

STUDY OF TERNARY GLASS SPHERICAL PARTICLE BEDS: POROSITY, TORTUOSITY AND PERMEABILITY

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Ternary mixtures of glass beads were constructed as a model of granular packing. Porosity and permeability were experimentally determined in a wide range of mixture composition. Based on experimental data, tortuosity was calculated using the Kozeny-Carman equation. Applying the conventional relation that expresses tortuosity as the inverse of the power order of porosity, it was found that the parameter n varies with the fraction content. The observed phenomenon was explained by wall effects between particles of different sizes.

Keywords: ternary mixed bed; porosity; tortuosity; permeability.

INTRODUCTION

Mixed particle beds have wide application in industry and, in particular, play a significant role in solid-liquid separation. Therefore, the understanding and description of mixed bed properties are important and have still to be the subject of investigation under different conditions¹⁻⁹.

The relation between permeability, porosity and tortuosity of ternary mixtures is found in practice for many separation processes (sedimentation and centrifugation^{10,11}, filtration^{1,9}, cake washing etc.) when particle fractions different in size form a dispersed system. For instance, in a dispersed system of a filter aid, cells and cell debris will define a ternary mixture of particles significantly different in size. The permeability k is inversely proportional to the specific cake resistance, which is the main characteristic of a filter cake.

To simplify the experimental study of ternary systems, ternary mixtures of glass beads can be used to simulate the properties of a particular system. Moreover, ternary glass mixtures are quite suitable to build porous media with pre-defined properties, since:

1. the amount of each particle fraction gives the possibility to easily predict and control the mixture porosity;
2. as the mixtures can vary in terms of particle size ratio and proportion of each fraction, three glass bead sizes are usually sufficient to get a wide range of pore size, porosity, permeability, etc.

Transition from uniform particle packing to the multi-component mixed bed of different particle size makes the porous medium properties more complicated but more "flexible" as a subject of control. A granular system may exhibit a number of different microscopic states at fixed macroscopic densities¹², hence, at the same porosity, a granular bed may have, for example, different permeability or diffusivity.

Ternary packing beds have previously been investigated in numerous works^{2-4,8,13,14,14-20} either experimentally or by

means of modelling.

The fluid flow velocity u through a granular bed under an applied pressure p is described by the well-known Kozeny-Carman equation²¹⁻²⁴

$$u = k \left(\frac{p}{\mu L} \right) \quad (1)$$

where L is the bed thickness and μ the liquid viscosity. Permeability k [m²] depends, for a mixed bed of spherical particles, on the average particle diameter d_{av} , on the overall packing porosity ε and on tortuosity T :

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

where K_0 is a shape coefficient (factor) dependent on a cross-section capillary pore shape (usually assumed to be $K_0 = 2.0$)²¹, $T = L_e/L_0$ the tortuosity, L_e the effective pathway length and L_0 the bed thickness. The ratio k/d_{av}^2 is only a function of the bed porosity and tortuosity if K_0 is assumed constant.

According to Equation (2), the ratio ε/T plays an important role in the permeability value for constant or variable T (Figure 1). Moreover, Mota *et al.*³ analysed the effect of the ratio ε/T in terms of the fraction of coarse particles (largest discs) in the packing. It was found that ε/T is sensitive to the largest/smallest particle size ratio and to the packing fractional content. To validate the observed effect, additional experimental investigation of ternary packing is necessary.

In some models the expression $36K_0T^2$ in Equation (2) is replaced by a numerical coefficient²⁵⁻²⁸, the most usual being $36K_0T^2 = 180$. In this case from Equation (2) it follows that

