COMPUTATIONAL ANALYSIS OF DISPLACEMENT OF PARTICLES WITH GIVEN SIZE ON THE NONSTATIONARY BULGING MEMBRANE AS A THEORETICAL MODEL OF MEMBRANE FOULING

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A simple model of behaviour of a single particle on the bulging membrane was presented. As a result of numerical solution of a motion equation the influence of the amplitude and frequency of bulging as well as the particle size on particle behaviour, especially its downstream velocity was investigated. It was found that the bulging of a membrane may increase the mean velocity of a particle or reinforce its diffusive behaviour, dependeing on the permeation velocity. The obtained results may help to design new production methods of highly fouling-resistant membranes.

Keywords: membrane; fouling; membrane cleaning; numerical modelling

1. INTRODUCTION

Rapid developments in membrane filtration technology have resulted in an increased number of industrial applications including dairy, beverages, brewery, food, pharmaceutical, wastewater, and desalination process industries. For the long and efficient work of membrane filtration it is important to adopt certain methods or techniques to reduce membrane fouling. The fouling potential always exists in the process of 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 is due to the deposition of small colloidal particles on inner walls of membrane pores: standard blocking, blocking of membrane pore opening, complete blocking and buildup of particles in the form of a cake layer on membrane surface. Fouling due to blocking and cake formation is assumed to be the predominant mechanism in UF and MF filtration, whereas in RO operation cake filtration is the predominant effect for the reduction of the permeate flux through the membrane (Boerlage et al., 2002; Mousa and Al-Hitmi, 2007).

Filtration-induced macrosolute or particle deposition is often reversible fouling. There are several methods to reduce this type of fouling for a range of different applications: addition of coagulants for the formation of layer particles, which are easily swept off the membrane surface (Al-Malack and Anderson, 1996), use of dispersed phase to disrupt concentration polarisation (Paravatjar, 1996), introduction of flow instability by low-frequency pressure and velocity pulsing (Zahka and Leary, 1985), cross-flushing and backwashing (Kroner et al., 1984), and many others.

In water-treatment applications involving colloids, microbes and un-dissolved hydrocarbons, foulants are often adhesive and cause irreversible fouling due to hydrophobic interaction, hydrogen bounding, van der Waals attractions, extracellular macromolecular interactions and other effects.

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Reduction or elimination of adhesive fouling requires more sophisticated methods, which involve the local effects of interaction between particles and the membrane surface. These methods include: 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 for the reduction of adhesive fouling on a membrane surface is the excitation of membrane surface through local surface pulsing. It could be achieved by the interaction of electromagnetic particles immersed under a membrane surface with an external electromagnetic field.

The aim of this paper is to present the introductory theoretical consideration of such effects as a potential solution against membrane fouling. A simple model of interaction between a particle and a membrane was developed and the results of modelling for a single particle as well as for a few particles (early stage of fouling) will be shown. The final result of our simulation is determination of conditions at which particle removal from a membrane surface is most efficient.

2. THE MODEL

Let a spherical rigid particle of radius R interact with a flat membrane and a surrounding fluid. There are four forces that act on the particle (Henry et al., 2012; Quasirani and Samhaber, 2011): an adhesive force F_a , a drag force due to cross-flow F_d , a lift force F_l and a Brownian force F_b . These forces are depicted in Fig. 1.





The potential of adhesive van der Waals interactions between a particle and membrane has a form (Parsegian 2006):

$$U_{pm}^{vW} = -\frac{A_H}{6} \left[\frac{R}{\delta} + \frac{R}{2R + \delta} + \ln \frac{\delta}{2R + \delta} \right]$$
(1)

where δ is a distance between surfaces of particle and membrane and A_H is a Hamaker constant. The force acting on particle is thus given as:

$$F_{vW} = -\frac{\partial U_{pm}^{vW}}{\partial \delta} = -\frac{A_H}{6} \left[\frac{R - \delta}{\delta^2} - \frac{3R + \delta}{(2R + \delta)^2} \right]$$
(2)

Another force acting between a particle and a membrane comes from the presence of a double electric layer. The potential energy of this force has a form (Senger et al., 1994):

$$U_{pm}^{dl} = \pi \varepsilon_r \varepsilon_0 R \psi_s^2 \ln[1 - \exp(-2\kappa\delta)]$$
(3)

where κ denotes the reciprocal of Debye length and ψ_s^2 is an electric potential of a particle.

