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Gas transport and scale effect on gas transport parameters in differently textured porous media

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Abstract

The main gas transport parameters, air permeability (k_a) and gas diffusion coefficient (D_p) and their variations with porous media type and air-filled porosity play a key role in gas emission problems including volatilization of toxic chemicals at polluted sites and the production and emission of greenhouse gases. In this study, gas transport parameters (k_a and D_p) were investigated using Toyoura sand and granulated molten slag as fine and coarse materials respectively under different moisture content conditions. The results show that large pore continuous network of porous media enhances gas advection resulting in higher k_a values for molten slag compared to Toyoura sand. However, gas diffusion coefficient was controlled by air filled porosity for large core samples and for small core samples, D_p was dominated by both air filled porosity and tortuosity of pore structure in two porous media. Besides, scale effect of gas transport parameters were investigated using two standard size samples (large core, 2120 cm^3 and small core, 100 cm^3). Measured air permeability and gas diffusion coefficient of Toyoura sand show no scale dependency as little or no variation of k_a and D_p values were observed in this study for two standard size samples. However, coarser molten slag showed considerable scale effect (large difference in measured k_a and D_p values for large core and small core samples) for both the gas transport parameters (k_a and D_p) at different water or air content conditions. For molten slag, small core samples give higher k_a and D_p values compared to large core samples may be due to side wall effect for small core samples which might enhances gas advection and diffusion during measurement.

Keywords: Air permeability, gas diffusion coefficient, pore characteristics indices and scale effect.

1. Introduction

The fate, transport and emission of gases in the soil are mainly controlled by advection and gas diffusion phenomena. Advection is driven by pressure gradient, while gas diffusion is controlled by concentration gradient across the sample. The air permeability (k_a) and gas diffusion coefficient (D_p) are the governing gas transport parameters for gas advection and diffusion processes respectively. Previously, numerous studies were performed on gas transport parameters (k_a and D_p) to determine their controlling factors (Hamamoto et al., 2008; 2009; Rouf et al., 2010) using different porous media. Hamamoto et al. (2008) showed that gas diffusion coefficient was dominated by air filled porosity (ε) only but air permeability was controlled by ε as well as pore structure of porous media. Moreover, Rouf et al. (2010) measured k_a and D_p of six mixtures of molten slag and volcanic ash soil and observed that gas diffusion coefficient was dominated by ε and pore tortuosity of porous media, and air permeability was controlled by large pore continuous network of porous media.

To measure the gas transport parameters accurately, knowledge and understanding of standard size of sample is very necessary for different particle size of porous media. In the past studies, different sizes of samples with differently textured material were used to measure gas transport parameters (k_a and D_p). For example, 100 cm^3 samples were used to measure k_a (Kawamoto et al., 2006; Poulsen et al., 2008) and D_p (Kawamoto et al., 2006; Thorbjørn et al., 2008 and Kristensen et al., 2010) using different types of soil. Fujikawa and Miyazaki (2005) measured gas diffusion coefficient of 100 cm^3 as well as 40 cm^3 samples in two different locations of Japan using sandy and clayey loam soil as porous media. Poulsen et al. (2006) and Hamamoto et al. (2009) measured k_a and D_p of sand materials using 100 cm^3 volume small core samples, while Rouf et al. (2010) measured k_a and D_p using 2120 cm^3 volume sample of six mixtures of molten slag and volcanic ash soil. Moreover, Kawamoto et al. (2010) measured k_a, D_p and soil water characteristics curves for different porous media using 2120 cm^3 volume sample. However, very few study on scale effect (or sample size dependency) of k_a and D_p was performed previously. To the best of our knowledge, only Iversen et al. (2001) conducted measurement of air permeability using two different size samples (100 cm^3 and 3140 cm^3 volume) for in-situ, onsite and laboratory samples and found that structured soils have scale effect for air permeability using two different size samples for in situ measurements, while onsite and laboratory measurement showed very little difference in k_a values. They also suggested that for fine material small volume of samples (100 cm^3) and for coarse material large volume of samples are required to measure the gas transport parameters. Though scale dependency of k_a and D_p for porous media using coarser material is very much challenging, no studies were performed so far on scale effect using coarser material. Therefore, in this study measurement of k_a and D_p for fine Toyoura sand (D_{50} = 0.18 mm) and coarser molten slag (D_{50} = 1.20 mm) was done to investigate the scale effect of these two gas transport parameters. Additionally, the gas transport mechanism through porous media was investigated considering pore characteristics indices calculated from measured gas transport parameters.

