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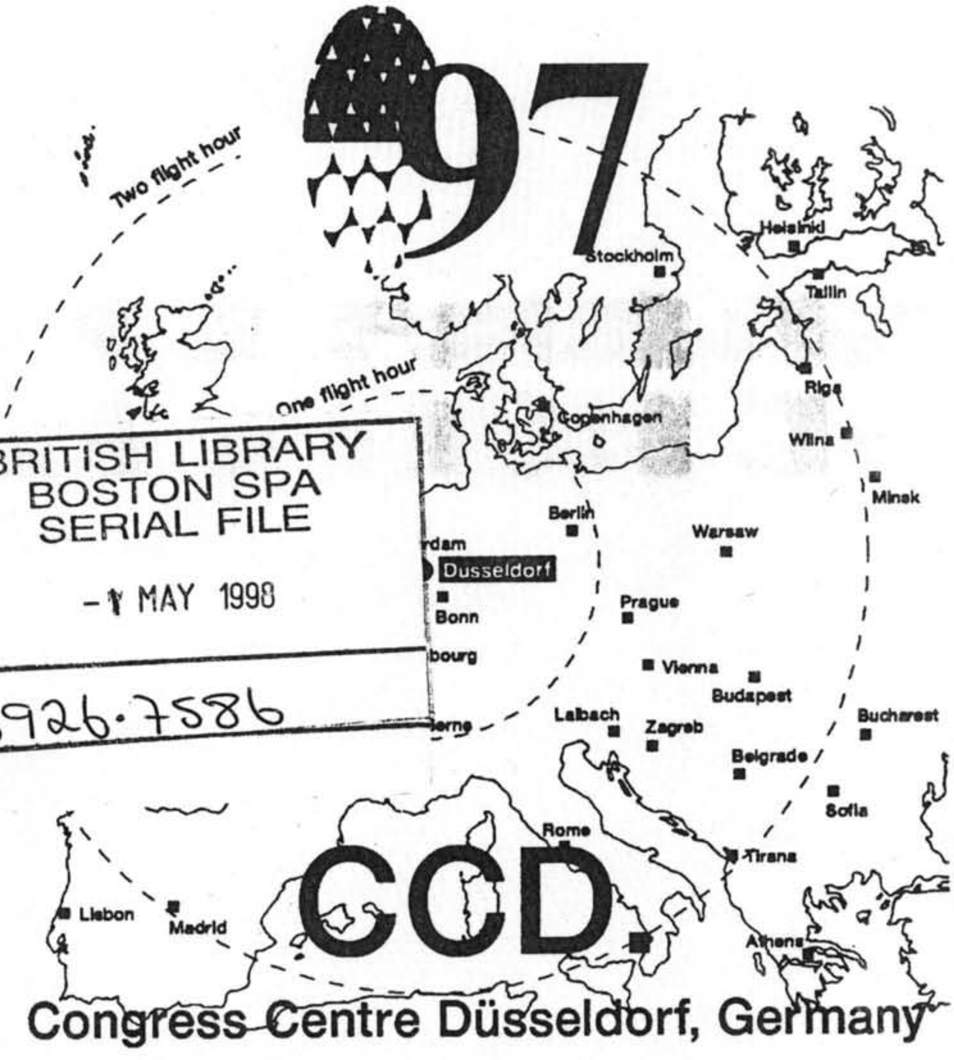

Congress Centre Düsseldorf, Germany

October 14 - 16 1997

Organised by the Filtration Society

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POROUS MEDIA BEHAVIOURS MODELLING AND ANALYSIS IN SEPARATION PROCESSES

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The main parameters are used to define effective diffusion coefficient and permeability of the medium is porosity, ε, tortuosity, T, and particle diameter. The qualitative model of the porosity and tortuosity relationship was proposed on the basis of experimental data for binary mixtures of spherical particles. Two-dimensional (2-D) image modelling of binary mixtures gave the possibility of separating impact of ε and T on effective diffusivity and permeability. Both, ε and T pass over extremum (ε has minimum, and T has a maximum), which do not coincide with each other in the case of 2-D model. The range of x_D, where the tortuosity maximum observed, corresponds to the transition zone between unimodal and bi-modal pore size distribution. Compressibility of the layer formed by aggregated particles in simplified case may be consider on the basis of the binary mixture if primary suspended particles have narrow size distribution and are built compact aggregates. The tortuosity variation in the binary mixture has significant impact on permeability and diffusivity and must be taken in consideration for interpretation of experimental permeability or diffusivity data as well as when granular porous media model is used for predicting porous media properties.

