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Electro-microfiltration of Colloidal Suspensions

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ABSTRACT

Crossflow microfiltration experiments using polymethyl methacrylate (PMMA) colloidal suspensions by a flat-channel electro-filter were performed to investigate the effects of electric field strength applied and the surface properties of particles, such as zeta potential and electric double-layer thickness, on the filtration rate, average specific cake filtration resistance, and cake porosity. Mathematical models for relating the quasi–steady state filtration rate and the separated distance between deposited particles on cake surface to the operating parameters were also presented. Both the experimental data and the theoretical model indicated that the filtration rate–electric field strength dependence was linear when the filter was operated in regions under the critical electric field strength. However, when the electric field was greater than the critical strength, the filtration rate was obtained by the addition of the electro-osmotic flux and the hydraulic permeate flux through the membrane. Due to the electro-osmotic flow in the membrane having the same order of magnitude as

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the filtration rate, it was found that no correction of the filtration rate with the electro-osmotic flux would lead to underdetermined values of the average specific cake filtration resistance. The effect of electrolyte concentration on the cake characteristics was also analyzed theoretically and experimentally.

Key Words: Crossflow microfiltration; Electric field; Electrophoresis; Electroosmosis.

INTRODUCTION

Crossflow microfiltration is a method widely used for the separation of fine particles from suspensions in many industrial sectors. However, the flux decline due to membrane fouling is generally a serious problem, especially for separating colloidal and proteinaceous solutions. Even if only a very thin gel or cake layer is formed by these materials, the high permeation resistance results in an unacceptable filtration rate and frequently renders the process uneconomical. To maintain a high filtration rate for extended periods of time, therefore, it is necessary to limit the buildup of cake or gel layer on the filter surface. With the demands for improved separation technologies, the electric field used as a combined technique with crossflow filtration to control membrane fouling has been attracting an increasing amount of attention recently.[1]

The so-called crossflow electrofiltration (CFEF) is a process developed to assist crossflow filtration in minimizing membrane fouling by applying a DC electric field across the filter medium surface to alter the motion of the particles, with the electrophoretic migration away from the membrane surface. When the strength of the applied electric field reaches a critical value where there is a balance between the particle's convective velocity due to bulk flow toward the filter media and the particle's velocity away from the medium due to the electrophoretic and shear migration effects, no cake is formed in the case. The second electrokinetic effect involved in electrofiltrations is the electro-osmosis which may occur within the cake layer and medium, which generally would allow for more fluid to flow across the membrane. Since most fine particles acquire a surface charge when in contact with a polar medium, generally negative charged, and the electrokinetic phenomena, being surface processes, are relatively insensitive to the particle size, it thus seems that the techniques can provide an alternative method for separating materials that are difficult to filter. In practice, there are considerable promises from experimental results for the application of an electric field to improve the filtration of fine particles.[2–5]

In conjunction with conventional crossflow filtrations, Henry et al.[6] have described the filtrate flux in terms of a series resistance due to media, cake layer, and the concentration polarization layer with a modification of the resistances of membrane and cake to the filtrate in terms of electro-osmotic effects. By taking a balance of forces acting on a particle perpendicular to the membrane surface under steady state, a relationship implying that the flux enhancement due to the electrical force is additional to the flux obtained without an electric field has also been proposed by Okada et al.[7]

It was known that the mechanism of particle deposition and the specific resistance of the deposited cake play the major roles in the filtration rate. Therefore, to know how the electrokinetic effects coupled with the shearing action affect the deposition of particles and the filterability of the deposited cake layer is the essential step in understanding the performance of CFEF. When an electric field is imposed upon a filter, the particle motion through the filter chamber and its deposition on the medium is not only influenced by the fluid velocity profile but will also be further affected by the electrophoresis. The hydrodynamic behavior and the particle's trajectory in such filters have also been discussed.[8–9]When a given electric field strength is applied, the electrophoretic velocity of suspended particles and the electro-osmotic flow in its deposited cake layer are mainly determined by the properties such as zeta potential and the electrical double layer around the particle. Besides, the physiochemical properties of suspensions may also significantly influence the electrostatic repulsive forces between particles and the electro-osmotic flow in the membrane. The former is an important factor in controlling the structure of deposited cake,[10–11] and the latter may give a considerable enhancement of the filtration rate. Therefore, the pH and ionic strength of the suspension, which are the main factors in determining the surface properties of particles, give a strong influence on the performance of fine particle filtration. Although it was known that the electrokinetic behaviors of suspended particles and in cake layers are dependent on the physiochemical properties of suspensions, attention was seldom paid to a quantitative analysis of the effect of such properties on the particle deposition and the filterability of cake in CFEF.

In the present work, the characteristics of the filtration rate, the specific cake resistance, and cake porosity for filtering fine particles in CFEF are investigated. The effect of electric field action on the filtration are analyzed with a concept of force balance model for depositing particles to relate the steady-state filtration rate to the electrophoretic velocity of particles and the operating parameters. Emphasis is placed on the effects of electric field strength and the electrolyte concentration on the filtration behavior. Additionally, the role of electro-osmosis in the filtration is also examined.

THEORY

Figure 1 depicts the deposition of suspended particles onto a cake surface in a crossflow filtration system, where particle B is a particle already stably deposited on the cake surface and particle A is a particle just arriving at the cake surface. The angle β is known as the "angle of friction."[12] The forces exerted on particle A include the drag generated by the fluid flow over the particle, F_h (by tangential flow parallel to the filter septum), F_p (by permeate flow normal to the filter septum), and F_l (called lift force); the net gravity force, F_g; the interparticle force, F_i; and the electrophoretic force, F_e, induced by the applied electric field. The force due to Brownian motion of particles is neglected in this study since the diameter of the particles used is larger than 1.0 μm.