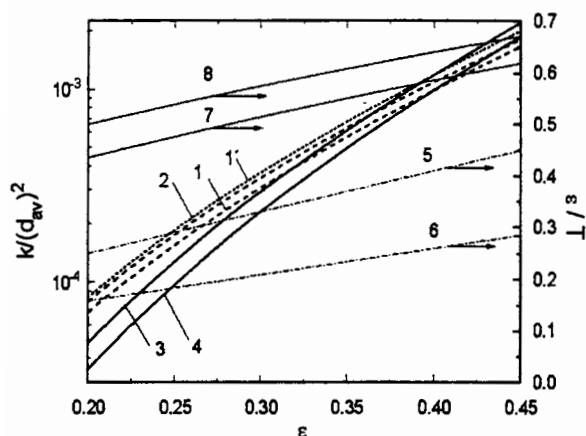


Figure 1: Dependence of k/d_{av}^2 (curves 1 – 4) and ϵ/T (curves 5 – 8) on the porosity ϵ for cases of constant and variable with the porosity T . Dimensionless permeability k/d_{av}^2 1 and 1' – Equation (3) at $36K_0T^2 = 180$ and 150; 2 – Equation (4); 3 – Equation (3) at $T = 1/\epsilon^{0.4}$; 4 – Equation (3) at $T = 1/\epsilon^{0.5}$. Complex ϵ/T : 5 – Tortuosity $T = 1.0$; 6 – $T = 1.58$; 7 – $T = 1/\epsilon^{0.4}$; 8 – $T = 1/\epsilon^{0.5}$.

$$\frac{k}{d_{av}^2} = \frac{1}{36K_0T^2} \left(\frac{\epsilon^3}{(1-\epsilon)^2} \right) = \frac{1}{180} \left(\frac{\epsilon^3}{(1-\epsilon)^2} \right) \quad (3)$$

A coefficient of 180 infers the assumption of a constant tortuosity. A similar relation was used by MacDonald *et al.*¹⁶ to investigate the porosity and permeability of a spherical ternary packing for a ratio of coarse d_C to fine d_F particles of 2.37 and 5.08. Assuming $K_0 = 2.0$, the tortuosity in this case is $T = 1.58$ (Figure 1, curves 1 and 6).

In fact, the tortuosity, even in a narrow range of porosity variation, is not constant and depends on the particles size distribution, packing type, etc. Values of T ranging from 1.27 up to 1.58 can be found in several works²⁹⁻³⁵. As an example, curves 5 and 6 in Figure 1 are given for ϵ/T at $T = 1$ and 1.58 respectively.

Hamilton³⁶ made an attempt to avoid the constant 1/180 in the complex k/d_{av}^2 by using an approach based on spheres assembling²⁴ (Figure 1, curve 2):

$$\frac{k}{d_{av}^2} = \frac{1}{12} \left(\frac{3(1-\epsilon)^{5/3} - 3(1-\epsilon)^{1/3} - 2(1-\epsilon)^2 + 2}{(1-\epsilon)[2(1-\epsilon)^{5/3} + 3]} \right) \quad (4)$$

However, in the porosity range 0.2 – 0.45 the relation in Equation (4) is quite close to the function with constant tortuosity and k/d_{av}^2 accommodates coefficients $36K_0T^2$ between 180 and 150, respectively (curves 1 and 1').

More productive seems an approach where tortuosity is considered as a function of the porous media porosity^{7,22,23,31,37-48}. For granular beds it is often presented in the form

$$T = \frac{1}{\epsilon^n} \quad (5)$$

where n is usually assumed to be 0.4 or 0.5. It was shown that the exponent index n for granular beds ranges from 0.4 (loose packing) to 0.5 (dense packing)⁴⁹ and for granular mixed beds this exponent index is related to the fractional content of a packing bed.

In Figure 1 we show dependences of both k/d_{av}^2 and ϵ/T when the tortuosity is considered to be constant or is a function of the porosity, curves 3, 4 and 7, 8. Calculations clearly demonstrate that ignoring the variation of T vs. ϵ results in differences in the modelled permeability.

The fundamentals of bed porosity can be found in numerous publications^{2,8,13,14} where ternary mixtures are usually represented by a triangular diagram. It has been observed^{2,4,8,13-19} that a region of lowest porosity is located near to a binary mixture axis that represents the coarse d_C and fine d_F particle sizes. As an example, the sketch of the triangular diagram from⁸ is shown in Figure 2 for a ternary mixture of spheres. The packing density iso-curves are presented in the form 1- ϵ ; F , M and C represent fine, medium, and coarse particle fractions of size $d_F = 7$ mm, $d_M = 14$ mm and $d_C = 28$ mm respectively.

