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BINARY SPHERICAL PARTICLE MIXED BEDS: POROSITY AND PERMEABILITY RELATIONSHIP MEASUREMENT

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Abstract - A continuous function relating the porosity of a mixture, ε, and the large particle volume fraction, x_D, of binary mixtures of spherical particles has been established. The incorporation of the proposed porosity model into the conventional Kozeny-Carman equation gives a good agreement between the measured and the predicted permeability vs. x_D. Based on this relationship and on the dependence of ε vs. x_D a model predicting tortuosity and permeability was obtained, having demonstrated that the tortuosity may significantly alter the permeability of a mixed bed. The proposed model shows that the simulated permeability curve presents a minimum for x_D values that are specified by the particle size ratios. The proposed model may be useful for the analysis of transport phenomena in granular beds as well as in engineering applications.

Keywords: Mixed beds; porosity; tortuosity; permeability; modelling; experiment.

INTRODUCTION

Mixed particle beds have a wide application in industry and play a significant role in nature. A better understanding of the relationship between porosity, permeability and particle composition may improve mass transfer processes in granular beds.

The flow velocity, u of a fluid through a granular bed under an applied pressure, p is described by the well-known Kozeny-Carman equation[1-3]:

$$u = \frac{\varepsilon^3 d_p^2}{36K(1-\varepsilon)^2} \cdot \frac{p}{\mu L} \tag{1}$$

where L is the bed thickness; μ is the liquid viscosity; and permeability k [m^2] is defined according to Equation (2). For a bed of spherical particles k depends on particle diameter, d_p, porosity, ε, and tortuosity, T:

$$k = \frac{\varepsilon^3 d_p^2}{36K(1-\varepsilon)^2} = \left(\frac{\varepsilon}{T}\right)^2 \left(\frac{\varepsilon d_p^2}{36(1-\varepsilon)^2 K_0}\right) \tag{2}$$

where, $K = K_0 T^2$ - the Kozeny's coefficient, for granular beds[4] is K = 4.2 - 5.0; K_0 is a shape coefficient (factor) depending on a cross-section capillary pore shape (for a cylindrical pore K_0 = 2.0). Variation of K depends, mainly, on the packing density and, hence, the tortuosity variation[5-8]. According to Bear[1], $T = L_e/L_0$ and L_e is the real pathway length and L_0 the bed thickness. In an ideal porous medium with tortuous non-intersecting capillaries the pathway tortuosity of flow streamlines would match to the geometrical tortuosity of a defined porous medium.

If the granular beds are considered as isotropic porous media, well-established data for granular bed tortuosity and spherical particle beds can be used for model verification. Geometrical tortuosity values[6,9] obtained from granular bed models ($T = \pi/2$ = 1.571 or $T = 2^{0.5}$ = 1.4142) are close to the measured T values in flow experiments[2-7].

When catalyst pellets with bidisperse pore structure were simulated by packed beds with a blend of microporous ion exchange resin and glass particles of different size[10] the tortuosity factor obtained was in the range 1.27 – 1.58. For random bed spherical particles and Knudsen diffusion regime Huizenga and Smith[11] measured T = 1.47, whereas, Wright *et al.*[12] reported a value of T = 1.48, respectively. In ordered sphere packings[13], the measured T was in the range 1.31 – 1.36. For mixtures of glass beads of particle size ratio up to 10.22 the geometrical tortuosity varies from around T = 1.45 (monosize packing) up to around 1.7 (binary mixture)[7]. If T = 1.45 is assumed for packed beds of mono-size spherical particles, then $K = K_0 T^2$ = 4.205.

Due to the random nature of mixed beds[8] as well as to the different preparation methods applied, they may exhibit large variations in porosity and in tortuosity[5,14-17]. A discussion on the fundamentals of bed porosity can be found, for instance, in Yu *et al.*[18].

The tortuosity depends on porosity[1,2,19] $T(\varepsilon)$ and often the correct presentation of T as the function of ε, is correlated to a model parameter[20,21]. For granular mixed beds and binary mixtures, in particular, both T and ε are related to the volume fraction of different size particles (for binary mixtures, the volume fraction of large particle x_D is often used). Therefore, to analyse mixed bed permeability in all ranges of x_D values (from 0 to 1.0), we need to know the relationships $\varepsilon = \varepsilon(x_D, \delta)$, $T = T(\varepsilon)$ and $d_p = f(d, D, x_D)$, where ε is the overall mixture porosity; δ is the particle size ratio, $\delta = d/D$; d and D are the size of the small and large particles in the mixture.

