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Permeability analysis in bisized porous media: Wall effect between particles of different size

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Summary The permeability of binary packings of glass beads with different size ratio — 13.3, 20, and 26.7, was investigated. In the Kozeny–Carman equation, the dependence of the tortuosity τ on the mixture porosity $\varepsilon(x_D)$ was described according to $\tau = 1/\varepsilon^n$ for different volume fraction of large particles in the mixture, x_D. Obtained data on packing permeability shows that the parameter n is a function of the volume fraction and particle size ratio, with values between 0.5 and 0.4. This can be explained by the wall effect resulting from the arrangement of the small particles occurring near the large particle surface. A model taking in account this effect was suggested that can be useful in the characterization of transport phenomena in granular beds as well as in engineering applications.

Introduction

Permeability is defined by the porous media structure including porosity, pore size, shape, and tortuosity, etc., being a key parameter on the development of models for fluid flow and mass transfer in porous media. Even for the simple case of binary packing, formed by particles that differ in particle size, the influence of porosity and related pore tortuosity on the bed permeability must be taken in account (Mota et al., 2001).

Binary packing porosity has been extensively investigated in numerous works (Yu and Standish, 1988) and it has been observed that most of the granular packings found in nature have a random structure (Cheng et al., 2000). An analytical–parametric theory of the random packing of particles developed by Yu and Standish (1988) shows the existence of two packing mechanisms: the filling mechanism and the occupation mechanism. The interplay of different packing mechanisms at different particle size ratio and binary mixture composition together with packing imperfections

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creates a gap in the packing porosity between random loose and dense packings (Nolan and Kavanagh, 1994) and, consequently, in the permeability (Standish and Mellor, 1980; Standish and Collins, 1983). For a granular packing, a flow velocity u is well described by the Hazen–Darcy equation and can be calculated by

$$u = k \cdot \Delta p/(\mu L) \quad (1)$$

where k is the permeability, Δp is the pressure drop, L is the bed thickness; μ is the liquid viscosity. Permeability can be estimated by the Kozeny–Carman equation (Bear, 1972):

$$k = d_{av}^2 \varepsilon^3/(36K_0\tau^2(1-\varepsilon)^2) \quad (2)$$

where d_{av} is the average particle diameter in the mixture of large D and small d size particles: $1/d_{av} = x_D/D + (1-x_D)/d$; x_D is the volume fraction of large particles in the binary mixture; the complex $K_0\tau^2 = K$ is the Kozeny's coefficient and for granular beds K = 4.2–5.0; K_0 is a shape factor that depends on the cross-section shape of the capillaries present in the granular beds, being K_0 = 2.0 for a packing of spheres (Mota et al., 2001; Dias et al. 2007).

The tortuosity coefficient plays an important role on the rates of heat and mass transfer obtained in porous media (Dias et al., 2007; Boving and Grathwohl, 2001) and in other areas (Fernandes et al., 2007) being defined as $\tau = L_e/L$, where L_e is the overall pathway length and L the bed thickness (Bear, 1972).

In the Kozeny–Carman Eq. (2) a proportionality coefficient $1/(36K_0\tau^2)$ is often assumed as a constant 1/150 or 1/180 (Jeschar, 1964; Dullien, 1975; MacDonald et al., 1991; Thies-Weesie and Philipse, 1994; Wu et al., 2003; Stewart et al., 2006). This assumption is acceptable in the packing porosity range ~0.36–0.4 where the tortuosity variation does not exceed 15–20% and can be, therefore, assumed to be a constant. Beyond this range a dependence of the tortuosity on porosity must be taken into consideration (Mota et al., 2001; Dias et al. 2006, 2007) and τ must considered as a variable.

The tortuosity coefficient depends on the overall porosity of the system ε, being both a function of the fraction of large particles present in the binary mixture $\tau = \tau(\varepsilon)$ and $\varepsilon = \varepsilon(x_D)$ (Mota et al., 2001). A power law function is the most frequently used to describe the relation between τ and ε (Ho and Strieder, 1981; Pape et al., 1987; Riley et al., 1996; Mauret and Renaud, 1997; Mota et al., 2001; Boving and Grathwohl, 2001):

$$\tau = 1/\varepsilon^n \quad (3)$$

where n is a coefficient dependent on the binary packing properties and ranges from 0.4 (loose packing) to 0.5 (dense packing) (Klusácek and Schneider, 1981; Millington and Quirk, 1961; Zhang and Bishop, 1994; Mota et al., 1998; Dias et al., 2006).