The ternary mixture porosity is well investigated, whereas available information on the tortuosity of the ternary packing is scarce. The purpose of the present research is to determine tortuosity of mixed beds of spherical particles and to analyse the influence of the tortuosity T , particle size ratio, and fractional content on ternary bed permeability.

EXPERIMENTAL BACKGROUND

As tortuosity can be defined from the permeability, some experiments were performed on the porosity and permeability of the ternary mixture modelled by glass beads of different sizes.

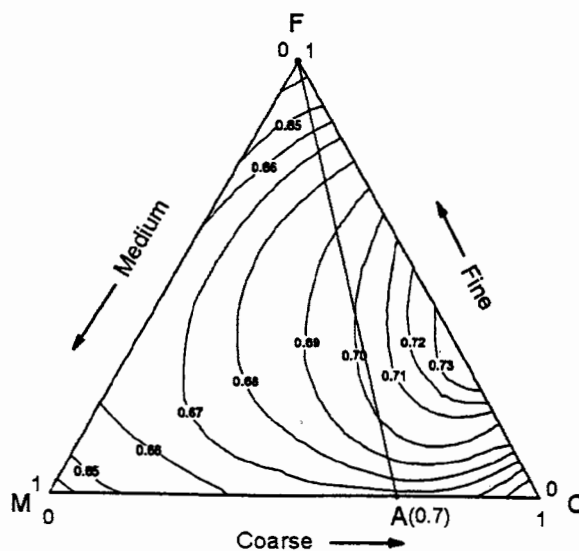


Figure 2: Sketch of the triangular diagram⁸ for a ternary mixture of coarse C , medium M , and fine F particle sizes. The packing density iso-curves is presented in the form 1- ϵ and represent fine, medium, and coarse particle fractions of size $d_F = 7$ mm, $d_M = 14$ mm, and $d_C = 28$ mm respectively.

As can be seen from Figure 2, the most interesting region for further analysis is the region of low porosity near the axis of the binary mixture $F - C$. Ternary mixtures directly bordering binary mixtures have properties close to the binary packing, whereas the region of mixtures (M) enriched with medium size particles have small variation in packing density and, for the example in Figure 2, have $1 - \varepsilon = 0.65 - 0.69$. Based on data presented in previous works^{2,4,8,13-19}, for further investigation we chose compositions defined by line $F - A$ in Figure 2. At point A the composition is 30% medium and 70% of coarse particles. Compositions along the line $F - A$ elicit the analysis of the entire span of porosities available in a ternary mixture.

When the particle size ratios between fractions $\delta_{MC} = d_M/d_C$ and $\delta_{FM} = d_F/d_M$ are small and approach zero ($\delta_{MC} \rightarrow 0$ and $\delta_{FM} \rightarrow 0$), we can, by analogy, apply the binary model in the form of a linear approach, for example^{7,9}.

The volume fraction of fine particles in the ternary mixture may be represented as $v_F = \varepsilon_{C+M} (1 - \varepsilon_F^0)$ where $\varepsilon_{C+M}^0 = \varepsilon_C^0 \varepsilon_M^0$ and hence the volume fraction of coarse and medium particles, by analogy with a binary mixture, may be written as

$$X_{C+M} = \frac{v_{C+M}}{v_{C+M} + v_F} = \frac{1 - \varepsilon_{C+M}}{1 - \varepsilon_{C+M} \varepsilon_F^0} \quad (6)$$

The overall packing porosity then becomes

$$\varepsilon = \varepsilon_{C+M} \varepsilon_F^0 = \varepsilon_F^0 (1 - X_{C+M}) / (1 - X_{C+M} \varepsilon_F^0), \quad (7)$$

$$X_{C+M} \in [0, X_{(C+M)\min}]$$

$$\varepsilon = 1 - (1 - \varepsilon_{C+M}^0) / X_{C+M}, \quad X_{C+M} \in [X_{(C+M)\min}, 1] \quad (8)$$

with the minimum porosity $\varepsilon_{\min} = \varepsilon_F^0 \varepsilon_{C+M}^0 = \varepsilon_C^0 \varepsilon_F^0 \varepsilon_M^0$ corresponding to the fractional content

$$X_{(C+M)\min} = \frac{1 - \varepsilon_{C+M}^0}{1 - \varepsilon_C^0 \varepsilon_F^0 \varepsilon_M^0} \quad (9)$$

Here ε_C^0 , ε_F^0 and ε_M^0 are the porosity of coarse, fine, and medium particle size fractions, respectively.

