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

R. DIAS[2], M. MOTA[1], J.A. TEIXEIRA[1] and A. YELSHIN[1]

[1]*Centro de Eng. Biológica, University of Minho, Campus de Gualtar, 4710-057 Braga, Portugal.*
[2]*Departamento de Tecnologia Química, Polytechnic Institute of Bragança, Campus de Sta. Apolónia, Ap. 134, 5301-857 Bragança, Portugal.*

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 conditions[1-9].

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 predefined 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 densities[12], 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 equation[21-24]

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

where L is the bed thickness and μ the liquid viscosity. Permeability k [m^2] 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:

$$k = \frac{d_{av}^2 \varepsilon^3}{36K_0T^2(1-\varepsilon)^2} = \frac{\varepsilon d_{av}^2}{36K_0(1-\varepsilon)^2}\left(\frac{\varepsilon}{T}\right)^2 \tag{2}$$

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 coefficient[25-28], the most usual being $36K_0T^2 = 180$. In this case from Equation (2) it follows that

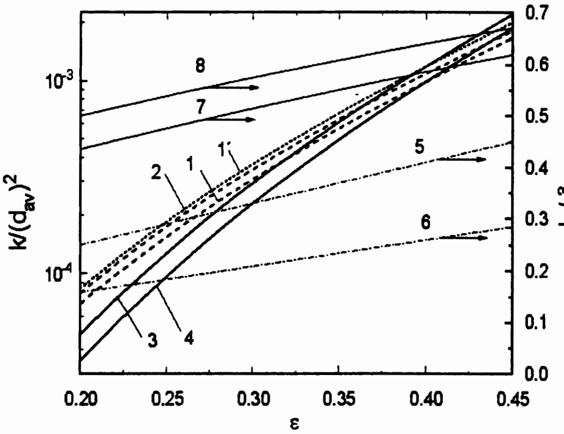

Figure 1: Dependence of k/d_{av}^2 (curves 1 – 4) and ε/T (curves 5 - 8) on the porosity ε for cases of constant and variable with the porosity T. Dimensionless permeability k/d_{av}^2 1 and 1′ – Equation (3) at $36K_0T^2 = 180$ and 150; 2 – Equation (4); 3 – Equation (3) at $T = 1/\varepsilon^{0.4}$; 4 – Equation (3) at $T = 1/\varepsilon^{0.5}$. Complex ε/T: 5 – Tortuosity $T = 1.0$; 6 – $T = 1.58$; 7 – $T = 1/\varepsilon^{0.4}$; 8 – $T = 1/\varepsilon^{0.5}$.

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

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 works[29-35]. As an example, curves 5 and 6 in Figure 1 are given for ε/T at T = 1 and 1.58 respectively.

Hamilton[36] made an attempt to avoid the constant 1/180 in the complex k/d_{av}^2 by using an approach based on spheres assembling[24] (Figure 1, curve 2):

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

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

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

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 from[8] 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.

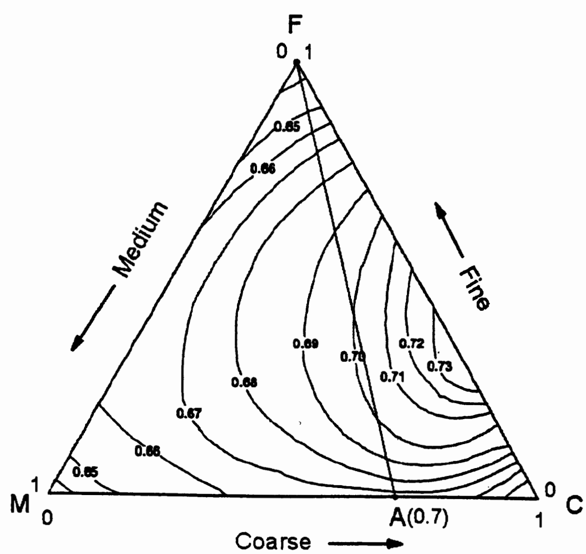

Figure 2: Sketch of the triangular diagram[8] for a ternary mixture of coarse C, medium M, and fine F particle sizes. The packing density iso-curves is presented in the form 1 - ε and represent fine, medium, and coarse particle fractions of size d_F = 7 mm, d_M = 14 mm, and d_C = 28 mm respectively.

