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Numerical Study on the Relationship among Air Permeability, Pore Size and Pressure Assuming Molecular and Viscous Flows

Yuya Sakai1*

Abstract

The relationship among air permeability, pressure, and pore size from a viscous- to a molecular-flow region is not well understood. In this work, air permeability in a straight circular pipe was studied considering viscous and molecular flows. I learned that the air-permeability coefficient and intrinsic air-permeability coefficient exhibit contrasting pressure dependence: that of the air-permeability coefficient is larger in a larger pore, whereas that of the intrinsic air-permeability coefficient is larger in a smaller pore. I thus proposed a method to obtain the air-permeability coefficient at atmospheric pressure from that measured under vacuum or pressurised condition. From the Reynolds number study, turbulent flow study is unnecessary in air flow in concrete.

1. Introduction

The surface air-permeability test proposed by Torrent to evaluate the quality of a covercrete is a non-destructive test (Torrent 1992, 2009). Good correlations between the surface air-permeability coefficient and various test parameters such as water absorption rate (Beglarigale et al. 2014), carbonation depth (Neves et al. 2015), and diffusion coefficient of a chloride ion have been reported (Andrade et al. 2000). Switzerland has established a specification that requires the evaluation of mass-transfer resistance in covercrete by performing certain tests, including the Torrent method (AGB 2007/007). Although the Torrent method has been used worldwide, the relationship among air permeability, pressure P, porosity ε, and pore size r of concrete is not very well understood. For example, intrinsic air-permeability coefficient $k'$ (m²) has been reported to be affected by mean pressure (Monlouis-Bonnaire et al. 2004; Picandet et al. 2009; Villar et al. 2015), but $k'$ measured by the Torrent and Cembureau methods (Kollek 1989) agreed well even though the pressure P values at the concrete surface in these tests were different, which were 30 and 3000 hPa at the most (Ebensingperger and Torrent 2012). One of the effective approaches to examine the relationship among air permeability, P, ε, and r is to perform a theoretical calculation. In previous papers, viscous flow was assumed, and $k'$ or air-permeability coefficient k (m/s) was formulated to be proportional to the square of the pore size (Dullien 1975; Gueguen and Dienes 1989; Roy et al. 1993). However, as discussed in Section 2.2, air permeability can differ by 10 or 100 times depending on whether viscous or molecular flow is assumed (collision between the air molecule and pore wall is dominant in the molecular flow). The air permeability and these parameters must be properly correlated to quantitatively investigate the air transport in concrete because the dominant flow can vary with $P_m$ and r. Klinkenberg (1941) proposed a correction to consider the effect of slippage on the pore wall, but the effect of the pore size or porosity was not clear. I have conducted numerical simulation assuming a molecular flow to study the relationship among $k'$, ε, and r because molecular flow becomes dominant in a small space such as a pore in concrete (Sakai and Kishi 2016). However, air flow could be a viscous or transition flow in a large pore or under high pressure. In the present study, first, the equations that correlate k, $k'$, P, and r from a viscous to molecular flow will be derived, assuming air flow in a straight circular tube. The validity of this assumption will be discussed later. Then, the effect of P and r on k and $k'$ will be discussed using the derived equations.

2. Calculation method of air flow in a straight circular tube

2.1 Viscous and molecular flows

Viscous flow, where the collision among molecules is dominant, occurs when the space is sufficiently large compared with mean free path $\lambda$, which is the mean distance before a gas molecule collides with another one. On the other hand, molecular flow, where the collision between a molecule and the pore wall is dominant, occurs when $\lambda$ is close to space size $L_s$ in a small pore or under vacuum. The dominant flow is determined by Knudsen number $K_n$, which is the ratio between $\lambda$ and $L_s$ (Kumagai et al. 1970). $\lambda$ and $K_n$ are calculated as

$$\lambda = \frac{k_b T}{\sqrt{2\pi P d^2}}$$  (1)
\[ K_s = \frac{\lambda}{L_s} \]  

(2)

where \( \lambda \): mean free path (m), \( k_B \): Boltzmann constant \((=1.3807 \times 10^{-23} \text{ N·m/K})\), \( T \): temperature (K), \( P \): pressure (Pa), \( d\): molecular diameter (m) and \( L_s \): space size (m).

