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Application of an Electric Field for the Reduction of Particle and Colloidal Membrane Fouling in Crossflow Microfiltration

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Abstract

In crossflow microfiltration applications, the solution to be treated is generally made up of a wide range of particles and colloids. The presence of colloids complicates this process due to its intrinsic characteristics such as high surface charge, high stability, high specific surface area, and relative small size in comparison to the membrane pores. This permits them to create a film of colloids on the external and internal membrane surface due to the interfacial phenomenon effects (electrokinetic and adsorption). This colloidal fouling plays a significant role in increasing the resistance to the filtration flux in comparison to the external membrane fouling created only by the particles. The application of an electric field helps to reduce the membrane fouling due to colloids and particles, and thus results in an eventual improvement of the filtration flux.

INTRODUCTION

Crossflow electrofiltration (CF/EF) is a technique developed to minimize accumulation of particles and colloids present on the membrane surface in a crossflow microfiltration (CFMF) operation. This hybrid physical operation process, combining both the CFMF and electrophoretic separation techniques, is achieved by placing the membrane between two electrodes and, as in CFMF, the suspension to be

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filtered is circulated parallel to the membrane. By applying a dc electric field of sufficient strength and proper polarity, the fine colloids and particles can be made to migrate from the membrane surface. This mechanism of migration in the direction opposite to the convective pressure driven force will result in a clear boundary layer at the membrane/solution interface and will eventually lead to higher particle-free filtration flux.

A number of researchers have extensively investigated CF/EF in the solid/liquid separation process. Moulik et al. (1, 2) studied the separation of suspended bentonite particles in an electrofiltration cell and modified the conventional pressure filtration model equation by taking into account the change of filter medium resistance in an electrofilter under the influence of filter pore and cake electroosmotic effects. Later, Henry et al. (3) investigated the application of CF/EF for the suspension of kaolin clay and chemically destabilized oil and water emulsion, and they developed a mathematical model for CF/EF considering the total resistance to the flow as a function of film, medium, and cake resistance and distinguishing between the influence of shear force and electrical fields on the transport mechanisms. This reviewed model is generally considered to be the first approximate model for CF/EF. Alex Kuo (4) included the effect of particle radial migration in Henry's model.

Radovich et al. (5, 6) used a plate-and-frame-type electroultrafiltration module for the recovery of paints from rinse streams by modifying Henry's model for EUF flux involving gel layer formation in the module. However, Henry's model does not analyze the film resistance in terms of the formation of a clear boundary layer at sufficiently high electric field strength. Lee et al. (7, 8) developed another model for CF/EF by considering this clear boundary layer effect.

These previous studies with CF/EF using only particles in the feed solution have been done in the regime where there is no resistance to the permeate flux due to concentration and particle polarization, namely in the regime of $E > E_c$. This condition could be realized in microfiltration by working at low pressure, low crossflow velocity, and high E values: in this case the electrophoretic flow is of the same magnitude as the convective pressure-driven flow.

However, CFMF is usually operated in the pressure range of 0.25 to 2.0 bars within a crossflow velocity range of 0.5 to 5 m/s. Apart from this, in almost all practical application of CFMF a wide range of particles and colloids is present in the feed solution (9, 10). In this working condition, a high electric field value is needed in order to have electrophoretic flow in the same magnitude of convective pressure-driven flow, whereas it is laborious to work at $E > E_c$ without electroheating of the CF/EF cell.

The primary objective of this study is thus to examine the total resistance to the filtration flux in detail by taking into consideration the resistances due to the membrane, colloids, and particles separately. This will result in knowledge of the significance of each of the above-mentioned resistances present in the total resistance equation. In addition, we have studied the different operating parameters such as pressure, crossflow velocity, and electric field strength E in the $E < E_c$ regime of CF/EF, which will lead to a reduction of particle and colloidal membrane fouling and thus to an eventual increase in the flux. The work has been done in the $E < E_c$ regime due to the practical difficulties mentioned above.

The feed solution used in CF/EF is made up of both particles and colloids.

THEORY OF CROSSFLOW ELECTROFILTRATION

A. Resistance Analysis

The CF/EF flux J_{ef} can be represented by the sum of three independent filtration fluxes as

$$J_{ef} = J_{cf} + J_{ep} + J_{eo} \quad (1)$$

where J_{cf} is the CF/EF flux in the absence of an electric field ($E = 0$ V/cm).

