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HOLLOW FIBRE MEMBRANE OSCILLATIONS AGAINST FOULING PROCESS – A NUMERICAL STUDY

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The modelling of colloidal fouling and defouling of hollow fibre membranes in the presence of membrane oscillations is analysed by means of numerical simulations as an effect of complex coupling between hydrodynamic and surface forces. To describe the latter the Derjaguin-Landau-Vervey-Overbeek (DLVO) model has been employed. We have investigated the influence of various parameters of the process like flow rate, mean particle diameter, amplitude and frequency of the oscillations, and others, on the efficiency of the defouling process. The investigated parameters is close to that of a silica suspension in , a typical system modelling used to investigate membrane separation. On the basis of numerical simulation results e have defined an optimal set of parameters preventing membrane fouling.

Keywords: hollow fibre membrane; fouling; numerical modelling; DLVO model

1. INTRODUCTION

The membrane separation technology has undergone intensive development during the last few decades. This has resulted in an increased number of applications in pharmaceutical and food industry, wastewater and desalination processes and many others.

However, besides many advantages of membrane separation processes, there are some drawbacks to their day-to-day operation that reduce the applicability of these processes or increase their economic costs. The most important of these difficulties is membrane fouling. It usually appears in such processes as ultrafiltration (UF), microfiltration (MF), and reverse osmosis (RO) as an inherited and avoidable element of the process of membrane separation (Chang et al., 1995; Quasirani and Samhaber, 2011). Fouling caused by the deposition of small colloidal particles on the inner walls of membrane pores, the blocking of membrane pore opening and the buildup of particles in the form of a cake layer on the membrane surface. The effect of permeation flux reduction due to fouling is twofold. First of all, pore blocking and cake formation lead to the increase in flow resistance. After that, the presence of colloidal particles deposited on the membrane surface hampers liquid mixing. Thus, a relatively high concentration of solutes persists near the membrane surface which causes the reduction of the solvent flux crossing the membrane (Henry et al., 2012).

We usually distinguish between the non-adhesive (reversible) and adhesive (irreversible) fouling. The former is the effect of filtration-induced macrosolutes or particles deposition. The methods of reducing this kind of fouling are: cross-flushing and backwashing (Kroner et al., 1984), the application of low-frequency pressure and velocity pulsing leading to flow instability (Zahka and Leahy, 1985), the

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addition of coagulants for the formation of layer particles, which are easily swept off the membrane surface (Al-Malack and Anderson, 1996), and many others.

The second kind of fouling (irreversible) is caused by van der Waals attractions, hydrogen bounding, hydrophobic interaction, extracellular macromolecular interactions and other effects.

The reduction or elimination of adhesive fouling is usually more difficult than the reduction of non-adhesive one. This could be achieved by physically coating water-soluble polymers or charged surfactants onto the membrane surface (Jonsson and Jonsson, 1991), coating of hydrophilic polymers on the membrane (Stengaards, 1988), grafting monomers to membranes by electron beam irradiation (Kim et al., 1991), and a combined method of back-pulsing and membrane surface modification with photo-induced grafting (Ma et al., 2000).

Another possibility of reducing adhesive fouling on a membrane surface is its excitation through local surface pulsing. It could be achieved by the interaction of electromagnetic particles immersed under a membrane surface with an external electromagnetic field.

Membrane oscillation as a method of preventing colloidal fouling was considered in our previous work (Gac and Gradoń, 2013). In that paper we analysed the embedding of magnetic nanoparticles in the membrane. Placing such a membrane in an oscillating magnetic or electromagnetic field results in the oscillations of magnetic nanoparticles which leads to membrane bulging. We also showed that such bulging hampers the deposition of particles on the membrane and facilitates the detachment of deposited particles. In some cases, however, the periodic magnetic field may lead not to bulging but to the oscillations of the whole membrane. This is expected, e.g. in the case of hollow fibre membranes. Indeed, because of their characteristic cross-section, such membranes are usually very hard to deform. Thus, if magnetic nanoparticles are immersed into a hollow fibre membrane, the periodic magnetic field leads to the oscillations of the whole fibre. We expect that these oscillations may also help us to prevent the membrane from colloidal fouling.