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 $\delta_{M/C} = d_M/d_C$ and $\delta_{F/M} = d_F/d_M$ are small and approach zero ($\delta_{M/C} \to 0$ and $\delta_{F/M} \to 0$), 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 $v_F = \varepsilon_{C+M}\left(1-\varepsilon_F^0\right)$ 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

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

The overall packing porosity then becomes

$$\varepsilon = \varepsilon_{C+M}\varepsilon_F^0 = \varepsilon_F^0\left(1-X_{C+M}\right)/\left(1-X_{C+M}\varepsilon_F^0\right), \quad X_{C+M} \in \left[0, X_{(C+M)\min}\right] \tag{7}$$

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

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

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

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 $\delta_{M/C} \to 0$ and $\delta_{F/M} \to 0$ or $\delta_{F/C} = d_F/d_C \to 0$. 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 $\delta_{M/C} \to 0$, $\delta_{F/M} \to 0$ the tortuosity T can be presented as[50]

$$T = T_C^0 T_F^0 T_M^0 \tag{10}$$

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 content[49]. This fact can be taken into consideration by incorporating some correction functions in the model. For simplicity we use the correction function φ as follows[49]:

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

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/m^3 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 work[51] 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 to[52]

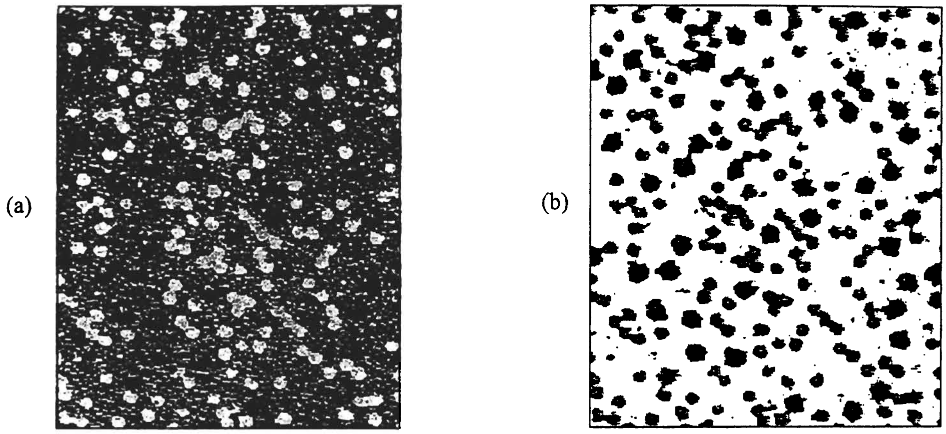

Figure 3: Example of a ternary packing of spheres: d_C = 4 mm (black) 25%; d_M = 2 mm (grey) 25%; d_F = 0.875 mm (white) 50 %. (a) – original image; (b) – treated image.

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

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

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

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), assuming K_0 = 2.0.