At a given applied pressure P , J_{cf} can be written as (3)

$$J_{cf} = P/(\mu R_t) \quad (2)$$

R_t is the total resistance to filtration flux, expressed as the sum of three independent resistances, such as resistance of the filter medium (R_m), resistance due to external fouling of colloids (due to the concentration polarization of colloids and adsorption/deposition of colloids on the membrane surface) (R_{ef}), and resistance due to the deposition and formation of particle polarization on the membrane surface R_p . Thus

$$R_t = R_{ef} + R_m + R_p \quad (3)$$

R_m is the resistance of the membrane which depends on the characteristics of the filter medium at the initial stage of the filtration cycle. To take

into account the effect of internal membrane fouling due to fine colloids in a microfiltration, another resistance factor R_{if} is added to Eq. (3), which results in

$$R_t = R_{ef} + R_{if} + R_m + R_p \quad (4)$$

J_{ep} is the increase in filtration flux in a CF/EF due to the application of an electric field. This increment of flux is equal to the rate of electrophoretic migration of particles and fine colloids away from the membrane. It can be represented by

$$J_{ep} = U_{ep}E \quad (5)$$

In a CF/EF operation the electrophoretic migration of particles and colloids will lead to a reduction of R_{ef} , R_{if} , and R_p values and hence that of R_t . At the condition $R_t = R_m$, the applied electric field E is called the critical voltage, E_c . At E_c , the rate of electrophoretic migration of particles and colloids from the membrane is equal to the rate of migration of particles and colloids toward the membrane due to the convective pressure-driven force.

J_{eo} is the increment of filtration flux in a CF/EF due to the secondary effects such as electroosmosis, which can be expressed as

$$J_{eo} = (K_m E) + (K_c E) + (K_{if} E) \quad (6)$$

Here, K_m , K_c , and K_{if} are the electroosmotic constants of the filter medium, cake (due to the deposition of particles on the membrane surface), and internal colloidal fouling layer. These electroosmotic constants can have either positive or negative values depending on the surface charge of medium, cake, and internal fouling layer. However, in most practical applications of CF/EF, these constants are small, and the total J_{eo} flux is negligible in comparison to J_{ef} and J_{ep} .

B. Particle Trajectory Analysis

CF/EF could be operated in three regimes: 1) $E = E_c$, 2) $E > E_c$, and 3) $E < E_c$. The regime $E = E_c$ is obtained when the rate of migration of particles and colloids toward the membrane due to pressure-driven convective force is equal to the rate of migration of particles and colloids away from the membrane due to electrophoretic migration (neglecting the flux due to the electroosmotic effect).

In this working condition, as there will not be any concentration (due to colloids) or particle polarization at the membrane surface, there will be no diffusive transport. Figure 1 shows the different forces acting on a particle/colloid in a CF/EF at three different regimes of operation. At $E = E_c$, the convective pressure-driven force F_c is equal to the electrophoretic migration force F_{ep} . As a result, the particle/colloid will move in the direction of the shear force due to crossflow velocity F_t , which means that the particle/colloid will never be deposited on the membrane surface.

In the regime $E > E_c$, more of the particles and colloids will migrate away from the membrane, and the concentration at the membrane will be much less than that of the bulk solution. Hence, the diffusive transport (F_d) will be toward the membrane and because $F_{ep} > (F_c + F_d)$, the resultant particle trajectory will be away from the membrane.

At $E < E_c$, in contrast to the regime $E > E_c$, there will be a concentration and particle polarization at the membrane surface. In this condition the diffusive transport will be away from the membrane, and because this working condition will lead to $F_c > (F_{ep} + F_d)$, the resultant