2. Materials and Methods

2.1 Materials

Two different porous media Toyoura sand and molten slag were used as testing material to measure the air permeability (k_a) and gas diffusion coefficient (D_p). The Toyoura sand was taken by passing 0.425 mm mesh, washed and air dried. On the other hand, the used molten slag was collected from Centre for Environmental Science Saitama (CESS), Japan and sieved by 2 mm passing and 0.106 mm retained, washed and air dried. The physical properties of Toyoura sand and molten slag were shown in Table 1.

Table 1. Physical properties for Toyoura sand and molten slag.

Parameters	Toyoura Sand	Molten Slag
Particle density (g cm^{-3})	2.64	2.88
Uniformity coefficient	1.50	2.64
D_{50} (mm)	0.18	1.20
Dry bulk density (g cm^{-3})	1.58	1.70
Total porosity (m^{3}m^{-3})	0.40	0.40

2.2 Sample preparation and gas transport parameters measurement

Collected Toyoura sand and molten slag was used for sample preparation by adding water with oven dry materials to make samples at three different moisture content conditions. The thoroughly mixed samples were kept in air tight plastic bag for about one week to make uniform distribution of moisture. Then the prepared samples were packed in large cores (15 cm in inner diameter and 12 cm in height; 2120 cm^3) and small cores (5.6 cm in inner diameter and 4.05 cm in height; 100 cm^3) to measure the gas transport parameters for each porous media. A little exception was made when measurement was done for Toyoura sand with large core samples. Instead of using different samples at different moisture content condition, a unified measurement system with suction control (UMS_SC) as described by (Kawamoto et al., 2010) was used for gas transport parameters measurement by application of suction head taking one 2120 cm^3 sample only.

The packed large core and small core samples dry bulk density was kept almost constant at 1.58 g cm^{-3} (Toyoura sand) and 1.70 g cm^{-3}(molten slag) for each moisture content condition. Small core samples were made in duplicate for measurement of k_a and D_p while single sample measurement was done for large core samples.

2.2.1 Air permeability (k_a)

The air permeability was measured by the modified field air permeameter of (Iversen et al., 2001) as discussed in (Ball and Schjønning, 2002). The air permeameter used in this study is shown in Fig. 1a. This device included a bank of flow meter connected to a stopcock, a digital pressure gauge or water manometer to read pressure difference across the soil sample, an air compressor, two stands and two plates to hold the samples with bolting facility. The volumetric flow rate of air, Q (m^3 s^{-1}), flowing across the soil sample was read from the flow meters, and the applied pressure difference, ΔP (kPa), was recorded using the digital air pressure gauge. To prevent preferential air flow and to ensure no air leaks along the periphery of the steel core, the edges of the soil sample were carefully kneaded and O-ring was used at the top plate. The top plate also has facility to insert two tubes for measuring pressure difference across the sample. In this study, the airflow rate through the porous media was relatively low, so the gas slippage at the grain boundary can be ignored. Under these conditions, Darcy's Law applies, which enables air permeability (k_a, m^2) to be described by equation 1.

$$k_a = \frac{\mu_{vis} Q}{A} \frac{\Delta L}{\Delta P} \tag{1}$$

where μ_{vis} is air dynamic viscosity (Pa s), Q is volumetric airflow rate (m^3 s^{-1}), A is the cross-sectional column area (m^2), ΔL is the sample length (m), and ΔP is the differential pressure (kg m^{-1} s^{-2}) across ΔL . The experiments were done in a climate controlled experimental room with a constant temperature of 20°C and a relative humidity of about 60% so variation in μ_{vis} is negligible.