Introduction

Porous media play an important role in nature and technology. There are several models and approaches that describe porous media properties and associated mass transfer phenomena (Bear, 1972; Dullien, 1975; Suzuki, 1990). In general, the major porous media properties are expressed in two parameters - the permeability coefficient (flow phenomena) and the effective diffusion coefficient (mass transfer phenomena). Both coefficients include two main characteristics of porous media: porosity, ε, and tortuosity, T.

The flow of fluid through porous media may be described by the Kozeny-Carman model (Bear, 1972):

$$u = k\frac{\Delta p}{\mu L}, \text{ where } k = \varepsilon^3 d_s^2 / 36K(1-\varepsilon)^2 = \left(\frac{\varepsilon}{T}\right)^2 \frac{\varepsilon \cdot d_s^2}{36K_0(1-\varepsilon)^2}. \tag{1}$$

Here u is fluid flow velocity; k, the permeability; Δp, the pressure drop; μ, the fluid viscosity; L, the media thickness; ε is the porosity, and d_s is the particle equivalent diameter. The Kozeny coefficient, K, may be expressed as $K = K_0T^2$, where K_0 is a constant that equals to 2 in most cases.

Mass transfer phenomena depends on the effective diffusion coefficient, $D_e = (D_0\varepsilon)/T$, or a ratio

$$\eta = D_e / D_0 = \varepsilon / T, \tag{2}$$

where D_e is the effective diffusion coefficient in porous media, and D_0 is the diffusion coefficient in a bulk liquid.

As can be seen, mass transfer as well as flow phenomena depends, respectively, on ε / T and $(\varepsilon / T)^2$. But tortuosity T depends on porosity $T(\varepsilon)$, also (Bear, 1972; Dullien, 1975; Suzuki, 1990; McCune, et al., 1979). For granular mixed beds, both tortuosity and porosity are related to the volume fraction of different size particles in the mixture. A background for the description of the porosity of beds was developed and well discussed in (Yu, et al., 1996; Zou and Yu, 1996) but a problem of the tortuosity relationship with porosity and particle size distribution still actual for mixed granular beds.

At the fall spring of 1996 by Internet was held a discussion about a definition of a tortuosity. The discussion showed cordially different points of view on the subject. Here are some opinions:

- "If you consider that a porous media is just a porous media, tortuosity is not even part of the vocabulary".
- "I believe it is $(L/L_e)^2$". Here L_e and L is the pore length and the porous medium thickness, respectively.
- "...There is usually no need whatsoever to view or define a tortuosity by a ratio of lengths (or square of lengths). This is a "pedagogical" way of representing things, which is, sadly, wrong (cf. Constrictions, network effects, (fractal) morphological aspects)..."
- "I have yet to find a single answer for tortuosity".

As we can see the problem still actual as it was some decades ago. The tortuosity is evident or latent in all models of mass transfer in porous media.

Permeability as well as effective diffusivity contain ε and T in the form of ratio ε / T, as it was mentioned above, and both parameters are dependent on many factors, for instance, particles size distribution, a particle shape factor, the porous medium scale, mode of the porous media preparation, etc. Moreover, the porous medium parameters often depend on processes occurring during mass transfer: porous media compressing or expanding; particles or macromolecules deposition inside pore channel up to pore blocking phenomena, etc.

The relationship between porosity and tortuosity analyses is the subject of this work.

Porous media gradation

First of all let us consider a porous media gradation that can be a basis for further development some general approach for the porous medium characterisation. Behaviours of porous media for separation processes and mass transfer are important both static conditions (when media are unstressed or under static stress) as well as "in a process" under dynamic flow stress and due to comprising or expanding.