It is also worth noting that the oscillations of hollow fibre membranes can be induced not only due to their interaction with the oscillating external field but also due to mechanical oscillations of membrane caused by other effects (e.g. shaking of membrane module). If the oscillation frequency is high enough (over 100 Hz as we show above) this mechanical stimulation should lead to defouling in the same manner as in the case of oscillating external field.

The aim of this paper is to investigate the colloidal fouling of a hollow fibre membrane. By means of numerical simulations we answer the question within which parameters of these oscillations a significant decrease of fouling can be observed.

This paper is organised as follows: in Section 2 we present the problem statement, its geometrical conditions and mathematical description; in Section 3 we present the results of modelling for various parameters of the system and the features of the oscillations; finally, in Section 4 we give the conclusions of simulation results concerning the optimal parameters of membrane process operation.

2. NUMERICAL MODEL

In this paper we consider the system presented in Fig. 1. We analyse a fragment of a single large hollow fibre membrane with the outer diameter equal to 100 to 200 μm and the thickness 20 to 40 μm. In our computation we take into account a fragment of fibre with the length equal to three lengths of the diameter. The flow of liquid is directed from the outside into the inner space of the membrane. On both ends the periodic boundary conditions are applied.

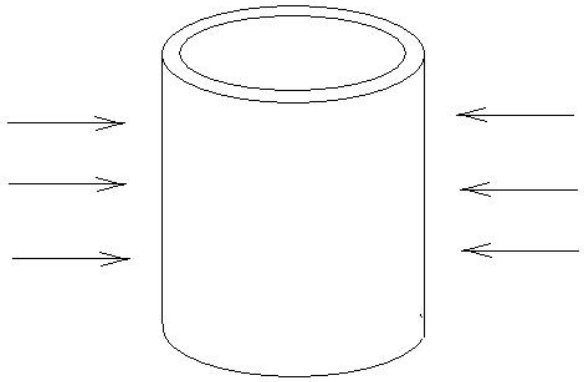

Fig. 1. The scheme of hollow fibre membrane

To describe the interactions between colloidal particles and the membrane as well as other particles we apply the well-known Derjaguin-Landau-Vervey-Overbeek (DLVO) model (Derjaguin and Landau, 1941, Vervey and Overbeek, 1948). According to this model, a single colloidal particle interacts with other particles and the membrane by means of van der Waals forces and electric double layer (EDL) forces. The van der Waals force is an effect of the interactions between single molecules forming two particles. This force is usually attractive, independent of the distance between particles. In contrast, the electric double layer force, which is an effect of interactions between electric charges deposited on the surfaces of particles and surrounding them, may be either attractive or repulsive, dependent on the signs of electric potentials of both particles.

The van der Waals force between two spherical particles with radii R_1 and R_2 is given by means of the formula (Parsegian, 2006):

$$F_{vdW} = -\frac{A_H R_1 R_2}{6(R_1 + R_2)\delta^2} \tag{1}$$

This formula has been chosen as recommended for the interactions between two spheres at relatively small distances (Parsegian, 2006; Ruggiero et al., 1999; Senger et al., 1994).

The EDL force between two spherical particles is given in the form:

$$F_{EDL} = 2\pi\varepsilon_0\varepsilon_r \frac{R_1 R_2}{R_1 + R_2} \frac{\kappa}{1 - e^{-2\kappa\delta}} \left(2\psi_1\psi_2 e^{-\kappa\delta} - \left(\psi_1^2 + \psi_2^2\right)e^{-2\kappa\delta}\right) \tag{2}$$

where κ is the inverse of Debye length and is given as:

$$\kappa = \sqrt{\frac{2e^2 I}{\varepsilon_0\varepsilon_r k_B T}} \tag{3}$$

There are several other formulas to be found in the literature. However, Equation (2) is developed under assumptions of constant charge of particles, small particle potentials and small separation distances. The first two assumptions are justified by the small diameters of considered particles (about 1μm). The small separation distances between interacting bodies arises from the small value of Debye length which in turn is an effect of relatively large ionic strength of the solution at the vicinity of membrane surface.