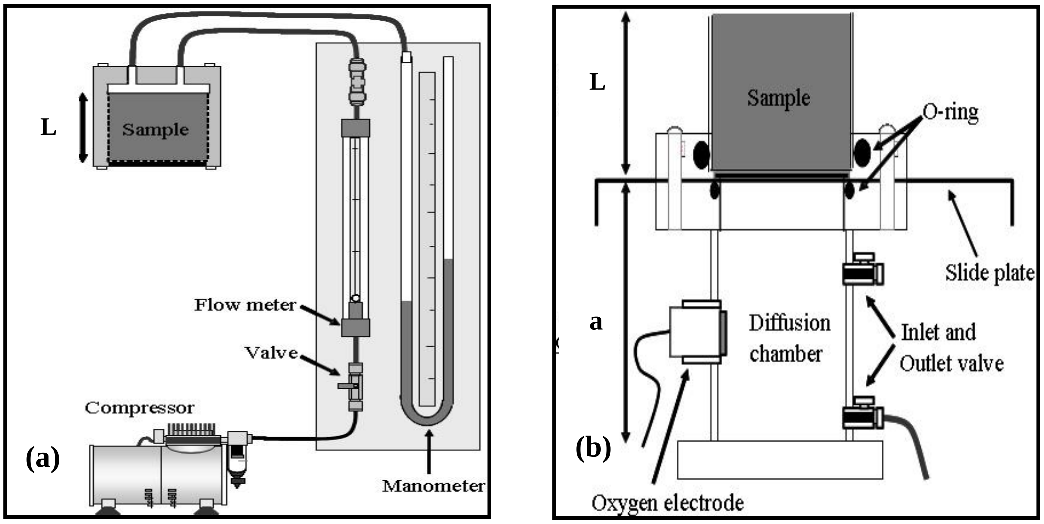

Figure 1. (a) Air permeability (k_a) measuring apparatus, (b) Diffusion chamber to measure gas diffusion coefficient (D_p).

2.2.2 Gas diffusion coefficient (d_p)

The soil-gas diffusion coefficient (D_p) was measured by using the diffusion chamber developed by (Currie, 1960) as shown in Fig. 1b, and recommended by (Rolston and Moldrup, 2002). At first, keeping the soil core in place in the sample holder, the slide was closed to establish no contact between the soil sample and the diffusion chamber. The diffusion chamber was initially flushed with 100 % N_2 gas through the inlet valve and allowed to mix within the diffusion chamber at a substantial time. It was assumed that the gas pressure within the pressure chamber was same as the atmospheric pressure since the same volume of air was flushed out through the outlet port. Contact between the soil core and the diffusion chamber is then established by opening the slide. The upper end of the soil core is exposed to the atmosphere. Binary diffusion of oxygen occurred from the atmosphere through the soil core toward the diffusion chamber while nitrogen diffused from the diffusion chamber to the atmospheres at a closed system (total pressure gradient is constant). Microbial oxygen consumption in the soil samples can be considered negligible for the short measurement time. Mixing of air within the small diffusion chamber was assumed to occur instantly. The oxygen concentration inside the diffusion chamber was measured every minute using an O_2 electrode sensor connected to a CR10X data logger (Campbell Scienctific, USA).

The unsteady state diffusion equation was used to describe the movement of oxygen to the diffusion chamber, and is given by the combination of Fick's first law of diffusion and the continuity equation as,

$$\varepsilon \frac{\partial C_g}{\partial t} = D_p \frac{\partial^2 C_g}{\partial x^2} \tag{2}$$

where C_g is the oxygen concentration inside the chamber at time t, ε is the soil-air filled porosity, x is the distance from the top of the soil core, and D_p is the soil-gas diffusion coefficient. A solution of the diffusion equation at the initial and boundary conditions was given by Carlslaw and Jaeger (1959) for the relative concentration in the chamber (C_r).

$$C_r = \frac{C_g - C_0}{C_i - C_0} = \sum_{n=1}^{\infty} \frac{2h \exp(-D_p \alpha_n^2 t / \varepsilon)}{L(\alpha_n^2 + h^2) + h} \tag{3}$$

Where, C_o is the oxygen concentration at atmosphere and C_i is the initial concentration of oxygen inside the diffusion chamber, $h = \varepsilon / a$ (a is the length of the diffusion chamber), and α is the positive roots of the equation 4 with n = 1, 2, 3…. .