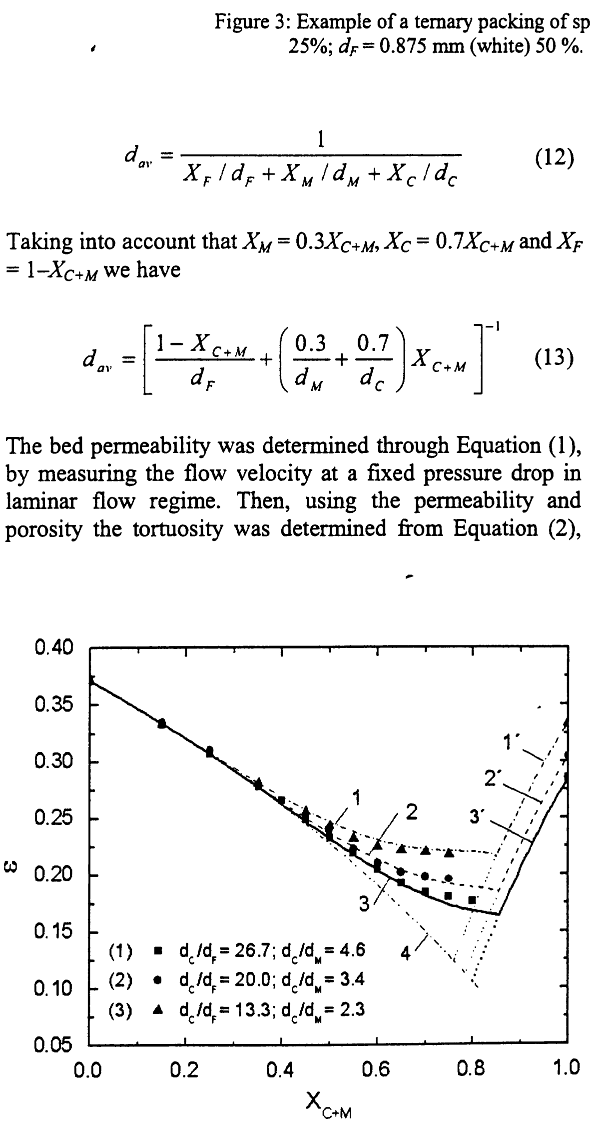

Figure 4: Dependence of ternary packing porosity ε on volume fraction X_{C+M}. Curves 1 – 3 calculated by Equation (14). Curves 4 characterise the linear model limit $\delta_{M/C} \to 0$, $\delta_{F/M} \to 0$. Curves 1′ – 3′ represent the linear approach, Equation (8), when $\varepsilon^0_{C+M} = \varepsilon^0_C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

RESULTS AND DISCUSSION

The results obtained and correlation dependences are given below.

Porosity

Monosize packing porosity measurements give the following values: for fine particle packing ε^0_F = 0.371, and for coarse and medium size mono packings $\varepsilon^0_M = \varepsilon^0_C$ = 0.4 ± 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 $\varepsilon^0_{M+C} = \varepsilon^0_C \varepsilon^0_M$ is calculated by the experimentally measured porosities of the binary mixture ε^0_{M+C}. Values of ε^0_{M+C} 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 $\delta_{M/C} \to 0$, $\delta_{F/M} \to 0$ for the chosen mixtures and has ε^0_F = 0.371.

Comparing experimental data in Figure 4 with curve 4, it is seen that displacement and wall effects between particles of different sizes[49] 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

$$\varphi = \exp\left(2.0989 X_{C+M}^{2/\sqrt[4]{\delta_{M/C}\delta_{F/C}}}\right)$$

and the ternary packing porosity was modelled by the relation

$$\varepsilon = \exp\left(2.0989 X_{M+C}^{2/\left(\sqrt[4]{\delta_{M/C}\delta_{F/C}}\right)}\right)\varepsilon^0_F \frac{1-X_{C+M}}{1-X_{C+M}\varepsilon^0_F} \qquad (14)$$

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 $n = n(K_0, d_{av}, k, \varepsilon, X_{C+M})$. 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.

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

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

1. value n in relation $T = 1/\varepsilon^n$ is the function defined by Equation (15), curves 1 – 3
2. value n is the constant assumed to be 0.5, curves 1′- 3′.

