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EFFECT OF THE PARTICLE SHAPE ON FLOW THROUGH POROUS MEDIA

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ABSTRACT

In order to study the performance of shaped particles flow in porous media, filtration of two different shape - spherical and rod-like – micro particles was performed through a porous bed. Filtration was investigated at a constant flow rate of 0.04 cm/s with yeast cells, diameter 5 microns, micro spheres, diameter 1 micron, and rod-like bacilli *Lactobacillus bulgaricus* with 6 microns average length and 0.5 micron diameter. Yeast diameter is close to the bacillus length and micro-sphere diameter is in the scale of the bacillus diameter. All particles have similar density. For the packing, the following glass beads were used: coarse particles, size 1.125 mm; fine particles, size 0.1115 mm. Experiments were carried out using a column loaded with a binary packing (volume fraction of coarse particles in the mixture 0.7) or with a monosize packing with the same amount of coarse or fine particles as used in the binary packing. The analysis of the experimental results was based on two models: pure exclusion effect and hydrodynamic separation model (HDC). Results for spheres show that the classic HDC model (B = 1.0) fits well the data whenever the ratio of particle size to the bend scale is high (~ 1/100, as for micro spheres). However, if this ratio increases and becomes ~ 1/20, the HDC model needs to be corrected due to the effect of channel wall curvature on the exclusion effect. This assumption leads to a modified HDC equation - $R = B / (1 + 2\lambda - 2.8\lambda^2)$, where $B \geq 1$ and λ represents the ratio of microparticle size to the pore size. The effect of pore topology plays an important role in the separation of shaped particles when the aspect ratio λ approaches 0.1 and, in the case of bacillus, separation occurs by an exclusion mechanism. For the binary packing, the rod-like particles behave differently from the spherical particles having a length or a diameter in the same scale of bacillus length and diameter. The explanation is the interference of rod-like particles with the pore topology. The exclusion model for particles was formulated in a general form as $R = A / (1 - \lambda)^z$, where A is a coefficient proportional to the tortuosity and parameter z = 1, 2 or 3 depends mainly on the pore shape. For instance, in a parallel-plate channel flow: $R \sim 1/(1 - \lambda)$, for a cylindrical pore $R \sim 1/(1 - \lambda)^2$, and for 3-D pore $R \sim 1/(1 - \lambda)^3$. Further investigation is needed to clarify the particle – pore topology interaction and its effect on particle separation.

KEYWORDS

Porous media, filtration, particle shape, particle separation, flow in complex structure

INTRODUCTION

Motion of colloids and micro-organisms in porous media is a phenomenon widely spread in nature and industry and it is important to know how particle size and shape affect particulate matter dispersion during flow through porous media with tortuous pores.

In general, the tortuosity factor combines both the tortuosity of the pathway and the shape factor (constraints) of the pore channel. We may speculate that in appropriate conditions a tortuous pathway for a rod-like object may create a disorder momentum. This fact may reduce the rod-like micro particle flow velocity as compared with the flow of spherical particles of similar weight. The influence of tortuosity on rod-like particles flowing through porous media is less investigated than others factors and was analysed in present work.

Micro particle separation under non-equilibrium conditions during a dispersion flow in a porous media may be performed by hydrodynamic chromatography (HDC) based on a void excluding effect and flow velocity profile or slalom chromatography (SC) related with a hindrance effect in the tortuous channel.

Hydrodynamic chromatography as a size separation technique is applied in two main forms: capillaries and columns packed with monosize non-porous particles. It must be pointed out that most of HDC theories have been derived from a model of the flow of the dispersed phase in open capillaries [1] where the tortuosity has no influence. The migration of a polymer or a micro particle is characterised by a retention value $R = t_m / t_0$ as a function of an aspect ratio $\lambda = d_e / d_{por}$ in the form:

$$R = B / (1 + 2\lambda - C \cdot \lambda^2) \tag{1}$$

where B = 1.0; t_m and t_0 are the migration time of the micro particle and of an infinite small sized marker, respectively; C is a coefficient; d_e is the micro particle effective diameter; and d_{por} is the diameter of the capillar or the pore.

