Iranian Food Science and Technology Research Journal
Vol. 8, No. 1, 2012, p. 91-102

نشریه پژوهشهای علوم و صنایع غذایی ایران
جلد ۸، شماره ۱، بهار ۱۳۹۱، ص. ۱۰۲-۹۱

Modeling and Simulation of Limiting Flux in Membrane Filtration of Skimmed Milk

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Received: 08-08-2010
Accepted: 16-11-2011

Abstract

In this research, modeling of limiting flux in membrane filtration of skimmed milk as well as the results of simulation by MATLAB is presented. The presented model is actually a force balance approach and consists of drag, gravitational and lift forces, and forces between colloidal particles (electrostatic repulsive and van der Waals attractive forces). To consider the back diffusion phenomenon in modeling, the Brownian and shear-induced diffusions have also been considered.To evaluate the ability of the presented model in this study for prediction of limiting flux of membrane filtration of skimmed milk, comparisons between the results of the present model and experimental data and the results of the model presented by Samuelsson et al. have also been made in this research. By surveying some statistical parameters obtained from these comparisons, it was found that in order to predict the limiting flux in membrane filtration of skimmed milk, interaction forces between particles should be considered.

Keywords: limiting flux; skimmed milk; van der Waals attraction; electrostatic repulsion; membrane filtration

Introduction

The application of membrane filtration process in different fields of industry is increasingly developing. One of the uses of this process in food industry is the separation of colloidal particles from solutions. A significant and inevitable phenomenon in the separation of colloidal particles from solutions by membrane is membrane fouling which causes the membrane surface to be covered with colloidal particles, so the membrane permeate flux is decreased. In this situation, if the transmembrane pressure is increased, the permeate flux will increase as well. However, with increasing transmembrane pressure, the amount of colloidal particles sedimentation on the membrane surface is also increased. In this way the filtration system reaches a point where the permeate flux is not increased with the increase in transmembrane pressure, and the whole surface of the membrane is covered with a dense layer of particles. In this situation, the formed layer on the membrane is called the cake (gel) layer and the membrane permeate flux is called limiting flux. At the limiting flux level, the membrane system experiences limitation and it is possible for the flux to be reduced to the extent that the membrane separation cannot be justified economically. On the other hand, since this flux, is independent of transmembrane pressure, any increase in the transmembrane pressure does not cause a remarkable change in it and therefore with the awareness of the amount of limiting flux, the increase in the transmembrane pressure is not necessary after reaching the limiting flux. Therefore, the prediction of limiting flux has a significant importance.

The existence of limiting flux was reported as early as 1970. Many studies have been done to examine and justify this phenomenon, and the effective parameters on it have been the subject of many researches. Many factors including temperature, colloidal solution properties, the shape and geometry of membrane modules and hydrodynamic parameters affect the limiting flux (Samuelsson et al., 1997). Limiting flux is also dependent on the crossflow velocity and the higher the velocity, the more limiting flux (De et al., 1999).

Many different models and theories have been suggested to explain the limiting flux. Belfort et al. (1994) examined the limiting flux by describing the concentration polarization layer and the particle behavior near the membrane surface.

In some researches it has been assumed that a cake layer is formed on the membrane surface and the particles flow toward the membrane because of convection and are moved away from the membrane surface by a phenomenon called back diffusion. To predict the limiting flux by considering the back diffusion phenomenon, four models were examined by Samuelsson et al. for microfiltration of skimmed milk. These four models include Brownian diffusion, shear-induced diffusion, inertial lift and surface transport. They showed that the shear-induced model predicts the limiting flux value near the experimental amounts and if the Brownian diffusion model is combined with shear-induced diffusion, a better estimation for limiting flux can be obtained (Samuelsson et al., 1997). Also, Huisman et al. (1999) and Yoon et al. (1999) considered

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Brownian diffusion and shear induced diffusion to consider the back diffusion phenomenon.

Many studies have shown that the interactions between particles have a remarkable effect on filtration flux. Yoon et al. (1999) presented a three dimensional simulation to investigate the reduction of flux with time in microfiltration of iron oxide particles with different sizes. They focused on the particle transport mechanism as a function of particle size to consider the particles sedimentation rate. They revealed that the charge repulsion was the most important mechanism of cake formation reduction in the conditions of their research. Huisamn et al. (1999) showed that the effect of these interactions on the filtration flux could be defined by introducing a new diffusion coefficient for particles interactions. They calculated the limiting flux in microfiltration of the particles suspension by a numerical solution of governing convection-diffusion equation in the concentration polarization conditions. They only considered electrostatic repulsive force and ignored the gravity and van der Waals attractive forces. The result of this modeling showed that the effect of particle surface potential on permeate flux was numerically less than the predicted amount.

Bacchin et al. (2006) developed a complicated numerical simulation of momentum transfer and mass transport using the CFD modeling to describe the latex particles accumulation on the porous surfaces during the membrane filtration in crossflow condition. The limiting flux was not examined in the research and only the physical and chemical effects resulted from the colloidal surface interactions on particle accumulation near the membrane surface were investigated. Also, Bacchin et al. (1995) presented a theoretical model for the description of colloidal sedimentation on the membrane surface with consideration of surface interactions. In this model, a mass transport equation relates the sedimentation rate to the physical and chemical properties of the suspension.

To calculate the effect of particle size, the distribution of the particle size and surface potential on the structure of cake, Fu et al. (1998) developed a force balance model. The model suggests that the steady cake is formed in the low surface potentials. In this research, only the interaction forces and diffusion forces were taken into consideration to study the effect of particle size and charge on the structure of the cake.

Blake et al. (1992) presented a general model of frictional force balance for a latex particle on the cake surface to predict the steady filtration flux. They assumed that if the proportion of net axial force to the net vertical force exceeds friction coefficient, sedimentation does not occur. The presented model is too general and does not involve the effects of repulsive electrostatic force and Brownian diffusion on the limiting flux. However, the effect of van der Waals attractive force has been included.