$$(\alpha L)\tan(\alpha L) = hL \tag{4}$$

Neglecting the terms for $n \geq 2$, yields,

$$\frac{C_g - C_0}{C_i - C_0} = \frac{2h \exp(-D_p \alpha_1^2 t / \varepsilon)}{L(\alpha_1^2 + h^2) + h} \tag{5}$$

and expressing the equation into a linear form, yields,

$$\ln\left(\frac{C_g - C_0}{C_i - C_0}\right) = -\frac{D_p \alpha_1^2}{\varepsilon} t + \ln\left(\frac{2h}{L(\alpha_1^2 + h^2) + h}\right) \tag{6}$$

The soil-gas diffusion coefficient (D_p) can be derived from the slope of the plot of ln(C_r) versus time t which becomes linear with slope $-D_p\alpha_1^2/\varepsilon$ for a sufficiently large time (t). Gas diffusivity was calculated by the ratio of D_p/D_0 where D_0 is the gas diffusion coefficient of O_2 in free air at 20^0c equal to 0.20 cm^2s^{-1} (Currie, 1960).

2.2.3 Calculation of Pore Characteristics Indices

The pore characteristics of both porous media were calculated by applying the tube model of (Ball, 1981). He assumed pores of porous media as uniform tortuous and jointed tubes of similar diameter and expressed the tortuosity (τ) (L/L_s, the ratio of tube length (L) to sample length (L_s)) and the equivalent pore diameter (d_{eq}, μm), the effective diameter of the drained pores active in flowing air through the sample, by combining Fick's law and Poiseuille's law as

$$\tau = \sqrt{\frac{\varepsilon}{D_p / D_0}} \tag{7}$$

$$d_{eq} = \sqrt{\frac{32 * k_a}{D_p / D_0}} \tag{8}$$

where k_a is air permeability, D_p/D_0 is gas diffusivity and ε is air filled porosity of porous media.

3. Results and Discussion

3.1 Measured gas transport parameters

Figure 2 shows measured gas transport parameters of Toyoura sand and molten slag at different air or water filled conditions using large and small core samples. The convective (pressure driven) gas transport parameter air permeability (k_a) for both materials was plotted against ε using large core samples (Fig. 2a) and small core samples (Fig. 2b). Calculated gas diffusivity (D_p/D_0) was plotted against air filled porosity (ε) similar to k_a using both Toyoura sand and molten slag for large core (Fig. 2c) and small core (Fig.2d) samples.