The aim of the present work is to study the influence of the volume fraction and particle size ratio on the coefficient n, due to the importance of the tortuosity coefficient in the Kozeny–Carman model.

Materials and methods

Binary mixtures had a particle size ratio inferior to 0.1 in all the experiments and the following glass beads were used: Beads with diameter 2, 3 and 4 mm from *Simax*. Beads from *Sigmund Lindner* with average diameter 0.15 mm. The density of the glass beads was 2500 kg/m^3.

Since the porosity of a binary mixture is significantly affected by the packing conditions, a method to obtain controlled binary packing beds, based on the application of a previous investigation, was applied. A water–glycerol solution was used as a binder between the different sized particles and the uniform distribution of the different size spheres within the packing was checked by image analysis (Dias et al., 2004b). In all experiments, the packing height varied between 10 and 15 cm. Mixtures with particle ratio $1/\delta = D/d$ = 13.3, 20 and 26.7 were investigated.

The permeability was calculated by Eq. (1), using the measured flow velocity at fixed pressure drop in laminar regime, as described by Dias et al. (2007). The Reynolds number was less than 0.1 in all the experiments. Using the experimental permeability, porosity and d_{av}, the tortuosity was calculated using Eq. (2).

With the tested particulate packing beds the minimum porosity, ε_{min}, was achieved at a volume fraction of large particles $x_{Dmin} \cong 0.7$. For mixtures with $x_D > x_{Dmin}$ the amount of small size particles was insufficient to fill all large particle skeleton voids and this gave rise to bi-layer particulate packing beds (Dias et al., 2004a, b). Consequently, the behaviour of parameter n was only studied in the range $x_D \leqslant x_{Dmin}$ where no segregation effect occurs. The coefficient n was estimated by

$$n = \frac{\ln\{36kK_0(1-\varepsilon)^2/(\varepsilon^3 d_{av}^2)\}}{2\ln(\varepsilon)} \quad (4)$$

this relation being obtained introducing Eq. (3) in Eq. (2).

Results and discussion

Porosity

The overall packing porosity of the present particulate binary mixtures was analysed in the work from Dias et al. (2004a), being described by the relation:

$$\varepsilon = \frac{\varepsilon_d^0(1-x_D)}{(1-\varepsilon_d^0 x_D)}\phi \quad (5)$$

this model being applicable in the range $x_D \leqslant x_{Dmin}$. The correction function ϕ is given by

$$\phi = \exp\left(1.2264 x_D^{1/\sqrt{\delta}}\right) \quad (6)$$

Eq. (5) describes well the porosity in the range of particle size ratios used on the present investigation (maximum deviation of 1.9%) and respects different boundary conditions. If x_D = 0, the mono-size packing porosity, ε_d^0, of the fraction with a smaller diameter is obtained (0.371 on the present investigation). When $\delta \to 0$, the well known (Yu and Standish, 1991) linear mixture porosity model (for $x_D \leqslant x_{Dmin}$) is obtained since $\phi \to 1$, this model representing the lowest porosity that can be achieved with particulate binary mixtures of spheres, for a given x_D. When $\delta \to 0$ and for $x_D = x_{Dmin}$ (region of minimum porosity) the amount of large particles is enough to build up a skeleton in the packing, the voids of the referred skeleton containing the small particles.

Tortuosity and permeability

In Fig. 1, experimental and calculated permeability, Eq. (2), are compared. The experimental data is located between the simulation results obtained with $n = 0.4$ and $n = 0.5$. For $x_D < 0.3$, experimental and simulated values are closer when $n = 0.5$, while for $x_D > 0.5$ the model fits better the data with $n = 0.4$. Therefore, the assumption of a fixed n is incorrect for particulate binary mixtures and confirms that n is the coefficient dependent from the binary packing content and particle size ratio, ranging from 0.4 (loose packing) to 0.5 (dense packing) (Klusácek and Schneider, 1981; Millington and Quirk, 1961; Zhang and Bishop, 1994; Mota et al., 1998; Dias et al., 2006).