As it was mentioned before, in the limiting flux conditions, the surface of the membrane is covered by a layer of particles and therefore the properties of the membrane cannot have any remarkable effect on the limiting flux (Tang et al., 2009).

A lot of research has also been done on the effective parameters on limiting flux in colloidal solution filtration in the laboratory scale. In a research on milk casein micelles, a linear relationship between the limiting flux and pH was observed. Since the zeta potential has linear relationship to pH, it is possible to see a linear relationship between the limiting flux and the zeta potential of casein micelles. Thus the less the zeta potential of casein micelle is, the less the limiting flux will be. It was also observed that the limiting flux of the skimmed milk was dependent on the interparticle electrostatic repulsive force to the extent that with the increase in interparticle repulsion, the limiting flux was increased (Baudry et al., 2005).

Although, many researchers have studied cases such as accumulation of particles near the membrane, the reduction of flux with time and the cake structure, however, the limiting flux and the effect of different factors on it have not been studied directly. In spite of developments in different models and their good general agreement with experimental data, different models give contradictory interpretations of experimental results. This shows that the mechanism of reaching the limiting flux in a membrane filtration system with crossflow condition has not been correctly understood and the agreement with experimental data does not necessarily accept the existence of hypotheses in these models.

In the present research, the limiting flux has been modeled by the use of a comprehensive model of force balancing and according to the majority of governing forces on the movement of particles including electrostatic repulsive, van der Waals attractive, gravitational and lift forces. Two factors of Brownian diffusion and shear-induced diffusion have been taken into consideration for the effect of back diffusion phenomenon too. It is worthy of mention that so far no similar model has been proposed to include all of the above mentioned cases for the calculation of limiting flux for the membrane filtration of skimmed milk.

Material and methods

Modeling

A tubular membrane has been considered for modeling with co-ordinates as to Fig. 1.

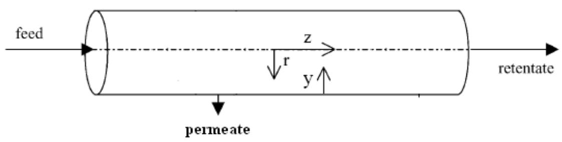

Fig.1 Co-ordinates in crossflow filtration in a tubular membrane

The force balance approach is considered in this modeling. To study the forces, the given particle has been noted in the concentration polarization layer and close to the surface of the cake. In formation of fouling, the imposed forces on the particle in the vertical direction to the membrane surface have greater effect on the particle than the forces parallel to the surface. Therefore, to study the limiting flux, the vertical forces on the particle that is in the concentration polarization layer and near the cake surface are considered. In the limiting flux conditions the summation of these forces is equal to zero:

$$\sum F_r = 0 \quad (1)$$

In general, the mentioned vertical forces are as follows:

1. Drag force that draws the particle toward the membrane
2. Gravitational and buoyancy forces (because of gravitational acceleration on the particle and the difference in density between the particle and solvent)
3. The lift force which is exerted on the particle from the moving fluid and its orientation is toward the bulk
4. The electrostatic repulsive force between colloidal particles which repels the particle from the membrane
5. The attractive van der Waals force which draws the particle toward the deposited particles on the membrane surface (cake)

In addition to the above mentioned forces, some other forces such as the hydrophilic and hydrophobic forces have been mentioned in some articles which won't be dealt with here because of their minor effects and their restriction to special cases. So it can be written:

$$\sum F_r = 0 \longrightarrow F_{lift} + F_{gravity} + F_{vdW,pp} + F_{EDL,pp} + F_{drag} = 0 \quad (2)$$

Where F_{lift} is the lift force, $F_{gravity}$ is the force resulted from weight and floating of the particle in the solvent, $F_{vdW,pp}$ is the van der Waals attractive force, $F_{EDL,pp}$ is the electrostatic repulsive force between the particles, and F_{drag} is the drag force.

For lift force, the following equation is considered (Vyas et al., 2001):

$$F_{lift} = 0.761\frac{\tau_w^{1.5} d_p^3 \rho^{0.5}}{\mu} \quad (3)$$

Where τ_w is the wall shear stress, d_p is the diameter of the particle, and ρ and μ are the density and viscosity of solvent, respectively.

In addition, the following equation exists for gravitational force (Vyas et al., 2001):

$$F_{gravity} = \frac{1}{6}\pi d_p^3(\rho_p - \rho_f)g \quad (4)$$

Where g represents the acceleration of gravity, ρ_p is the particle density and ρ_f is the density of fluid.

One of the most important parameters affecting the limiting flux in the separation of colloidal suspensions is the interaction forces of colloidal particles. The interactions between the colloidal particles are the results of different forces which the most important ones are van der Waals attraction and electrostatic repulsion (Shaw, 1980).

Many methods of calculating the van der Waals interaction forces have been reported in different researches. For two spheres with equal radius, the following equation for van der Waals attraction exists (Liang et al., 2007):

$$V_A(h) = -\frac{A_H}{6}\left\{\frac{2a^2}{h^2+4ah} + \frac{2a^2}{(h+2a)^2} + \ln\left[1 - \frac{4a^2}{(h+2a)^2}\right]\right\} \quad (5)$$

Where a is the particle radius, h is the distance between the particles and A_H is the Hamaker constant.