Coefficient C in Eq. (1) is a constant ranging between 0.5 and 5 [2]. In the packed particulate bed, recommended values of C are in the range: C = 2.698 – 2.8.

For both flexible and stiff macromolecules, it is shown [3] that the flow rate dependence of macromolecules of different size can be explained in terms of deformation and orientation of simple macromolecular models in steady uniaxial elongation. The strength of the flow can often be correlated with Deborah number, De: a ratio of the hydrodynamic forces to the Brownian forces. Significant molecular stretching in steady flows occurs only when De > 0.5.

Contrary to flexible macromolecules, rigid rod-like macromolecules are able to orient themselves in strong flow and although not causing the dramatic rheological effects observed with flexible macromolecules, the orientation of rod-like macromolecules during flow in the small tortuous channels of the column can also be detected as a flow-dependent elution behaviour [3].

Further investigations show that for rigid and semi-flexible macromolecules HDC can be transformed, for certain values of λ, into the separation mechanism called slalom chromatography (SC) [4].

The slalom chromatography method is based on the effect of channel topology on macromolecules motion in a pore channel in the slalom-like manner [5]. Theoretical background of SC is still giving its first steps and the transient condition between HDC and SC is under investigation. In both cases (HDC and SC) the effect of tortuosity and pore topology on separation is insufficiently known.

Depending on the mechanism controlling a separation process, the following series can be obtained according to the size of sample components appearance: 1) large size objects appear first and smallest later – size exclusion chromatography (SEC) and HDC; 2) small size objects appear first and largest later – SC and hindered diffusion.

In order to study the performance of shaped particles flow in porous media, filtration of two different shape - spherical and rod-like – micro particles was performed through a porous bed. Monosize beds form by coarse and fine spheres as well as a binary packing were used.

THEORETICAL BACKGROUND

For separation, rod-like and linear species, behave differently from sphere shape particles, even when other physico-chemical effects are omitted. In appropriate conditions, the tortuous pathway, especially in the form of bends, may create a disorder momentum for the rod-like object. In Fig. 1, a sketch of a rod particle motion in a tortuous channel is shown.

Each time a rod-like particle goes through a constraint in the bend form, this constrain acts as a disordering factor of the particle axial orientation relative to the flow streamlines. Micro particle motion in the pore channel may be considered as a "slalom" regime where the main retardation effect is the friction force when the particle passes the bend region with a change occurring in its spatial orientation. The most "favourable" orientation of rod-like particles is along streamlines but in the bend region streamlines and main particle axis directions are changed: rod-like particles expose a higher cross-section area that increases the friction force and results in the retardation phenomenon.

A relaxation effect in the macromolecular flow is characterised by the Deborah number that can be presented as the ratio of a moving object relaxation time θ to the time when the object was exposed to deformation θ_p

$$De = \theta / \theta_p \tag{2}$$

For macromolecules θ is directly proportional to the boundary viscosity at zero shear velocity and to the molecular weight and is inversely proportional to the molecular concentration [6] whereas θ_p is defined by porous media properties: $\theta_p = a \cdot \varepsilon \cdot d_p / u$, where ε is the bed porosity, d_p is the particle diameter, and a is a numerical coefficient. When a is assumed to be 1.0, the Deborah number becomes

$$De = u\theta /(\varepsilon \cdot d_p) \tag{3}$$

For rod-like particles, the estimation of the relaxation time, θ, is important and can be made using some analogy between flexible and rigid macromolecules. Although rigid rod-like macromolecules do not cause the dramatic rheological effects seen with flexible

polymers, the orientation of rod-like macromolecules during flow in the pore channel is characterised by the rotational relaxation time [3].
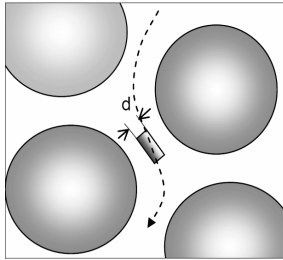