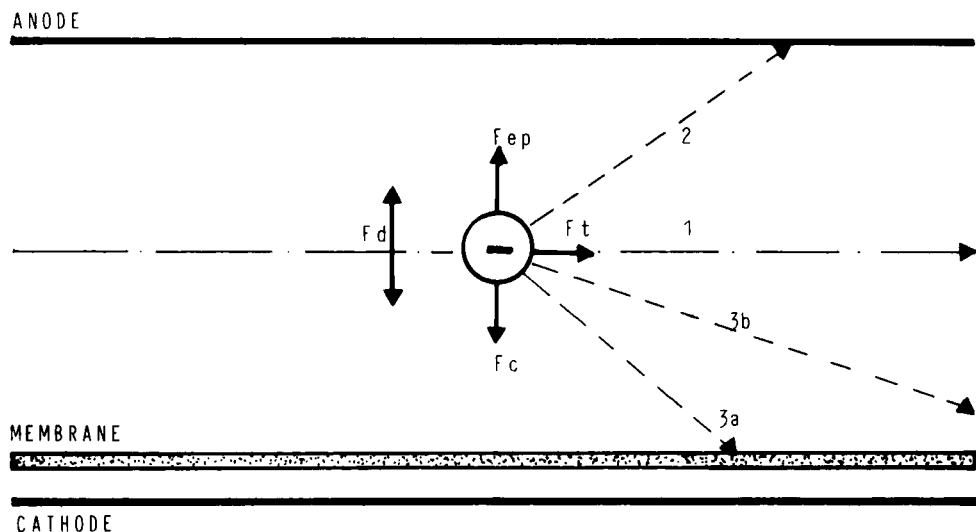


FIG. 1. Particle trajectory in a CF/EF cell. Trajectory 1: At $E = E_c$ where $F_c = F_{ep}$, $F_d = 0$. Trajectory 2: At $E > E_c$ where $F_{ep} > (F_c + F_d)$. Trajectory 3a: At $E < E_c$ where $F_c > (F_{ep} + F_d)$. Trajectory 3b: At $E < E_c$ where $F_c > (F_{ep} + F_d)$ but $F_t(3b) \gg F_t(3a)$ and $F_c(3b) \ll F_c(3a)$.

particle trajectory will be toward the membrane. However, by adopting appropriate working conditions such as high crossflow velocity (F_t) or reduced pressure (F_c), we can further reduce the particle/colloids deposition on the membrane surface as shown by trajectory 3b in Fig. 1.

EXPERIMENTAL

The experiments were carried out with a plate-and-frame-type CF/EF module, and it is pictured in Fig. 2. This module was constructed of Plexiglas, with a flow channel depth, width, and length of 0.5, 4, and 20 cm, respectively, and with platinum-coated titanium electrodes. All experiments were carried out using a Versapor-200 membrane (Gelman Science) with a pore size of 0.2 μm and a filtration surface area of 80 cm^2 . The model feed suspensions were made up with water from a Mill-Q (Millipore) system (with a resistivity of 18 Mohm \cdot cm and organic matter, particle, and colloid free water), with a known ionic strength containing 1.2 g/L Na_2HPO_4 . Here 3.0 g/L Na_2HPO_4 with a conductivity of 1.56×10^{-3} ohm \cdot cm was used as electrolyte.

RESULTS AND DISCUSSION

A. Total Resistance Analysis

In order to analyze R_t in terms of individual resistances, the experiments were carried out with the following four feed solutions.

(a) Mill-Q water at a pressure of 0.038 bar and a crossflow velocity of 0.5 m/s. This experiment permits calculation of the membrane resistance R_m , as there will not be any internal membrane fouling and particle and concentration polarization. The filtration flux is then obtained from the expression

$$J_{cf} = P/(\mu R_m) \quad (7)$$

(b) 100 mg/L colloid-free granular rounded silica particles (Serva Feinbiochemica) with a average diameter of 3.0 μm and negatively charged at pH = 9.45. The flux from these experiments can be calculated by