Using the energy equation in the following manner, it is possible to obtain the attraction force between two colloidal particles (Liang et al., 2007):

$$F_A = -\frac{\partial V_A}{\partial h} \quad (6)$$

Consequently, combining equations (5) and (6), equation (7) is obtained for the van der Waals attraction force between two particles:

$$F_{vdW,pp} = \frac{A_H}{6}\left\{\frac{-2a^2(2h+4a)}{(h^2+4ah)^2} + \frac{-4a^2}{(h+2a)^2} + \frac{8a^2}{(h+2a)\left[(h+2a)^2 - 4a^2\right]}\right\} \quad (7)$$

Considering the variety and limitation of the existing parameters in the skimmed milk, in the present research, the following equation which is the result of Derjaguin approximation method have been used for the repulsive electrostatic energy between two spherical particles (Shaw, 1980).

$$V_{EDL,pp} = 2\pi\varepsilon_r\varepsilon_0 a\psi_0^2 \ln(1+\exp(-\kappa h)) \quad (8)$$

In this equation ε_r is the relative dielectric constant, ε_0 is Vacuum permittivity (8.8542×10^{-12} C^2 N^{-1} m^{-1}), ψ_0 is surface potential. κ *is* the Debye length with the following equation (Kühnl et al., 2010):

$$\kappa = \sqrt{\frac{2N_A e^2 I}{\varepsilon_r\varepsilon_0 k_B T}} \quad (9)$$

Where N_A is the Avogadro number, e is the electron charge (1.602×10^{-19} C). I is the ionic strength, k_B is the Boltzmann constant (1.38×10^{-23} JK^{-1}) and T is the Kelvin temperature (K).

The electrostatic force can also be estimated through the energy equation (Eq. 6). Therefore, the following equation exists for the electrostatic force between the particles of two spheres:

$$F_{EDL,pp} = \frac{2\pi a\varepsilon_0\varepsilon_r\psi_0^2\kappa\exp(-\kappa h)}{1+\exp(-\kappa h)} \quad (10)$$

The following equation exists for the drag force (Huisman et al., 1999).

$$F_{drag} = 6\pi\mu a(v - v_p)(1-\varphi)^{-6.55} \quad (11)$$

In this equation, v is the fluid velocity, v_p is the particle velocity in the vertical direction toward the membrane surface and φ is the volume fraction of the particle.

The fluid velocity in vertical direction to the membrane represents limiting flux which is the main unknown. Assuming the particle close enough to the cake surface in steady state condition, particle acceleration can be neglected. Therefore, particle vertical velocity is considered constant. This velocity can be determined by the following equation (Huisman et al., 1999):

$$v_p \varphi = D \frac{\partial \varphi}{\partial r} \quad (12)$$

It is sufficient to integrate the above equation to obtain the amount of v_p:

$$v_p dr = \frac{D}{\varphi} d\varphi \xrightarrow{\int} -\int_{R_0}^{(R_0-\delta)} v_p dr = \int_{\varphi_{max}}^{\varphi_{bulk}} \frac{D}{\varphi} d\varphi \quad (13)$$

Where δ is the thickness of mass transport boundary layer, R_0 is the radius of membrane, φ_{bulk} is the feed concentration and φ_{max} is the concentration near the membrane surface.

On the other hand, regarding the back diffusion phenomenon, parameter D can be considered equal to the sum of Brownian diffusion coefficient, D_{Br} and the shear-induced diffusion coefficient D_{SI}(Samuelsson et al., 1997; Huisman et al., 1999).

$$D = D_{Br} + D_{SI} \quad (14)$$

$$D_{Br} = \frac{k_B T (1-\varphi)^{6.55}}{6\pi\mu a} \quad (15)$$

$$D_{SI} = \frac{0.33 \tau_w a^2 \varphi^2 (1 - 0.5 \exp(8.8\varphi))}{\mu} \quad (16)$$

An important parameter in this modeling is the thickness of mass transfer boundary layer. In this modeling, the flow inside the tube has been considered to be turbulent and the following equations have been used to estimate the thickness of mass transfer boundary layer (Munir, 1998).

$$Sh = 0.023 (Re)^{0.8} (Sc)^{0.33} \quad (17)$$

In this equation Sh is the Sherwood number, Re is the Reynolds number and Sc is the Schmitz number. Considering the relationship between Sherwood number and the mass transfer coefficient (k), it can be written that:

$$\left.\begin{aligned} Sh &= \frac{kd_h}{D} \\ k &= \frac{D}{\delta} \end{aligned}\right\} \Rightarrow Sh = \frac{d_h}{\delta} \Rightarrow \frac{d_h}{\delta} = 0.023 (Re)^{0.8} (Sc)^{0.33} \quad (18)$$

Where d_h is the hydraulic diameter of the channel.

In the limiting flux conditions, the membrane surface is covered with fouled particles. Therefore, to estimate the forces between the particles, it is necessary to consider the forces exerted on the particle from the particles near to it. In this way the modeling considers that each deposited particle is near to four particles that have already been precipitated and therefore the exerted force has been noted to be from four mentioned sides. The angle between the vertical element of force and the interaction force between two particles in this condition has been assumed 54.74° (Huisman et al., 1999).

Simulation

To solve the model presented in this research, the MATLAB software was used. The calculation procedure of this program is shown in Fig. 2. After giving the input data such as physical properties of the solution, the size of particles, bulk and wall concentration, etc., the program respectively calculates the values of Schmitz number, thickness of mass transport boundary layer, and the particle velocity in the vertical direction to the membrane based on the input values. Then it calculates the amount of the exerted forces on the particle and finally it calculates the limiting flux.

As it was mentioned, this study is aimed at the modeling and simulation of limiting flux of skimmed milk. Skimmed milk which is the low fat milk has got a lot of casein that are scattered in the continuous phase in the form of micelles. Since the other proteins in the milk, compared with casein have more solvability in water, only the casein particles that are separated by the membrane have been considered in this research.

The needed parameters for the solution of equations of the model taken from different articles have been presented in table 1. Parameters such as the density, viscosity and ionic strength of skimmed milk and density, surface potential and Hamaker constant of casein have been presented in this table. Since the presented model is very sensitive to the parameters, diligent care has been taken in the selection of parameters.

Considering the parameters in table 1 and solving the equations, one can obtain some diagrams for the variations of limiting flux associated with changes of different parameters.