Generally, a flow with \( K_s < 0.01 \) is considered to be a viscous flow, that with \( K_s > 0.3 \) is considered to be a molecular flow, and that with \( 0.01 \leq K_s \leq 0.3 \) is considered to be a transient flow (Hayashi 1987). In the Torrent method, a concrete surface is vacuumed up to 30 hPa. Assuming \( T = 293 \text{ K}, P = 30 \text{ hPa}, \) and \( d = 0.38 \text{ nm} \) (diameter of a nitrogen molecule), the mean free path is 2100 nm. Under this condition, according to the above-mentioned classification, viscous flow is dominant only in a space larger than 0.2 mm, such as a macroscopic crack. Under atmospheric pressure \((P = 1013 \text{ hPa})\), viscous flow is dominant only in a space larger than 6200 nm, which corresponds to very coarse pores in concrete. Under \( P = 3000 \text{ hPa}, \) which is a common condition, viscous flow is dominant in a space larger than 2100 nm.

In both viscous and molecular flows, the ratio of the mass flow to the pressure difference is called conductance (Kumagai et al. 1970).

\[ Q = C \cdot \Delta P \]  

(3)

where, \( Q \): mass flow \((\text{Pa·m}^3/\text{s})\), \( \Delta P \): pressure difference (Pa) and \( C \): conductance \((\text{m}^3/\text{s})\).

The conductance values in viscous and molecular flows along a cylindrical tube of radius \( r \) are calculated using the following equations:

\[ C_v = \frac{r^4 \pi P_m}{8 \mu d} \]  

(4)

\[ C_m = \frac{2 \pi r^3 \sigma}{3 l} \]  

(5)

\[ \sigma = \sqrt{\frac{3RT}{M}} \]  

(6)

where \( C_v \): conductance in viscous flow \((\text{m}^3/\text{s})\), \( C_m \): conductance in molecular flow \((\text{m}^3/\text{s})\), \( r \): tube radius (m), \( P_m \): mean pressure of two points (Pa), \( \mu \): viscosity (Pa·s), \( l \): distance between two points (m), \( \sigma \): mean velocity of gas molecules \((\text{m/s})\), \( R \): gas constant \((= 8.31 \text{ J·K}^{-1} \cdot \text{mol}^{-1})\) and \( M \): molar mass \((\text{kg/mol})\).

2.2 Theory of air- and intrinsic air-permeability coefficients

In the current research, air flow in a straight circular tube was studied. The mass flow of air molecules between two points in a circular tube was calculated using the following equations derived from Eqs. (3)-(5).

\[ Q_v = C_v \cdot \Delta P = \frac{A r^2 P_m \Delta P}{8 \mu l} \]  

(7)

\[ Q_m = C_m \cdot \Delta P = \frac{2 \pi r^2 \gamma \Delta P}{3 l} \]  

(8)

where \( Q_v \): mass flow for viscous flow \((\text{Pa·m}^3/\text{s})\), \( A \): cross-sectional area \((\text{m}^2)\) and \( Q_m \): mass flow for molecular flow \((\text{Pa·m}^3/\text{s})\).

The theoretical air-permeability coefficient of a viscous flow in a circular pipe was derived as follows. First, Darcy’s law is expressed by Eq. (9).

\[ v = k \cdot i_d \]  

(9)

Under atmospheric pressure \((P = 1013 \text{ hPa})\), viscous flow is dominant only in a space larger than 6200 nm, which corresponds to very coarse pores in concrete. Under \( P = 3000 \text{ hPa}, \) which is a common condition, viscous flow is dominant in a space larger than 2100 nm.

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where \( Q_v \): mass flow for viscous flow \((\text{Pa·m}^3/\text{s})\), \( A \): cross-sectional area \((\text{m}^2)\) and \( Q_m \): mass flow for molecular flow \((\text{Pa·m}^3/\text{s})\).