The force coming from the double layer is again a derivative of a potential:

$$F_{dl} = -\frac{\partial U_{pm}^{dl}}{\partial \delta} = -2\pi\varepsilon_r \varepsilon_0 R \psi_s^2 \frac{\exp(-2\kappa\delta)}{\left[1 - \exp(-2\kappa\delta)\right]}$$
(4)

The drag force due to fluid flow is given by means of a simple Stokes formula:

$$F_d = 6\pi\mu R u \tag{5}$$

where u is the instantaneous streamwise velocity difference between particle and fluid defined at the particle center and μ is the fluid viscosity.

The lift force is a result of a vertical gradient of fluid velocity. It acts in the direction of this gradient. So, in a case of a particle deposited on a membrane this force facilitates particle detachment as can be seen it has been marked in Fig. 1. For a case of a constant velocity gradient near a the wall, this force has a form (Wang et al., 1997):

$$F_{l} = -6.46\,\mu R^{2}u\,\mathrm{sgn}(G)\sqrt{\frac{|G|}{v}}\tag{6}$$

where G is a velocity gradient.

The last force acting on a particle is random force originating in collisions between particles and molecules of a fluid called also known as a Brownian force. This force manifests itself in random Brownian displacement of a particle. Usually it is assumed that the random force acting on a particle has a form:

$$F_B = \frac{4\pi}{3} \rho_s R^3 A(t) \tag{7}$$

where A(t) is the Brownian random force exerted on per unit mass of the particle, and it follows a Gaussian distribution. The Brownian displacement within a time period Δt is then expressed as a random Gaussian value with the mean value equal to zero and the variance equal to $\sqrt{2D_0\Delta t}$.

It is, however clear that in a real system the presence of a membrane disturbs the random walk of a particle (Lin et al., 2000). Thus, the diffusion coefficient depends on the distance between a particle and a membrane. This effect, usually neglected, is expected to play a significant role in our study when a particle spends a long period of time in the vicinity of a membrane.

The dependence of a diffusion coefficient on the distance between a particle and a membrane is rather poorly understood (Lin et al., 2000). In the present work we will use a simple model presented in (Happel and Brenner, 1983). According to this model, diffusion coefficients for distinguished directions are as follows:

$$D_{\parallel} = \frac{k_B T}{6\pi\mu\lambda_{\parallel}a} = \lambda_{\parallel}^{-1}D_0 \tag{8}$$

$$D_{\perp} = \frac{k_B T}{6\pi\mu\lambda_{\perp}a} = \lambda_{\perp}^{-1}D_0 \tag{9}$$

111

where D_0 denotes the diffusion coefficient far from membrane surface and:

$$\lambda_{\parallel}^{-1} = \frac{D_{\parallel}}{D_0} \cong 1 - \frac{9}{16} \left(\frac{R}{z}\right) + O\left(\frac{R}{z}\right)^3$$
(10)

$$\lambda_{\perp}^{-1} = \frac{D_{\perp}}{D_0} \cong 1 - \frac{9}{8} \left(\frac{R}{z}\right) + O\left(\frac{R}{z}\right)^3 \tag{11}$$

In the case of an oscillating membrane we assume that the oscillating bulge has a shape of a hemisphere as shown in Fig. 2. This shape for any asperities of membranes is widely used in many theoretical considerations of particle-membrane interactions (Henry et al., 2011; Henry et al., 2012). The radius of this hemisphere depends on time in the following form:

$$R_{H}(t) = R_{\max}\left(1 - \cos\left(\frac{2\pi t}{T}\right)\right)$$
(12)

i.e. it changes harmonically between zero and R_{max} with the period T.