The torque balance of particle A about the pivot position (labeled G in Fig. 1) at the critical state, for which the particle will deposit immediately when it arrives at the cake surface, is

$$F_n \cos\beta = F_n \sin\beta \tag{1}$$

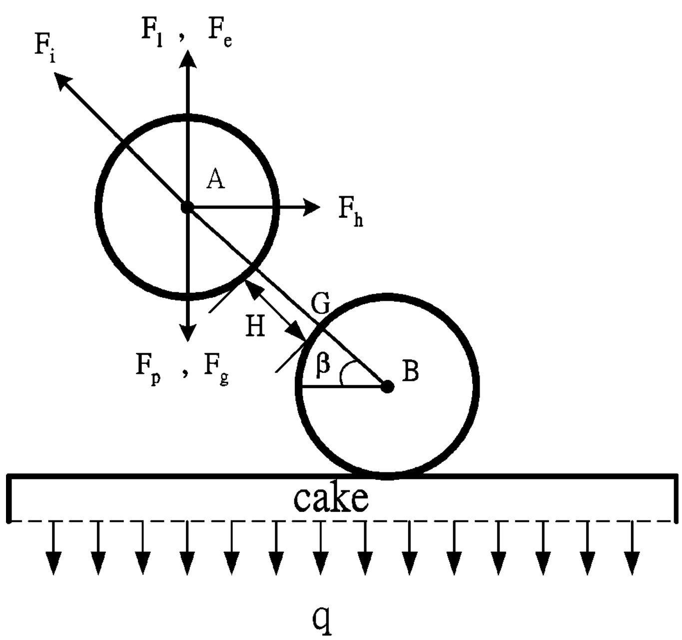

Figure 1. Forces exerted on the depositing particles.

where $F_n(= F_p + F_g - F_l - F_e)$ is the net force normal to the filter septum. A critical condition such as $F_n = 0$ has also been adopted for developing the steady state filtration flux model.[13] By knowing the operating conditions, F_h, F_p, F_l, and F_g can be calculated from the expressions relating the forces to the particle diameter D_p, wall shear stress τ_w and the filtration rate, q, as $F_h = a_0\tau_w D_p^2$, $F_p = a_1 q D_p$, $F_g = a_2 D_p^3$, and $F_l = a_3\tau_w D_p^4$, where the coefficients a_0, a_1, and a_3 are determined by considering the modification of the Stoke's law for fluid flow very near a wall.[14–15] If the electrophoretic velocity of the particle is denoted as v_e, the force F_e can be given similar to the expression of F_p, that is, $F_e = a_1 v_e D_p$. The expression for electrophoretic velocity of particles is generally given by

$$v_e = C\frac{\varepsilon\zeta E}{\mu} \tag{2}$$

where E is the electric field strength, ζ is the zeta potential of the particle, and C is a constant that depends on the magnitude of κR, a product of the reciprocal of the thickness of the double layer and the radius of the particle. For aqueous solutions at 25°C, the value of κ can be estimated as $\kappa = 3.59 \times 10^9\, I^{1/2}\, \mathrm{m}^{-1}$, where I is the ionic strength. The values of C are in the range from 2/3 for small values of κR as $\kappa R < 0.1$ to 1.0 for $\kappa R > 100$.

Substituting those forces described above into Eq. (2), the relationship between filtration rate q and the operating parameters for just arriving at a stable deposition of particles in diameter D_p can be represented by

$$q = v_e + \frac{a_0\tau_w \sin\beta}{a_1\cos\beta}D_p - \frac{a_2}{a_1}D_p^2 + \frac{a_3\tau_w^2}{a_1}D_p^3 \tag{3}$$

Based on the operating conditions and experimental results of quasi-steady filtration rate, q_s, the critical angle of friction,[12] β_c, for depositing particles can be estimated from this relationship. If the effect of electric field strength on the values of β_c and the coefficients a_0–a_3 are negligible, the relationship implies that the steady-state filtration rate in CFEF is the electrophoretic velocity of particles in addition to the filtration rate obtained only by cross-flow shearing action.

$$q_s = v_e + q_{E=0} \tag{4}$$

The interaction force between particles can be estimated by using the DLVO theory. When two particles move toward each other, there exist two long-range interaction forces, the electrostatic force, F_{es}, and the van der Waals force, F_v, acting on the particles. If two equal-sized spherical particles of low potential approach each other under the condition of constant potential and the retardation effect is not considered, the net interaction force, F_i, obtained by addition of F_{es} and F_v for the case of $D_p \gg H$ and $\kappa D_p > 20$, yields.[16]

$$F_i = \frac{-A_h D_p}{24H^2} + \pi\varepsilon D_p \psi^2 \frac{\kappa \exp(-\kappa H)}{1 + \exp(-\kappa H)} \tag{5}$$

Here, A_h is the Hamaker constant, which is about 1.05×10^{-20} J for the PMMA particles used in the study; ε is the permittivity of the fluid filled in the gap of the particles, and Ψ is the stern potential. The measurable value, zeta potential (ζ), can be used for most conditions instead of Ψ. Figure 2 shows the evaluated values of F_i between the PMMA particles, which are suspended in an aqueous solution with various KCl concentrations. Since the repulsion between the particles is strong, it can be seen that the net interaction forces are repulsive even at a very small separated distance when two particles nearly contact. At sufficiently low κH values, the F_i curves each appear at