The van der Waals and EDL forces between a spherical particle and a hollow cylinder may be computed by means of the exact formula given in work (Li and Chen, 2012). However, if the diameter of the cylinder is much bigger than the diameter of particle, the surface of the hollow fibre may be approached by means of a flat surface. Precisely speaking, from the results of the computations of Li

and Chen (2012) it appears that the error made with this approximation is of order of a few per cent when the particle to cylinder diameter ratio is about 1/10. In our simulations we apply the outer diameter of the membrane equal to 80 μm and the particle diameter up to 5 μm that gives the cylinder to membrane ratio equal to 1/16. Thus, in our computations we approach the hollow fibre membrane as a flat surface. The forces between the particle and the membrane are then obtained from Eqs. (1-3) putting $R_2 \to \infty$.

Where two particles or a particle and a membrane overlap, we assume they interact by means of elastic forces F_H described by Herz equation:

$$F_H = \frac{4}{3} E \sqrt{R|\delta|^3} \tag{4}$$

Finally, the equation of motion of a single particle with mass m has the form:

$$m \frac{dv}{dt} = \sum F \tag{5}$$

where $\sum F$ is the sum of all the forces described above (DLVO or Herz interactions) and the drag force given by:

$$F_d = 6\pi\mu R(u - \mathrm{v}_\perp) \tag{6}$$

The fluid flow is considered as radial, i.e.:

$$F_d = 6\pi\mu R(u - \mathrm{v}_\perp) \tag{7}$$

and the transversal as well as axial ones are equal to zero. While the concentration of solid particles is very small in most part of the whole numerical domain, the influence of these particles on flow field has been neglected. The only region where the concentration of particles is high enough to have a significant influence on the flow field is the fully formed filtration cake in the late stage of simulations. For this case we use the Carman-Kozeny formula to describe the fluid flow. This huge simplification is justified since for particles in the filtration cake the contact forces from other particles are much greater than those coming from fluid flow and thus detailed knowledge about the fluid flow is not necessary while analysing fouling and defouling processes.

Membrane oscillations are of two kinds. The first kind represents the oscillations of the whole membrane. We consider oscillations perpendicular to the fibre axis only. The oscillations in a parallel direction should not have a significant impact on fouling or defouling dynamics if we assume a smooth surface of the particles and the membrane. Indeed, the tangent force between the particle and the membrane is very low and may be neglected.

The second kind of oscillations causes deformation of the surface or bulging of the membrane. The effect of these oscillations was investigated in our previous work and will not be considered in the present paper.

3. RESULTS OF SIMULATIONS

Firstly, let us consider the dynamics of the fouling of a static hollow fibre membrane. Fig. 2 presents snapshots of membrane fouling during the filtration of a solution containing colloidal particles with a diameter equal to 3-5 μm. The outer radius of the hollow fibre is equal to 80 μm, the thickness of the membrane − 20 μm, and the initial flow rate of the solution is 0.75 cm^3/s per running meter of asingle hollow fibre length. That means water velocity at the surface of membrane is equal to 14.9 cm/s. The

value of Hamaker constant is equal to that for silica particles, i.e. 0.7×10^{-20} J (Rhodes, 2008). The Debye length is equal to 2.5 nm (value computed for the ionic strength equal to 0.01 mol/dm^3). The particle and surface potential are set as control parameters and in our simulation vary between -25 mV and +25 mV as these values are reported in literature to be the limits of applicability of classical DLVO theory (Ruggiero et al., 1999; Senger et al., 1994).

Equation (5) has been solved numerically for every particle by means of Verlet scheme. To speed up the simulations we apply two time steps. If the distance between a given particle and any other exceeds its diameter we assume that this particle interacts with the fluid only (the range of DLVO interactions is up to hundred nanometers) and thus we apply the bigger time step equal to 1.0 μs. When a particle approaches any other and the distance between them is less than its diameter we switch the value of time step to 0.01 μs. The distance travelled during the longer time step is then of the order of 0.15 μm. As a measure of fouling we choose two packing densities of the layer of width 10 μm around the fibre defined as a fraction of the space occupied by particles in this layer.