Granular media vs. particle size. Particle size ratio in the granular mixture together with their volume fraction distribution can radically change the porous medium structure. In the simplest case of a binary mixture of spherical particles the porosity is smaller than the porosity of mono-sized bed (Yu et al., 1996). The follow gradation of granular porous media is proposed on Fig. 1. Gradations may be used for porous media with deposited during separation processes particles or substrates on and inside media.

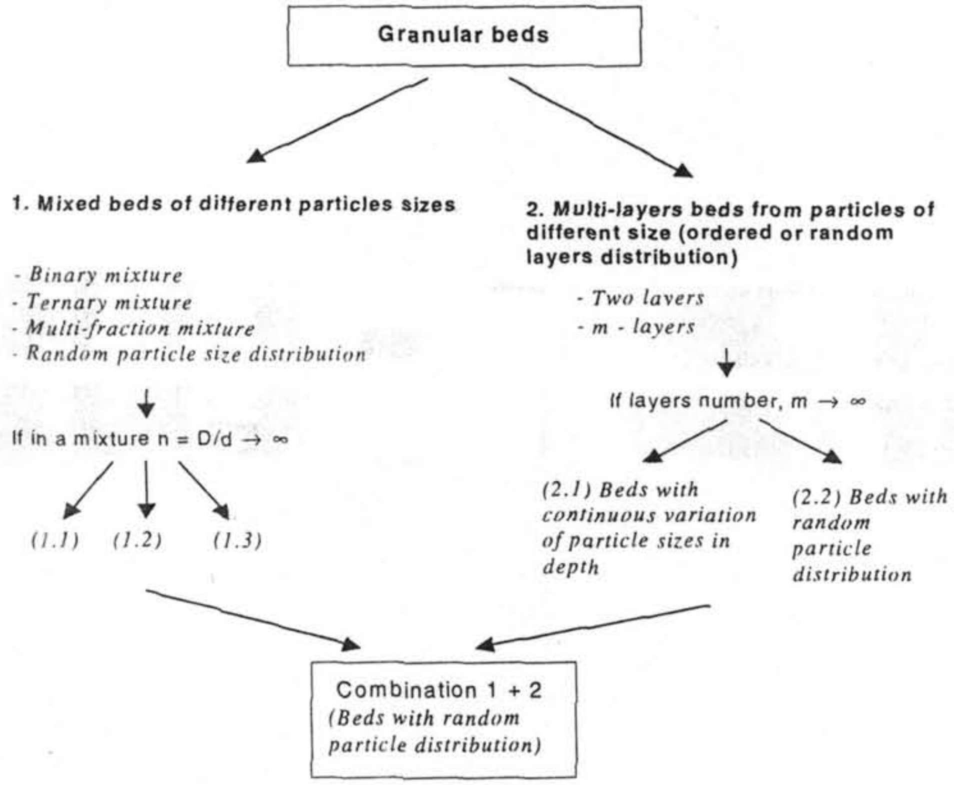

Figure 1. Scheme of the granular media gradation. Here D is the largest particle diameter, and d is the smallest particle fraction diameter. (1.1) – Porous media enrich of smallest particles. Large particles immobilised inside the small particle matrix. (1.2) – Intermediate porous media when the porosity and tortuosity are approach to minimal and maximal values, respectively. (1.3) – Porous media enrich of large particles. Small particles play a role of fillers of a void space between large particles.

In some cases the total volume of the porous medium is constant but the redistribution of small size particles inside the medium void space can be held during separation process. If the averaged by the medium total volume values d_s and ε are used in equations 1 and 2, they are indifferent to this kind of redistribution, but the tortuosity is still sensitive, Fig.2. On Fig. 2 is clear seen that different arrangement of small particles leads to different path tortuosity: in the case (a) the tortuosity is higher than in the case (b).

