Relationship between air diffusivity and permeability coefficients of cementitious materials

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Abstract

In this study, the relationship between air diffusivity and permeability in cementitious materials was investigated. First, we derived an equation to correlate air diffusivity and permeability in a straight circular tube. Then, we reviewed existing studies that measured both air diffusivity and permeability and compared reported data and calculated values to verify the applicability of the derived equation to cementitious materials. Although a correction factor was not used, the two sets of data showed good agreement quantitatively. This indicates that the derived equation can be applied to cementitious materials including concrete, and measured air diffusivity can be converted to permeability of concrete and vice versa using the derived equation.

Keywords: C. Diffusion, C. Durability, C. Permeability, C. Transport Properties

1. Introduction

The evaluation of concrete durability is becoming more important for rational design and maintenance. Carbon dioxide and oxygen are important deterioration factors of concrete structures as they cause carbonation of concrete and corrosion of reinforcement [1]. Therefore, the appropriate evaluation of resistance against air penetration should be performed to estimate the durability of concrete structures. In general, air diffusivity test [2, 3] or air permeability test [4-6] are conducted to evaluate air penetration in concrete. The driving force of the former is the concentration gradient, whereas that of the latter is the pressure gradient. The condition in a diffusivity test is closer to the real condition of oxygen and carbon dioxide penetration in concrete. However, the experimental setup in a diffusivity test is complicated because the pressure on the two flat surfaces of the sample plate should be kept the same and the concentration of air should to be monitored during the test. On the other hand, in air permeability test, the volume of air penetrating through concrete due to pressure gradient is determined by measuring the volume of air penetration or air pressure, and this test can be conducted using a relatively simple setup. Lately, devices for in-situ investigation of air permeability have been developed [7-10]; however, it is unclear if the actual penetration of oxygen or carbon dioxide due to diffusion can be evaluated using the air permeability test. Correlation between air diffusivity and permeability coefficients has been reported [11-14], but a method to convert air permeability to diffusivity has not yet been established. Once the relationship between air diffusivity and permeability is established, the penetration of carbon dioxide and oxygen can be estimated from air permeability measured with a simple experimental setup or even non-destructive testing.

In this study, a straight circular tube was used to theoretically investigate the relationship between diffusion and air permeability coefficients. Then, studies that measured both the diffusion and permeability coefficients of concrete, mortar and paste were reviewed and a comparison of reported and calculated values was carried out to confirm if the obtained relationship is applicable to actual cementitious materials with complicated pore structure.

2. Derivation of theoretical equation

Air flow can be roughly divided into two types: molecular flow and viscous flow. In molecular flow, collision between air molecules and wall is dominant and this occurs in a small space or depressurized condition. In viscous flow, collision between air molecules is dominant and this occurs in a large space or pressurized condition. The dominant flow can be determined by examining the Knudsen number calculated by the following equation.

$$K_n = \frac{\lambda}{L_s} \quad (1)$$

where λ is the mean free path (m) and L_s is the space size (m). λ can be calculated by the following equation.

$$\lambda = \frac{k_B T}{\sqrt{2}\pi P d^2} \quad (2)$$

where k_B is the Boltzmann constant (= 1.3807×10^{-23} N·m/K), T is the temperature (K), P is the pressure (Pa), and d is the molecular diameter (m). In general, a flow with $K_n < 0.01$ is considered to be viscous flow, that with $K_n > 1$ is considered to be molecular flow, and that with $0.01 \leq K_n \leq 1$ is considered to be transient flow [15, 16]. When molecular flow is dominant, the diffusion coefficient is expressed as follows [17]:

$$D_m = \frac{C_m l}{A + \frac{1}{4}\beta H s \bar{\upsilon} \tau} \quad (3)$$

where C_m is the conductance in molecular flow (m^3/s), l is the distance between two points (m), A is the inner cross-sectional area of a circular tube (m^2), β is the coefficient of surface roughness, H is the tube perimeter (m), s is a constant (less than 1; $1 - s$ indicates the fraction of a specular reflected molecule), $\bar{\upsilon}$ is the root mean velocity of a gas molecule (m/s), and τ is the mean sojourn time of molecules absorbed on a tube surface. The second term in the denominator of Eq. 3 can be ignored because air consists of mostly nitrogen, which has a very short τ of 10^{-12} s, and β and s in concrete have not been established quantitatively. The effect of this term was estimated by Sakai and Kishi [18] and was not large when $D_m > 10^{-5}$ m^2/s. In a straight circular tube, C_m is expressed as follows:

$$C_m = \frac{2\pi r^3 \overline{\upsilon}}{3l} \quad (4)$$

where r is the tube radius (m). The air permeability coefficient when molecular flow is dominant is expressed as follows [19]:

$$k_m = \frac{2\overline{\upsilon}\mu}{3P} r \quad (5)$$

where μ is the viscosity of air (= 2.0 × 10^{-5} Pa·s). By combining Eqs. 3–5, the relationship between air diffusion and permeability coefficients when molecular flow is dominant is obtained as follows:

$$D_m = \frac{P}{\mu} k_m \times 10^4 . \ (6)$$

On the other hand, in a large space where viscous flow is dominant, the diffusion coefficient is equal to that in balk. In this case, the diffusion coefficient is expressed as follows:

$$D_v = D_0 \quad (7)$$

where D_0 is the diffusion coefficient in balk (m^2/s). The diffusion coefficient considering molecular flow, viscous flow and transient flow is expressed as follows:

$$\frac{1}{D} = \frac{1}{D_v} + \frac{1}{D_m} . \ (8)$$

Combining Eqs. 6–8, D can be expressed as follows:

$$D = \frac{D_0 P k_m \times 10^4}{D_0 \mu + P k_m \times 10^4} . \ (9)$$

Fig. 1 shows the relationship between air diffusion and permeability coefficients calculated by Eq. 9 assuming P = 100 kPa, μ = 0.000018 Pa·s, D_0 = 0.7 cm^2/s (diffusion coefficient of nitrogen molecules in air). It can be observed that D is a curve that connects D_m and D_v smoothly. Fig. 2 shows the relationship between air permeability and diffusion coefficients with D_0 = 0.1 cm^2/s (diffusion coefficient of methane molecules in air) and 0.7 cm^2/s and P = 3, 100 and 350 kPa. The pressure of 350 kPa corresponds to the highest absolute pressure applied in the Cembureau method [5] and 3 kPa corresponds to the initial absolute pressure in the Torrent method [7]. It can be observed that D_0 changes the diffusion coefficient in the region with large air permeability. A change in P shifts the curves horizontally.