The van der Waals and double layer interactions between a particle and a hemispheric bulge are assumed to have the same form as the interaction between two spheres i.e. van der Waals potentials are given by the formula (Senger et al., 1994):

$$U_{pp}^{\nu W} = -\frac{A_H}{3} \left[\frac{R_1 R_2}{z^2 - (R_1 + R_2)^2} + \frac{R_1 R_2}{z^2 - (R_1 - R_2)^2} + \frac{1}{2} \ln \frac{z^2 - (R_1 + R_2)^2}{z^2 - (R_1 - R_2)^2} \right]$$
(13)

where $z = R_1 + R_2 + \delta$.

The van der Waals force is then given as a spatial derivative:

$$F'_{vW} = -\frac{\partial U^{vW}_{pp}}{\partial \delta}$$
(14)

The double layer potential and force for the interaction between sphere and hemisphere have the same form as (3) and (4) with the particle radius *R* replaced by $\frac{R_1R_2}{R_1 + R_2}$ (Ruggiero et al., 1999).



Fig. 2. Forces acting on a particle deposited on a membrane in the vicinity of a bulge

Interactions between various particles deposited on a membrane have the same form as interactions between a single particle and a bulge while we assume that all the particles are spherical.

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

Computational analysis of displacement of particles with given size on the nonstationary bulging membrane...

$$m\frac{dv}{dt} = \sum F \tag{15}$$

where ΣF is the sum of all the forces described above. This equation may be simplified if we assume a relatively high value of fluid viscosity which takes place especially in membrane filtration of liquids. Then (12) is approximated by means of the first-order Langevin equation:

$$\frac{dx}{dt} = \frac{1}{6\pi\mu R} \sum_{\text{det}} F + \sigma\xi$$
(16)

with the Gaussian random noise value ξ . In (16) $\sum_{det} F$ denotes the sum of all deterministic forces

acting on a particle, i.e. the sum of forces given by Eqs. (2), (4-6) and (13).

This Equation (16) may be solved numerically by means of the simple Euler scheme (Mannella and Palleschi, 1989):

$$x(t + \Delta t) = x(t) + \left(\frac{1}{\mu} \sum_{\det} F(t)\right) \Delta t + \sigma(t) \xi \sqrt{\Delta t}$$
(17)

3. RESULTS OF SIMULATIONS

To investigate the influence of membrane oscillations on particle dynamics, we analysed the following model. A spherical rigid particle is originally deposited on a membrane. We analysed particles having radius in a range of 0.2-4.0 μ m. The interactions of such a single particle with a membrane are described by means of forces (1-4). We assume a Hamaker constant equal to 10^{-20} J which is a typical value for common materials of membranes and particles (Parsegian, 2006) and the electric potential equal to 10 mV. The fluid surrounding a particle is water having dynamic viscosity of 890 mPas.

As to fluid flow, we set the mean shear rate as equal to 10^{-2} s^{-1} and the velocity of permeating water as equal to 10^{-5}m/s (which means the productivity of 100 cm³/s of filtrate from the membrane having a surface equal to 10 m²). We assume the initial concentration of particles on the membrane as equal to 12500 particles/mm².

Let us first investigate the dynamics of a single particle on a stationary membrane. In Fig. 3 we present sample trajectories of single particles of various radii. It is to be recognised that the bigger particle having a radius equal to 4 μ m slides in the direction of water flow with a nearly constant velocity. The effect of a random Brownian force is very poorly visible for this particle. On the other hand, a small particle with a radius equal to 0.2 μ m displays a random motion with a very weak drift. A conclusion can be drawn that the small particles are the hardest to remove. By analysing a few hundred trajectories of particles with various radii did not observe any case of particle detachment. Indeed, the attractive force coming from membrane together with a force coming from permeating flow exceeds the lift force. Taking this result into account we may conclude that the main way of fouling prevention in such a system is to force the rolling or sliding of the particles on a membrane.