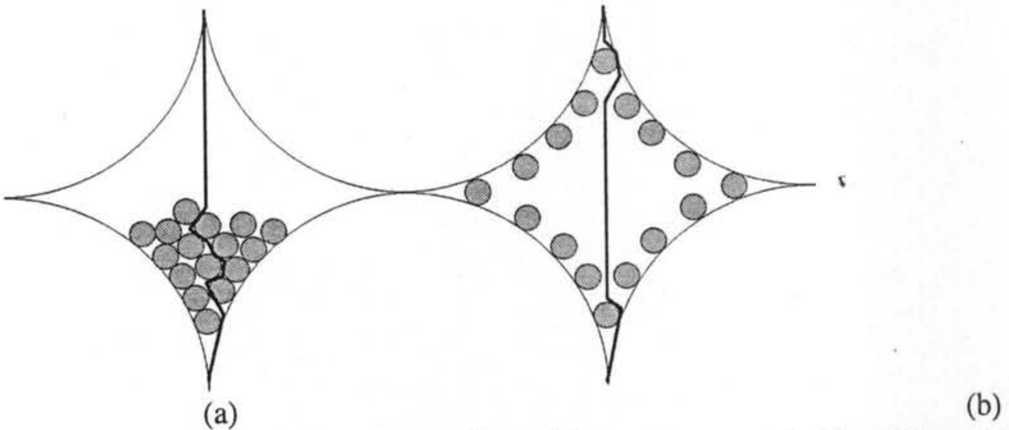

Figure 2. Scheme of two different types of 16 small particles arrangement inside of the void space of large particles and tortuosity of path.

It is known that the tortuosity, T, is the function of porosity, ε, and increase when the porosity decrease. Different empirical equations are used (Suzuki, 1990, Zhang et al., 1994). One of them is (Zhang et al., 1994)

$$T = 1/\varepsilon^{0.5}. \tag{3}$$

Since in binary mixture the porosity pass over a minimum when volume fraction of large particle in the mixture, x_D, increase from 0 to 1, we may expect that the tortuosity must pass over a maximum. Hence, both the porosity and tortuosity variation must be considered for interpretation of experimental permeability or diffusivity data as well as when granular porous media model is used.

Granular media deformation. The tortuosity changes with porous media deformation (compressing or expanding). This fact can play significant role in a high pressure separation processes. The ability to deformation depends on porous media nature (Yelshin, 1994).

The compressible cake phenomena under pressure drop and frictional drag acting on the cake particles can be divided as the following: 1). Rearranging of particles in the cake under the applied stress. 2). Matrix compression in a gel-like cakes. 3). Complex case when both effects take place in the cake, for instance in membrane separation of biological matters. The main factors of cake compressibility are decreasing porosity and pores size due to cake structure collapse or comprise, and increase the pore tortuosity associated with porosity.

During filtration this primary layer loaded the largest stress force instead the cake layers that formed later. So, the primary cake layer compression determined further output value of processes.

The complexity of the compressibility phenomena in cakes or boundary deposit layers in the microfiltration related with small cake thickness, colloidal or gel-like nature of the cake and unlimited variety of possible physico-chemical factors. Nevertheless, the further research should be direct towards understanding the relationships between the porosity, pore size, and tortuosity.

Difficulties of some experimental data interpretation appear when the tortuosity does not take to consideration. Decreasing comprised porous media permeability or diffusivity in that case exceeds expected values, calculated on the basis of d_s and ε reducing, only. For example, according to equation 1, when the tortuosity increased 20%, the permeability decreased 1.44 times, respectively.

Some examples of porous media in biotechnology are given in the Table. Often we meet compressible porous medium "sandwich" type, for instance in membrane bioreactors. This list is incomplete, since it does not comprise all possible causes of cake deformation.

Table. Examples of porous media in biotechnology

Type of porous media	Examples
Granular layer	Chromatography
Compactable layers	Flocs of biological substances; deposits/sediments
Deformable particles	Biofilms and biomaterials under high pressure
Non-granular layers, rigid skeleton	Biomaterial immobilised on porous support
Non-granular layers, deformable skeleton	Bioreactors with immobilised cell on gel or non-woven matrix
Mixed layers, rigid skeleton and deformable filler particles	Gel - like deposits in granular bed
Mixed layers, deformable skeleton and rigid filler particles	Biofilms with mineral particles distributed inside
Mixed layers, deformable skeleton and filler particles	Composite bio-sediments (flocs + separate cells)

Tortuosity of the granular mixed bed

A simplified case of the tortuosity interpretation like a geometrical ratio of channel length, L_e, to the porous medium thickness, L, will be consider below, $T = L_e / L$. Even this approach reflects the relationship of T with porous medium structure.

