for permeability prediction of cement paste, Image Analysis & Stereology 22(2) (2011)135–139.

[31] H. Ma, Mercury intrusion porosimetry in concrete technology: tips in measurement, pore structure parameter acquisition and application, J Porous Mat. 21(2) (2014) 207–215.

[32] Y. Sakai, C. Nakamura, T. Kishi, Threshold pore radius of concrete obtained with two novel methods, in: D. Bjegović, H. Beushausen, M. Serdar (Eds.) RILEM International workshop on performance-based specification and control of concrete durability, Zagreb, Croatia (2014).

[33] H. Scher, R. Zallen, Critical Density in Percolation Processes, J ChemPhys. 53(9) (1970) 3759–3761.

[34] D.P. Bentz, E.J. Garboczi, Percolation of phases in a three-dimensional cement paste microstructural model, Cem Concr Res. 21(2) (1991) 325–344.

[35] A.H. Thompson, A.J. Katz, C.E. Krohn, The microgeometry and transport properties of sedimentary rock, Adv Phys. 36(5) (1987) 625–694.

[36] D.P. Bentz, M.A. Ehlen, C.F. Ferraris, J.A. Winpigler, Service life prediction based on sorptivity for highway concrete exposed to sulfate attack and freeze-thaw conditions, National Institute of Standards and Technology, FHWA Report FHWA-RD-01-162, Federal Highway Administration, Washington, DC (2002).