Mixed beds are widely used in numerous applications and there is a well-established theoretical background[14,18,22-31] describing the relationship of ε vs. x_D. As was mentioned above, the tortuosity of a binary mixture is significantly affected by the packing conditions. To avoid problems related with the packing procedure and with the selection among numerous theoretical models, new approaches are needed. As there is, theoretically, the possibility of making an infinite number of binary mixtures, the present approach was based on the assumption of ε vs. x_D being a continuous function in the range of x_D that might be incorporated in the permeability Equation (2).

The available information on tortuosity is scarce and the purpose of this research is to determine the tortuosity of mixed beds of spherical particles and to analyse the influence that porosity, ε, tortuosity, T, particle size ratio, δ and volume fraction, x_D may have on bed permeability.

EXPERIMENTAL

Porosity Measurement

The following types of glass beads were used for mixed particle beds: Beads of Glen Mills Inc., code 4512, diameter 3.3 - 3.6 mm and code 4504, diameter 1.0 - 1.25 mm; Beads of Sigmund Lindner, code 4508, diameter 2.0 - 2.3 mm; Beads of Sovitec Iberica, s.a., Microperl, diameter 0.5 - 0.84 mm; Beads of Potters-Ballotini, s.a., Visibead, diameter 0.25 - 0.425 mm.

Samples of beads were kindly provided by the above mentioned companies. For all samples more than 80% of the particles correspond to the average particle size. Particle samples listed above were selected to avoid any overlap in size.

In all cases, beads of 0.25 - 0.425 mm with an average diameter, d = 0.3375 mm were used as the smallest particles. A particle ratio of D/d = 10.22 was obtained by mixing these small particles with particles having an average diameter of 3.45 mm, D/d = 6.37, with 2.15 mm beads, D/d = 3.33, with 1.125 mm beads, and D/d = 1.985, with 0.67 mm beads, respectively.

In this study a mixing procedure similar to the one described by Suzuki[22] was used. The procedure gives reproducible mixtures and is carried out as follows. A 45 mm diameter glass cylinder for which the ratio (cylinder diameter)/(largest particles diameter) was >10 to minimise wall effects was used to form a bed of particles. The particles required for a defined mixture were weighed and poured into the cylinder in the following order. Firstly, large particles were placed into the cylinder. The small beads were introduced by uniform spraying of the primary bed top surface. The particles were then vigorously shaken. The cylinder was tipped horizontally, slowly rotated about its axis and gradually returned to the vertical position to produce the final packing. The bed thickness was 50 – 60 mm. By filling the mixture with distilled water the void space volume was measured. In every case, the porosity was determined as the average of three experiments. The porosity values stayed within a 5% range of values.

Based on experimental data, the relationship between mixture porosity, ε and x_D for different particle size ratios was found.

Permeability Measurement

For the permeability determination, three types of mixtures were chosen: D/d = 3.33, 6.37 and 10.22. The permeability was measured using the filtration procedure described below.

Filter paper placed on the perforated bottom of a cylinder with a diameter of 45 mm was used as the layer support. A glass fibre filter paper for qualitative analysis (type MN, Macherey-Nagel GmbH & Co.) was used. The mixed bed was formed on the layer support as previously described and porosity determinations were made by filling the bed with distilled water up to the top surface via an inlet at the bottom. After porosity determination, the cylinder was filled with water for the filtration experiments. A wire mesh was placed on the top of the bed, to protect the bed surface during the filling procedure. The bed thickness was in the range 50 – 60 mm. A vacuum was applied to create a pressure drop of Δp of 13 kPa across the mixed bed, that was kept constant throughout the filtration experiments. This pressure drop allowed the error due to the height of water column to be maintained below 5%. During filtration, the flow velocity, $u = V/(Ft)$ was measured for further calculation of the bed permeability (V is the filtrate volume, F is the filtration area and t is the filtration time). The hydraulic resistance of the layer support was checked before each filtration test. For each bed a new support was used.

The mixed bed permeability was calculated using the Kozeny-Carman model (1), bearing in mind that u depends on the hydraulic resistance of the bed, L/k and on the support medium resistance, R_m:

$$u = \frac{\Delta p}{\mu\left(L/k + R_m\right)} \tag{3}$$

Hence, the permeability was calculated as:

$$k = \frac{L}{\left(\frac{\Delta p}{\mu u} - R_m\right)} \tag{4}$$

where, R_m was determined in a filtration experiment without a bed (L = 0), Equation (3).