Figure 1. Sketch of a rod micro particle motion in a granular bed. The trajectory of the rod particle in the tortuous channel between spheres is shown by the dashed curve.
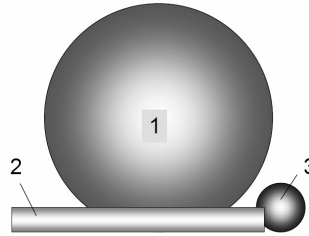
Figure. 2. Sketch of comparative sizes of particles used as the dispersed phase: 1 – *S. cerevisiae*, 2 – *L. bulgaricus*, and 3 – Micro sphere.

The effective size of the rigid dumb-bell during flow through a pore will be roughly related to the projection of the orientation distribution function into a plane perpendicular to the direction of flow. The maximum value of this projection will qualitatively describe the transverse extent of the oriented dumbbell. The model [3] of flow flexible macromolecules leads to a theoretical prediction for d_e

$$d_e = L, \ \theta \cdot \Gamma < 1/9 \qquad \text{or} \qquad d_e = L(9 \cdot \theta \cdot \Gamma)^{-1/2}, \ \theta \cdot \Gamma > 1/9 \qquad (4)$$

where L is the length of the dumbbell and $\Gamma = 1/\theta_p = au/d_p$, a = 6.

We shall now consider a packing consisting of spherical particles of size d_p and that the effect of pore channels tortuosity on micro particle flow is described by means of the pathway bends, Fig. 1. This allows for the estimation of the number of bends. Scale parameter for packing is chosen as the particle size d_p proportional to the number of bends.

Another parameter associated with the micro particle is the ratio λ of micro particle "effective" size d_e to the pore size d_{por}. For rods, as follows from Eq. (4), the effective size can be within the range of minimum and maximum geometrical scales (diameter and length) and depends on flow velocity. Moreover, in the complex pore topology d_e may be different from the predicted by the model (4).

Apparently, flowing rod-like micro particles enable the filling of the pore topology only in a limited range of λ values that, by preliminary fractal analysis of two-dimensional (2D) model of disc packing was defined as $0.08 > \lambda > 0.01$ for a compact micro particle shape.

The influence of the pore topology through the bend number can be presented as it follows - if h is the bed thickness and the tortuosity T of the average pathway of length L_e is $T = L_e / h$, then the bend number n_b is

$$n_b = L_e / d_p = T \cdot h / d_p \qquad (5)$$

For example, based on the data used in the below described experiments, in a monosized coarse sphere packing where h = 0.3 m and $d_p = d_c$ = 1125 microns the number of bends is n_b = 373 assuming T as 1.4.

The number of bends can be increased with the application of a mixed particle bed. For a binary mixture of particles significantly different in size, the overall packing tortuosity T becomes a product of micro- and macro-scale tortuosity T_f and T_c, respectively: $T = T_f \cdot T_c$. For a binary packing, equation (5) can be written as

$$n_b = T(h / d_p) = T_f \cdot T_c (h / d_f) \qquad (6)$$

where $d_f = d_p$ is the diameter of fine spheres in the packing.

Macro-scale tortuosity T_c (coarse particle fraction of size d_c) increases the pathway length per column, whereas the micro-scale tortuosity T_f (fine particle fraction of size d_f) is responsible for the number of bends per pore unit length or bed thickness. For h = 0.3 m and d_f = 111.5 microns, the bends number may be written as $n_b = 2.7 \cdot 10^3 \cdot T$. For coarse particle size ratio $d_c / d_f \geq 10$, it is possible to assume in a binary sphere packing that $T_f = T_c \sim 1.45$ and the overall packing tortuosity becomes $T = (1.45)^2$ = 2.1, Hence, a $n_{b(bin)}$ value 1.45 times higher than for equal bed thickness of pure fine particles packing and 15 times higher than for equal thickness of the coarse particles packing is obtained From this estimation, it is possible to assume a larger retardation effect for rod-like micro particles in a binary packing.