Equations (7) – (9) are boundary conditions when $\delta_{MC} \rightarrow 0$ and $\delta_{FM} \rightarrow 0$ or $\delta_{FC} = d_F/d_C \rightarrow 0$. In the range $x_{C+M} \in [X_{(C+M)\min}, 1]$ for significant particle size ratio between fractions a segregation effect is observed. Therefore, dependence (7) has a great practical interest and will be the subject of a further investigation.

For the boundary conditions $\delta_{MC} \rightarrow 0$, $\delta_{FM} \rightarrow 0$ the tortuosity T can be presented as⁵⁰

$$T = T_C^0 T_F^0 T_M^0 \quad (10)$$

where T_F^0 , T_M^0 and T_C^0 are the mono-size tortuosities of fine, medium, and coarse particle packings, respectively.

In a real mixture, each fraction will affect the others by

displacement and wall effects which are dependent on particles size ratios and composition content⁴⁹. This fact can be taken into consideration by incorporating some correction functions in the model. For simplicity we use the correction function φ as follows⁴⁹:

$$\varepsilon = \varepsilon_{C+M} \varepsilon_F^0 = \varphi \varepsilon_F^0 \frac{1 - X_{C+M}}{1 - X_{C+M} \varepsilon_F^0} \quad (11)$$

Hence, if the dependences of ε and the permeability k on X_{C+M} are determined in the experiments, tortuosity values can be obtained and analysed through Equation (2).

EXPERIMENTS

The following types of glass beads were used for mixed particle beds: Beads with diameter 2 ± 0.2 mm, 3 ± 0.2 mm and 4 ± 0.3 mm from *Simax*. Glass beads from *Sigmund Lindner*, code 4501, diameter $0.1 - 0.2$ mm (average diameter 0.15 mm) and code 4503, diameter 0.75 – 1.0 mm (average diameter 0.875 mm). Particle density was 2500 kg/m³ in every case. The following ternary packings were investigated: $d_C = 4$ mm, 3 mm and 2 mm, $d_M = 0.875$ mm, and $d_F = 0.15$ mm.

To minimise the possible segregation effect of a ternary particle mixture during the packing procedure a method described in a previous work⁵¹ was used. The method of mixing/preparing binary and ternary mixtures of glass beads was developed using a viscous solution of glycerol in water. After mixing, the mixture was transferred to a prismatic vessel (column) and glycerol was washed out. The square column used in experiments had an inner cross-section of 55 cm. The height of the ternary bed was from 10 to 15 cm. The different sized beads were differently coloured and digital pictures taken from each face were automatically treated by image analysis to determine the coloured fraction present in each face. Statistical analysis showed that no significant deviation existed in the colour distribution of each of the four faces. A chi-square test showed that a uniform distribution could be accepted for the beads. The two-dimensional picture obtained by image analysis was converted to the corresponding three-dimensional distribution, from which the bed porosity was inferred. The estimated porosity was compared with the experimental bed porosity determined by a gravimetric method. No significant deviations were found, thereby proving that the mixing method developed was reliable.

An example of ternary packing with the corresponding treated image for analysis is shown in Figure 3.

Ternary mixtures with compositions defined by line $F - A$ in Figure 2 were investigated. At the point A there is a binary composition of medium X_M and coarse X_C particles, therefore, the binary mixture content X_{C+M} is equal to $X_C + X_M = X_{C+M}$. By fixing binary particle composition content at the point A as 30% of medium ($X_M = 0.3X_{C+M}$) and 70% of coarse particles ($X_C = 0.7X_{C+M}$) a ternary mixture composition along the line $F - A$ may be calculated as $X_F = 1 - X_{C+M}$, where $X_C + X_M + X_F = 1.0$.