Let us consider the dynamics of the fouling of a hollow fibre membrane at the absence of the oscillations. Fig. 3 presents the dependence of packing density of the colloidal cake on time, for a case of separation of the suspension of silica particles having the diameter of 5 μm for various values of potentials of the particles and the membrane. It is a specific property that the low value of final packing density has a value of about 0.3, which suggests that deposited particles form a lattice close to a tetrahedral one. For particles with a diameter of about 3 μm, the final packing density is even smaller and equal to 0.2. From the comparison of these two values we recognize that the cake is rather porous. It could be observed in Fig. 4. This result is consistent with those described in (Henry et al., 2012).

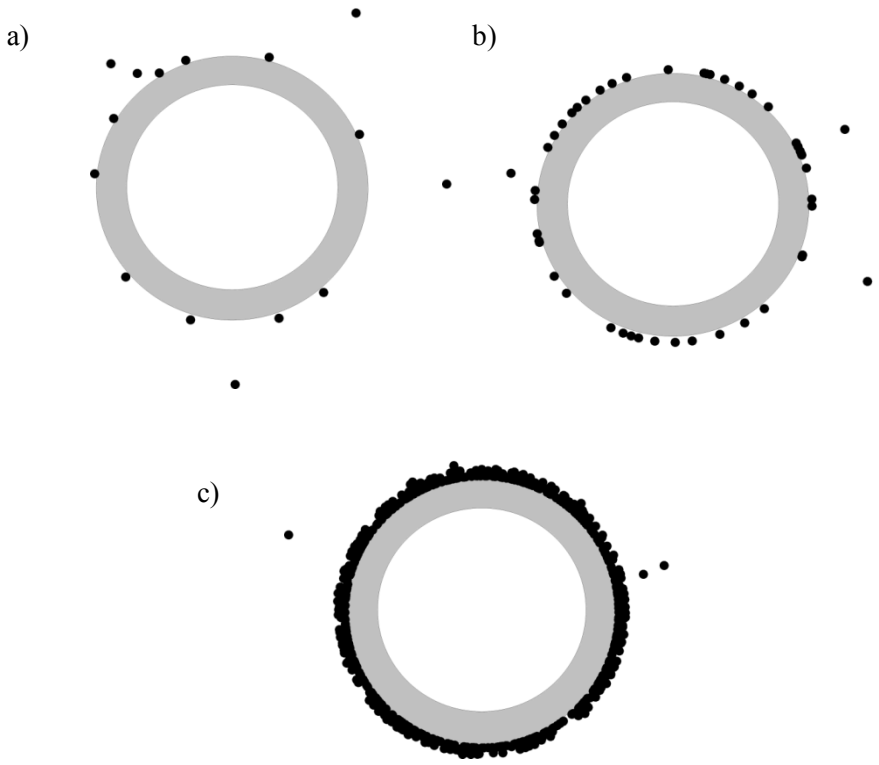

Fig. 2. Snapshots of the cake formation during membrane filtration of colloidal particles (diameter equal to 5 μm) on a hollow fibre membrane after a) 5 ms, b) 50 ms, c) 250 ms

Another conclusion drawn from Fig. 3 is that in the conditions used in the present study (especially in the range of particle and surface potentials between -25 mV and 25 mV with a given Hamaker constant), packing density appears to be independent of the magnitude or on the sign of the electric

potentials of particles and membranes. Further analyses show that the packing density of the filtration cake does not depend on Debye length either, and it is very weakly dependent on the value of Hamaker constant.