MATERIALS AND METHODS

To build a column packing it is necessary, at first, to choose the micro particles for the experiments. As the rod-like micro particles *Lactobacillus bulgaricus* cells of length ~ 5 – 6 and diameter ~ 0.5 microns were used. Its flow behaviour through porous media was compared with the flow behaviour of yeast cells (*Saccharomyces cerevisiae*, size ~ 5 micron) and micro spheres of size 1 micron. Both yeast and micro spheres have size close to the bacillus length and diameter, respectively, Fig. 2.

The dispersed phase flow was investigated for three types of bed packing: fine particles, coarse particles, and a mixture of fine and coarse particles. The ratio of pore size to the dispersed particles was chosen in the range recommended for HDC as well as in the range of pore topology sensitivity defined by previously performed 2D fractal analysis. Assuming that the ratio of the dispersed particle size d_e to the pore size d_{por}

must be $\lambda = d_e / d_{por}$ ~ 0.01, then for the fine particle packing, at d_e = 5 micron, the particle size would be around 0.1 mm. Glass beads of size d_f = 0.1115 mm were chosen as the fine particles.

To obtain the maximum effect of the bend approach, the binary packing must be in the range of minimum porosity that corresponds to a coarse particles volume in the mixture of ~ 0.7. An additional condition was imposed: the permeability of fine particles and binary packing must be close to each other. Binary packing of $\delta = d_f / d_c$ ~ 0.1 fulfils the imposed condition and glass beads of size d_c = 1.125 mm were chosen for the coarse particle fraction. The binary packing pore tortuosity was around 1.83 against T = 1.49 for the fine packing.

The binary packing height was ~ 15 cm. The microbial solutions used on filtration tests had a cell concentration on the order of 2.80×10^6 cells/mL. All particles have similar density ~ 1.05 – 1.1 g/cm^3. Dextran blue was used as a tracer for checking the column packing integrity. A constant flow rate of 0.04 cm/s was used in all experiments.

RESULTS AND DISCUSSION

Typical results obtained in experiments are given below in the form of a normalised concentration C_n that represents the ratio of eluted concentration to the maximum measured eluted concentration. Micro particles separation on the binary packing column is shown in Fig. 3.

In the mixed column spherical microparticles of different sizes (microspheres and yeast) do not follow the HDC law, indicating the existence of the hindrance effect. Moreover, for rod-like bacillus the retardation effect is most pronounced.
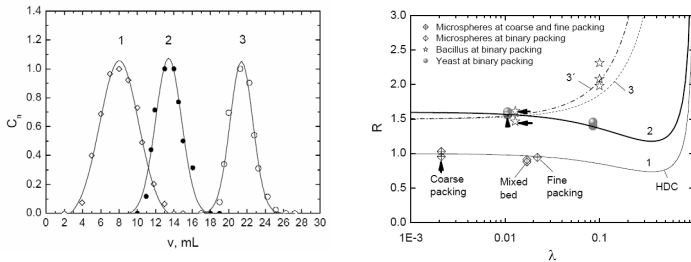

Figure 3. Normalised concentration C_n breakthrough curves vs. elution volume: 1 - Micro spheres; 2 - *S.cerevisiae*; 3 - *L.bulgaricus*. Curves represent a Gaussian distribution fitting function.

Figure 4. Results obtained on the separation of particles on different packings. Curves represent equations: 1 – $R = 1/(1 + 2\lambda - 2.8\lambda^2)$; 2 – $R = 1.6/(1 + 2\lambda - 2.8\lambda^2)$; 3 – $R = 1.5/(1-\lambda)^2$; and 3′ – $R = 1.5/(1-\lambda)^3$. Points marked by thick arrows (◄) correspond to experiments with the coarse particle packing.