Let us now turn to the considerations of the efficiency of particles removal from a membrane. While the residence time of a particle in a membrane module is inversely proportional to velocity and, following that, to the downstream displacement of the particle at a given time period - in our further considerations shall use the distribution of downstream displacement to describe the efficiency of particles removal from a membrane. The mean particle displacement provides information about the mean tangent velocity of a particle and the standard deviation informs about the deviation of this particle.



Fig. 3. Trajectories of particles of various radii deposited on a membrane surface

Fig. 4 presents distributions of the mean displacement of particles in the direction parallel to a fluid flow. It can be seen that the displacement is well described by means of a Gaussian distribution. The mean standard deviation of a displacement is nearly the same for all three radii of particles. That allows us to conclude that the main influence on particle motion comes from particle-membrane interactions and fluid flow but not from diffusion while it is very weak in the presence of a membrane.

Now, let us take into consideration oscillating particles embedded in a membrane. The new parameters describing a membrane with these particles are: the amplitude of bulging (dependent on an embedded particle radius), the frequency of oscillations and surface density of particles.



Fig. 4. Relative frequency of downstream displacement (after 5 s) of particles for three various radii

Fig. 5 we presents a distribution of displacement of particles with a radius of 4 μ m (Fig. 5a) and 1 μ m (Fig. 5b) at various amplitudes of shaking. Other parameters are as follows: the frequency of oscillations equal to 50 μ s and the surface density of embedded particles equal to 2500 mm⁻². It can be observed that the amplitude of shaking influences the dispersion of this distribution. We may say that the great amplitude of shaking improves the diffusion of particles and prevents the formation of agglomerates deposited. This tendency may have its part in membrane fouling prevention. Indeed, as

stated above, the main mechanism of deposited particles removal is to force particles roll or slide on membrane surface. This effect is easier achieved in the case of a single particle than in the case of an aggregate when adhesion forces are weaker for a single particle.

Comparing the plots presented in Fig. 5a and 5b it can be seen that in the presence of membrane oscillations distributions become asymmetric. It is interesting that for $R = 4 \mu m$ the distribution has a positive skewness while for $R = 1 \mu m$ – negative skewness. The origin of such a behavior is the presence of an oscillating particle, which locally disturbs the motion of single particles. Most particles spend a lot of time in the vicinity of these points. The maximum of distribution is "pinned" for a long time in the vicinity of an oscillating particle. This effect is presented in Fig. 6. For various time moments negative (Fig. 6a) or positive skewness of the distribution for the same conditions was observed.



Fig. 5. Relative frequency of downstream displacement (after 5 s) of (a) 4 µm and (b) 1 µm particles for static membrane and two amplitudes of oscillations

Fig. 7 presents the distribution of a displacement of a particle ($R = 1 \mu m$) for various values of oscillation period. It can be noted that a decrease of oscillation period causes an increase of the mean standard deviation of a distribution. Physically, that means that fast oscillations cause a "diffusion-like" motion, which is the result of common Brownian motion and the interaction (attraction or repulsion) with a bulge of varying radii. This effect denotes that at an early state of microfiltration particles do not form aggregates deposited on a membrane but they preferentially exist rather as single deposited particles which could be easier to remove. This is one of the ways to prevent membrane fouling.

Fig. 8 presents the distribution of a displacement of particles in the presence of various values of permeation velocity. It can be noted that all these distributions have nearly the same value of standard deviation. However, they differ in the position of a maximum and consequently in the mean particle velocity. The result of differences of the values of the mean velocity of particles is the fact that at low permeation velocity a temporal detachment of particles can occur and, in consequence, a decrease of attractive forces between a particle and a membrane.



Fig. 6. Relative frequency of a downstream displacement of 4 µm particles after (a) 4.75 s and (b) 5.0 s



Fig. 7. Relative frequency of downstream displacement (after 5 s) of 1 µm particles for static membrane and two values of a period of oscillations



Fig. 8. Relative frequency of downstream displacement (after 5 s) of 1 µm particles for three different permeation velocities

Computational analysis of displacement of particles with given size on the nonstationary bulging membrane...