The observed discrepancy between predicted and measured permeability at $x_D > 0.2$—0.3 shows that the real average flow pathway is shorter than the theoretically expected ($n = 0.5$). The reason for this includes two effects: (1) Non-homogeneity of the packing when particles differ insignificantly in size, thus creating conditions for pore channelling. (2) For particles of significant difference in size, the wall effect near the large particle surface causes a bypass of a fraction of the liquid through the less dense packing nearby the surface (distortion effect). For both situations, the values of tortuosity calculated using the experimental permeability give rise to smaller tortuosity coefficients than the predicted with a constant $n = 0.5$ in Eq. (3).

The decreasing of n from 0.5 to 0.4 when x_D approach x_{Dmin} may therefore be explained by the increase of the total surface area of large particles present in the mixtures and thus by the increase of the fraction of porous media involved in the wall effect. The fraction of surface area of large particles, F_D, in the mixture can be easily deduced:

$$F_D = \frac{x_D}{x_D + (1 - x_D)(1/\delta)} \tag{7}$$

The values of n for particulate binary mixtures with $1/\delta$ = 13.3, 20, and 26.7, calculated using the measured permeability, Eq. (4), are shown in Fig. 2. The values of parameter n are located in the range 0.4—0.5 and it was found that the experimental values of n are well described by:

$$n = 0.5 - \varphi F_D \tag{8}$$

Function φ was found using the experimental values of n ($r^2 = 0.945$), Fig. 2, and tacking in account that $n = 0.5$ for $\delta = 1$:

$$\varphi = 0.062(\delta^{-1} - 1) \tag{9}$$

In Table 1, the experimental values of φ and given by Eq. (9) are compared.

In the present δ region, Eqs. (8) and (9) predicts that the influence of the wall effect on n is more pronounced for lower values of δ this fact being more obvious in Fig. 2 if the data for $1/\delta = 13.3$ is compared with the remaining data, when x_D approach x_{Dmin}.

Eqs. (7)—(9) demonstrate the influence on the tortuosity of the fraction of surface area of large particles. By increasing x_D the number of small size particles involved in the wall effect increases. In addition, reduction of δ reduces accordingly the dead volume (zones free of small particles) close to the contact points of large particles. By decreasing the dead volume, the amount of small particles involved in the wall effect increases. Interplay of both components may be seen in Fig. 2.

If the ratio δ is lower than 0.03, function φ will be different from Eq. (9) since in this δ region the influence of the wall effect on n will decrease with the decrease of the particle size ratio (Dias et al., 2006).

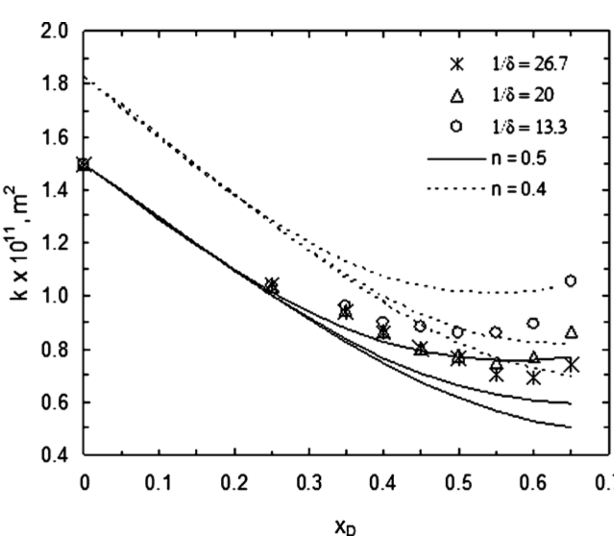

Figure 1 Experimental data and simulation results of $k(x_D)$ for particulate binary mixtures with $1/\delta$ = 13.3, 20, and 26.7. Simulated results were obtained by applying Eqs. (2), (3), and (5) with $n = 0.4$ and 0.5.