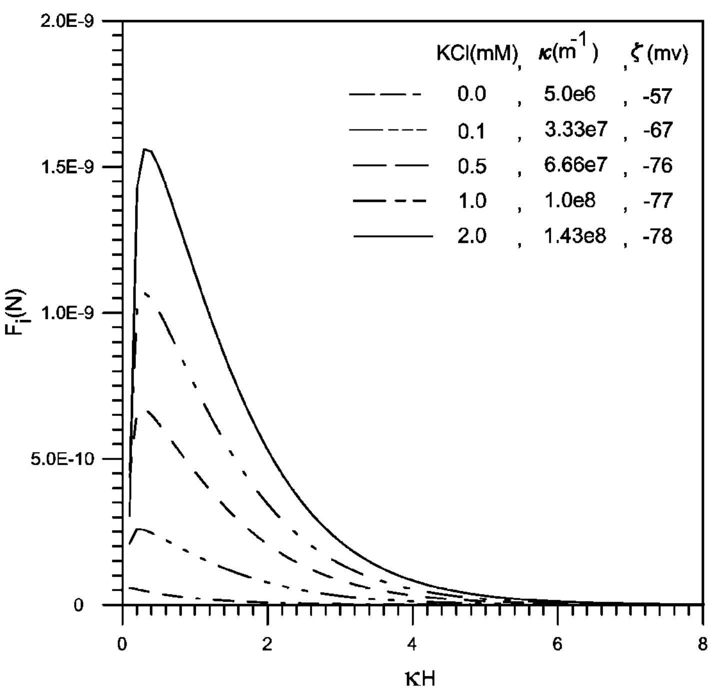

Figure 2. Variation of F_i distribution with κH for various KCl concentrations.

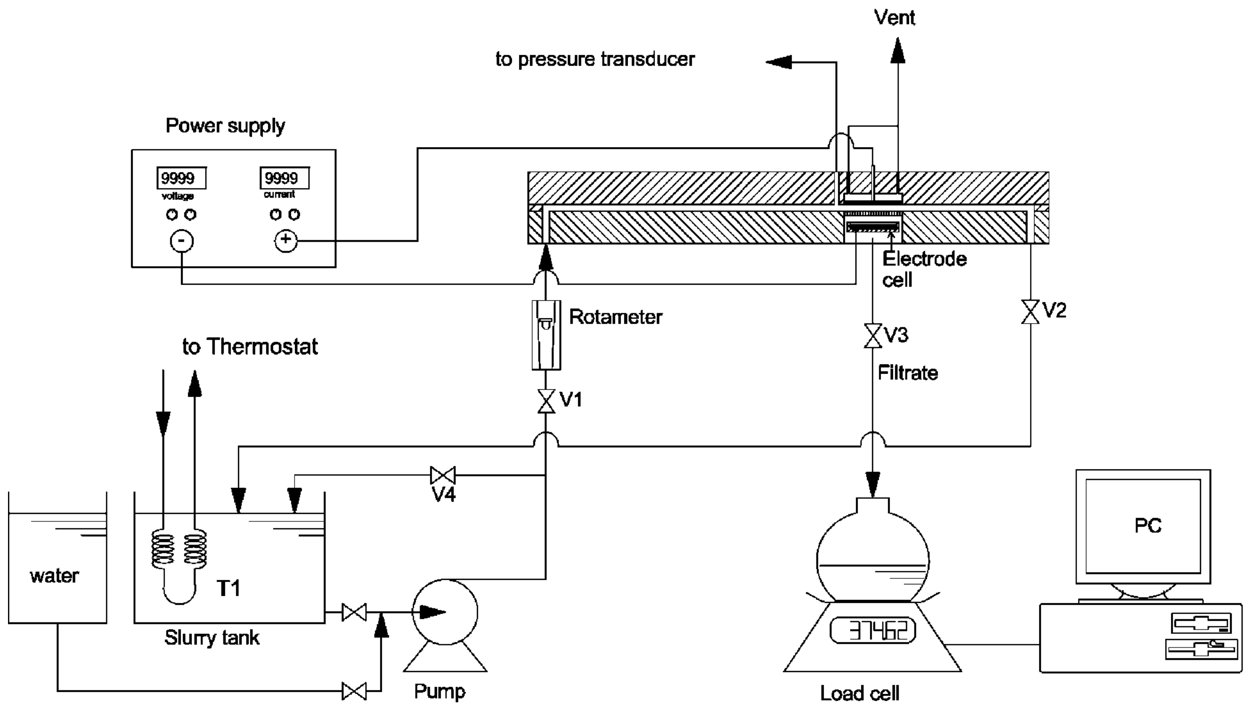

Figure 3. A schematic diagram of crossflow electrofiltration apparatus.