Results and discussion

Fig. 3 shows the changes of limiting flux versus wall shear stress. The increase in the wall shear stress causes the particles to get away from the membrane surface and so prevents fouling formation. When the fouling on the membrane surface is little, the permeate flux over the membrane will be more and consequently the limiting flux will also increase. As illustrated in Fig. 3, limiting flux is increased with the increase in wall shear stress (Samuelsson et al., 1997).

The increase of casein particles in the bulk is an important factor in the formation of fouling and it is predicted that with increasing bulk concentration, limiting flux will be decreased. The changes of limiting flux in terms of different amounts of bulk concentration have been illustrated in Fig. 4. As it is seen, in a particular shear stress, limiting flux is decreased with increase in bulk concentration.

One of the effective parameters on the limiting flux is the particle size. The size of casein particles depends on many factors such as pH and temperature and considering the distribution of other particles in the solvent, an average value is considered for them

(Samuelsson et al., 1997; Kühnl et al., 2010).

Fig. 5 illustrates the variations of limiting flux upon changes in the particle radius. As it is seen, with the increase in size, limiting flux is increased. This means that in the intended physical and chemical conditions, the increase of size between 60 to 100 nanometers results in the increase of the resultant vertical forces exerted on the particle, so it gets away from the membrane. In this way the fouling is reduced and so limiting flux is increased.

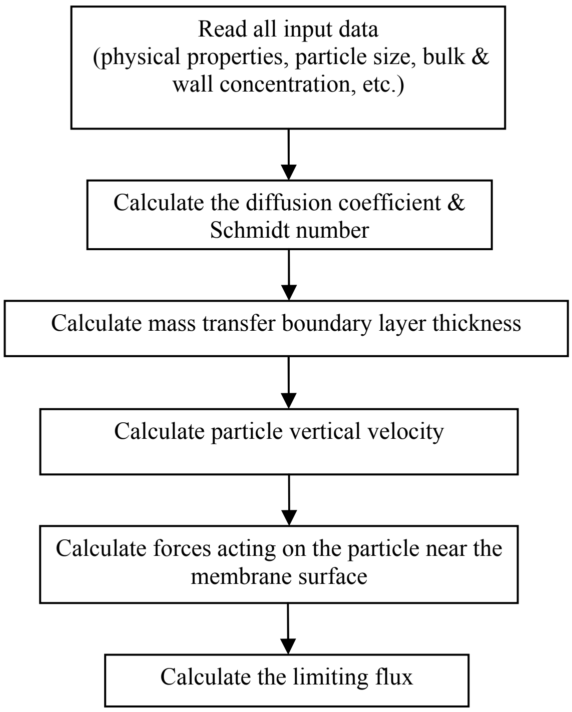

Fig. 2 Limiting flux calculation procedure with MATLAB

Table 1 Simulation data for skimmed milk

Parameter	Value	Reference
ε_r	80	(Kühnl et al., 2010)
a (nm)	60, 90, 100	(Samuelsson et al., 1997; Kühnl et al., 2010)
ψ_0 (mV)	17, 20, 22	(Yourvaong et al., 2002; Kühnl et al., 2010)
I (M)	0.08	(Fox et al., 1998; Tuinier et al., 2002)
ρ (at T=328 K) (kg/m^3)	1018.2	(Samuelsson et al., 1997)
μ (at T=328 K) (Pa. s)	0.755e-3	(Samuelsson et al., 1997)
ρ (at T=288 K) (kg/m^3)	1034.4	(Samuelsson et al., 1997)
μ (at T=288 K) (Pa. s)	1.99e-3	(Samuelsson et al., 1997)
ρ_p(kg/m^3)	1.2×1000	(Morr et al., 1970)
A_H (J)	0.298e-20	(Kühnl et al., 2010)
φ_{max} (dimensionless)	0.6792	(Huisman et al., 1999)