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 $T = 1/\varepsilon^n$ with n = constant. Nevertheless, at $\delta_{M/C} \to 0$ and $\delta_{F/M} \to 0$ the tortuosity behaviour will be close to the case of n = 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 $(\varepsilon/T)/(\varepsilon_F^0/T_F)$ 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 $T = 1/\varepsilon^n$ 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

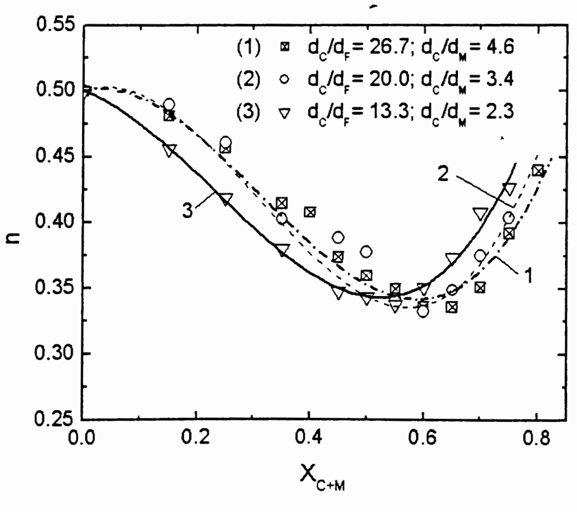

Figure 5: Dependence of calculated $n = n(d_{av}, k, \varepsilon)$ on X_{C+M} and fitting functions 1 – 3 as defined by Equation (15):

$$1: \ n = 0.5 + 0.1034X_{C+M} - 1.7157X_{C+M}^2 + 1.8417X_{C+M}^3$$

$$2: \ n = 0.5 + 0.156X_{C+M} - 2.05X_{C+M}^2 + 2.23X_{C+M}^3$$

$$3: \ n = 0.5 + 0.153X_{C+M} - 1.13X_{C+M}^2 + 1.625X_{C+M}^3$$

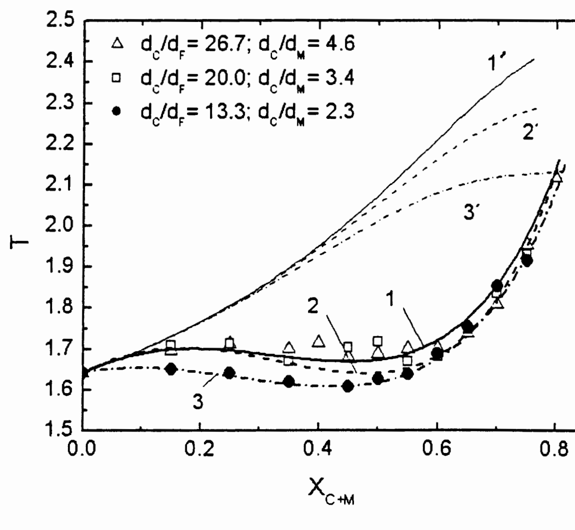

Figure 6: Dependence of on X_{C+M}. Curves 1 – 3: value n in $T = 1/\varepsilon^n$ is the function defined by Equation (15), expressions displayed in Figure 5. Curves 1′ – 3′: n is assumed to be $n = 0.5$.

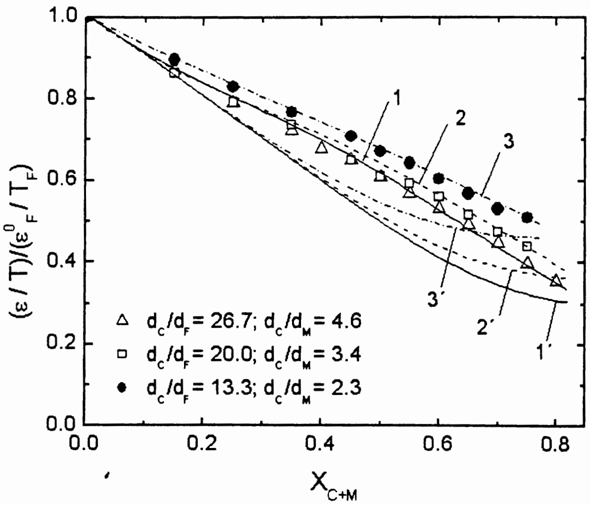

Figure 7: Normalised dependence of $\left(\varepsilon / T\right)\left(\varepsilon_F^0 / T_F\right)$ on X_{C+M}. Curves 1 – 3: value n in $T = 1/\varepsilon^n$ is the function defined by Equation (15), expressions displayed in Figure 5. Curves 1′ – 3′: n is assumed to be 0.5.