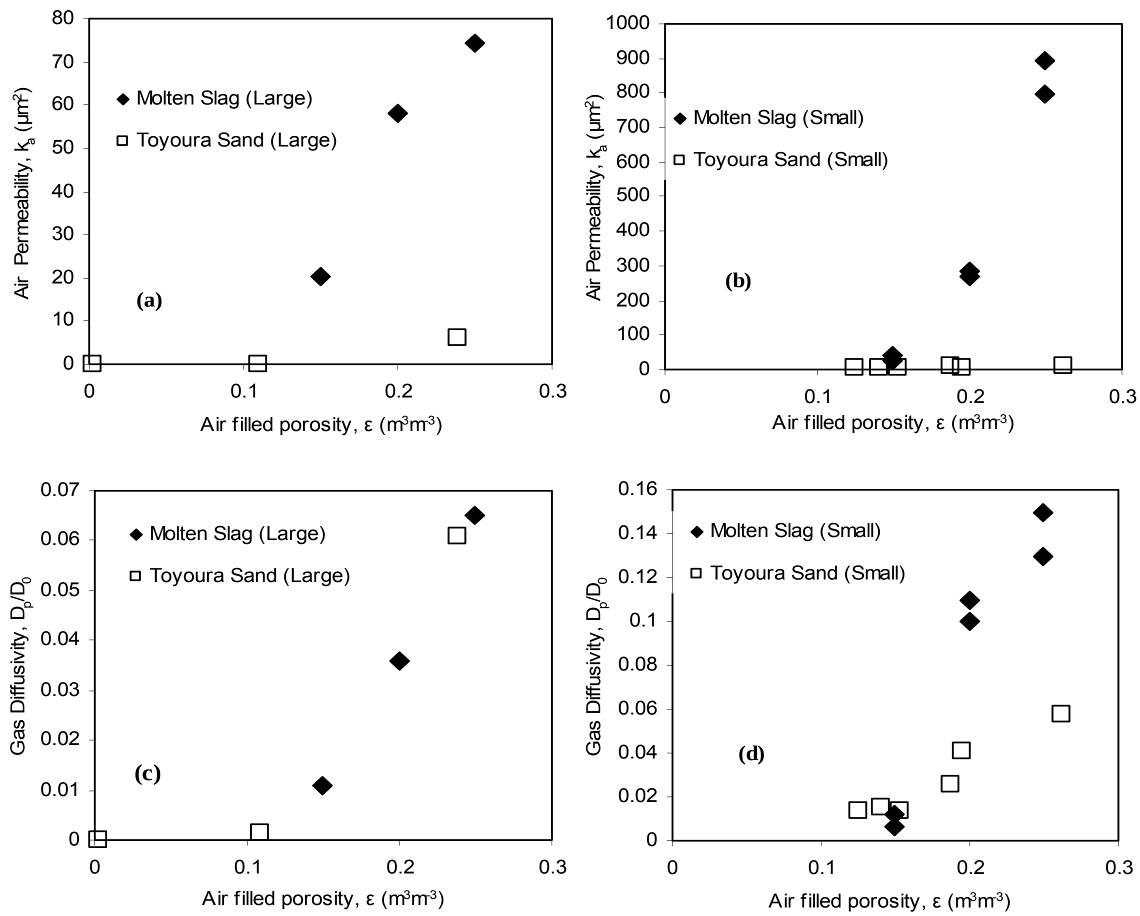

Figure 2. Air permeability (k_a) of (a) large core samples, (b) small core samples; gas diffusivity (D_p/D_0) of (c) large core samples, (d) small core samples for molten slag and Toyoura sand.

In Figure 2a air permeability (k_a) of large core samples were plotted for molten slag (closed diamond) and Toyoura sand (open rectangle) against air filled porosity. For small core samples, measured k_a values were shown in Figure 2b as a function of ε. The closed diamonds denote the k_a for molten slag while open rectangles denote k_a for Toyoura sand in Fig 2b similar to Fig 2a using small core samples. The plotted k_a results in Fig. 2a and 2b show that for both material k_a increases with ε of the sample. However, rate of k_a increase is not same; rather the rate is rapid in case of molten slag compared to

Toyoura sand. It indicates that not only air filled porosity but also pore structure of tested material played a significant role for measured k_a values as described by (Hamamoto et al., 2008). The mechanism of k_a transport will be further discussed in section 3.2 using pore characteristics indices.

For large core samples gas diffusivity of molten slag (closed diamonds) and Toyoura sand (open rectangles) show that D_p/D_0 (Fig. 2c) is only dependent on ε rather than material type and pore structure of porous media which is supported by (Buckingham, 1904; Hamamoto et al., 2008). On the other hand, plotted gas diffusivity against ε (Fig. 2d) for small core samples using molten slag (closed diamonds) and Toyoura sand (open rectangles) show marked variation in results especially at high ε. The additional discussion on gas diffusion mechanism will be made considering pore characteristics indices (equivalent pore diameter and tortuosity) in section 3.2

3.2 Pore Characteristics Indices

To understand the controlling factor for gas transport parameters pore characteristics indices: equivalent pore diameter (d_{eq}) and tortuosity (τ) were plotted in Fig 3. Figure 3a (large core samples) and Fig. 3b (small core samples) show the plotted d_{eq} (μm^2) against ε for molten slag (closed diamonds) and Toyoura sand (open rectangles). In the similar manner, tortuosity (τ) was plotted against ε using large (Fig. 3c) and small core samples (Fig. 3d) for both material.

Figure 3a and 3b show that for both large and small core samples equivalent pore diameter is greater for coarser molten slag compared to finer Toyoura sand but tortuosity (Fig. 3c) does not change so much. Therefore, large pore continuous network in coarser molten slag enhances air flow resulting in higher k_a compared to Toyoura sand as discussed by (Rouf et al., 2010) for mixture of molten slag and volcanic ash soil.