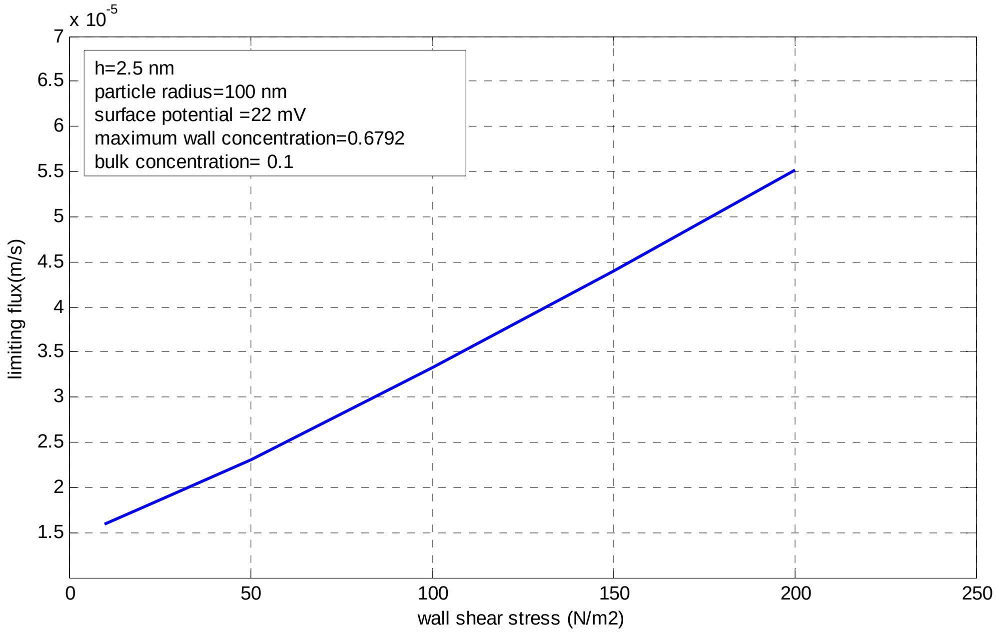

Fig. 3 Variation of limiting flux with wall shear stress

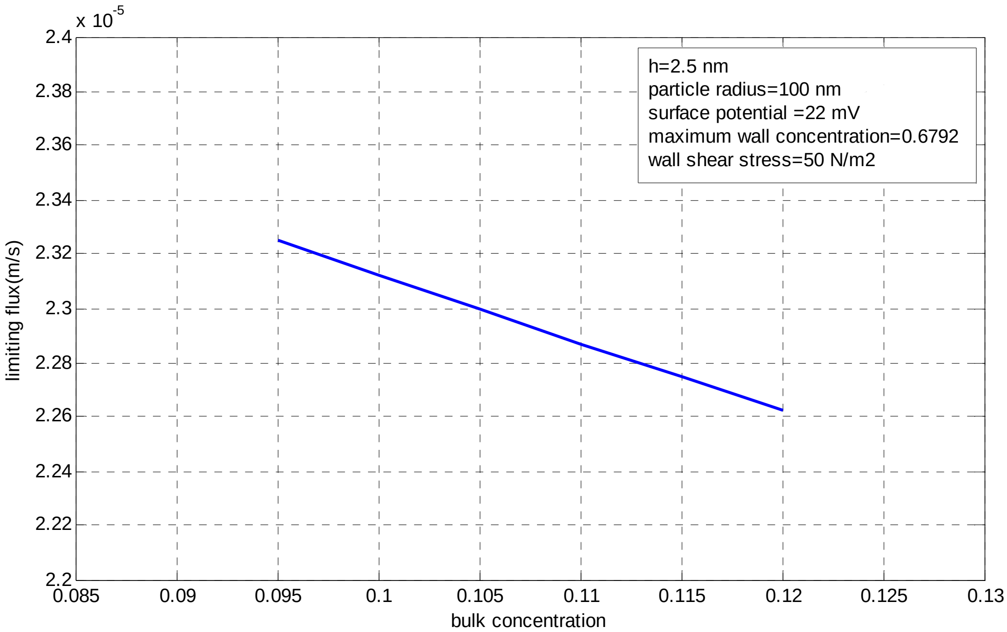

Fig. 4 Variation of limiting flux with bulk concentration

The effect of surface potential is shown in Fig. 6. The surface potential and the zeta potential have been considered the same in some researches (Yourvaong et al., 2002; Kühnl et al., 2010). Therefore, the effect of this parameter has been studied in Fig. 6. It can be seen that in a specific particle size, limiting flux is increased upon increase in surface potential, which consequently causes the repulsive electrostatic force to increase and results in distancing particles from the membrane surface which eventually leads to the increase in limiting flux (Baudry et al., 2005).

Another effective parameter on interparticle interaction forces is the distance of the particles from each other. These forces have a remarkable effect on each other when the particles are close together and because in the limiting flux conditions, the particles near the membrane surface have little distance from each other, these forces are significant. The effect of the particles distance on limiting flux has been accordingly studied in Figs. 7-9.