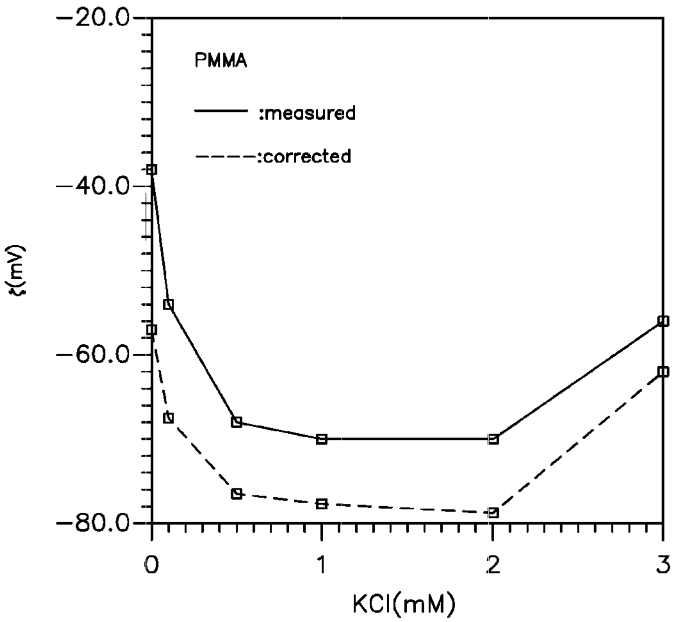

Figure 4. Effect of KCl concentration on zeta potential of PMMA particle.

a maximum, and its values are about 10^{-9}–10^{-11} N, depending on the KCl concentration.

If the total force exerted on particle *A* shown in Fig. 1, the direction of which is toward particle *B* and parallel to the line connecting the gravity centers of the two particles, is smaller than the repulsive force F_i, the particle *A* cannot connect precisely with particle *B* and will separate with an equilibrium distance when it arrives at a stable deposition. The equilibrium distance between the surfaces of neighboring particles can be determined by using the force balances under the critical state, i.e., the net force exerted on particle *A* in the direction toward *B* is equal to the interaction force.

$$F_h \cos\beta_c + F_n \sin\beta_c = F_i \tag{6}$$

On the other hand, if the total force in the left-hand side of Eq. (6) is larger than the value of maximum repulsive force, particle *A* will come into contact with particle *B* when the angle of friction β is less than the critical value, β_c.

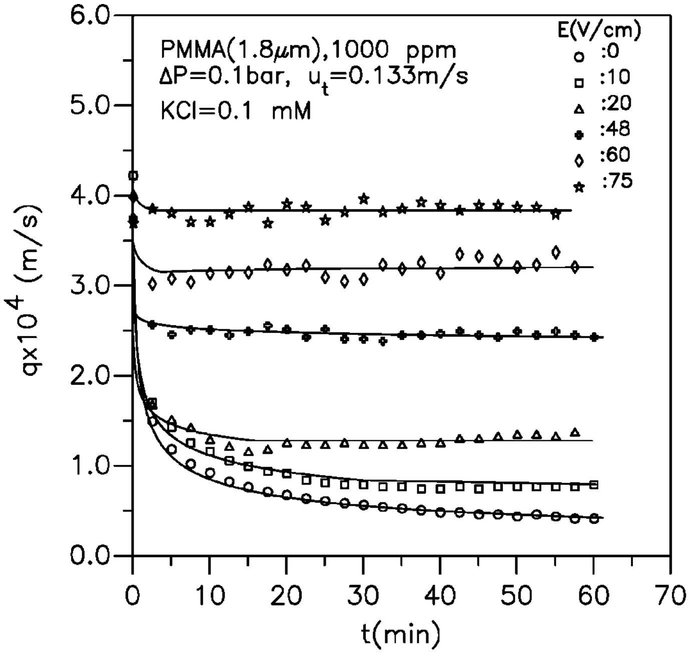

Figure 5. Effect of electric field strength on the filtration rate.

EXPERIMENTAL

The filtration experiments were carried out by using a flat-plate crossflow filter module on which a DC electric field is imposed, as shown in Fig. 3. A detailed design of the filter chamber was described elsewhere.[9] Experiments were performed using 500 ppm aqueous suspensions of PMMA latex. The particles have a median diameter of 1.8 μm and a density of 1200 kg/m^3. The electrolyte concentration of suspensions was adjusted by the addition of KCl solution. The values of zeta potential of the suspended particles were measured via MALVERN Zetasizer 3 and are shown in Fig. 4. The zeta potential obtained from the Zetasizer is based on substituting the measured electrophoretic velocity of particles into the electrokinetic relationship shown in Eq. (2) with C = 1.0 (Helmholtz–Smoluchowski model). However, since the values of C depend on κR, the zeta potentials used for evaluating the interaction force between particles were obtained by correcting the measured values with the consideration of the variation of C, which has values in a range

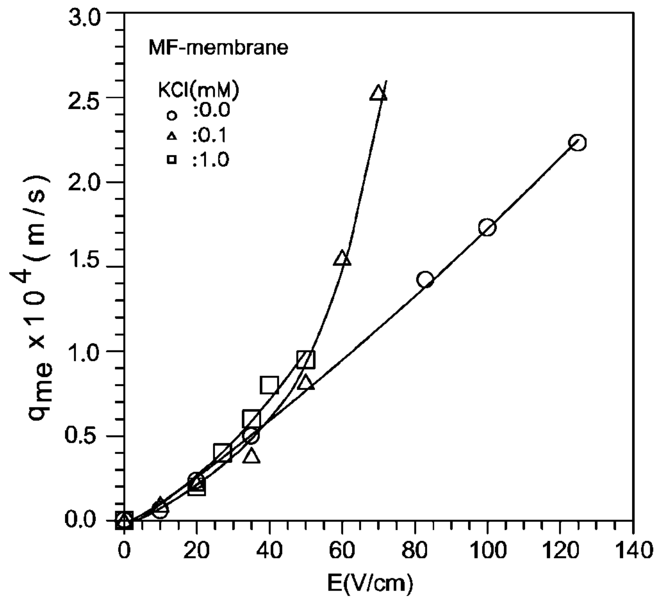

Figure 6. The electro-osmotic flow rate through membrane with various KCl concentrations.

of 0.664–0.884 under the operating conditions in this study. Since all of the values of zeta potential were large enough, the occurrence of particle aggregation in the suspensions can be neglected.