$$J_{cf} = P/[\mu(R_m + R_p)] \quad (8)$$

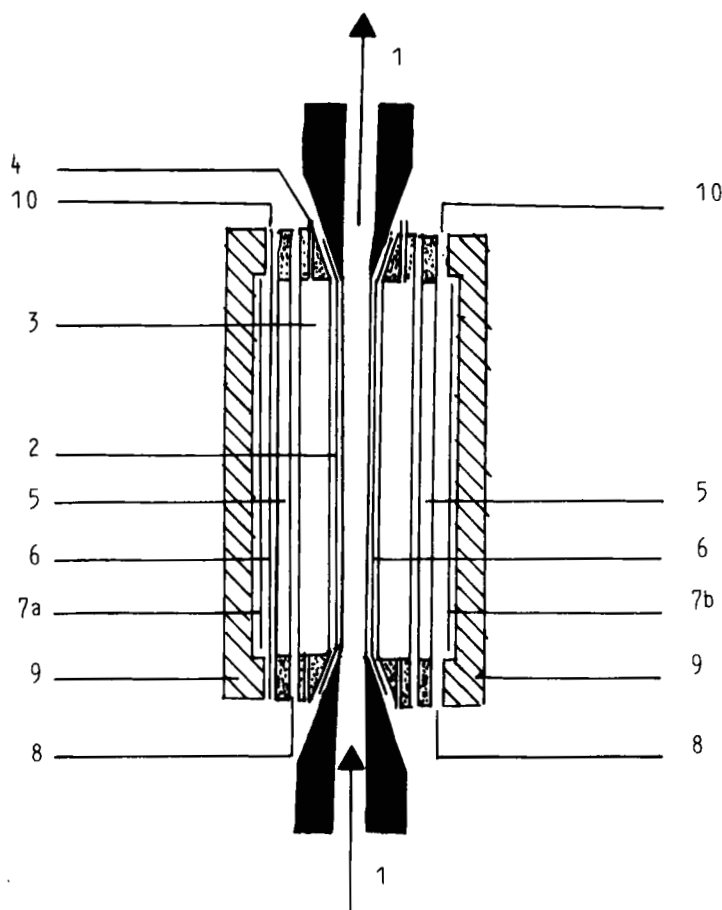


FIG. 2. Crossflow electrofiltration cell assembly. 1: Feed flow channel. 2: Microfiltration membrane. 3: Filtrate collection chamber. 4: Filtrate output. 5: Intermediate spacer. 6: Ion permeable cellulose membrane. 7a: Cathode. 7b: Anode. 8: Electrolyte input. 9: Electrode supporting plate. 10: Electrolyte output.

(c) 1.0 g/L Ludox HS-40% (E. I. du Pont de Nemours & Co.) colloids, with an average diameter of 12 nm, and negatively charged at pH = 9.45. As these colloids appear as nearly spherical in shape and very small in comparison to the membrane pore size, a fraction of the colloids is carried into the pores and thereby is deposited/adsorbed on its walls. So, in this condition the filtration flux can be given as

$$J_{cf} = P/[\mu(R_m + R_{ef} + R_{if})] \quad (9)$$

(d) 100 mg/L Silica particles and 1.0 g/L Ludox fine colloids. In this set of experiments all four independent resistances, R_m , R_{ef} , R_{if} , and R_p , contribute to the total resistance, and the equation for the filtration flux is

$$J_{cf} = P/[\mu(R_m + R_{ef} + R_{if} + R_p)] \quad (10)$$

Figure 3 shows the importance of the presence of colloids in the feed solution in CFMF. Note that the presence of fine colloids and particles in the feed solution increases R_t 3 to 4 times in comparison to the case when only particles are present in the feed. This phenomenon explains the significance of R_{if} and R_{ef} in the R_t equation. However, earlier studies (11) indicated that the internal fouling R_{if} is relatively insignificant in comparison to R_{ef} ($R_{if}/R_{ef} < 10\%$).

Table 1 presents the value of R_t calculated from the first three feed solutions and the experimental values determined from the fourth feed solution. Good correlation is found to exist between the calculated and the observed R_t values as shown in Fig. 4. It is important to note that $(R_{ef} + R_{if})$ contributes more than 70% to the R_t value in a CFMF operation, and that the resistance due to external membrane fouling of colloids (due to concentration polarization and deposition/adsorption of colloids) contributes a major part in the reduction of permeate flux. It indicates that in a CF/EF at $E < E_c$, where there will be external membrane fouling at the membrane surface, due importance should be given to the reduction of R_{ef} in order to increase the permeate flux.

B. Results of Crossflow Electrofiltration (CF/EF)

(a) Effect of Pressure and Crossflow Velocity on Filtration Flux

Figure 5 shows the relation between the permeate flux and pressure for different electric field strengths E . The increment of pressure for a given E