An average mixture particle diameter d_{av} of ternary mixture is equal to⁵²

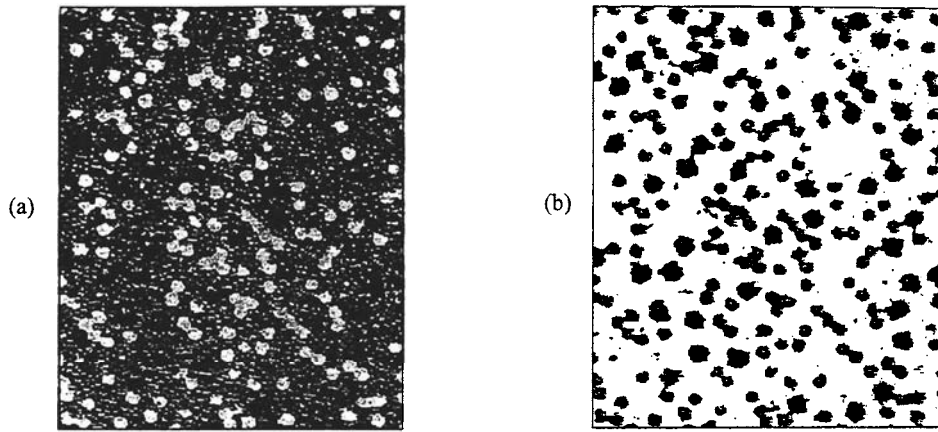


Figure 3: Example of a ternary packing of spheres: $d_C = 4$ mm (black) 25%; $d_M = 2$ mm (grey) 25%; $d_F = 0.875$ mm (white) 50 %. (a) – original image; (b) – treated image.

$$d_{av} = \frac{1}{X_F/d_F + X_M/d_M + X_C/d_C} \quad (12)$$

Taking into account that $X_M = 0.3X_{C+M}$, $X_C = 0.7X_{C+M}$ and $X_F = 1 - X_{C+M}$ we have

$$d_{av} = \left[\frac{1 - X_{C+M}}{d_F} + \left(\frac{0.3}{d_M} + \frac{0.7}{d_C} \right) X_{C+M} \right]^{-1} \quad (13)$$

The bed permeability was determined through Equation (1), by measuring the flow velocity at a fixed pressure drop in laminar flow regime. Then, using the permeability and porosity the tortuosity was determined from Equation (2),

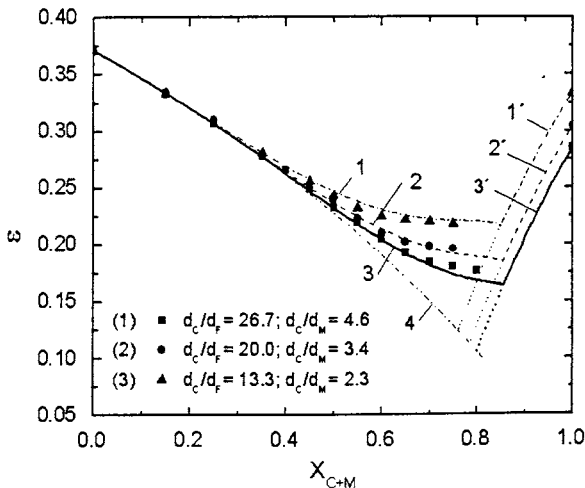


Figure 4: Dependence of ternary packing porosity ε on volume fraction X_{C+M} . Curves 1 – 3 calculated by Equation (14). Curve 4 characterise the linear model limit $\delta_{M/C} \rightarrow 0$, $\delta_{F/M} \rightarrow 0$. Curves 1' – 3' represent the linear approach, Equation (8), when $\varepsilon_{C+M}^0 = \varepsilon_C^0 \varepsilon_M^0$ are calculated based on experimentally measured porosities of the binary mixture.

assuming $K_0 = 2.0$.