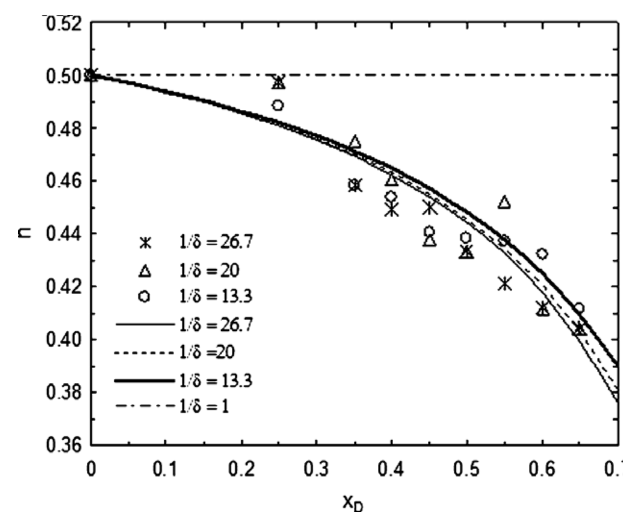

Figure 2 Dependence of n (calculated by Eq. (4)) on x_D for $1/\delta$ = 13.3, 20, and 26.7. Lines represent Eq. (8).

Table 1 Dependence of the function φ on the particle size ratio δ

δ	φ, Experimental	φ, Eq. (9)
1	0	0
0.075	0.765	0.775
0.05	1.178	1.163
0.0375	1.591	1.651

When δ reaches a value of 0.0035 (Dias et al., 2004a) the wall effect between the different particles becomes negligible and therefore the small particle packing present inside the skeleton ($x_D = x_{Dmin}$) formed by large particles approaches a regular packing, being expected that n returns to the value of 0.5 in the referred δ region.

Additionally, for all values of x_D located in the range $x_D \leqslant x_{Dmin}$ when $\delta \to 0$ the fraction of surface area of large particles — available for the wall effect — in the mixture falls to zero, as expressed by Eq. (7).

Permeability values obtained with Eq. (2), where the porosity is defined by Eq. (5) and the tortuosity is calculated using Eq. (3) with the parameter n modelled by Eq. (8) are shown in Fig. 3. The comparison of the permeability profile in Figs. 3 and 1 allows us to conclude on the importance of the $n = n(\delta, x_D)$ approach for modelling the permeability of particulate binary mixtures. In Fig. 3, it is also shown the limiting ($\delta \to 0$) permeability using n = 0.5.

Since minimum porosity is achieved for $x_D \cong 0.7$, the obtained results confirm the conclusion from Mota et al. (1998) and Mota et al. (2001) that permeability and porosity reach a minimum for different values of x_D, and that the curves $k(x_D)$ become more concave with decreasing δ.

When $\delta \to 0.1$ and for $x_D \cong 0.7$, the permeability of the binary mixture of glass spheres approaches the permeability of the packing containing the fraction with the smallest diameter alone (mono-size packing) (Dias et al., 2007). The effective thermal conductivity and transversal dispersion conductivity of the former packing is higher than the observed in the latter packing, due to the porosity decrease and tortuosity increase on the transition from the mono-size packing to the binary mixture (Dias et al., 2007).

The behaviour of the packing tortuosity with x_D is shown in Fig. 4. In spite of the scattering of the experimental data, the proposed model gives a good trend for the dependence of tortuosity τ vs. x_D. Due to the wall effect between particles of different size and consequent bypass of a fraction of liquid — explained above —, the tortuosity estimations using n = 0.5 are higher than the experimental values and, therefore, higher than the tortuosity values provided by the proposed model.