Let us consider how the tortuosity is change in the binary mixture of spherical particles when the volume fraction x_D is change from 0 to 1. Assuming the tortuosity $T = 1/\varepsilon^{0.5}$ and using data of Yu et al., 1996, a normalised tortuosity T / T_0 can be calculated as

$$T / T_0 = (\varepsilon_0 / \varepsilon)^{0.5}, \tag{4}$$

where $\varepsilon / \varepsilon_0$ is normalised porosity, T and ε is the tortuosity and porosity of the binary mixture, respectively, T_0 and ε_0 is the tortuosity and porosity of the monosized layer, respectively.

Calculated values are plotted on Fig. 3 and marked arrow. As we can see the tortuosity has maximum which depend on the particle size ratio D / d, where D and d is diameter of large and small particles, respectively.

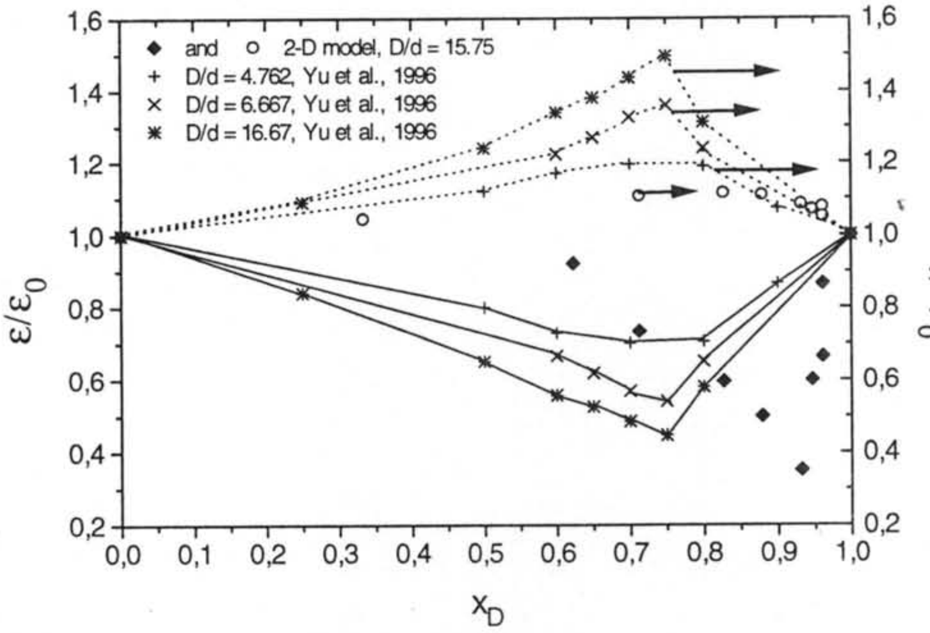

Figure 3. Dependence of the normalised porosity, $\varepsilon / \varepsilon_0$, (left axis) and normalised tortuosity, T / T_0, (right axis) on x_D. Values of T / T_0 for data of Yu et al., 1996, calculated by formula 4.

Since physical modelling of the tortuosity in mixed beds is rather complicated, two-dimensional modelling (2-D model) of mixed beds was done for testing this approach. The 2-D model of the binary mixture in the range of large particle volume fraction $x_D = 0 \div 1.0$ was build by method similar to the described by Suzuki et al., 1981, and Tory et al., 1973. 2-D model was based on building a dense packing and measuring of 2-D porosity and a minimal 2-D tortuosity the model image. As an example, the model data for binary mixture with particle size ratio $D / d = 15.75$ are shown on Fig. 3.

The model data are confirmed that the tortuosity of binary mixture is the function of x_D, and the dependence of the tortuosity on x_D is passed over maximum. Hence, the ratio ε / T, which is a part of the permeability and effective diffusivity, also passed through a minimum. Therefore, for example, by varying x_D the effective diffusivity in the mixture may decrease up to 70% of the monosized porous medium.