RESULTS AND DISCUSSION

The results obtained and correlation dependences are given below.

Porosity

Mono-size packing porosity measurements give the following values: for fine particle packing $\varepsilon_F^0 = 0.371$, and for coarse and medium size mono packings $\varepsilon_M^0 = \varepsilon_C^0 = 0.4 \pm 0.002$. The dependence of ternary packing porosity on volume fraction X_{C+M} is shown in Figure 4 together with the obtained correlation functions.

In Figure 4, curves 1' – 3' represent the linear approach, Equation (8), where $\varepsilon_{M+C}^0 = \varepsilon_C^0 \varepsilon_M^0$ is calculated by the experimentally measured porosities of the binary mixture ε_{M+C}^0 . Values of ε_{M+C}^0 measured for $d_C/d_M = 4.6, 3.4$ and 2.3 , were respectively $0.285, 0.304$ and 0.334 . Curve 4 characterises the linear model limit $\delta_{M/C} \rightarrow 0$, $\delta_{F/M} \rightarrow 0$ for the chosen mixtures and has $\varepsilon_F^0 = 0.371$.

Comparing experimental data in Figure 4 with curve 4, it is seen that displacement and wall effects between particles of different sizes⁴⁹ gave rise to significant deviations of the experimental porosity from the linear model in the region of minimum porosity. Using a fitting procedure the correction function φ in Equation (11) was defined as

$$\varphi = \exp\left(2.0989 X_{C+M}^{2/\sqrt{\delta_{M/C} \delta_{F/C}}}\right)$$

and the ternary packing porosity was modelled by the relation

$$\varepsilon = \exp\left(2.0989 X_{M+C}^{2/\sqrt{\delta_{M/C} \delta_{F/C}}}\right) \varepsilon_F^0 \frac{1 - X_{C+M}}{1 - X_{C+M} \varepsilon_F^0} \quad (14)$$

for the region $X_{M+C} \in [0, X_{(C+M)\min}]$ (curves 1 – 3, Figure 4).

Tortuosity

To analyse the influence of the porosity deviation from the

linear model on the tortuosity, Equation (5) was substituted in the permeability formula (Equation (2)). To account for the distortion effects present in a real ternary packing, parameter n can be considered as a complex variable $n = n(K_0, d_{av}, k, \epsilon, X_{C+M})$. If K_0 is assumed to be constant for granular beds and if d_{av} is defined by Equation (13) then, using measured k and ϵ at a defined X_{C+M} , parameter n may be calculated as shown in Figure 5, where curves 1 – 3 are fitting polynomial functions for the calculated n .

As can be seen in Figure 5, parameter n is around 0.5 for the mono-size packing but is variable in the ternary mixture. In the region of $X_{(M+C)min}$ the parameter ranges from 0.4 to 0.45. Anomalous low values of n were observed for a volume fraction of fine particles $1 - X_{C+M} \approx 0.4$ that coincides with the region of near maximum dense packing as shown in Figure 2. In this region the amount of fine particles is small enough to wedge into the coarse-medium particle skeleton and fill its free space. Therefore, the distortion effect of coarse and medium particles on the fine particle arrangement reaches its maximum. The main conclusion from obtained data is that the power order n in Equation (5) depends on the mixture fractional content and, hence, the tortuosity becomes a complex function of the porosity.

Together with the calculated values of n in Figure 5, fitting functions in the form of Equation (15) are shown as curves 1 – 3.

$$n = 0.5 + aX_{C+M} - bX_{C+M}^2 + cX_{C+M}^3 \quad (15)$$

Dependence of the tortuosity in ternary mixtures is shown in Figure 6 for two cases:

1. value n in relation $T = 1/\epsilon^n$ is the function defined by Equation (15), curves 1 – 3
2. value n is the constant assumed to be 0.5, curves 1' – 3'.