In each experiment, the crossflow velocity was kept at 0.133 m/sec (wall shear stress $= 0.102\,\mathrm{N/m^2}$) and the filtration pressure was maintained at 0.1 bar. Nylon 66 membrane (Pall Corp.) with a cutoff of 0.2 μm was used as the filter medium. The concentrated suspension was recycled back to the suspension tank and the amount of filtrate was detected by a load cell and recorded on a personal computer. As the filtrations were terminated after about 1 hr of operation, the cake that formed on the filter membrane was carefully scraped and sent to determine its wet and dry mass.

RESULTS AND DISCUSSIONS

Figure 5 shows the plots of filtration rate versus time for experiments with various electric field strengths. The membrane used in the filtrations has

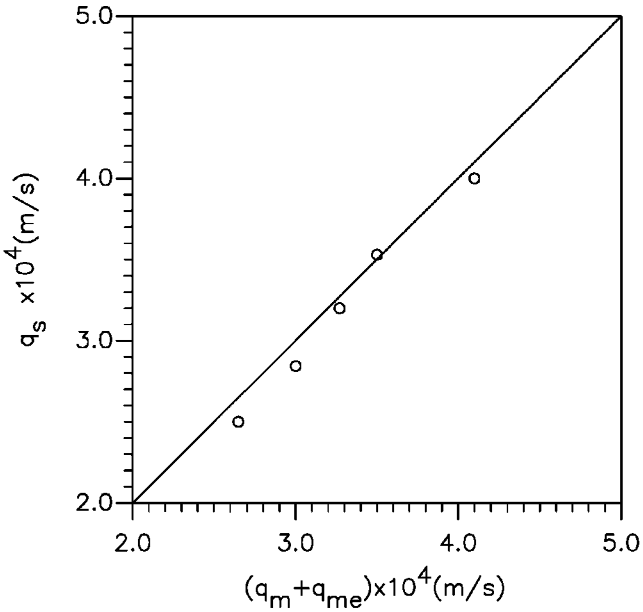

Figure 7. q_s vs. $q_m + q_{me}$ for electric field greater than the critical strength.

a resistance of $5.6 \times 10^{10}\,\text{m}^{-1}$ and hence, the permeation rate for water through the virginal membrane at the filtration pressure of 0.1 bar is $1.77 \times 10^{-4}\,\text{m/sec}$. According to the definition of critical electric field strength, E_c, at which the net particle migration velocity toward the membrane is zero, the value of E_c is about 48 V/cm for providing an electrophoretic velocity equal to the permeation rate of $1.77 \times 10^{-4}\,\text{m/sec}$. However, as the electro-osmosis induced in the membrane plays a significant role in enhancing the filtration rate, the critical strength will be obviously greater than the value estimated only concerning the hydraulic permeation rate through the membrane. This implies that the values of E_c can't be foreknown except with the measurements of the electro-osmotic flow through the membranes. The results in Fig. 5 indicate clearly that, with no electric field imposed, the filtration rate is low and still has an obvious decline even after 1 hr of operation. As an electric field is coupled with the crossflow filtration, the filtration rate increases and a shorter running time is required to approach the quasi-steady-state when a higher electric field strength is provided. Since only a very small amount of particle deposition on the septum was observed after the termination of the filtrations with E greater than 48 V/cm, the filtration resistance from its cake

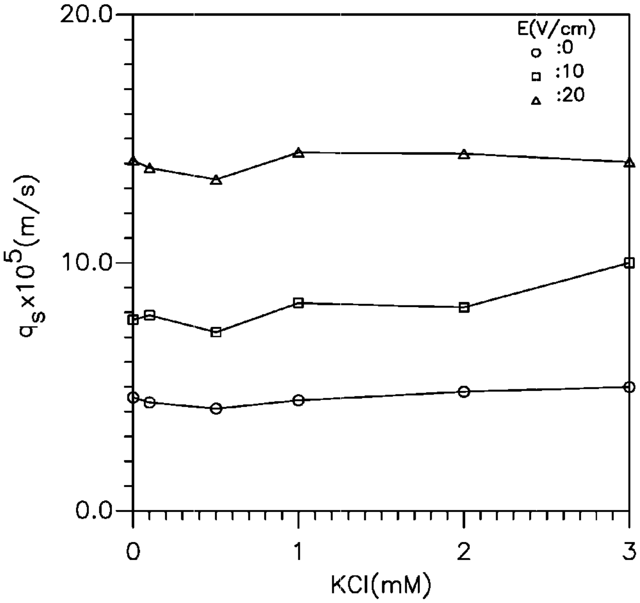

Figure 8. Effect of electric field strength on the quasi-steady-state filtration rate at $E < E_c$.

layer is negligible. Therefore, the significant increase in filtration rate when E is further increased might be due to the addition of electro-osmosis through the membrane.