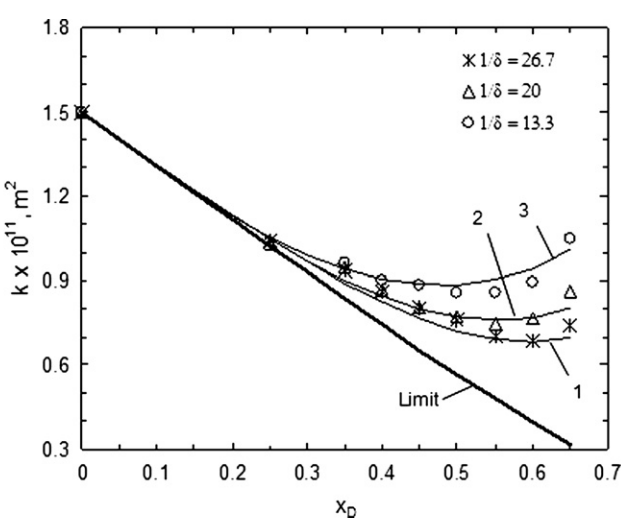

Figure 3 Experimental and simulated (Eq. (2)) values of k vs. x_D calculated using Eq. (3) together with Eqs. (5) and (8) (lines 1–3). Line (▬): Permeability when $\delta \to 0$ and n = 0.5.

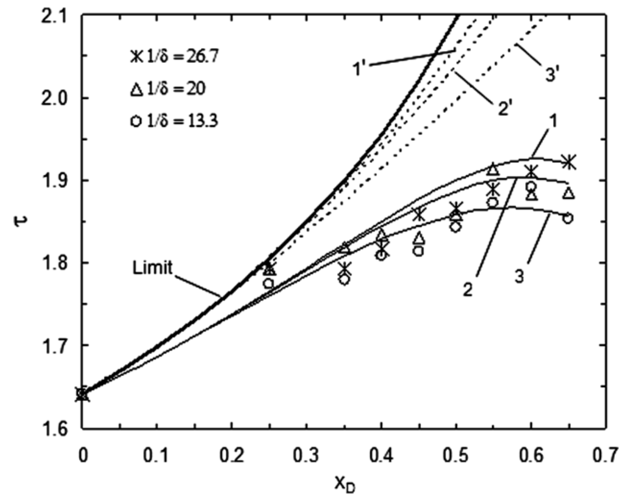

Figure 4 Experimental and simulated values of τ vs. x_D calculated using Eq. (3) together with Eqs. (5) and (8), used for porosity and n estimation, respectively (lines 1–3). Lines 1′–3′: corresponding to lines 1–3, respectively, but using n = 0.5 in Eq. (3). Line (▬): Tortuosity when $\delta \to 0$ and n = 0.5.

In the present work the porosity varied between 0.371 (mono-size packing) and 0.199 (binary packing with $1/\delta = 26.7$ and x_D = 0.65) being obtained tortuosity values between 1.641 and 1.925, respectively. Using spheres, sand, spheres mixtures, sand mixtures and spheres/sand mixtures, Currie (1960) found porosities between 0.424 (sand) and 0.171 (spheres/sand mixture) and tortuosities between 1.549 (n = 0.51 in Eq. (3)) and 2.175 (n = 0.44), respectively. Currie (1960) measured the effective diffusion coefficients of hydrogen in dry samples of the above referred materials, the main part of the n values being also located in the approximate range 0.4—0.5.

In Figs. 3 and 4, the limiting permeability and limiting tortuosity were calculated using n = 0.5 since for $\delta \to 0$ the wall effect can be neglected. A less pronounced wall effect and therefore a reduced liquid flow bypass through the zone close to the large particles surface may be obtained using non-spherical particles, for instance rod-like.

If the ratio δ is small enough, the introduction into the binary mixture of a limited amount of a third particle fraction with size small enough to fill the void of the binary mixture may diminish the porosity irregularity. In this case, the mixture becomes ternary (Mota et al., 1999). Using different concretes, similar results were observed in a cement paste—aggregate interfacial transition zone (Bentz and Garboczi, 1991; Garboczi and Bentz, 1991).

Conclusions

Binary mixtures permeability modelled by the Kozeny—Carman equation can differ substantially (50%) from the experimental values, if a fixed n is assumed in the relation $\tau = 1/\varepsilon^n$. The larger differences were observed in the region of minimum porosity of the binary packing beds.

Experimental results obtained for permeability and tortuosity show that the parameter n, is a function of the packing content, x_D, and particle size ratio, δ. The reason for *n* variation was explained by the wall effect of the small particles arrangement occurring near the large particle surface. A model accounting for this effect was proposed and may be useful for transport phenomena analysis in granular beds as well as in engineering applications.

Additional theoretical and experimental investigations must be undertaken, in order to fully understand the evolution of the wall effect with the decrease of the particle size ratio.

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