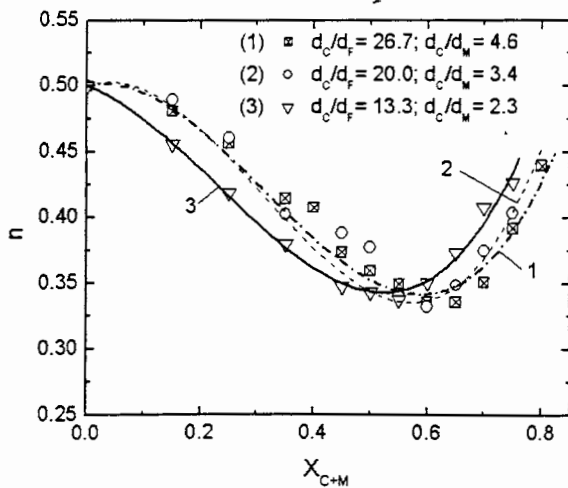


Figure 5: Dependence of calculated $n = n(d_{av}, k, \epsilon)$ on X_{C+M} and fitting functions 1 – 3 as defined by Equation (15):

- 1: $n = 0.5 + 0.1034X_{C+M} - 1.7157X_{C+M}^2 + 1.8417X_{C+M}^3$
- 2: $n = 0.5 + 0.156X_{C+M} - 2.05X_{C+M}^2 + 2.23X_{C+M}^3$
- 3: $n = 0.5 + 0.153X_{C+M} - 1.13X_{C+M}^2 + 1.625X_{C+M}^3$

Application of Equation (14) in Equation (5) predicts the tortuosity with a maximum deviation of 3% for the obtained experimental data.

It can be concluded that real ternary packings within the investigated range have properties significantly different due to distortion effects in the packing as compared with the values obtained by the expected model $T = 1/\epsilon^n$ with $n = \text{constant}$. Nevertheless, at $\delta_{MC} \rightarrow 0$ and $\delta_{FM} \rightarrow 0$ the tortuosity behaviour will be close to the case of $n = \text{constant}$ since the vast majority of fine particles become arranged as a dense packing.

Permeability

As mentioned above, permeability includes a complex factor ϵ/T that characterises the interplay between packing porosity and tortuosity. In Figure 7 the normalised dependence of ϵ/T expressed as $(\epsilon/T)/(\epsilon_F^0/T_F)$ is shown where T_F is the tortuosity of fine particles packing. Similar to the tortuosity case (Figure 6), the porosity was described by Equation (14) and curves 1 – 3 represent tortuosity $T = 1/\epsilon^n$ where n is given by Equation (15). Curves 1' – 3' were obtained for the condition $n = 0.5$.

The permeability is also affected by the packing distortion effects and significantly differs from the prediction for constant n (Figure 8, curves 1' – 3'). Conditions of models plotted in Figure 8 are similar to those mentioned for Figure 6. Experimental data and modelling results show the complexity of the real ternary packing arrangement. It may be concluded that for the description of real porous media then experimental measurements are preferable to predict flow and mass transfer processes.

CONCLUSION

The dependence of the porosity on mixture composition

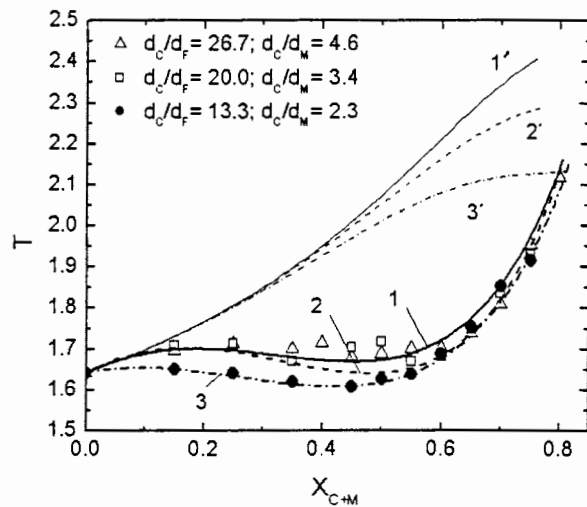


Figure 6: Dependence of T on X_{C+M} . Curves 1 – 3: value n in $T = 1/\epsilon^n$ is the function defined by Equation (15), expressions displayed in Figure 5. Curves 1' – 3': n is assumed to be $n = 0.5$.