In the study, the electro-osmotic flux through the virginal membrane was also measured and is shown in Fig. 6. The results reveal that the electro-osmotic flux is proportional to the electric field strength at very low electrolyte concentrations or $E < 60\,\mathrm{V/cm}$. As compared with the filtration rate data shown in Fig. 5, it appears that both values have the same order of magnitude. Apparently, the electro-osmotic flux through the septum plays an important role in the increase of the filtration rate. Since there is almost no particle deposition by the combined actions of cross flow and high electric field, only the hydraulic resistance and the electro-osmosis of the membrane must be considered in determining the quasi-steady-state filtration rate at $E > E_c$. A comparison of the measured filtration rates with the values estimated by adding the electro-osmotic flux to the hydraulic permeation flux through the clean membrane is shown in Fig. 7. The strong agreement clearly

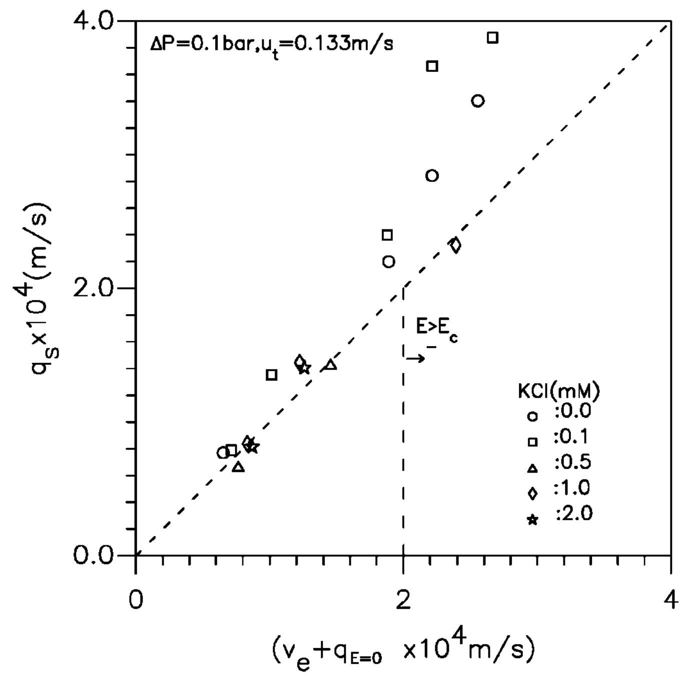

Figure 9. q_s vs. $v_e + q_{E=0}$ for various electric field strength.

demonstrates that the filtration rate at $E > E_c$ can be evaluated according to the filter medium resistance and its electro-osmotic flow.

Figure 8 shows how the KCl concentration in suspensions affects the quasi-steady state filtration rate at $E < E_c$. It is seen that in the range of 0–3 mM KCl used in the test the electrolyte concentration does not give a significant effect on the filtration rate. However, one can notice that for each electric field strength a minimum filtration rate appears at 0.5 mM KCl. As mentioned above in the theoretical analysis, if the effects of electric field strength on the critical frictional angle and the coefficients a_0–a_3 shown in Eq. (3) are negligible, the quasi-steady-state filtration rate, q_s, can be evaluated by addition of the electrophoretic velocity, v_e, of the particles and the filtration rate, $q_{E=0}$, obtained without electric field. Figure 9 shows a comparison between the measured q_s and the evaluated values from Eq. (4) for different KCl concentrations. A fairly good agreement is observed at $E < E_c$ where the cake is formed and its amount is controlled by the electrophoretic migration of the particles. As the applied electric field is greater than

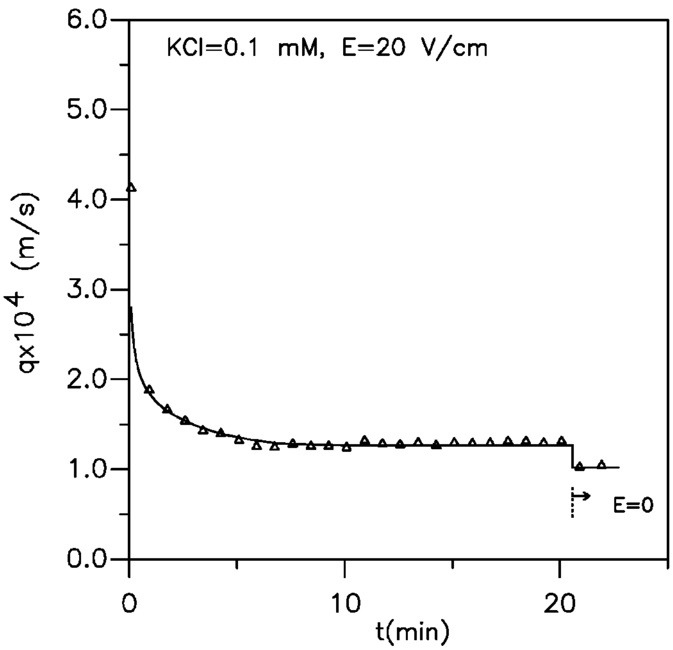

Figure 10. Variation of filtration rate with interruption of electric field.

the critical strength, the cake formation is completely limited and the measured filtration rate is much larger than the estimated values due to the electro-osmotic flux through the septum increases with the electric field strength.

In order to know the role that electro-osmosis plays during the filtration process, experiments that interrupted the electric field after a period of filtration were conducted under which an instantaneous drop of filtration rate occurred due to the stopping of electro-osmotic flow, as shown in Fig. 10. Such a filtration rate drop can be seen as the electro-osmotic flux induced in cake and medium during filtrations.