After that, the tortuosity was calculated using Equation (2), where the particle diameter, d_p was considered as the average particle diameter of a binary mixture, corresponding to the Sauter mean diameter (K_0 was assumed to be 2.0):

$$\frac{1}{d_p} = \frac{x_D}{D} + \frac{\left(1 - x_D\right)}{d} \tag{5}$$

RESULTS AND DISCUSSION

Porosity

The average measured porosity is shown in Figure 1 together with the modelling functions. The modelling functions were generated from the linear-mixed model[29-31], which is briefly described here.

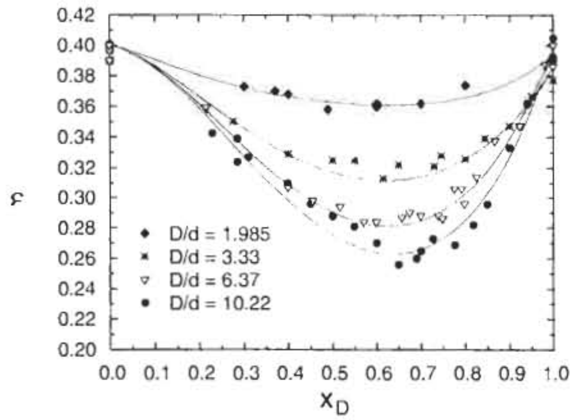

Figure 1: Experimental (symbols) and predicted (curves) dependencies of mixed bed porosity, ε on volume fraction of large particles, x_D. D/d is the ratio of large and small particles diameter. Full curves correspond to Equation (13).

In general, for a binary mixture, it may be stated that: the volume of total solid $(1-\varepsilon)$ = Volume of large particles $[1-\varepsilon_D(x_D)]$ + Volume of small particles $[1-\varepsilon_d(x_D)].\varepsilon_D(x_D)$ or:

$$\varepsilon = \varepsilon_D(x_D)\cdot\varepsilon_d(x_D) \qquad (6)$$

where, $\varepsilon_D(x_D)$ and $\varepsilon_d(x_D)$ are considered to be the individual porosities of the large and small particle fractions, respectively. In other words, ε_D is the specific void fraction of the large particles in total volume of the mixture, and ε_d is the specific void fraction of small particles in the remaining void volume of the mixture.

This equation implies that ε is a complex function depending on x_D, ε_d and ε_D. The boundary conditions for the above-mentioned function are:

$$\varepsilon_D(x_D)=\varepsilon_D^0,\ x_D=1 \text{ and } \varepsilon_D(x_D)=1,\ x_D=0 \qquad (7)$$

$$\varepsilon_d(x_D)=\varepsilon_d^0,\ x_D=0 \text{ and } \varepsilon_d(x_D)=1,\ x_D=1 \qquad (8)$$

where ε_D^0 and ε_d^0 are the porosities of a monosized beds of large and small particles respectively.

In a real mixture, the fraction of small particles will affect the large particle matrix structure by a displacement effect and by the ratio $\delta = d/D$. Therefore, some correction functions must be incorporated into the ideal mixture as proposed by Yu *et al.*[18]. Assuming for $\varepsilon_D(x_D)$ a correction function $F(\delta, x_D)$, then:

$$\varepsilon_D(x_D)=1-\left(1-\varepsilon_D^0\right)\cdot x_D\cdot F(\delta,x_D) \qquad (9)$$

On the other hand, the function $\varepsilon_d(x_D)$ can be written as:

$$\varepsilon_d(x_D)=\varepsilon_d^0+\left(1-\varepsilon_d^0\right)\cdot x_D\cdot f(\delta,x_D) \qquad (10)$$

where, $f(\delta, x_D)$ is the correction function in this case. Substituting Equations (9) and (10) in (6) gives the relationship $\varepsilon = \varepsilon(x_D, \delta)$.

The proposed functions (9) and (10) for calculating mixed bed porosity through Equation (6) include correction functions, which have to be determined experimentally. A large number of experimental measurements of the porosity were carried out for each set of diameter ratios. As was previously pointed out, for each volume fraction, x_D, three runs were performed and the average porosity was calculated (see Figure 1). On the assumption that the correction functions are complex functions of the particle size ratio $F(\delta)$, $f(\delta)$ and the large particle volume fraction, the following relationships were obtained:

$$F(\delta,x_D)=x_D^{[0.35-x_D F(\delta)]} \text{ and } f(\delta,x_D)=x_D^{f(\delta)-1}$$

Finally, according to Equations (9) and (10), it is obtained:

$$\begin{aligned}\varepsilon_D(x_D)&=1-\left(1-\varepsilon_D^0\right)x_D\cdot x_D^{[0.35-x_D F(\delta)]}\\&=1-\left(1-\varepsilon_D^0\right)x_D^{[1.35-x_D F(\delta)]}\end{aligned} \qquad (11)$$

$$\begin{aligned}\varepsilon_d(x_D)&=\varepsilon_d^0+\left(1-\varepsilon_d^0\right)x_D\cdot x_D^{f(\delta)-1}\\&=\varepsilon_d^0+\left(1-\varepsilon_d^0\right)x_D^{f(\delta)}\end{aligned} \qquad (12)$$