The theoretical air-permeability coefficient of a viscous flow in a circular pipe was derived as follows. First, Darcy’s law is expressed by Eq. (9).

\[ v = k \cdot i_d \]  

(9)

Dividing Eq. (7) by \( P_m \) and \( A \) yields the following equation:

\[ \frac{Q_v}{P_m A} = \frac{V_v}{A} = v_v = \frac{r^2 \gamma \Delta P}{8 \mu l} \]  

(12)

where \( v_v \): flow velocity \((\text{m/s})\), \( k \): air-permeability coefficient \((\text{m/s})\), \( i_d \): barometric gradient \((\text{m/m})\), \( \Delta h \): gas head (m) and \( \gamma \): unit weight of gas \((\text{N/m}^3)\).

Substituting Eq. (10) into Eq. (9) yields the following equation:

\[ v = \frac{k \Delta P}{\gamma l} \]  

(11)

Comparing Eqs. (11) and (12) yields

\[ k_v = \frac{r^2 \gamma}{8 \mu} \]  

(13)

where \( k_v \): air-permeability coefficient for viscous flow \((\text{m/s})\).

Eq. (13) agrees with the equation that calculates the air-permeability coefficient derived from the Hagen-Poiseuille equation. Previous studies used the equations in which \( k \) is proportional to the square of \( r \), such as Eq. (13) (Dullien 1975; Gueguen and Dienes 1989; Roy et al. 1993). The equation to calculate the air-permeability coefficient of a molecular flow is obtained as follows, which adopts the same procedure:

\[ k_m = \frac{2 \pi r^2 \gamma}{3 P_m} \]  

(14)

where \( k_m \): air-permeability coefficient for molecular flow \((\text{m/s})\).

Compared with Eq. (13), \( r \) has no exponent, but the denominator in Eq. (14) contains \( P_m \). Figure 1 shows the relationship between \( r \) and calculated \( k_v \) and \( k_m \) with \( P_m = \ldots \)
1013 hPa. \( k_0 \) becomes less than \( k_m \) when \( r \) is smaller than 300 nm; the gap is five times of \( k_0 \) at \( r = 100 \) nm and 50 times of \( k_0 \) at \( r = 10 \) nm. Klinkenberg (1941) discovered that the air and water permeabilities of the same specimen did not agree because the viscous flow could not describe the air permeability in a pore network composed of small pores, and the interaction between the gas molecules and the pore wall must be considered. This interaction is generally called as ‘Klinkenberg effect’. Klinkenberg proposed Eq. (15) to calculate the air permeability in a pore network composed of small pores.

\[
k_\mu = k_i \left(1 + \frac{b}{P}\right) = k_i + \frac{b}{P} K_i
\]

(15)

where \( k_\mu \): effective air permeability coefficient considering Klinkenberg effect, \( k_i \): air permeability coefficient of liquid or air in viscous flow, \( b \): Klinkenberg parameter and \( P \): pressure.

In Eq. (15), the first term is the air permeability coefficient of the viscous flow, and the second term is the air permeability considering the interaction between the gas molecules and the pore wall, i.e. molecular flow. This equation indicates that the summation of the air permeability coefficients of the viscous and molecular flows can describe both types of flows. Therefore, in this study, air-permeability coefficient \( k \) calculated using the following equation expresses the viscous and molecular flows, including the transition flow:

\[
k = k_i + k_m
\]

(16)

The unit of \( k \) is meters per second; however, intrinsic air-permeability coefficient \( k' \) has the unit of square meters, and \( k' \) is commonly used as applied in the Cembureau (Kollek 1989) or Torrent (Torrent 1992, 2009) methods. Therefore, \( k \), \( k_i \), and \( k_m \) were divided by unit weight \( \gamma \) and multiplied by \( \mu \) to convert them into \( k' \), \( k'_i \), and \( k'_m \), respectively.

\[
k' = k'_i + k'_m
\]

(17)

\[
k'_i = \frac{r^2}{8}
\]

(18)

where \( k' \): intrinsic air-permeability coefficient (m²), \( k'_i \): air-permeability coefficient for viscous flow (m²) and \( k'_m \): air-permeability coefficient for molecular flow (m²).