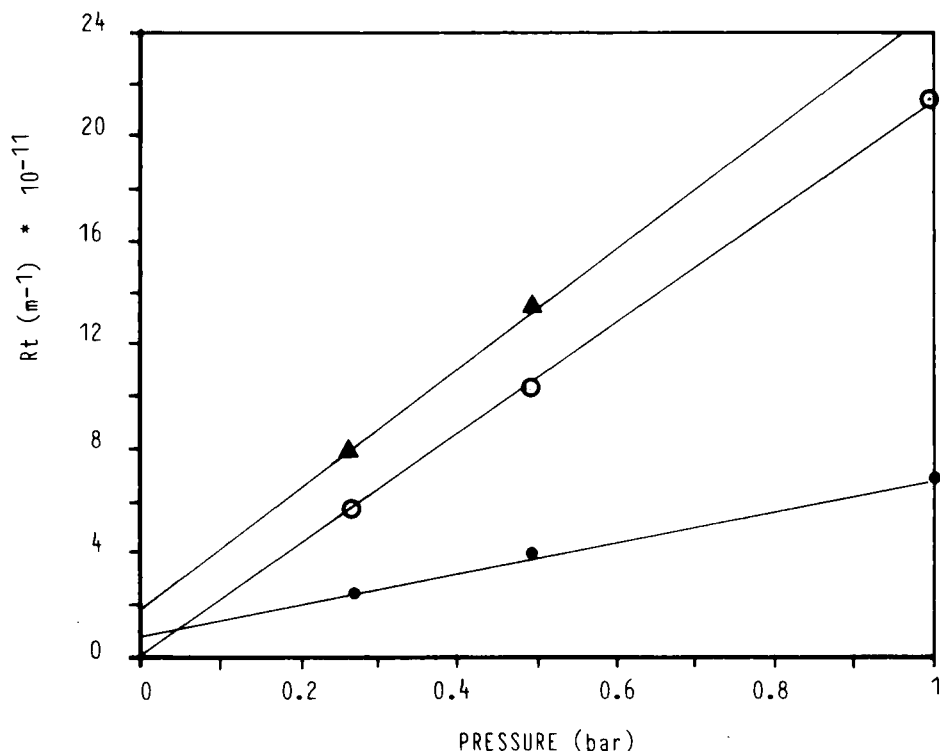


FIG. 3. Total resistance vs pressure for three different feed solutions of CFMF process ($V_f = 1.3$ m/s). (●) 100 mg/L silica particles, (○) 1.0 g/L Ludox colloids, (▲) 100 mg/L silica particles plus 1.0 g/L Ludox colloids.

value has no significant influence in the flux increment, indicating the presence of concentration/particle polarization on the membrane surface. The application of an electric field strength can lead to a reduction of these interfacial phenomena at the membrane/solution interface, and an eventual increment of filtration flux. Thus, as the field strength is increased from 0 to 14.45 V/cm, the flux is found to be increased three times. However, complete elimination of concentration/particle polarization was not achieved because the increase of electric field strength above 14.45 V/cm results in electroheating of the CF/EF cell.

The relation between crossflow velocity and filtration flux for different pressures is presented in Fig. 6. At $E = 0$ V/cm, the increase in pressure leads to a small increase in the filtration flux. On the other hand, when a certain electric field strength E is applied, the increase in pressure causes

TABLE 1
Calculated and Experimental R_t Values

Pressure (bar)	Crossflow velocity (m/s)	Resistance (m^{-1}) $\times 10^{-11}$							
		R_m^a (A)	$R_m + R_p^b$	R_p (B)	$R_m + R_{if} + R_{ef}^c$	$R_{if} + R_{ef}$ (C)	$R_t = R_m + R_{if} + R_{ef} + R_p$ (A + B + C)	Experimental R_t^d	$R_t/(R_{ef} + R_{if})$ (%)
0.27	0.5	0.3	2.76	2.46	9.17	8.87	11.63	13.5	76
	0.9	0.3	2.76	2.46	7.15	6.85	9.61	8.83	71.3
	1.3	0.3	2.33	2.03	5.68	5.38	7.71	7.8	69.8
0.5	0.5	0.3	4.88	4.58	16.67	16.47	21.35	23.38	78.4
	0.9	0.3	4.84	4.54	11.68	11.38	16.22	15.38	70.2
	1.3	0.3	3.89	3.59	10.23	9.93	13.82	13.53	71.9
1.0	0.5	0.3	9.05	8.75	33.64	33.34	42.39	43.37	78.7
	0.9	0.3	8.64	8.34	22.93	22.63	31.27	30.25	72.4
	1.3	0.3	6.75	6.45	21.3	21.0	27.75	24.8	75.7

^aFrom Eq. (7).

^bFrom Eq. (8).

^cFrom Eq. (9).

^dFrom Eq. (10).