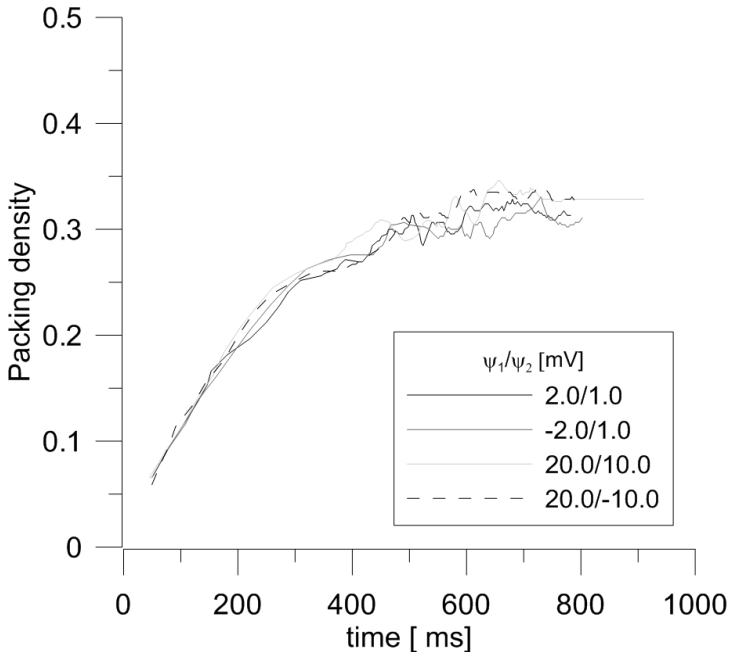

Fig. 3. Dependence of packing density on time for membrane filtration for various potentials of particles (ψ_1) and membrane (ψ_2). The diameter of particles is equal to 5 μm

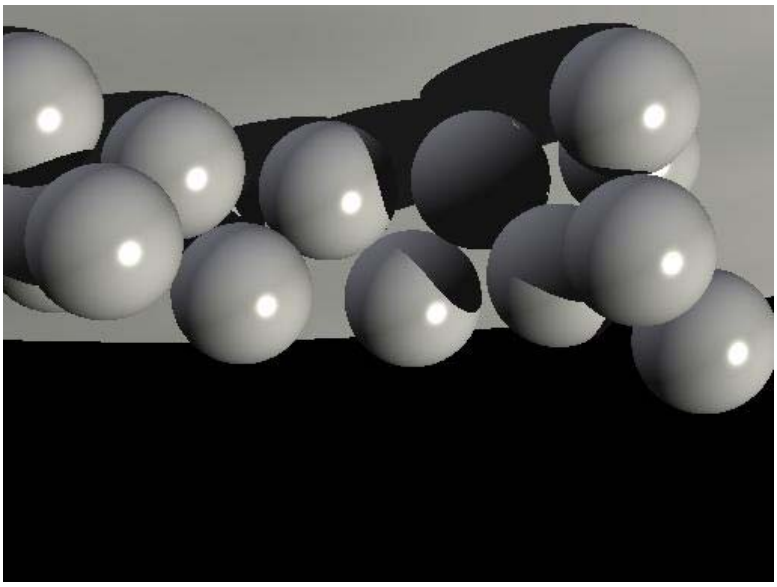

Fig. 4. Magnification of the fragment of filtration cake deposited on hollow fibre membrane. Note the high porosity of the cake

Now, let us turn to the case of the oscillating membrane. Fig. 5 presents the dependence of the packing density as a function of time for various frequencies of oscillations. The amplitude is equal to 10 μm in all cases. We observe that for a relatively low frequency (below 200 Hz) this dependence nearly overlaps with that obtained for the case without oscillations. For greater frequencies we recognise a decrease of the final value of the deposited mass.

Fig. 6 presents the dependence of the final (equilibrium) value of the packing density as a function of frequency of oscillations for two values of the flow rate. In both cases the same tendency is observed. At low values of frequency the final value of the packing density is nearly constant and equal to the value for a non-moving membrane. By increasing the frequency of oscillations we reach the value at which the increase of the packing density starts. As one can expect, this critical value of the frequency depends both on the flow rate and the amplitude of oscillations. According to Fig. 6 it appears that at a constant value of amplitude, the critical value of frequency decreases with the flow rate. By the extrapolation of the results we find that the value of frequency at which the final packing density is equal to zero (that means nearly no particle is permanently deposited on the membrane) is of the order of tens of kHz. This result would, of course, not have a significant practical meaning as such frequency of oscillations is completely unrealistic. However, it is clearly seen that for the frequencies up to 1 kHz the number of particles deposited on the membrane may be halved.