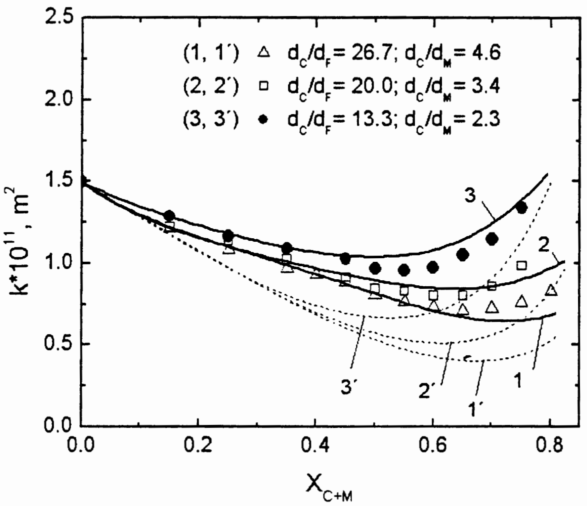

Figure 8: Dependence of the permeability k on X_{C+M}. Curves 1 – 3: value n in $T = 1/\varepsilon^n$ is the function defined by Equation (15), expressions displayed in Figure 5. Curves 1′ – 3′: n is assumed to be $n = 0.5$.

content was obtained by using the Kozeny-Carman equation to determine the packing tortuosity and permeability. Results show that in the conventional relation $T = 1/\varepsilon^n$ 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.

ACKNOWLEDGEMENT

The authors wish to thank Fundação para a Ciência e Tecnologia (FCT) for providing the funds to perform this work through the project POCTI/EQU/37500/2001, as well as for the grant awarded to A. Yelshin. This project was partially funded by FEDER.

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)

REFERENCES

1. Hoover F.W. and Iler R.K., 1977, US Patent 4060488, November 20, 1975.
2. Yu A.B., Zou R.P. and Standish N., 1996, Modifying the linear packing model for predicting the porosity of nonspherical particle mixtures, *Industrial Engineering Chemistry Research*, **35**(10), 3730-3741.
3. Mota M., Teixeira J.A. and Yelshin A., 1999, Image analysis of packed beds of spherical particles of different sizes, *Separation and Purification Technology*, **15**(15), 59-68.
4. Möhrmann C., Jung A. and Woldt E., 1999, The packing density of randomly packed spheres and the structural stability of metallic glasses, e-publication *http://andreas.welcomes-you.com/research/atompack/*, 1-14.
5. Yang R.Y., Zou R.P. and Yu A.B., 2000, Computer simulation of the packing of fine particles, *Physical Review E*, **62**(3), 3900-3908.
6. Venugopalan R., Sathiyamoorthy D. and Gadiyar H.S., 2001, Mathematical modelling of ternary particle system for determining the critical size ratio in a chemically reacting condition, *Powder Technology*, **114**, 229-236.