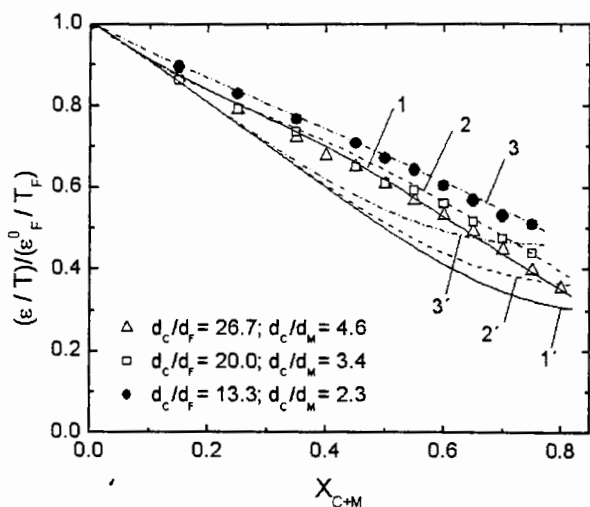


Figure 7: Normalised dependence of $(\epsilon/T)(\epsilon_F^0/T_F)$ on X_{C+M} . Curves 1 – 3: value n in $T = 1/\epsilon^n$ is the function defined by Equation (15), expressions displayed in Figure 5. Curves 1' – 3': n is assumed to be 0.5.

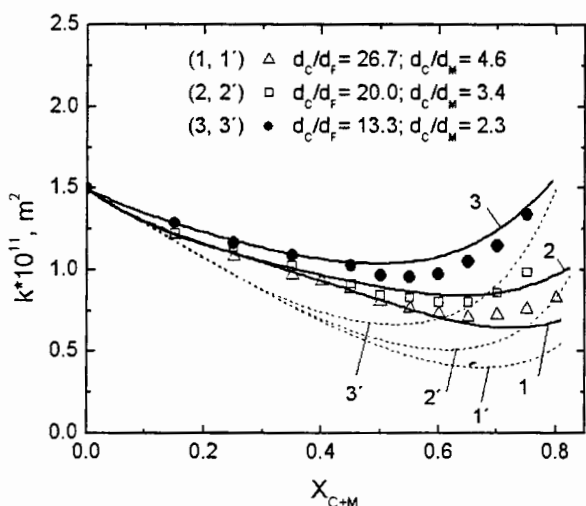


Figure 8: Dependence of the permeability k on X_{C+M} . Curves 1 – 3: value n in $T = 1/\epsilon^n$ is the function defined by Equation (15), expressions displayed in Figure 5. Curves 1' – 3': n is assumed to be $n = 0.5$.

content was obtained by using the Kozeny-Carman equation to determine the packing tortuosity and permeability. Results show that in the conventional relation $T = 1/\epsilon^n$ the parameter n varies with the fraction content. The observed phenomenon can be explained by wall and distortion effects between particles of different sizes. This fact significantly affects predicted permeability of ternary packing and must be taken into consideration in practical applications.

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NOMENCLATURE

- d_C average diameter of coarse particles (m)
- d_F average diameter of fine particles (m)
- d_M average diameter of medium size particles (m)
- d_{av} average particle diameter of a particle mixture (m)
- K_0 constant, assumed to be 2.0
- k permeability (m^2)
- L thickness of the bed (m)
- L_e average flow pathway in the bed (m)
- T tortuosity
- p pressure drop through packed bed (Pa)
- u flow velocity (m/s)
- X_C volume fraction of coarse particles in the mixture
- X_F volume fraction of fine particles in the mixture
- X_M volume fraction of medium size particles in the mixture
- X_{C+M} sum of coarse and medium volume fractions of particles in the mixture
- ϵ overall packing porosity
- ϵ_C^0 porosity of pure coarse particle packing
- ϵ_F^0 porosity of pure fine particle packing
- ϵ_M^0 porosity of pure medium size particle packing
- ϵ_{C+M} fractional porosity of coarse and medium size particles in the packing
- φ correction function
- μ viscosity (Pa s)

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