The flow velocity is calculated by the following equation and not by Eq. (11):

\[
v = \frac{k'}{\mu} = \frac{k' \Delta P}{\mu (l - d)}
\]

(20)

In this work, \( k \) and \( k' \) of a straight circular pipe with one end under atmospheric pressure (\( P = 1013 \) hPa) and the other end under pressure \( P \) were studied. In the calculation, \( M = 28.8 \times 10^{-3} \) kg/mol, \( T = 293 \) K, and \( \mu = 1.84 \times 10^{-5} \) Pa·s were used assuming air temperature of 20 °C. \( \gamma \) was calculated from the following equation, which is derived from the equation of an ideal gas state:

\[
\gamma = \frac{P_o g M}{RT}
\]

(21)

where \( g \) is the gravitational acceleration (= 9.8 m/s²).

We should note that the \( k \) and \( k' \) values calculated in this work do not quantitatively agree with the experimentally obtained results because the coefficients calculated in this work are the proportional constants between the pressure gradient and the flow volume divided by the ‘inner area of the pipe (pore)’, whereas the experimentally obtained coefficients are the proportional constants between the pressure gradient and the flow volume divided by the ‘measured area’.

In this study, each air permeability coefficient was calculated assuming single \( r \) and pressure. Actual concrete is composed of pores with various radii from the nanometer to the millimeter scale, and air cannot migrate through the saturated pore. Therefore, the \( r \) value used in this study corresponded to the pore radius that represented all unsaturated pores. We should keep in mind that the calculations in this study were performed assuming a straight circular tube, and the absolute values of the obtained results did not agree with those of actual concrete. This point will be discussed later.

3. Results and discussion

3.1 Relationship among air-permeability coefficient, intrinsic air-permeability coefficient, and pressure

An example of calculated \( k, k_i, \) and \( k_m \) with \( r = 100 \) nm is shown in Fig. 2(a), which shows that \( k \) smoothly connects \( k_i \) and \( k_m \). \( k_m \) does not change by \( P \), however, \( k_i \) increases with \( P \), and \( k \) increases with increasing \( P \). Figure 2(b) shows an example of calculated \( k', k'_i, \) and \( k'_m \) with \( r = 100 \) nm. \( k'_i \) does not change with \( P \); however, \( k'_m \) decreases with \( P \), and \( k' \) decreases with decreasing \( P \). This difference in the response of \( k \) and \( k' \) with \( P \) is attributed to the fact that \( k' \) was calculated by dividing \( k \) by
γ', which increases with P. As discussed in the previous section, the measured intrinsic air permeability has been reported to decrease with increasing pressure (Monlouis-Bonnaire et al. 2004, Picandet et al. 2009, Villar et al. 2015), and this trend agrees with the results shown in Fig. 2(b). From the calculation, this decrease in k' due to increasing P is attributed to the pressure dependence of k_m'.

3.2 Effect of pipe size and pressure
The changes in k and k' by P and r are shown in Fig. 3(a) with P as the horizontal axis. With smaller r, k' is more affected by P than k. This result indicates that the gap between k' under atmospheric pressure and k' under pressurised or vacuum condition increases in dense concrete. The Cembureau method increases the pressure up to 3000 hPa, and the measured result is expressed in terms of k'. The right plot in Fig. 3(a) shows that the Cembureau method underestimates k' compared with k' under atmospheric pressure. On the other hand, the Torrent method vacuums the concrete surface up to 30 hPa, and the measured k' is overestimated compared with k' under atmospheric pressure. These test methods still provide reasonable results because the gap between k' measured at these test pressure values and under atmospheric pressure is twice at the largest, and k' is generally expressed in a logarithmic scale. The above-mentioned trend is reversed when the results are expressed in terms of k (m/s). The left plot in Fig. 3(a) shows that the k values measured using pressurised test
overestimates $k$ under atmospheric pressure, and the $k$ values measured using vacuum test underestimates $k$ under atmospheric pressure. We should note that the calculations in this study were performed assuming air flow in a straight circular tube. The calculated air permeability cannot be applied to the actual air flow in concrete, which has a complicated pore structure. However, in the tendency shown in Fig. 3, for example, $k$ (m/s) with larger pressure dependence in a larger pore remains valid regardless of the complexity of the pore structure.