Hence, the porosity of a mixed bed may be defined by the following function:

$$\varepsilon=\left\{1-\left(1-\varepsilon_D^0\right)x_D^{[1.35-x_D F(\delta)]}\right\}\cdot\left[\varepsilon_D^0+\left(1-\varepsilon_D^0\right)x_D^{f(\delta)}\right] \qquad (13)$$

where $F(x_D)$ and $f(x_D)$ were found to be:

$$F(\delta)=\left\{0.27-\frac{1.55}{1+1/\exp\left(\dfrac{\delta+0.06}{0.27}\right)}\right\}^{-1} \qquad (13a)$$

$$f(\delta)=5-4\cdot\sqrt{\delta} \qquad (13b)$$

Equation (13), Figure 1, represents the continuous change in porosity of mixed beds and allows for the calculation of the overall mixture porosity knowing the particle size ratio, δ and x_D. The proposed model is easy to use for modelling properties such as permeability as is shown below.

Permeability

In mixed beds the tortuosity is often assumed to be constant[14,27]. However, when the porosity of the mixture approaches the region of minimal values, Figure 1, this assumption is not valid. For a granular bed, although losing some precision, the dependence of the tortuosity on porosity may be approximated by a simplified function[7]:

$$T=1/\varepsilon^{0.4} \qquad (14)$$

In this particular case, the value of the power proposed to be 0.5 by Zhang and Bishop[21] was less suitable. In this study a better correlation was found for a power of 0.4 (on average the deviation of calculated values from the experimental values was 30% lower than that obtained using a power of 0.5).

Based on the Kozeny's coefficient, K the tortuosity was calculated for several particle size ratios. The results obtained

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show that in the range of minimum porosity the measured tortuosity is larger. A value T_0 = 1.45, corresponding to the tortuosity of spherical monosized beds and a porosity, ε_0 = 0.4 were assumed for a uniform spherical particle bed.

In general, for mixed binary media ε, d_p and T depend on x_D. Equation (2) should thus, be written as:

$$k=\frac{\varepsilon^3\left(d_p\left(x_D\right)\right)^2}{36K_0\left(1-\varepsilon\right)^2\left(T\left(\varepsilon\right)\right)^2} \quad (15)$$

where, ε is defined by Equation (13). For beds of spherical particles, the equivalent average particle diameter mentioned in Equation (5) may be recalculated as:

$$d_p\left(x_D\right)=\frac{D}{n-\left(n-1\right)x_D} \quad (16)$$

where, $n = D/d$.

In the case of the mixture under discussion, taking account of Equation (6), the tortuosity becomes:

$$T=\frac{1}{\varepsilon^{0.4}}=\frac{1}{\left[\varepsilon_D\left(x_D\right)\varepsilon_d\left(x_D\right)\right]^{0.4}} \quad (17)$$

As mentioned above, the tortuosity values determined experimentally through Kozeny's coefficient, $K = K_0T^2$ are slightly different to those calculated by the porosity model, Equation (17), when the approach $T = 1/\varepsilon^{0.4}$ is used. This means that the nature of the tortuosity is somewhat more complicated than usually admitted. Differences between tortuosity values predicted by equation (17) and experimental values of T/T_0 across the range of x_D, do not exceed 10%. Hence, good results of the application of the proposed model can be expected.

The dependence of permeability on the volume fraction of large particles in the mixture, when the porosity is modelled by Equation (13) and the tortuosity by Equation (14), is shown together with experimental values in Figure 2. For the largest particle size ratio, the permeability is at a minimum when the volume fraction of large particles lies in the

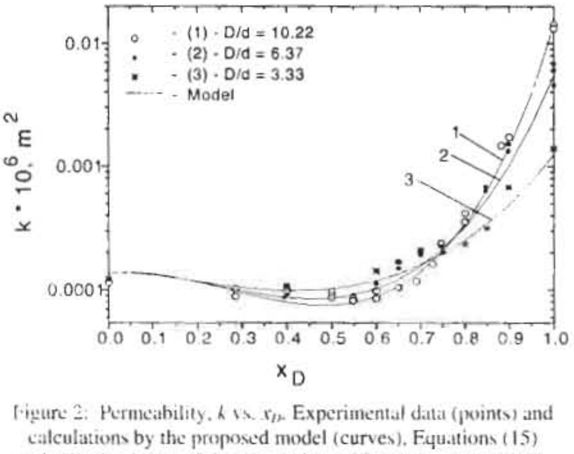

Figure 2: Permeability, k vs. x_D. Experimental data (points) and calculations by the proposed model (curves), Equations (15) and (17). 1 - D/d = 10.22, 2 - D/d = 6.37, and 3 - D/d = 3.33.

range 0.4 – 0.55. Prediction of the permeability is quite close to the experimentally determined values. It becomes clear that, by manipulating the volume fraction and the ratio D/d, the permeability may change by 2 orders of magnitude.