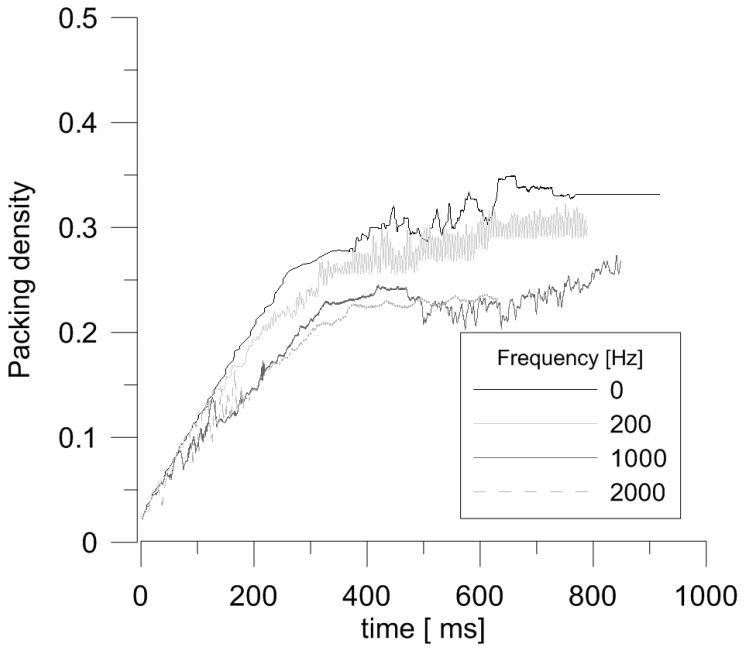

Fig. 5. Dependence of packing density on time for various values of hollow fibre oscillation frequency

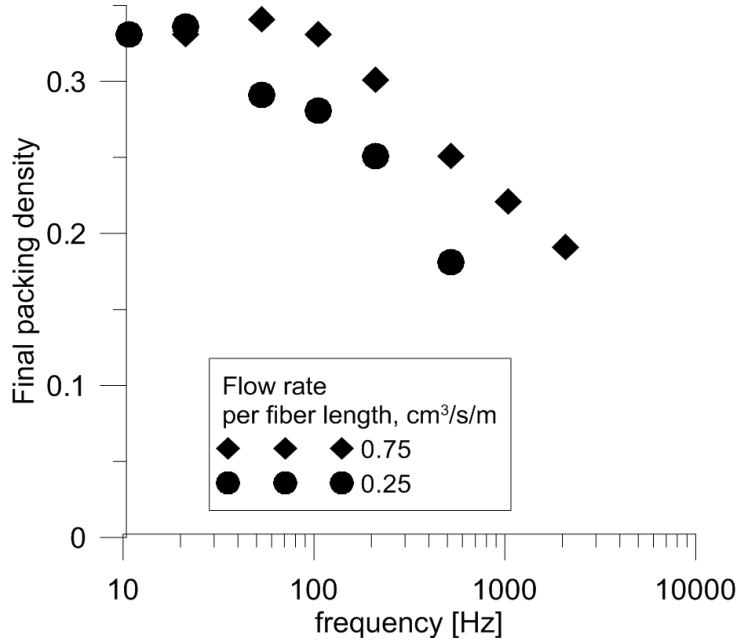

Fig. 6. Dependence of final packing density on the frequency of oscillations of a fibre for two values of low rate