The right plot in Fig. 3(a) shows that $k'$ measured by the Torrent method becomes four times larger than $k'$ measured by the Cembureau method due to the pressure difference during the test; however, similar values have been reported (Ebensperger and Torrent 2012). This contradiction can be explained if actual $k'$ measured by the Torrent method is larger than that measured by the Cembureau method due to the complexity of the pore structure. The $k'$ value measured by the Torrent method is larger than the actual value, and the calculated $k'$ value becomes similar to $k'$ using the Cembureau method. According to a previous study (Sakai and Kishi 2016), when $k'$ is calculated with $\varepsilon = 0.15$ while actual $\varepsilon = 0.03$, the calculated $k'$ value becomes one-fifth compared with that calculated using actual $\varepsilon$. Therefore, the overestimated porosity could have possibly cancelled the gap among the air permeabilities measured under different pressures. The measured total porosity of concrete can be approximately 0.15 or more (Neithalath et al. 2010). However, the volume of the active path for air migration can be smaller than the actual total pore volume because pores that are smaller than few nanometers, such as the interlayer space (Prasuhn 1995), have large resistance, and these pores may contribute less to air migration. Of course the differences in the specimen condition (e.g. moisture content and pore structure, among others) or the testing condition could have probably caused the agreement between the test results measured using the Torrent and Cembureau methods even though the testing pressures were different. The important point is that even if the pore structure and water content are the same and uniform, theoretically, the test results from the Torrent and Cembureau methods do not agree due to the pressure difference in these tests.

The change in $k$ and $k'$ by different $P$ is shown in Fig. 3(b) with $r$ along the abscissa. The change in $k$ and $k'$ varies according to $r$. These results indicate that $r$ can be obtained by conducting air-permeability test using different $P$ values. However, to be precise, $r$ cannot be calculated from the results of the Torrent and Cembureau methods because, as explained earlier, $k'$ calculated by the Torrent method includes the effect of assumed $\varepsilon$, and $k'$ obtained by the Cembureau method includes the effect of actual $\varepsilon$. In addition, the Torrent method is not affected by the tortuosity of the pore network when the concrete is sufficiently thick because the effective depth, which is used in the calculation of air permeability, agrees with the actual length. On the other hand, the Cembureau method is affected by the tortuosity of the pore network because $k'$ is calculated by multiplying the specimen thickness, which does not consider the tortuosity. Therefore, the pressure needs to be varied in the same test method to obtain $r$ from the air-permeability test.

Figure 3 shows that the air-permeability test using pressurised air is better than that under vacuum because the change in $k$ or $k'$ due to $r$ is larger under higher pressure. Figure 4 shows the relationship between $r$ and the ratio of $k'$ at different $P$ values. This result shows that a larger pressure difference is favourable to obtain $r$ due to the larger change in the ratio.

### 3.3 Calculation of $k'$ at atmospheric pressure and $\varepsilon$

$\varepsilon$ can be calculated using $r$ obtained from the air-permeability tests under different pressures. The equations to obtain $\varepsilon$ are expressed below.

In the Cembureau method, $k'_c$ is calculated by the following equation (Kollek 1989):

$$k'_c = \frac{2P_c Q_c \mu}{A (P_c^2 - P_a^2)}$$

(22)

where $k'_c$: intrinsic air-permeability coefficient measured by the Cembureau method (m²), $Q_c$: average flow rate (m³/s), $l$: specimen thickness (m) and $P_c$: applied pressure (Pa).

When straight circular tubes are assumed, $k'_c$ is calculated using Eqs. (17)-(19).

$$k'_c = \left(\frac{r^2}{8} + \frac{27\mu r}{3P_m}\right) \times \varepsilon_a$$

(23)

Here, the right-hand side member is multiplied by $\varepsilon_a$ because, as explained in the last section, $k'$ is the proportional constant between the pressure gradient and the flow volume divided by the ‘inner area of the pipe (pore)’, whereas measured $k$ is the proportional constant between the pressure gradient and flow volume divided by the ‘measured area’. Rearranging Eq. (23) yields the following equation:

$$\varepsilon_a = \frac{24P_m}{3P_m r^2 + 167\mu r} k'_c$$

(24)