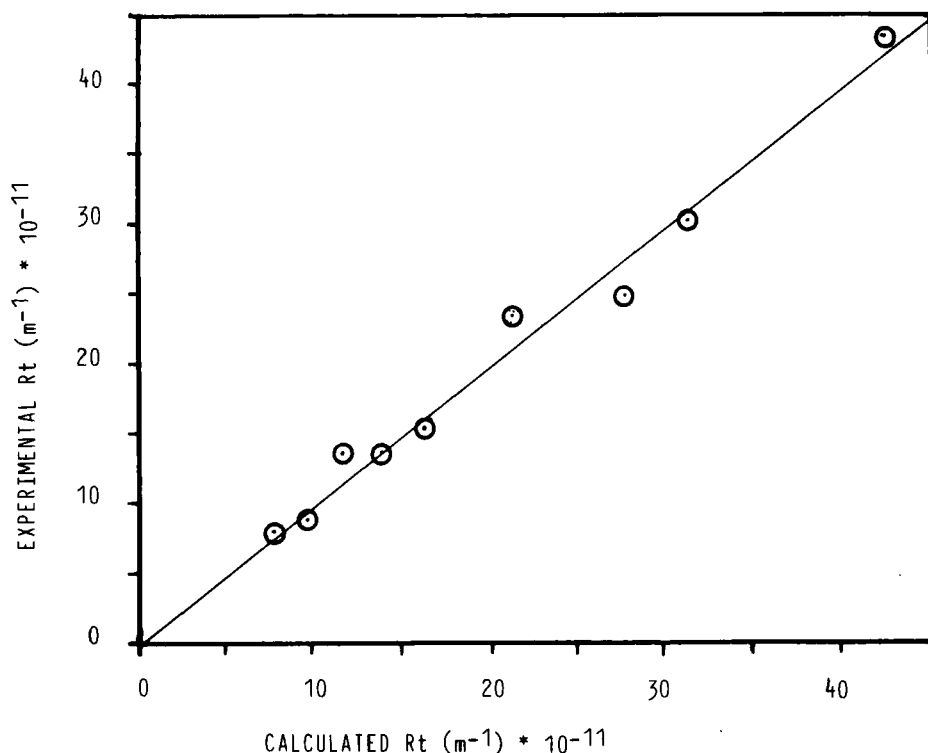


FIG. 4. The relation between calculated and experimental R_t values.

a small decrease in filtration flux. This reduction of flux can be explained by particle trajectory analysis (Fig. 1). For a constant crossflow velocity (F_t is constant) and a fixed E value (F_{ep} remains constant), the increase in pressure results in a higher convective pressure force F_c . In this case, more of the particles and colloids tend to move toward the membrane as the pressure is increased. This phenomenon permits an increase in the concentration and particle polarization, and an eventual increase of R_{ef} and R_p .

(b) Effect of Electric Field Strength on Filtration Flux

Figure 7 shows the effect of electric field strength at a particular pressure on the filtration flux for a feed solution of 100 mg/L silica

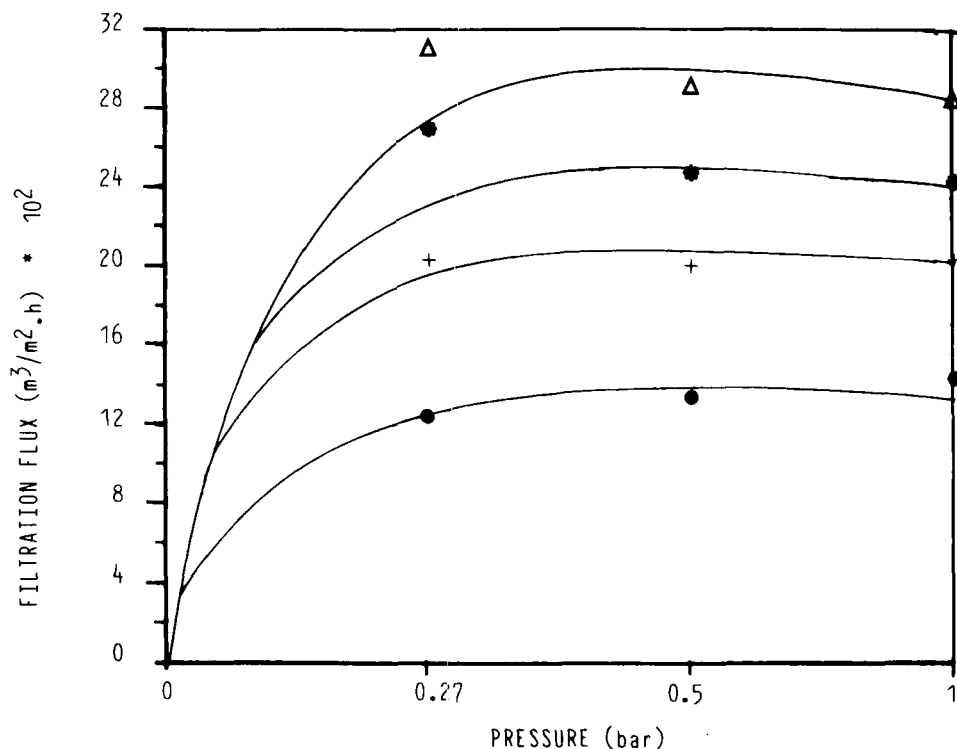


FIG. 5. Filtration flux vs pressure for different electric field strengths for a feed solution containing colloids and particles. $V_f = 1.3$ m/s. (●) 0 V/cm, (+) 7.22 V/cm, (*) 10.84 V/cm, (Δ) 14.45 V/cm.

particles plus 1.0 g/L Ludox colloids. A linear relation exists between E and the filtration flux for a particular crossflow velocity, but the increment of crossflow velocity further increases the filtration flux while the slope remains almost constant. At a working pressure of 0.27 bar, an increase in E from 0 to 14.45 V/cm increases the flux by four times, whereas an increase in crossflow velocity from 0.5 to 1.3 m/s leads to an increase of the flux of only 1.5 times.