In cake filtrations, the average specific cake resistance, α_{av}, can be determined from the data of the filtration rate, q, the mass of dry cake, w_c, and the filter medium resistance, R_m, by using the equation of $\Delta P = \mu q[\alpha_{av} w_c + R_m]$. In Fig. 11, the effect of electric field strength on the α_{av} is shown. Since the α_{av} means the hydraulic resistance for fluid flow through the cakes, the filtration rate adopted in determining the α_{av} should not involve

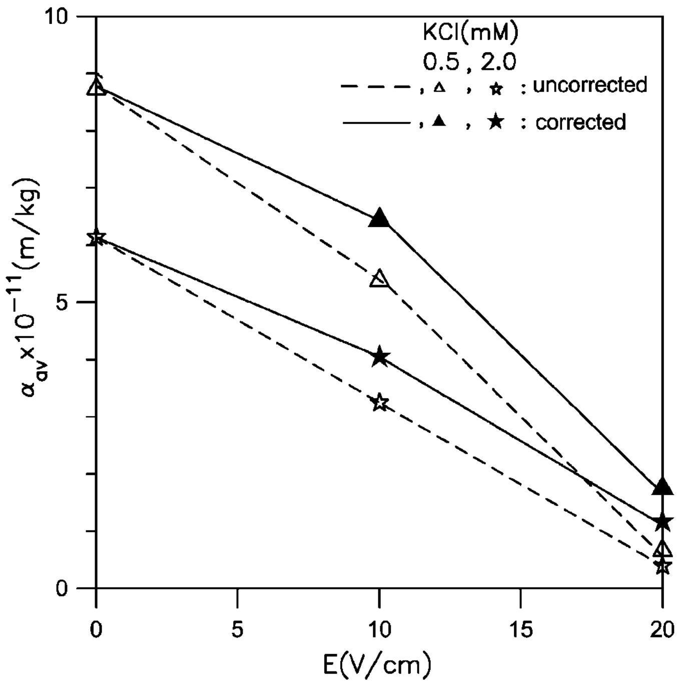

Figure 11. Effect of electric field strength on the average specific filtration resistance.

the component from electro-osmotic flow. A comparison of the α_{av} determined by the measured filtration rate and the corrected α_{av} determined from filtration rate without containing the electro-osmotic flux term is also shown in the figure. It can be found that the uncorrected α_{av} has a smaller value, and the difference between both increases when an increase in *E* due to the electro-osmotic flux becomes more dominant in the filtration rate. Note that as *E* is increased to 20 V/cm the uncorrected α_{av} are only about 30–40% of the corrected values. The results show that the values of α_{av} decrease significantly with the increase of *E*, and, within the KCl concentrations tested in the study, a higher value of α_{av} appears at 0.5 mM.

The measured results of average cake porosity, ε_{av}, are shown in Fig. 12. Because the application of electric field leads to a more open structured filter cake, the ε_{av} obtained increases obviously with the electric field strength. It is

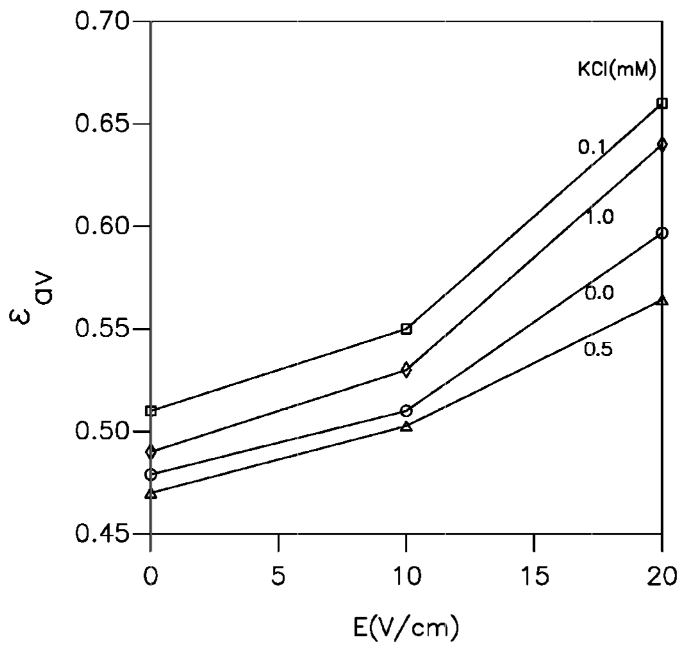

Figure 12. Effects of electric field strength and KCl concentrations on the average cake porosity.

interesting to note that, in the range of KCl concentrations tested, a denser packing of cake is obtained at 0.5 mM KCl, which is coincided with the expectation from the α_{av} results. It was known that the structure of cake depends strongly on the critical frictional angle, β_c, of the depositing particles and the equilibrium distance between particles, the lower values of β_c or the larger values of equilibrium-separated distance will form a more porous structure of cake.[12] By using the data of the quasi-steady-state filtration rate, the α_{av} and the operating parameters such as wall shear stress, τ_w, and particles diameter, D_p, the values of β_c determined from Eq. (3) are plotted against E in Fig. 13. As can be seen, the increase in E leads to a smaller β_c and, when an electric field is applied, the values of β_c decrease with the increase of KCl concentration.