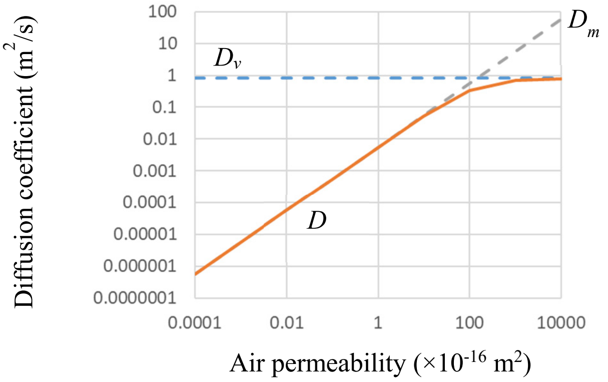

Fig. 1 Relationship between air diffusivity and permeability described by Eq. 9

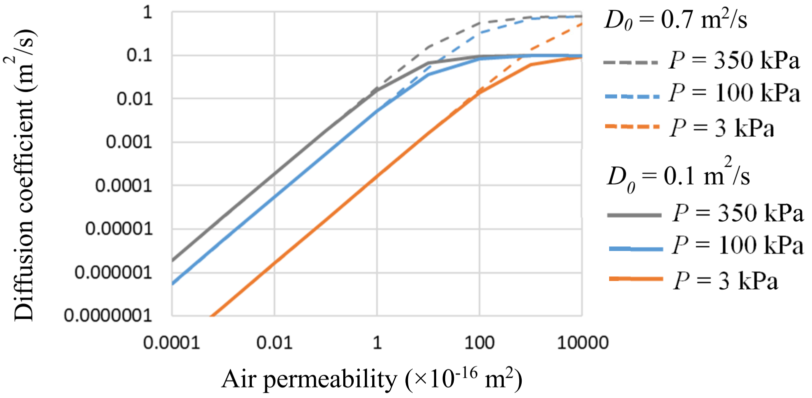

Fig. 2 Relationship between air diffusivity and permeability described by Eq. 9 with different D_0 and P

3. Data from existing studies

Related studies on air diffusivity and permeability were reviewed as listed Table 1. In the table, the type of sample, gas types in diffusion test, as well as gas types, monitored items, units and test pressures in permeability test are presented. The units of air diffusivity and permeability were converted to cm^2/s and m^2 when other units were used. When the reported air permeability value has a unit of m/s, it is multiplied by μ (0.000018 Pa·s) / γ (12.7 N/m^3) to convert to m^2. When the reported air permeability value has a unit of $cm^4/(s \cdot N)$, it is multiplied by $100^{-4} \times \mu$ (0.000018 Pa·s) to convert to m^2. The equations for calculating air diffusivity and permeability were not the same in the studies reviewed; however, the reported values were adopted as they were because the original data were not available and hence, it is impossible to calculate the air diffusivity and permeability using the same approach. In the studies in [12, 21], data of various moisture contents were presented. Fly ash and granulated blast furnace slag were used in [11] and [14], respectively. The samples in [22] were immersed in different aqueous solutions before drying, whereas the samples in [20, 23] were conditioned at various humidity and temperature values.

Table 1 Related works reviewed in this study

References	Specimen	Diffusion test		Permeability test			
		Gas 1	Gas 2	Gas	Monitor	Unit	Pressure (kPa)
[11]	Mortar	CH_4	Air	Air	Flow rate	cm^2	N/A
[12]	Concrete	O_2	N_2	Air	Flow rate	$cm^4/(s \cdot N)$	200
[13]	Concrete	O_2	N_2	Air	Pressure	m^2	3
			N_2	O_2	Concentration	m^2	20-110
[14]	Paste	CO_2	CO_2	Air	Pressure	m^2	150-250
[20]	Concrete	O_2	N_2	O_2	Pressure	m/s	100 → Lower
[21]	Concrete	O_2	N_2	Air	Flow rate	$cm^4/(s \cdot N)$	200
[22]	Mortar	O_2	N_2	O_2	Pressure	m/s	100 → 50
[23]	Mortar	O_2	N_2	O_2	Flow rate	m^2	50-250

4. Results and discussion

Fig. 3 shows the calculated and reported air diffusivity and permeability. The reported data are distributed on two lines with different slopes; these data were obtained for concrete as well as mortar and cement paste. This indicates that a strong correlation exists between air diffusion and permeability coefficients. The calculated values were obtained assuming P = 100 kPa, μ = 0.000018 Pa·s and D_0 = 0.1 cm^2/s instead of 0.7 cm^2/s because Fig. 2 shows that D_0 affects the diffusion coefficient when the air permeability is approximately few m^2 and Sasaki and Miyakoshi [11] obtained the data in this region using methane. Although a correction factor was not used in the calculation, the calculated and reported values show good agreement quantitatively. This indicates that Eq. 9 can describe the relationship between diffusivity coefficient and air permeability in cementitious material. Most of the calculated diffusion coefficients are larger than the reported values. The reasons for this gap are not clear, but possible reasons include the assumptions made in the calculation of the diffusion coefficients and air permeabilities in existing studies and experimental error.