The large core samples tortuosity (Fig. 3c) does not vary so much (except at high water content or low air content condition due to high water blockage effect) and follow a general trend as observed in (Moldrup, 2001) for wet undisturbed soil. Therefore, large core samples gas diffusivity (Fig. 2c) is dominated by ε of the porous media only and there is no (or little) effect of pore tortuosity of the porous media on D_p/D_0. For small core samples tortuosity of molten slag vary a lot against ε, while the tortuosity of Toyoura sand vary a little against air filled porosity. Therefore, tortuosity of molten slag at high ε is less compared to Toyoura sand (Fig. 3d) indicating higher gas diffusion resulting in more gas diffusivity for molten slag compared to Toyoura sand (Fig 2d) which also agrees with (Rouf et al., 2010) measured gas diffusivity.

3.3 Scale Dependency on Gas Transport Parameters

Figure 4 shows the gas transport parameters for same material at the same dry bulk density using two different sample sizes (100 cm^3 and 2120 cm^3). Air permeability (k_a) and gas diffusivity (D_p/D_0) of Toyoura sand are plotted against air filled porosity in Fig. 4a and 4c respectively for both large and small core samples. Similarly, the values of k_a (Fig. 4b) and D_p/D_o (Fig. 4d) were plotted against air filled porosity for molten slag using large and small core samples at different moisture content conditions.

Figure 4a represents measured k_a against air filled porosity at different moisture content conditions using two standard size samples for Toyoura sand. The closed circles show the large core samples (2120 cm^3) k_a data measured in this study, while the open circles show the small core samples (100 cm^3) (Hamamoto et al., 2009) measured k_a values for Toyoura sand at same dry bulk density and total porosity. It shows that k_a increases with the increase of ε for