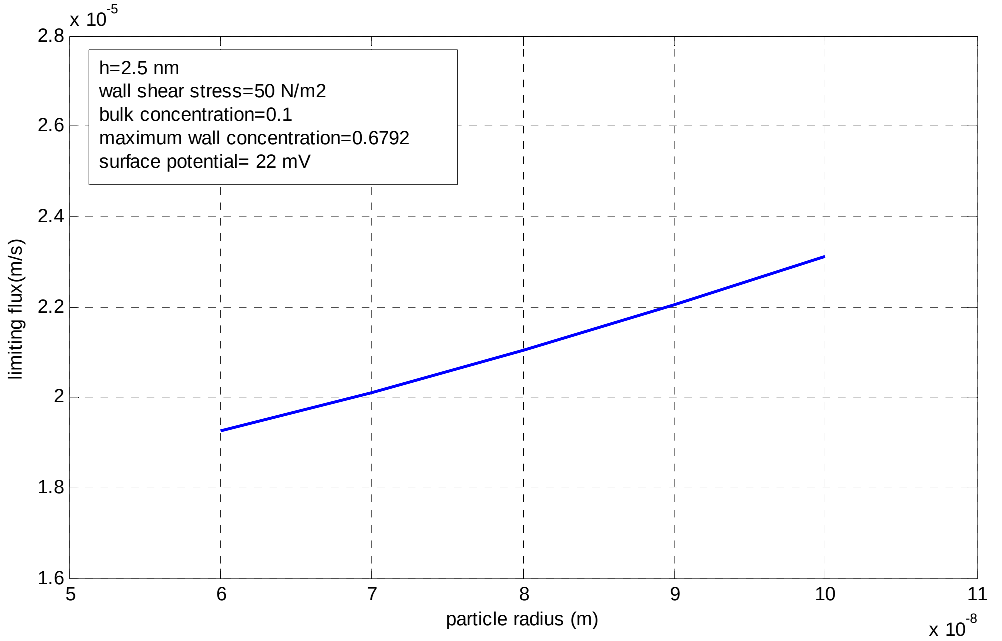

Fig. 5 Variation of limiting flux with particle size

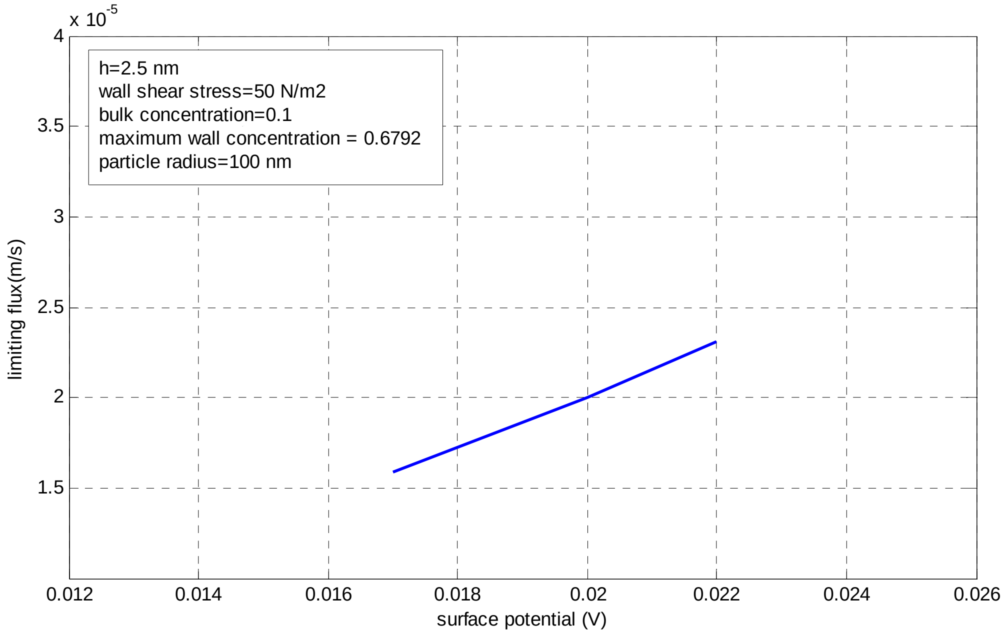

Fig. 6 Variation of limiting flux with surface potential

As expected, the increase of the distance between particles causes the limiting flux to decrease since the increase in distance reduces the interaction forces. If the distance between the particles is more than 10 nm, the effect of these forces is negligible(Fu et al., 1998). As in larger distances, colloidal forces do not have considerable effect on limiting flux, it can be concluded the particles in the bulk cannot affect the limiting flux and so the particles near the membrane surface (i.e. the mass transfer boundary layer) are more effective on limiting flux.

An effective parameter in the repulsive electrostatic force is the surface potential of the particle, the effect of this parameter together with the effect of distance between particles has been shown in Fig. 8. In a specific distance between particles (less than 10 nm).

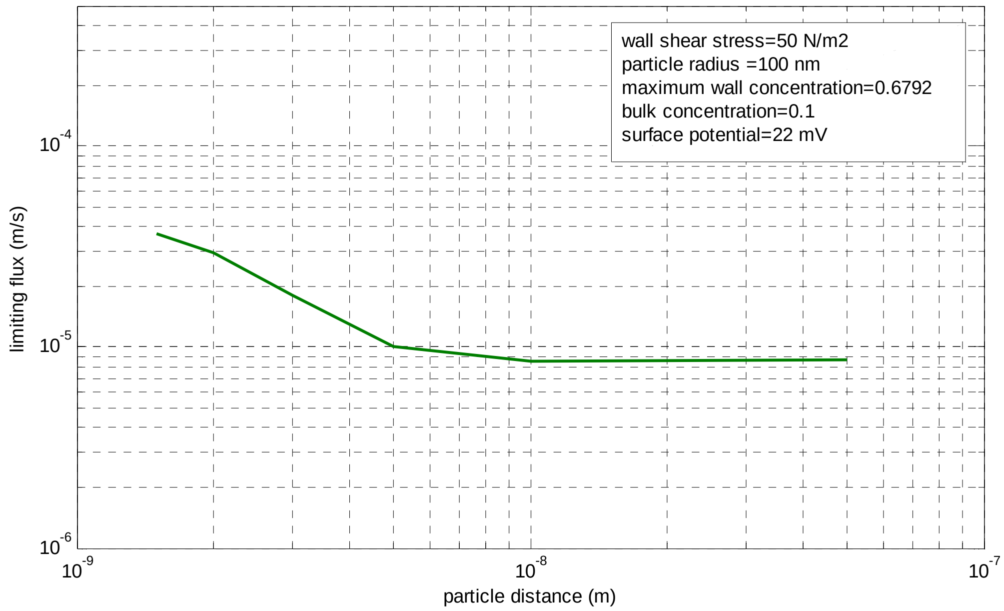

Fig. 7 Variation of limiting flux with particle distance

with the increase in surface potential, limiting flux is increased, because the increase in surface potential increases the repulsive electrostatic force and so the particles repel each other and get away from the membrane surface and consequently the limiting flux increases. However, the increase in limiting flux is limited in short distances and concerning Fig. 8, this factor (surface potential of the particle) is significant in short distances as with the increase of particles distance, the effect of surface potential on the limiting flux becomes trivial.

In Fig. 9 the limiting flux variations based on the particle distance together with the study of the effect of particle size is shown. As it was seen before, the increase in particle size in the intended physical and chemical conditions causes the limiting flux to increase. This result is clearly seen in Fig. 9 too. The diagrams on the left side of the Fig. 9 are closer to each other and their difference in short distances is less.