7. Mota M., Teixeira J.A. and Yelshin A., 2001, Binary spherical particle mixed beds porosity and permeability relationship measurement, *Transactions Filtration Society*, **1**(4), 101-106.
8. Liu S. and Ha Z., 2002, Prediction of random packing limit for multimodal particle mixtures, *Powder Technology*, **126**, 283-296.
9. Mota M., Teixeira J.A., Yelshin A. and Bowen W.R., 2003, Interference of coarse and fine particles of different shape in mixed porous beds and filter cakes, *Minerals Engineering*, **16**(2), 135-144.
10. Doheim M.A., Abu-Ali M.H. and Mabrouk S.A., 1997, Investigation and modelling of sedimentation of mixed particles, *Powder Technology*, **91**, 43-47.
11. Falk V. and D'Ortona U., 2002, A polydisperse sedimentation and polydisperse packing model, *Powder Technology*, **128**, 229-235.
12. Caglioti E., Coniglio A., Herrmann H.J., Loreto V. and Nicodemi M., 1998, Geometrical frustration: a dynamical motor for dry granular media, *Physica A*, **257**, 419-423.
13. Ouchiyama N. and Tanaka T., 1984, Porosity estimation for random packings of spherical particles, *Industrial Engineering Chemistry Research*, **23**(4), 490-493.
14. Suzuki M. and Oshima T., 1985, Verification of a model for estimating the void fraction in a three-component randomly packed bed, *Powder Technology*, **43**(2), 147-153.
15. Le Goff P., Leclerc D. and Dodds J., 1985, The structure of packed beds: Continuity of research in Nancy and some new results, *Powder Technology*, **42**, 47-53.
16. MacDonald M.J., Chu C.-F., Pierre, P.P. and Ng K.M., 1991, A generalized Blake-Kozeny equation for multisized spherical particles, *American Institute Chemical Engineers Journal*, **37**(10), 1583-1588.
17. Yu A.B. and Standish N., 1991, Estimation of the porosity of particle mixtures by a linear-mixture packing model, *Industrial Engineering Chemistry Research*, **30** (6), 1372-1385.
18. Yu A.B. and Standish N., 1993, A study of the packing of particles with a mixture size distribution, *Powder Technology*, **76**, 113-124.
19. Yu A.B., Bridgwater J. and Burbidge A., 1997, On the modelling of the packing of fine particles, *Powder Technology*, **92**, 185-194.
20. Mota M., Teixeira J.A. and Yelshin A., 1999, Image modelling of mixed granular porous media, *Fluid/ Particle Separation Journal*, **12**(2), 71-79.
21. Carman P.C., 1937, Fluid flow through granular beds, *Transactions Institution Chemical Engineers*, **15**, 150-166.
22. Bear J., 1972, *Dynamics of Fluids in Porous Media*, Dover Publications, New York.
23. Dullien F.A.L., 1975, Single phase flow through porous media and pore structure, *Chemical Engineering Journal*, **10**(1), 1-34.
24. Happel J. and Brenner H., 1965, *Low Reynolds Number Hydrodynamics*, Prentice-Hall, Englewood, New Jersey.
25. Schluep T. and Widmer F., 1996, Initial transient effects during cross flow microfiltration of yeast suspensions, *Journal of Membrane Science*, **115**(2), 133-145.
26. Fane A.G., Fell C.J.D., Hodgson P.H., Leslie G. and Marshall K.C., 1991, Microfiltration of biomass and biofluids: effects of membrane morphology and operating conditions, *Filtration and Separation*, **28**(5), 332-340.
27. Shimizu Y., Shimodera K.-I. and Watanabe A., 1993, Cross-flow microfiltration of bacterial cells, *J. Fermentation and Bioengineering*, **76**(6), 493-500.
28. Madaeni S.S., 1997, Mechanism of virus removal using membranes, *Filtration and Separation*, **34**(1), 61-65.
29. Kausch H.H., Fesko D.G. and Tshoegl N.W., 1971, The random packing of circles in a plane, *J. Colloid Interface Science,* **37**(3), 603-606.
30. Jones W.M., 1976, The flow of dilute aqueous solutions of macromolecules in various geometrics: IV. The Ergun and Jones equations for flow through consolidated beds, *J. Phys. D: Appl. Phys.*, **9**(5), 771.
31. Satterfield C.N., 1980, *Heterogeneous Catalysis in Practice*, McGraw-Hill, New York.
32. Klusácek K. and Schneider P., 1981, Effect of size and shape of catalyst microparticles on pellet pore structure and effectiveness, *Chemical Engineering Science*, **36**(3), 523-527.
33. Huizenga D.G. and Smith D.M., 1986, Knudsen diffusion in random assemblages of uniform spheres, *American Institute Chemical Engineers Journal*, **32**(1), 1-6.
34. Wright T., Smith D.M. and Stermer D.L., 1987, Knudsen diffusion in bidisperse mixtures of uniform spheres, *Industrial Engineering Chemistry Research*, **26**(6), 1227-1232.
34. Olague N.E., Smith D.M. and Ciftcioglu M., 1988, Knudsen diffusion in ordered sphere packings, *American Institute Chemical Engineers Journal*, **34**(11), 1907-1909.
35. Hamilton R.B., 1997, Darcy constant for multisized spheres with no arbitrary constant, *American Institute Chemical Engineers Journal*, **43**(3), 835-836.
36. Satterfield C.N., 1970, *Mass Transfer in Heterogeneous Catalysis*, M.I.T. Press, Cambridge.
37. Ho F.-G. and Strieder W., 1981, A variational calculation of the effective surface diffusion coefficient and tortuosity, *Chemical Engineering Science*, **36**, 253-258.
38. Johnston P.R., 1983, The most probable pore-size distribution in fluid filter media. II. Determining the size of the average pore from fluid permeability measurements, *J. Testing and Evaluation*, **11**(2), 122-125.
39. Suzuki M., 1990, *Adsorption Engineering*, Kodansha-Elsevier, Tokyo.