Fig. 4 $r$ and the ratio of $k'$ at different $P$ values.
In the study by Villar et al. (2015), pressure in the air-permeability test was varied from 3000 hPa to 1 MPa, and the ratio of \( k' \) at \( P = 3000 \) hPa and \( P = 1 \) MPa was 1.4. Figure 4 shows that the ratio of \( k' \) at \( P = 3000 \) hPa to 1 MPa is 1.4 when \( r = 250 \) nm, and Eq. (24) yields \( \varepsilon_a = 0.002 \) using this \( r \). Figure 5 shows the data measured by Villar et al. (2015). \( k' \) was calculated using Eq. (23) by varying \( r \) and \( \varepsilon_a \), and the best fit was attained when \( r = 250 \) nm and \( \varepsilon_a = 0.002 \), as shown in Fig. 5. The \( r \) and \( \varepsilon_a \) values that provided the best fit varies depending on the pore structure and water content. The agreement of the values that provided the best fit varies depending on the pore structure and water content. The agreement of the calculated curve and the distribution of the plots indicates the validity of Eqs. (17)-(19) for qualitative discussion. \( k' \) at atmospheric pressure is calculated by substituting \( \mu_n = 1013 \) hPa, \( r = 250 \) nm, \( \varepsilon = 0.002 \), and the other variables into Eq. (23), which yields \( k' = 4.6 \times 10^{-17} \) m\(^2\). The sample tested by Villar et al. (2015) had a water-to-cement ratio (w/c) of 0.4. According to Sakai et al. (2014), the threshold pore, which was regarded as the representative pore to describe the mass-transfer resistance, of concrete at w/c = 0.4 is 52.5 nm, which is one fifth of \( r \) obtained in this study. \( \varepsilon_a = 0.002 \) is very small compared with the typical values. The discrepancy in pore size and porosity indicates the limitation in assuming the pores as straight circular pipes to obtain accurate information about the pore structure by conducting air permeability test at different pressure values.

3.4 Reynolds number

Generally, turbulent flow occurs when \( Re \) is larger than 2000, and \( Re \) is calculated by the following equation (Prasuhn 1995):

\[
Re = \frac{Lv \rho}{\mu}
\]

(25)

where \( Re \): Reynolds number, \( L \): characteristic linear dimension (m) and \( \rho \): density (kg/m\(^3\)).

However, in soil physics, laminar flow occurs when \( Re \) is less than one with the mean grain diameter as \( L \) (Bear 1988). Fig. 6 shows calculated \( Re \) of the investigated cases in this study with \( l = 50 \) mm and \( L = 2r \). \( \nu \) was calculated using Eq. (20). None of the parameters exceeded \( Re = 2000 \) and \( Re = 1 \). The case with \( r = 10 \) \( \mu \)m and \( P = 3 \) MPa showed the highest \( Re \) but it was 0.98. Thus, Fig. 6 shows that the effect of turbulence does not need to be considered for air flow in general concrete.

4. Conclusions

In this study, the equations to calculate the air-permeability coefficient covering from viscous to molecular flow in a straight circular tube has been derived, and the relationship among air permeability, pore size, porosity, and pressure was studied. The following conclusions are obtained:

- The air-permeability coefficient with a unit in meters per second was numerically shown to increase with increasing pressure, whereas the intrinsic air-permeability coefficient with a unit in square meters decreased with increasing pressure.
- Previous studies reported decrease in the intrinsic air-permeability coefficient by increasing the pressure in the air-permeability test, and this decrease was attributed to the decrease in the intrinsic air-permeability coefficient for molecular flow by increasing the pressure.
- The air-permeability coefficient was numerically shown to be more affected by pressure in the concrete with a large pore, and the intrinsic air-permeability coefficient was more affected by the pressure in the concrete with a small pore.
- The procedure to convert the intrinsic air-permeability coefficient obtained in the air-permeability test under vacuum or pressurised condition into that under atmospheric pressure has been presented. This process may contribute to the quantitative discussion of air permeability in actual concrete.
- A procedure to obtain the pore radius and porosity by conducting air-permeability test at different pressure has been proposed, but the obtained values were not realistic, probably because the assumption of pores as straight circular pipes was too simple to obtain accurate pore size and porosity.
- In this research, the Reynolds number was more than one only when the pipe radius was 10 \( \mu \)m and the pressure was 3 MPa. This result shows that turbulence does not need to be considered to evaluate air migra-
tion in concrete because this pipe size is very large compared with the general pore size, and this pressure was very large compared with the atmospheric pressure or the pressure in the air-permeability tests.

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