The dependence of micro particles retention R on the aspect ratio λ for different column packings is given in Fig. 4, where points marked by thick arrows belong to the coarse particle packing. Fine and coarse particles amount in the monosized column was the same as in the previous binary packing. Due to the small pore size in the fine particle packing, around 43 microns, only dextran and micro spheres were able to pass throughout the column - 100% for dextran blue and 79.2% for micro spheres. No significant HDC effect is observed for micro spheres in comparison with dextran in both fine and coarse beds.

Separation of yeast and bacillus by the coarse bed shows, as in the binary column, that yeast appears later than micro spheres whereas the retardation effect of bacillus vanishes. Observed results indicate that together with the aspect ration λ other factors, such as tortuosity and non-HDC conditions, may affect separation.

For micro spheres, in all packing types the aspect ratio is higher than 0.025 and their behaviour corresponds to HDC in the binary packing ($\lambda = 0.017$) with R = 0.88 against the dextran blue macromolecules, being less pronounced at λ = 0.002 (for coarse packing), as the HDC lower limit is reached. Obtained R values for the different beds fit well with the HDC model (1) at C = 0.28, Fig. 4, curve 1.

Yeast *S. cerevisiae* spheroid cells behave on a different manner than micro spheres as the aspect ratio λ is close to the upper HDC limit. Anyway, Fig. 4, experimental points for binary and coarse packings are simulated by the HDC model (1) with displacement parameter B = 1.6: $R = 1.6/(1 + 2\lambda - 2.8\lambda^2)$. Results for spheres show that the classic HDC model (B = 1.0) fits well to the data whenever the ratio of particle size to the bend scale is high ~ 1/100 (fine particles) and particles scale is the Brownian scale (micro spheres). However, if the micro particles size increases (yeast), the HDC model needs to be corrected because of the decreasing particles spatial mobility (hindrance) as a result of the channel wall curvature, being this accounted for on the higher than 1 B value.

Two-dimension simulations mentioned above were provided with mono- and binary packing of disks. Pore area fractal analysis was performed by a test-box method where the box simulates a compact particle in pores. Measured fractal dimension D_F was compared with an aspect ratio λ' of the box size to the pore size showing the sensitivity of D_F on λ'. For a hexagonal monosize 2D packing, fractal analysis gives the following result: at $\lambda' \sim 0.01$, $D_F \sim 1.8$; at $\lambda' \sim 0.05$, $D_F \sim 1.4$; and at λ' = 0.1, $D_F \sim 1.1 \div 1.2$.

Transition to the binary packing with D/d = 15 with fine $\lambda' \sim 0.1$ ($D_F \sim 1.3$) and coarse $\lambda' \sim 0.01$ ($D_F \sim 1.7$) packing characteristics, resulted in a drastic reduction of D_F - from the coarse packing to the minimum porosity packing up to 1.5 with a further linear decreasing towards the fine packing D_F while the aspect ratio in the region of minimum porosity $\lambda' \sim 0.08$ was close to the fine packing value.

Changing in 2D fractal dimension from 2.0 towards 1.0 can, by analogy, in 3D space considered as a D_F reduction from 3.0 towards 2.0 and less. This explains yeast cells behaviour in coarse and binary packings: reduction in D_F means that the micro particle recognises the pore space as anisotropic and together with topological anisotropy imposes a limit on available for testing pore space, hence B in the model (1) becomes higher than unity.

Elution volumes of yeast (diameter ~ 5 microns) and bacillus (length ~ 5 micron) in coarse packing are close to each other indicating the occurrence of the rotational effect in the presence of rod-like particles. This is the main advantage of porous media flow as compared to capillary flow: due to the complex pore topology, stream perturbations do not allow rod-like micro particles to reach the position parallel to the streamlines.

The above mentioned definition is valid at λ around 0.01 (coarse particles packing) if assumed that, for bacillus, the effective diameter is equal to their length. However, as λ approaches 0.1 (binary packing), Fig. 4, the difference between yeast and bacillus retention is observed. Moreover, elution positions of yeast and bacillus becomes inverted.