4. DISCUSSION AND CONCLUSIONS

A simple mathematical model of a rigid particle interacting with a static or bulging membrane was built. This model, however, takes into account all the forces that act on a particle.

The main results of the simulations are as follows:

- The main parameters controlling the residence time of a particle at the membrane are: amplitude and frequency of bulging and permeation velocity; there is no significant influence of a density of particles embedded in a membrane on fouling prevention.
- At high permeation velocities, typical for membrane processes, the bulging of a membrane does not influence the velocity of a particle sliding on a membrane. However, a great influence of a displacement of a particle on the mean deviation was observed. This method of "improving diffusion" may play a significant role in the prevention of agglomerate formation and consequently membrane fouling.
- The effect of an increase of the mean deviation of a particle is stronger at high amplitudes and frequencies of bulging.
- At low permeation velocities bulging is conductive to the particle detachment from a membrane and thus increases the particle velocity.

Concluding, there are two main ways by which bulging prevents the membrane fouling. The first is to force deposited particles into a motion which decreases the risk of huge agglomerate formation. This mechanism is dominant at high permeation velocities, i.e. during the normal work of a membrane module. The second mechanism is the detachment of a particle as a result of collision with oscillating bulge. This mechanism may play a significant role at low permeation velocities, e.g. when a membrane is out of operation.

Taking the above facts into account a conclusion can be drawn that the membrane bulging prevents or slows down the fouling. Bulging may be forced e.g. by the embedding of magnetic nanoparticle in a membrane and placing this membrane into a nonstationary magnetic field as it was proposed in the Introduction. Tests on this subject are now in progress.

The considered model assumes that the surface of a membrane is ideally smooth (except of the bulges) so it has no asperities or pores. This assumption is a rather gross simplification, which limits the use of the model, especially for finer particles. Another simplification is that a membrane is ideally rigid (which may not be true especially at high-pressure differences on both sides of a membrane. However, the proposed model allows us to give at least a rough description of the mechanisms of particle detachment from a bulging membrane.

This work was supported by the grant Era-Net/MNT/NSFM/1/2011.

SYMBOLS

A(t)	random component of Brownian motion
A_H	Hamaker constant, J
$D_{ heta}$	Diffusion coefficient, m ² /s
$D_{ }$, D_{\perp}	Diffusion coefficients for a motion at the vicinity of the wall for parallel and perpendicular
	direction respectively, m ² /s
F_a	adhesive force, $F_a = F_{dl} + F_{vW}$, N
F_b	Brownian force, N
F_d	drag force, N

F_{dl}	electric double-layer force, N
F_{vW}	van der Waals force, N
G	velocity gradient, s ⁻¹
k_B	Boltzmann constant, J/K
т	mass of a particle, µg
R, R_1, R_2	radius of a particle, μm
Т	temperature, K
t	time, ms
Δt	time step in numerical schemes, ms
v	particle velocity, m/s
<i>x, y, z</i>	particle coordinates, µm
Greek symbols	
δ	particle-membrane or particle-particle distance, µm
\mathcal{E}_{0}	electric permittivity of free space, H/m
$\mathcal{E}_{ ho}$	relative permittivity (of water)
ĸ	reciprocal of Debye length, μm^{-1}
$\lambda_{\parallel},~\lambda_{\perp}$	coefficients of decreasing of diffusion in the vicinity of a wall
μ	dynamic viscosity of a fluid (water), mPas
ν	kinematic viscosity of a fluid (water), m ² /s
$ ho_{\sigma}$	density of a particle, kg/m ³
σ	noise amplitude (in numerical schemes)
ξ	random number (in numerical schemes)
ψ_{σ}	electric potential of particle/membrane surface, mV

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Received 06 November 2012 Received in revised form 18 January 2013 Accepted 21 January 2013