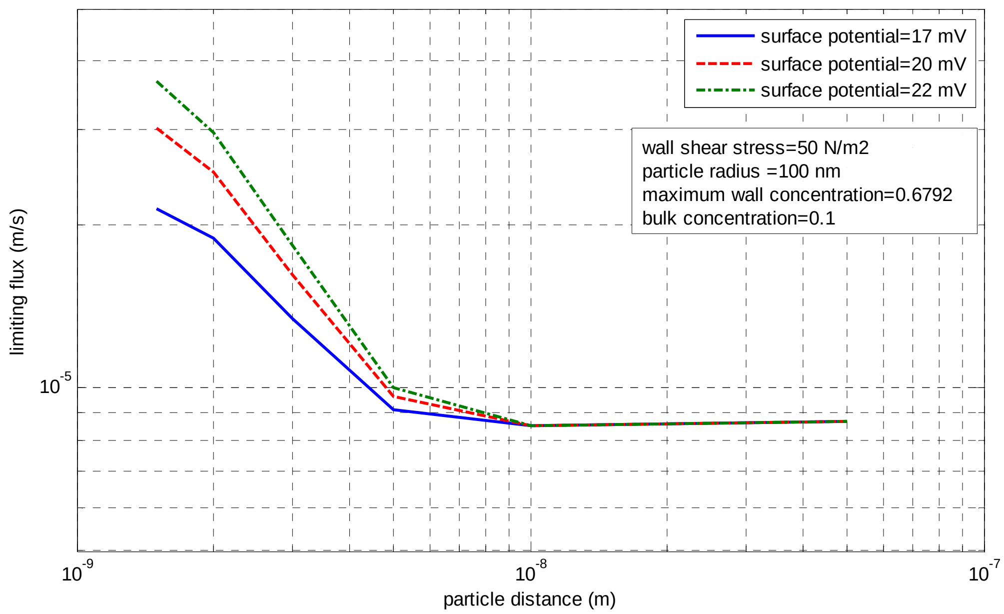

Fig. 8 Variation of limiting flux with particle distance and surface potential

In other words, in the short distances between the particles, the effective parameter on limiting flux is the distance of the particles from each other while in longer distances, the effective parameter on the limiting flux, is the particle size. According to both Figs. 8 and 9, it can be concluded that in short distances, surface potential and in long distances, particle size are the effective parameters on limiting flux.

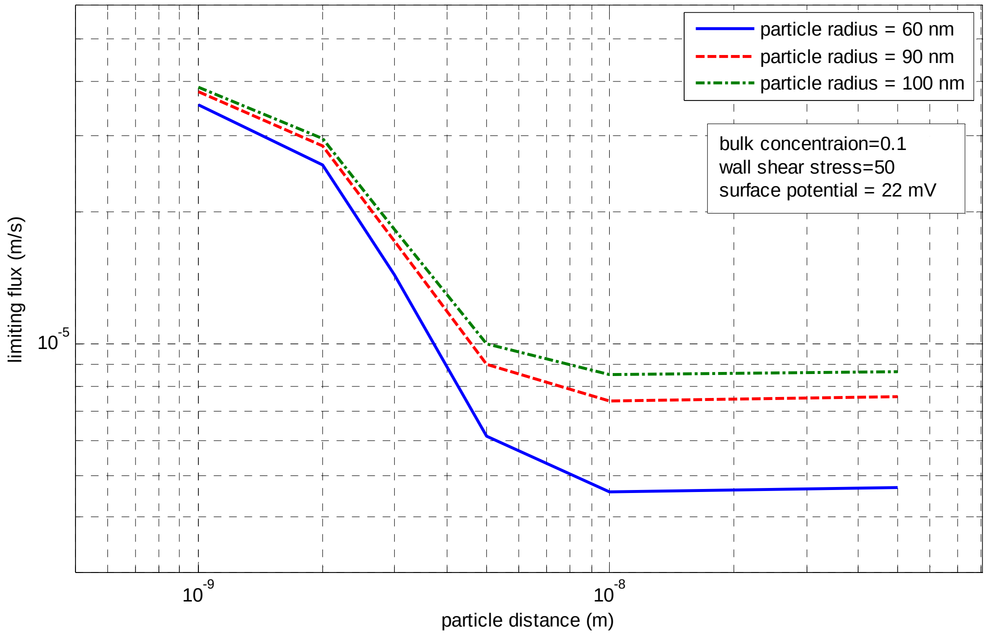

Fig. 9 Variation of limiting flux with particle distance and size

Another effective parameter on limiting flux is temperature. Samuelsson et al. have studied limiting flux variations with the wall shear stress in two different temperatures of 15°C and 55°C (Samuelsson et al., 1997). To calculate the limiting flux, they considered the back diffusion phenomenon with the combination of two Brownian and shear-induced diffusion. In Fig. 10 the limiting flux variations with the wall shear stress have been shown. With the increase of temperature, the limiting flux is increased. In this figure the points are the experimental data of limiting flux reported by Samuelsson et al.. The broken lines indicate the results of the combination of two diffusion factors by Samuelsson et al. The continuous lines in this figure indicate the presented modeling in this research. As it can be seen, the results of modeling of this research are closer to the experimental data. In the current model, the interactions between colloidal particles have also been taken into consideration in addition to the back diffusion phenomenon. Moreover, based on Fig. 10 and the similarity of the results of this modeling with experimental data, it can be concluded that one of the most effective factors on limiting flux in filtration of colloidal solutions is force between colloidal particles.

To have a better understanding of matching between the modeling and experimental data, some statistical parameters have been studied and the values of these parameters at two different temperatures, for the current model and the model reported by Samuelsson et al. were calculated, which compared in Tables 2 and 3. The following equations exist for these parameters:

$$NB = \sum_i^n \frac{(LF_{model,i} - LF_{exp,i})/LF_{exp,i}}{n} \times 100 \quad (19)$$

$$SSE = \sum_i^n (LF_{model,i} - LF_{exp,i})^2 \quad (20)$$

$$MSE = \frac{\sum_i^n (LF_{model,i} - LF_{exp,i})^2}{n} \quad (21)$$

$$RMSE = \sqrt{\frac{\sum_i^n (LF_{model,i} - LF_{exp,i})^2}{n}} \quad (22)$$

$$R^2 = \frac{\sum_i^n (LF_{exp,i} - LF_{model,mean})^2 - \sum_i^n (LF_{model,i} - LF_{exp,i})^2}{\sum_i^n (LF_{exp,i} - LF_{model,mean})^2} \quad (23)$$

In which *NB* is normalized bias, *SSE* is standard squared error, *MSE* is mean squared error, *RMSE* is root mean squared error, R^2 is squared correlation coefficient, LF_{exp} is experimental limiting flux, LF_{model} is limiting flux obtained by model and $LF_{model, mean}$ is mean limiting flux obtained by model.