Theoretically, a significant retardation of bacillus can be the result of the attachment (adhesion) – detachment effect in the porous media. However, the absence of a tail of the bacillus breakthrough curve allows excluding the mentioned reasons. Hence, we may speculate about the interference of rod-like particles with the pore topology.

Moreover, comparing curves 3 and 3′ it is possible to assume that rods, due to their similarity to a one-dimensional object (λ is defined as the ratio of length size to the pore size), recognise the pore channel as a three-dimensional space rather than a two-dimensional one as in the case of yeast.

This recognition, related with the pore space topology, may be supported by the following examples: in a parallel-plate channel flow [7]: $F(\lambda) \sim 1/(1-\lambda)$; for a cylindrical pore $F(\lambda) \sim 1/(1-\lambda)^2$, and for 3D pore $F(\lambda) \sim 1/(1-\lambda)^3$. Therefore, the exclusion model for non-Brownian particles based on the particle size to the pore size ratio can formulated in the general form

$$R = A/(1-\lambda)^z \qquad (7)$$

where A is a coefficient proportional to the tortuosity factor and the parameter z = 1, 2 or 3 depends on the spatial recognition by moving particles of the channel space in a porous medium. In Fig. 6 two modes of exclusion were compared: z = 2, curve 3, and $z = 3$, curve 3′, the best fit was obtained for $z = 3$.

Based on the bends model, Eq. (6), some estimation for the experimental conditions of binary and coarse packings will be made. Binary packing for h = 0.15 m gives the following bends number $n_{b(bin)}$ = 2825. The coarse packing, with the same amount of beads as the binary packing has $n_{b(c)}$ = 193 that is almost 15 times less than at the binary bed.

The time θ_p when the rod-like bacillus is exposed to the distortion effect when passing through the bend depends on the flow velocity and is for the binary packing $\theta_p \approx$ 0.064 s and 1.08 s for the coarse packing. It must be admitted that the rotational relaxation time θ of the rod-like particle in the complex geometry of a packed bed is difficult to measure and calculate and only preliminary estimations are made below.

If De = 0.5 is considered then from (2) the θ values are: for a binary packing, 0.031 s, and for a coarse packing 0.54 s. The rotational relaxation time for xanthan [3], considered as a stiff polymer, with a nominal molecular weight of $2 \cdot 10^6$ was measured to be θ = 0.014 s that is in the scale of the relaxation time for the binary packing.

Hence, the coarse packing value of 0.54 s is too large and, assuming θ = 0.031 s, recalculated De number of coarse packing becomes $De \approx 0.03$.

It is possible to speculate that for the increasing bends impact on shaped micro particle flow through a porous medium, the following consideration must be taken in account: an increase (contrary to HPLC) in overall tortuosity with bends number and flow velocity. However, the opposite effect may occur as the reduction in the bend scale in comparison with the particle size may result in channel surface roughness. Further investigation is needed to clarify the particle – pore topology mechanism of the interference hereby identified.

CONCLUSON

Theoretical estimations on the retardation effect of the packing tortuosity (and bends) on rod-like particles were confirmed experimentally

Results for spheres show that the classic HDC model (B = 1.0) fits well the data whenever the ratio of particle size to the bend scale is high (~ 1/100, micro spheres). However, if this ratio increases and becomes ~ 1/20, the HDC model needs to be corrected by $B \geq 1$ due to the effect of channel wall curvature on the exclusion effect. The effect of pore topology plays an important role in the separation of shaped particles when the aspect ratio approaches λ = 0.1 and for bacillus separation takes place by an exclusion mechanism. For binary packing, the rod-like particles behave differently from the spherical particles of the same size of bacillus length and diameter. This is explained by the interference of rod-like particles with the pore topology. The exclusion model for particles was formulated in the general form $R = A/(1-\lambda)^z$, where A is a coefficient proportional to the tortuosity.

The complexity observed in the retardation phenomenon raises the question of further investigation in this area. Obtained results are interesting for bio-separation, deep bed filtration, and motion of micro particles (viruses, etc.) in porous media.

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