Neglecting the electroosmotic effects, the electrophoretic mobility can be calculated from Eqs. (1) and (5). The electrophoretic mobility for Ludox plus silica particles has been calculated to be $3.827 \times 10^{-4} \text{ cm}^2/\text{V} \cdot \text{s}$. This value is close to that determined from the Zeta Meter

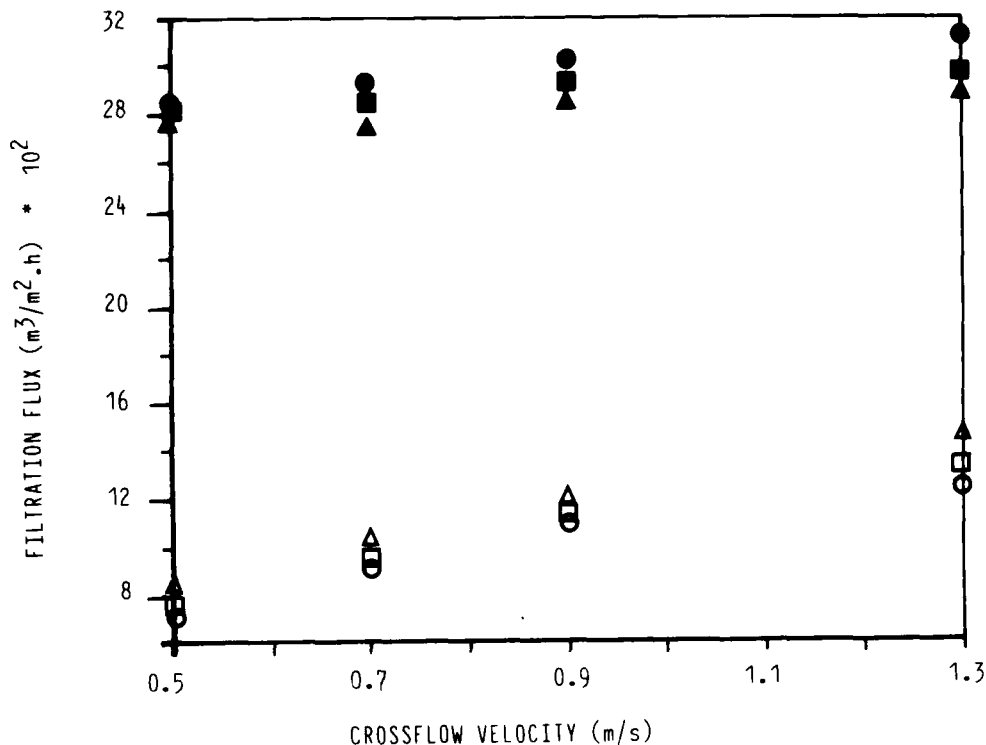


FIG. 6. Filtration flux vs crossflow velocity relation for different applied pressure. At $E = 0$ V/cm: (O) 0.27 bar, (□) 0.5 bar, (△) 1.0 bar. At $E = 14.45$ V/cm: (●) 0.27 bar, (■) 0.5 bar, (▲) 1.0 bar.

$(3.24 \times 10^{-4} \text{ cm}^2/\text{V} \cdot \text{s})$ while observing the movement of the silica particles only.

CONCLUSION

In the domain of membrane technology, CFMF is found to be an appropriate technology capable of replacing the conventional filtration process in the field of solid/liquid separation. However, at present the application of this technology is limited due to internal and external membrane fouling problems related to the colloids. The presence of colloids in the feed solution plays a significant role in flux reduction by