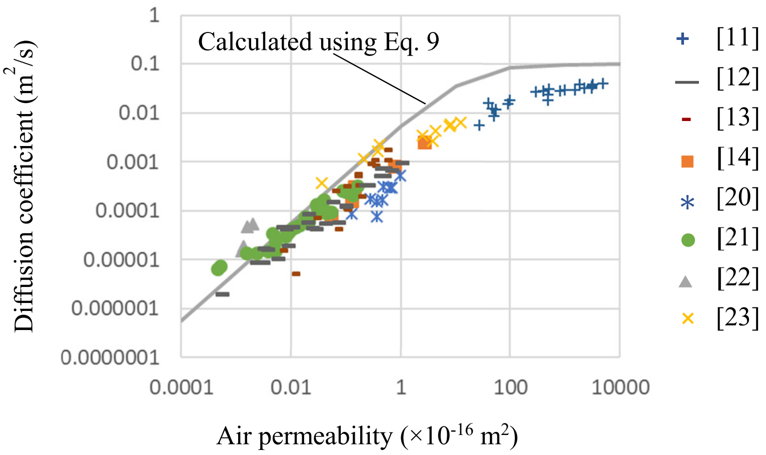

Fig. 3 Relationship between air diffusivity and permeability described by Eq. 9 with different D_0 and P

In both calculation and experiments, there is an inflection point at air permeability of approximately 50×10^{-16} m^2. This indicates that air permeability of 50×10^{-16} m^2 is the boundary between molecular flow and viscous flow. Using Eq. 2, the mean free path in atmospheric pressure (P = 100 kPa) is obtained as 62 nm assuming T = 293 K and d = 0.38 nm (diameter of a nitrogen molecule). Therefore, according to the thresholds described in Section 2, viscous flow becomes dominant when r = 6200 nm and molecular flow is dominant when r = 62 nm. In logarithmic scale, the midpoint of these radii is 620 nm and this radius is the boundary between viscous flow and molecular flow. Therefore, when the measured air permeability is 50×10^{-16} m^2, the representative pore radius in terms of air penetration is 620 nm. Sakai, Nakamura [24] proposed a relationship between air permeability and the representative pore radius as follows:

$$r\ (\text{nm}) = 46\sqrt{k(\times 10^{-16}\, m^2)}\ . \quad (10)$$

According to Eq. 10, when $k = 50\times10^{-16}$ m^2, r is 325 nm, which is close to 620 nm in logarithmic scale. This result further validates Eq. 10.

The agreement in Fig. 3 validates the conversion of air permeability to diffusion coefficient using Eq. 9. As introduced earlier, devices that can evaluate the air permeability of concrete in a non-destructive manner are presently available and we can now obtain the diffusion coefficient of concrete on site using such devices and Eq. 9. Furthermore, Fig. 3 indicates that we do not need to evaluate both air diffusivity and permeability because one of these can be obtained by conversion from the other one. The results obtained in this research will contribute to rational evaluation of the durability of concrete structures.

5. Conclusion

In this study, the relationship between air diffusivity and permeability was investigated using theoretical approach and literature survey. An equation that describes the relationship between air diffusivity and permeability in molecular flow, transition flow and viscous flow was derived. Although a straight circular tube was assumed in the derivation of the equation, the calculated values showed good agreement quantitatively with experimental data. This indicates that air diffusion and permeability are governed by the same factor, possibly the pore structure, and air diffusion can be converted to permeability coefficients and vice versa, using the equation derived in this paper. The studies reviewed in this paper already contain data for concrete, mortar and cement paste of various mix designs prepared under various conditions; however, further tests on samples prepared at extreme conditions are required to determine the limitation of the equation derived in this paper.

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Declarations of interest

None.

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