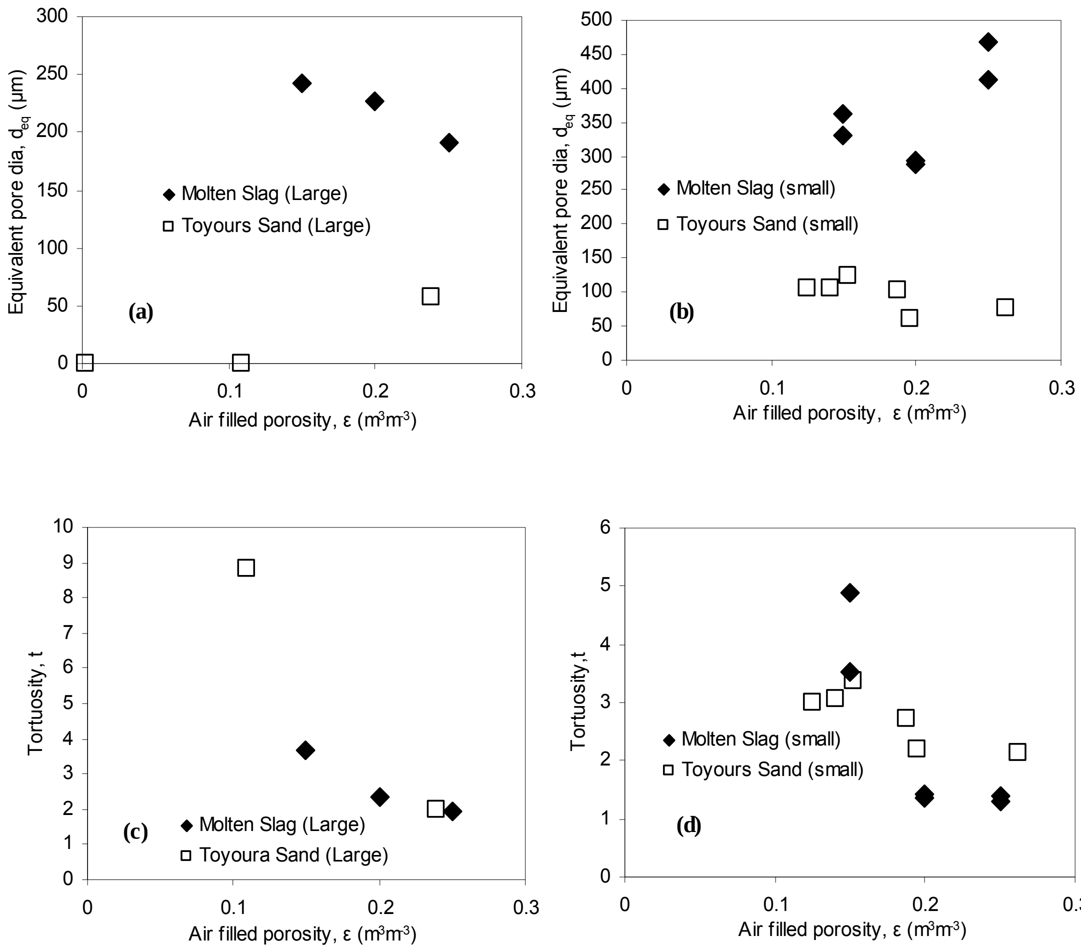

Figure 3. Equivalent pore diameter (d_{eq}) of molten slag and Toyoura sand for (a) large core samples, (b) small core samples; tortuosity (τ) of molten slag and Toyoura sand for (c) large core samples, (d) small core samples.

both small core and large core samples showing negligible variations of k_a at the same air or water filled porosity for these two traditionally used standard sample sizes. Therefore, Toyoura sand does not show scale effect on air permeability for different size samples rather it depends on air filled porosity and pore structure of porous media.

Again, for Toyoura sand, measured gas diffusivity (D_p/D_0) was plotted as a function of air filled porosity as shown in Figure 4c for small core (Hamamoto et al., 2009) and large core samples using almost similar dry bulk density (1.58 g cm^{-3}) and total porosity (0.40). The open diamonds show the small core samples gas diffusivity and closed diamonds show the large core samples gas diffusivity for Toyoura sand. Figure 4c shows that as the air filled porosity increases gas diffusivity increases for the large core and small core samples. Similar to k_a, D_p/D_0 presents the similar values (little or no variation) for two different size samples at same air filled porosity with similar dry bulk density showing negligible scale effect on gas diffusivity for fine Toyoura sand.