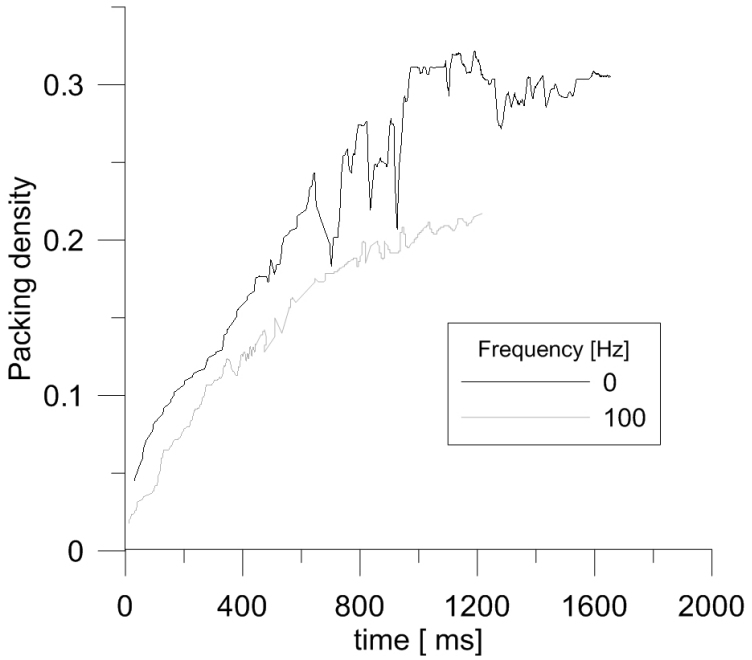

Fig. 7. Dependence of packing density on time for various values of hollow fibre oscillation frequency for the filtration of colloidal particles with a log-normal distributed diameter

Fig. 7 presents the same results for the case of non-uniform distribution of the diameter of particles which could cause membrane fouling. Here we assume that this distribution is given by means of log-normal distribution with the geometric mean equal to 5 μm and geometric standard deviation of 1 μm. We observe that in this case the packing density also decreases with oscillation frequency. This result allows us to expect that though our simulations are run for the particular values of parameters, their quantitative results are more general.

4. DISCUSSION AND CONCLUSIONS

We have analysed by means of numerical simulations the dynamics of colloidal fouling of hollow fibre membranes and their defouling as an effect of membrane oscillations. The DLVO model for the interactions between colloidal particles as well as the particles and the membrane has been used. The contact forces between the overlapping particles are computed by means of the Herz model.

We have observed that the application of membrane oscillations leads to a decrease of the packing density of the filtration cake. The value of the frequency above, the effect of which is clearly visible, depends on the amplitude of oscillations and the flow rate of water. It also depends on the distribution of the diameter of particles. The last parameter influences not only the dynamics of the packing density, but also its final equilibrium value.

On the other hand, these quantities are nearly independent on the physicochemical parameters of particles, the membrane and the solution, such as the Hamaker constant, the electric potential of both particles and the membrane, and Debye length.

In our investigations we have not taken into account many parameters which may occur in a real system. These are: the gradient of electrolyte concentration, the fluctuation of water velocity around the membrane (which is the result of the fluctuation of the packing density of the filtration cake) and many others. The influence of these parameters on membrane fouling will be the subject of forthcoming studies.

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SYMBOLS

A_H	Hamaker constant, J
E	Young modulus of particle material, Pa
F_d	drag force, µN
F_{dl}	electric double-layer force, µN
F_H	Herzian (elastic) force, µN
F_{vdW}	van der Waals force, µN
I	ionic strength, mol/dm^3
k_B	Boltzmann constant
m	mass of a particle, µg
Q	flow rate per unit length of hollow fibre, cm^3/s/m
R, R_1, R_2	radius of a particle, µm
$\vec{r}$	radial coordinate, µm
T	temperature, K
t	time, ms
u	fluid velocity, m/s
v	particle velocity, m/s
$v_\perp$	component of particle velocity perpendicular to the membrane, m/s
x, y, z	particle coordinates, µm

Greek symbols

δ	particle-membrane or particle-particle distance, µm
ε_0	electric permittivity of free space, H/m
ε_r	relative permittivity (of water)
κ	reciprocal of Debye length, µm^{-1}
μ	dynamic viscosity of a fluid (water), mPa·s
ψ_1, ψ_2	electric potential of particle or membrane surface, mV

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