40. Fan L., Yokota K. and Fujimoto K., 1992, Supercritical phase Fischer-Tropsch synthesis: Catalyst pore-size effect, *American Institute Chemical Engineers Journal*, **38**(10), 1639-1648.

41. Johnston P.R., 1992, *Fluid Sterilization by Filtration;* Interpharm Press: USA.

42. Choi K.J., Cecala D.M. and Yorbrough D.W., 1992, Determination of a fraction factor and permeability of non-woven filter media, *Fluid/Particle Separation Journal*, **5**(1), 10-13.

43. Delikouras E.A. and Perlmutter D.D., 1993, Combined effects of mass transfer and inaccessible porosity in gasification reactions, *American Institute Chemical Engineers Journal*, **39**(5), 829-836.

44. Zhang T.C. and Bishop P.L., 1994, Evaluation of tortuoşity factors and effective diffusivities in biofilms, *Water Research*, **28**(11), 2279-2287.

45. Sansone U. and Voitsekhovitch O., 1996, *Modelling and Study of the Mechanisms of the Transfer of Radioactive Material from Terrestrial Ecosystems to and in Water Bodies Around Chernobyl*, Final report EUR 16529 EN; European Commission, Luxembourg, Brassels.

46. Young D.F. and Ball W.P., 1998, Estimating diffusion coefficients in low-permeability porous media using a macropore column, *Environmental Science Technology*, **32**(17), 2578-2584.

47. Mota M., Teixeira J.A. and Yelshin A., 2001, Immobilized particles in gel matrix-type porous media. Homogeneous porous media model, *Biotechnology Progress,* **17**(5), 860-865.

48. Mota M., Teixeira J.A., Dias R. and Yelshin A., 2004, Effect of real particles packing with large size ratio on porosity and tortuosity of filter bed, *Proceedings of 9th World Filtration Congress*, pp.1-30, AFS, New Orleans, Louisiana.

49. Mota M., Teixeira J.A., Bowen R. and Yelshin A., 2000, Effect of tortuosity on transport properties of mixed granular beds, *Proceedings of 8th World Filtration Congress*, pp.57-60, The Filtration Society, Brighton, England.

50. Dias R., Teixeira J.A., Mota M. and Yelshin A., 2004, Preparation of controlled particulate mixtures with glass beads of different sizes, *Separation and Purification Technology*, **37**(1), 69-80.

51. Perry R.H., Green D.W. and Maloney J.O., 1988, *Perry's Chemical Engineers Handbook, 6th Ed.*, pp. 5.54, McGraw-Hill, New York.

ADDRESS

All correspondence concerning this paper should be addressed to Manuel Mota, Centro de Eng. Biológica, University of Minho, Campus de Gualtar, 4710-057 Braga Portugal. E-mail: mmota@reitoria.uminho.pt