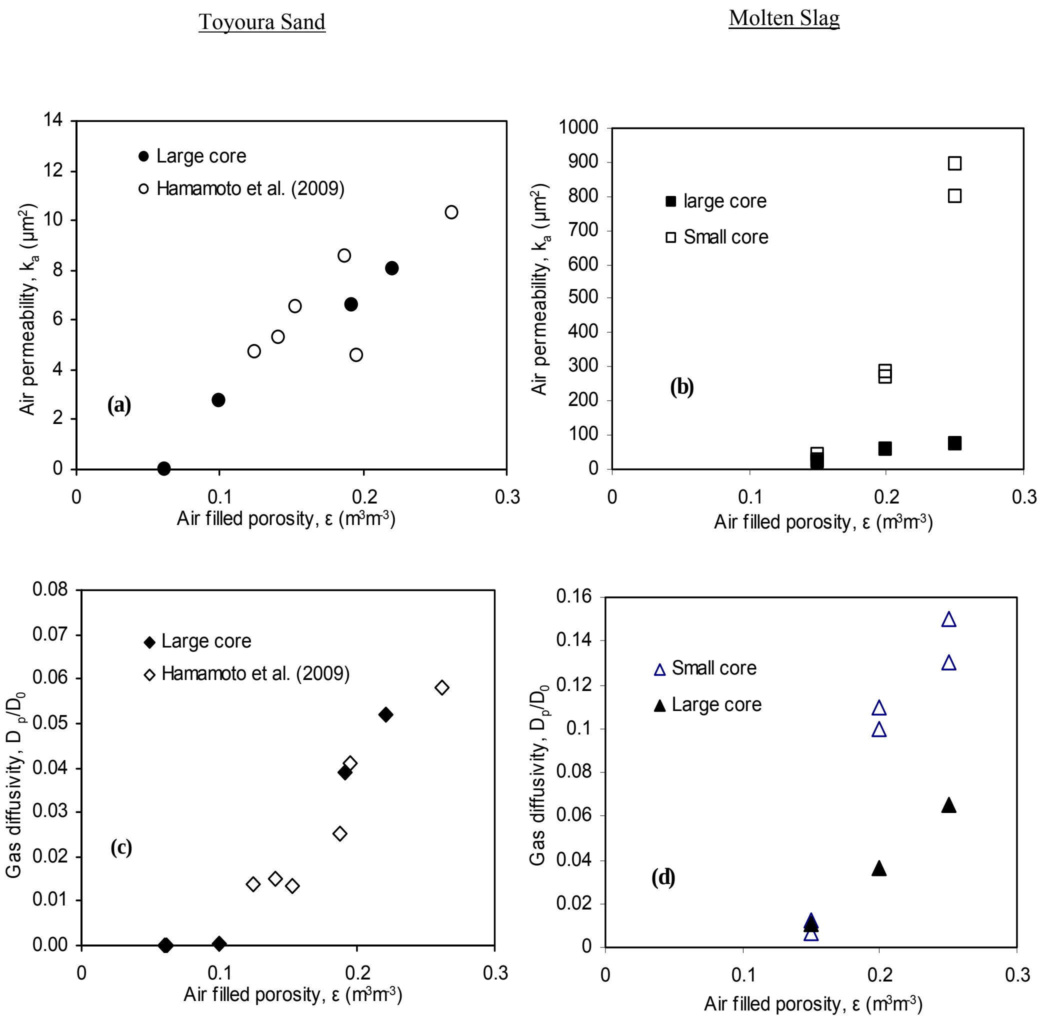

Figure 4. Air permeability of large and small core samples for (a) Toyoura sand, (b) molten slag; gas diffusivity of large and small core samples for (c) Toyoura sand, (d) molten slag.

For molten slag, measured k_a was plotted against air filled porosity using small and large core samples as shown in Figure 4b at dry bulk density of 1.70 g cm^{-3} and total porosity of 0.40 in different moisture content conditions. It shows that air permeability increases exponentially with ε for small core samples as shown in open rectangles in Fig. 4b. On the other hand, k_a increases gradually for large core samples shown in closed rectangles of Figure 4b. From Figure 4b it is also shown that at low air filled porosity both large and small core samples give similar results of k_a may be due to higher water blockage in pore spaces for both cases. However, at high air filled porosity ($\varepsilon \geq 0.20$) air permeability increases for small core samples compared to large core samples. The reason may be side wall effect. For example, preferential flow path might occurred during measurement of small core samples because of larger pore spaces inside the side wall along the periphery of the sample for coarser molten slag. It is evident from Figure

4b that there is a considerable scale effect on air permeability for coarse grained molten slag.

Figure 4d shows the gas diffusivity (D_p/D_0) against air filled porosity for molten slag using two standard sized samples at the same dry bulk density and total porosity as used for k_a measurements. The open triangle represents the small core samples D_p/D_0, while the closed triangles represent large core samples D_p/D_0. It also shows that at low air filled porosity both the samples show similar values of gas diffusivity due to the water blockage effect for both small and large core samples. On the other hand at high ε ($\varepsilon \geq 0.20$) the gas diffusivity of small core samples are grater than that of large core samples showing large variation in D_p/D_0 values for two different sample sizes at the same air filled porosity. This variation is may be due to the side wall effect as described for air permeability. Therefore, coarser molten slag shows considerable scale effect on gas diffusivity using two traditionally used sample size.

4. Conclusions and Recommendations

Gas transport parameters (k_a and D_p) of molten slag and Toyoura sand were investigated using different volume of samples at varied air or moisture content conditions. The plotting of measured air permeability was dominated by large pore continuous network of used porous media, while the plotted gas diffusivity was dominantly controlled by air filled porosity and pore tortuosity of the porous media. Moreover, the scale effect of k_a and D_p were also discussed in this paper. This research showed that for fine grained Toyoura sand there is no significant scale effect on gas transport parameters (k_a and D_p/D_0). However, coarser molten slag showed considerable scale effect especially at higher air filled porosity for both gas transport parameters (k_a and D_p). Therefore, further investigation is required to confirm the representative sample size for other coarser materials or structured soils, however, this study discussed the insight of gas transport mechanism and scale effect on gas transport parameters for porous media with both fine and coarse material.

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