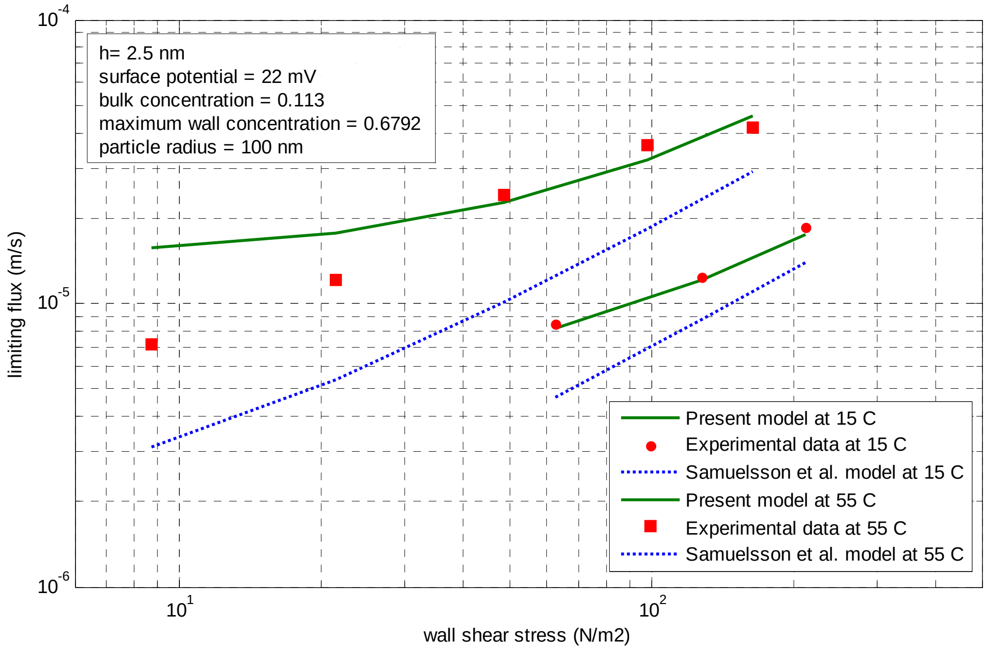

Fig. 10 Comparison of limiting flux variation with wall shear stress and temperature for two models with experimental data

Table 2. Statistical results at 15 °C

Statistical parameter	Samuelsson et al. model	Present model
NB	-67.5488	-5.6392
SSE	4.7642e-011	1.2578e-012
MSE	1.5881e-011	4.1927e-013
RMSE	3.9851e-006	6.4751e-007
R^2	0.5181	0.9761

Table 3. Statistical results at 55 °C

Statistical parameter	Samuelsson et al. model	Present model
NB	-121.5804	141.5340
SSE	7.2827e-010	1.3979e-010
MSE	1.4565e-010	2.7958e-011
RMSE	1.2069e-005	5.2876e-006
R^2	0.5130	0.8487

Based on Table 2, the values of NB parameter resulted from this research and Samuelsson's model are both negative, so they under predict the limiting flux but regarding these values, the under prediction is much less by the present model and its results are closer to the real values. SSE, MSE and RMSE parameters that indicate the deviation or error are less in the present model, which indicates a better prediction of limiting flux by present model. Comparing the values of R^2 for the two models at 15°C shows that using the presented model in this research at mentioned temperature caused that limiting flux values are fitted better.

At 55°C (Table 3), the values of NB parameter resulted from the present research and Samuelsson model are positive and negative, respectively which means using the Samuelsson model caused that the limiting flux is under predicted while it is over-predicted using the present model. It shows that Samuelsson model predicts the limiting flux amount less than the real amount while the present model predicts it more than the real one. The numerical amount of NB parameter in both models has a similar order in this temperature, so there is no noticeable difference between them in this case. However the three other parameters, SSE, MSE and RMSE are one order of magnitude less in the present model which indicates a better prediction of limiting flux by the present model. Therefore, using this model reduces the error value of limiting flux. On the other hand, it can also be seen that the amount of R^2 resulted from the present model is higher than it reported by Samuelsson et al. and therefore the experimental data is fitted better in this temperature and the presented model in this research can predict limiting flux better.

Conclusion

In the membrane filtration processes, the permeate flux is reduced by the formation of fouling on the membrane surface and reaches to a limited value called limiting flux. The prediction of this limiting flux can be useful in the design of membrane systems. In the present study, a comprehensive force balance model has been presented in which all of the exerted forces on the colloidal particle near the precipitated layer on the membrane surface in steady conditions have been considered. In addition, the model notes the back diffusion phenomenon by considering Brownian diffusion and shear-induced diffusion. The results obtained from this modeling indicate that with the increase in wall shear stress, particle size, and surface potential, limiting flux increases, and in a specific wall shear stress, with the increase in bulk concentration and interparticle distance, the limiting flux decreases. Interactive electrostatic repulsion and van der Waals attractive forces have been considered in calculating the limiting flux. In the short distances between particles, the effective parameter on limiting flux is the surface potential of particle,while in larger distances the effective parameter on limiting flux is particle size.

In this research a comparison between the experimental data, presented model in this study and the combined model presented by Samuelsson et al. has been also done and some statistical parameters have been calculated. The values of these parameters indicated that the present model can predict the limiting flux closer to the real values than the Samuelsson's model. In the Samuelsson et al. model, the limiting flux has been obtained only by considering the back diffusion phenomenon, while the physical and chemical properties of colloidal suspension of skimmed milk have not been taken into account. However, in the present model which is a force balance approach, in addition to back diffusion phenomenon, the interparticle colloidal forces e.g. repulsive electrostatic forces and van der Waals attraction have also been considered. Based on the above mentioned results, the importance of considering the interaction forces between particles in estimating the limiting flux in the membrane filtration of skimmed milk was observed and it is suggested that it is better to always take these forces into consideration to determine the limiting flux of membrane process for colloidal suspensions.

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