By substituting the values of β_c into Eq. (6), the equilibrium-separated distance between deposited particles on cake surface was evaluated and is shown in Fig. 14. It appears that the equilibrium-separated distance is very slightly

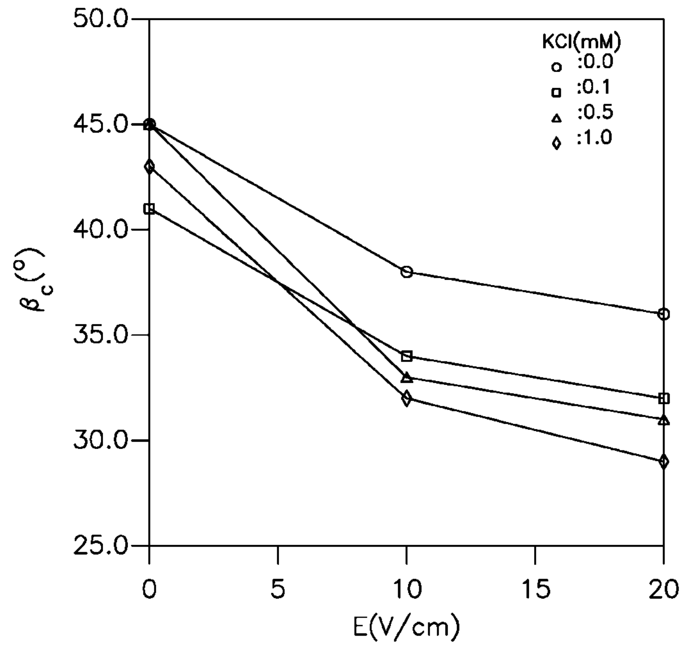

Figure 13. The critical friction angle of particles under various electric field strength for four kinds of KCl concentrations.

affected by electric field strength, whereas it is hardly affected by KCl concentration. Its values decrease sharply with the increase of KCl concentration ranging from 0 to 0.5 mM. However, with a further increase of the electrolyte concentration, there is only a slight decrease in the separated distance. Because the zeta potential of PMMA particles increases significantly along with the electrolyte concentration tested (see Fig. 4), the phenomena of decreasing separated distance results from the decreased repulsive force caused by the compression of the electric double layer. Based upon the results that there is only a slight decrease in separated distance as the KCl concentrations higher than 0.5 mM and the values of β_c at 0.5 mM are obviously larger compared to that at 1.0 mM, one can roughly predict that a denser packing of particles in cake will appear at 0.5 mM KCl concentration, which agrees with the measured results shown in Fig. 12.

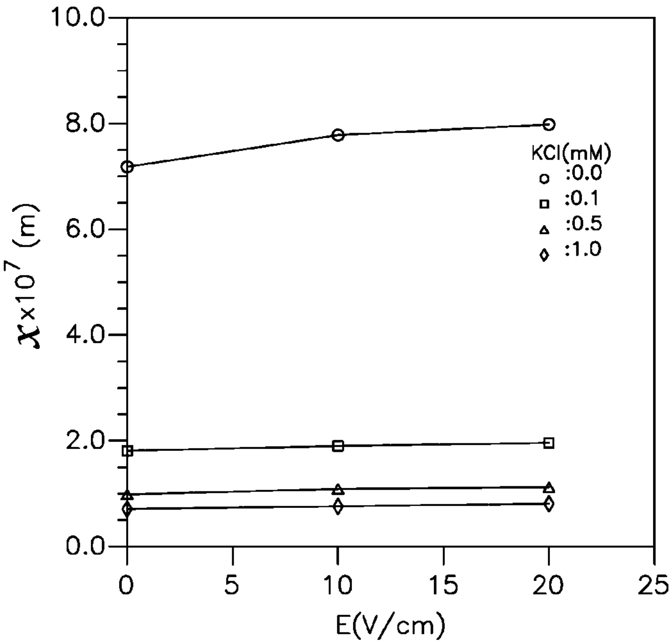

Figure 14. The separated distance between deposited particles under various KCl concentrations.

CONCLUSION

The characteristics of electrically enhanced crossflow microfiltration were analyzed and examined by using colloidal solutions under various electrolyte concentrations. It was shown theoretically and experimentally that in the region where the electric field is less than the critical strength, the filtration rate can be estimated by adding the electrophoretic velocity of the particles to the filtration rate obtained without an electric field. However, as the electric field applied is greater than the critical strength, the filtration rate is equal to the hydraulic permeation flux plus the electro-osmotic flux through the membrane.

Experimentally determined specific cake resistance showed that its values decrease with increasing the electric field strength due to a more porous structure of cake that is formed. Based on the models developed in the study for evaluating the critical friction angle and the equilibrium distance between particles deposited at cake surface, the prediction that a more compact cake would be formed at the 0.5 mM KCl agrees with the experimental results.

NOMENCLATURE

A_h	Hamaker constant (J)
D_p	particle diameter (m)
E	electric field strength (V/m)
F_e	electrophoretic force (kg m/s^2)
F_i	interaction force between particles (kg m/s^2)
H	separated distance between two approaching particles (m)
Δp	pressure drop (Pa)
q	filtration rate (m/s)
q_s	filtration rate at quasi-steady state (m/s)
v_e	electrophoretic velocity of particles (m/s)

Greek Letters

α_{av}	average specific filtration resistance of cake (m/kg)
β	angle of friction for depositing particles in crossflow filtration (rad.)
β_c	critical value for β (rad.)
χ	separated distance between deposited particles on cake surface (m)
ε	dielectric constant of liquid (F/m)
ε_{av}	average cake porosity
κ	Debye–Huckel parameter (m^{-1})
τ_w	shear stress acting on the filter surface (N/m^2)
ζ	zeta potential of the particles (V)
μ	viscosity (kg/m s)

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