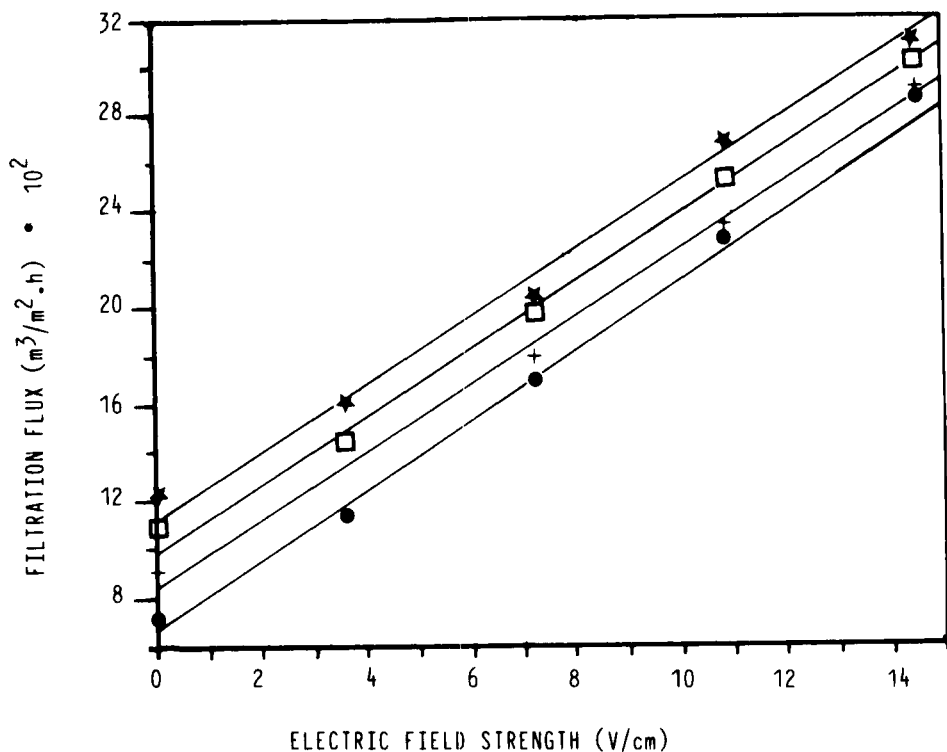


FIG. 7. Effect of filtration flux on crossflow velocity and electric field strength at an applied pressure of 0.27 bar. Feed solution containing colloids and particles: (●) 0.5 m/s, (+) 0.7 m/s, (□) 0.9 m/s, (*) 1.3 m/s.

contributing a major part in the increment of the total resistance to the flux. To have an economically optimal level of filtration flux in a CFMF system, the following antifouling techniques have to be considered: high crossflow velocity, regular membrane backwashing, application of an electric field (CF/EF), etc. Among these techniques, the CF/EF process offers great promise in reducing the fouling problems related to charged colloids and particles. In this process the filtration flux is found to increase linearly with the applied field strength, and it can be predicted by using the experimental zeta potential values of colloids and particles. In the regime of $E < E_c$ for a particular crossflow velocity and electric field strength value, an increase of pressure will lead to a small reduction

of the filtration flux. Hence, the optimum working condition in a CF/EF for a given crossflow velocity can be obtained in the $E < E_c$ regime with the highest permissible electric field strength value without electro-heating of the cell and at a low pressure.

SYMBOLS

CF/EF	crossflow electrofiltration
CFMF	crossflow microfiltration
E	electric field strength (V/m)
E_c	critical electric field strength (V/m)
F_c	convective pressure-driven force
F_d	diffusive transport force
F_{ep}	electrophoretic migration force
F_t	shear force due to crossflow velocity
J_{ef}	crossflow electrofiltration flux (m/s)
J_{ef}	crossflow electrofiltration flux at $E = 0$ V/cm (m/s)
J_{eo}	electroosmotic flux (m/s)
J_{ep}	electrophoretic flux (m/s)
K_m	electroosmotic constant of the filter medium ($\text{m}^2/\text{V} \cdot \text{s}$)
K_c	electroosmotic constant of the filter cake ($\text{m}^2/\text{V} \cdot \text{s}$)
K_{if}	electroosmotic constant of the internal colloidal fouling layer ($\text{m}^2/\text{V} \cdot \text{s}$)
P	pressure (N/m^2)
R_t	total resistance to the flux (m^{-1})
R_{ef}	external colloidal membrane fouling resistance (m^{-1})
R_{if}	internal colloidal membrane fouling resistance (m^{-1})
R_p	particle membrane fouling resistance (m^{-1})
R_m	initial membrane resistance (m^{-1})
U_{ep}	electrophoretic mobility ($\text{m}^2/\text{V} \cdot \text{s}$)
V_t	crossflow velocity (m/s)
μ	viscosity of the feed solution ($\text{kg}/\text{m} \cdot \text{s}$)

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