The influence of the ratio D/d on permeability for a fixed small particle diameter d = 0.3375 mm is shown in Figure 3. Increasing the ratio D/d leads to an increase in the curvature of the permeability curve and for D/d = 100, the minimal permeability is about twice as small as the permeability of the uniform small particle bed. This is an important outcome of the model because normally the effect of an increase in D/d on the permeability curve is not taken into account.

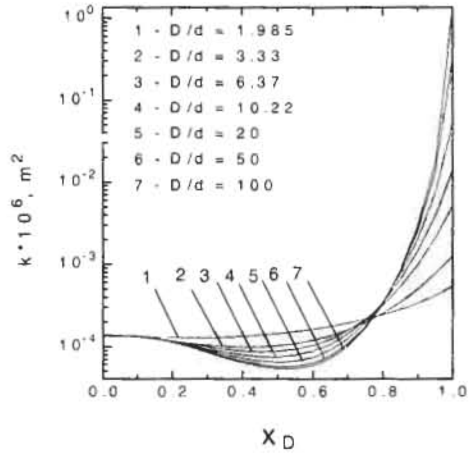

Figure 3: Dependence of k vs. x_D by the model, Equation (15), for different particle ratio, D/d. Small particle size assumed to be d = 0.3375 mm and $\varepsilon_D^0=\varepsilon_d^0=0.4$.

The analysis of the influence of D/d on k for a fixed value of large particle diameter shows that the permeability curve keeps the shape when D/d increases. Thus, for a mixed binary bed, in a range of x_D specified by the D/d value the permeability can be smaller than would be expected.

The results obtained clearly prove that the tortuosity variation must be taken into consideration when modelling transport phenomena in granular beds.

CONCLUSIONS

Based on experimental data and on a linear mixing model, the relationship of mixture porosity, ε vs. x_D for a binary mixture of spherical particles was established as a continuous function. The incorporation of the proposed porosity model into the conventional permeability model adequately described the relation between permeability k vs. x_D, as was confirmed by the good agreement between measured and predicted data.

The relationship of tortuosity, T with mixture porosity, ε was found to be $T = 1/\varepsilon^{0.4}$. Based on this relation and on the dependence of ε vs. x_D the model for predicting tortuosity and permeability was obtained.

Tortuosity is not significantly affected by the increase in D/d. Indeed, by applying Equation (14) for an extreme D/d = 10.22 the maximum tortuosity is 1.71, whereas, for a monosized bed, the tortuosity is 1.45. Nevertheless, it was shown that tortuosity might significantly change the permeability of a mixed bed. The proposed model gives the possibility to simulate the permeability curve profile that presents a minimum in specific ranges of x_D.

The proposed model may be useful for transport phenomena analysis in granular beds as well as in engineering applications, such as deep bed filtration used for drinking water purification.

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NOMENCLATURE

D	Average diameter of large particles (m)
d	Average diameter of small particles (m)
d_p	Average particle diameter of a binary mixture (m)
F	Filtration area (m^2)
k	Permeability (m^2)
K	Kozeny coefficient (K_0T^2)
K_0	Constant, assumed to be 2.0
L	Thickness of the bed (m)
n	Ratio D/d
p	Applied pressure (Pa)
R_m	Filter paper resistance (m^{-1})
t	Filtration time (s)
T	Tortuosity
T_0	Tortuosity of uniform spherical particle bed, T_0 = 1.45
u	Flow velocity (m s^{-1})
V	Filtrate volume (m^3)
V_t	Total mixture volume (m^3)
x_D	Volume fraction of large particles in the total volume of particles in the mixture
ε	Porosity of a mixed bed
ε_D^0	Porosity of a monosized bed of large particles, $\varepsilon_D^0 = 0.4$
ε_d^0	Porosity of a monosized bed of small particles, $\varepsilon_d^0 = 0.4$
δ	Particle size ratio, $\delta = 1/n = d/D$
μ	Viscosity (Pa s)
Δp	Pressure drop through packed bed (Pa)

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