Analysis of a frequency distribution of different pore size fractions created in 2-D binary mixture model shows that there is transition from bimodal to unimodal distribution of pore size, Fig. 4. The following groups of the pore fraction, enumerated on Fig. 4, were considered. Group 1, pores of a throat size between two small particles (particle diameter *d*). Group 2, pores of the throat size between of the small, *d*, and large, *D*, particles. Group 3, pores with intermediate size in the range between groups 1 and 2. Group 4, pores of a throat size between two large particles (particle diameter *D*). Group 5 includes pores with size large than group 4. Region of the tortuosity maximum location in 2-D model correlates well with mentioned above the transition zone of the pore size distribution.

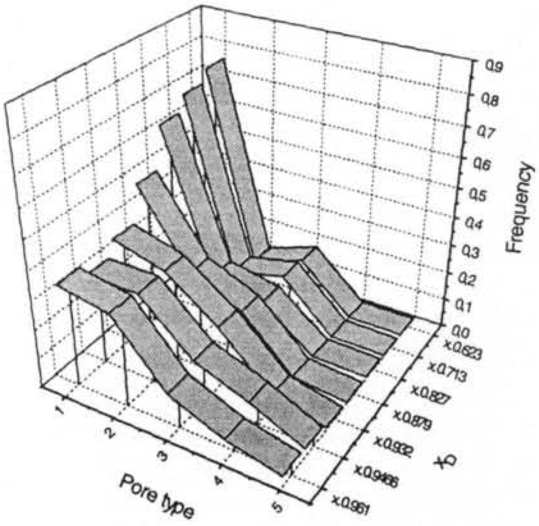

Figure 4. Example of a histogram of the pore fractions distribution in the binary mixture with $D / d = 15.75$ for different x_D. Types of the pore fractions see in the text.

In 2-D model the maximum of T does not coincide to minimum of ε but it locates in the region of x_D which is close to the minimal mixture porosity. Additional experiments with granular mixed beds are necessary for define x_D region corresponds to the maximal tortuosity.

As expected, the diffusivity and permeability are sensitive to tortuosity. Hence, the impact of tortuosity variation due to binary particle beds must be taken in account when modelling transport phenomena in granular beds. For the binary mixture two different packing may have the same porosity or tortuosity but different volume fraction, x_D, which means the existence of the hysteresis loop on the dependence of T vs. ε. This fact must be taken into consideration when binary packing data are represented in terms of T vs. ε.

The discussed above approach can be useful for interpretation cake compressibility when the cake compression effect defined by particles and aggregates rearrangement in the cake structure. Compressibility of the layer formed by aggregated particles in simplified case may be consider on the basis of the binary mixture if primary suspended particles have narrow size distribution and are built compact aggregates. In the comprising cake aggregates can be considered as the large size particles and primary particles as the small size particles. During the cake compression volume fraction of large particles (aggregates), x_D, decrease due to restructuring. Hence, the porosity reducing and the tortuosity increasing can be estimated by the discussed approach for granular binary mixtures. Besides the ratio D / d is decreased, also.

Conclusion

Investigation of mixed granular beds properties show the complicated relationship of the tortuosity and porosity, even in simplified geometrical interpretation. The qualitative model of the porosity and tortuosity relationship was proposed on the basis of experimental data for binary mixtures of spherical particles. Two-dimensional (2-D) image modelling of binary mixtures gave the possibility of separating impact of ε and T on effective diffusivity and permeability. Both, ε and T pass over extremum (ε has minimum, and T has a maximum), which do not coincide with each other in the case of 2-D model. The range of x_D, where the tortuosity maximum observed, corresponds to the transition zone between unimodal and bi-modal pore size distribution. The tortuosity variation in the binary mixture has significant impact on permeability and diffusivity and must be taken in consideration for interpretation of experimental permeability or diffusivity data as well as when granular porous media model is used for predicting porous media properties. Compressibility of the layer formed by aggregated particles in simplified case may be consider on the basis of the binary mixture if primary suspended particles have narrow size distribution and are built compact aggregates. It is necessarily more detailed investigation for establish functional dependence of the tortuosity on such parameters as porosity, porous media topology, particles size ratio, etc.

Acknowledgement

This work was funded by PRAXIS XXI under the grant number BBC/6440/95.

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