Pulsed Electrophoretic Filter-Cake Release in Dead-End Membrane Processes

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The application of electric fields can be a very effective means of removing deposits from membrane surfaces. Such a means of process improvement has usually been applied to cross-flow filtration, allowing efficient operation at low cross-flow velocities. In the limiting case of dead-end filtration (zero crossflow) it is possible to use electric-field pulses to release the filter cake for collection. Experimental data are presented for the dead-end ultrafiltration of silica colloids and the protein bovine serum albumin and for the dead-end microfiltration of titania dispersions taking as variables the magnitude of the applied field, the pulse interval, the pulse duration, and the feed conditions (pH, ionic strength, concentration). The data identify the conditions when pulsed electric fields can be used as an efficient means of releasing filter cakes for collection. A force balance model is developed to predict the filtration rate at the end of the release process, taking into account electrophoretic, electroosmotic, and hydrodynamic forces. The model shows excellent agreement with the experimental data for ultrafiltration and reasonable agreement with the experimental data for microfiltration. The use of pulsed electric fields to release filter cakes in dead-end membrane processes is a promising technique that is most likely to find application in the clarification of process feeds containing low dispersed solutes.

Introduction

Membrane microfiltration and membrane ultrafiltration are means of separating particles in the size ranges 10–1.0 μm and 0.1 μm–5nm, respectively. These processes find widespread industrial use in the processing of both inorganic and biological materials. Their classification shows that they are usually thought to separate materials according to size. However, all particles, colloids, and biological materials dispersed in solution acquire a surface charge due to ion dissociation, ion adsorption, or ion dissolution. Proton equilibria at the surface are especially important in aqueous solution. Application of applied electric fields can then allow manipulation of the relative movement of particles and solvent by means of electrophoresis or electroosmosis (Hunter, 1981). Electrophoresis is the transport of a charged surface relative to a liquid in an electric field, for example, the movement of particles. Electroosmosis is the transport of a liquid relative to an immobile charged surface by an electric field, for example, the movement of an electrolyte solution through a membrane pore or filter cake. Application of such fields during a filtration process has given rise to a family of processes that may be collectively termed electrofiltration.

Some of the earliest applications (Krishnaswary and Klinkowsi, 1986; Bowen, 1992; Jagannadh and Muralidhara, 1996) of electrofiltration were in "dead-end filtration." Here the deposition of particles was enhanced by electrophoretic deposition and/or the solids content of the filter cake was increased by electroosmotic dewatering. Such processes require the continuous application of electric fields. With the advent of cross-flow membrane filtration processes, the emphasis in the development of electrofiltration changed to the use of electrophoresis to reduce concentration polarization and membrane deposition (Henry et al., 1977). Such approaches are also applicable to biological materials (Yukawa et al., 1983; Iritani et al., 1992). However, the continuous application of electric fields also required with these processes has several disadvantages including a high-energy requirement, substantial heat production, and changes in the process stream due to reactions at the electrodes.

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Materials deposited on membrane surfaces often retain a surface charge. Therefore, it is possible to remove such materials from the surface by periodic application of electric-field pulses. Such techniques have been applied to cross-flow processes (Bowen and Sabuni, 1991, 1992; Robinson et al., 1993; Wakeman and Sabri, 1995). Application of electric-field pulses, often for only a small fraction of the overall process time, avoids many of the disadvantages of continuous-field application. Application of alternating or pulsed fields can also clean membranes in crossflow by inducing electroosmotic backwashing (Spiegler and Macleish, 1981; Visvanathan and Ben Aim, 1990; Bowen and Sabuni, 1992; Bowen and Sabuni, 1994). Once it has been demonstrated that application of electric fields can remove deposits from membrane surfaces effectively, it becomes possible to reduce the cross-flow velocity while still operating the process at acceptable filtration rates (Bowen and Sabuni, 1991). In effect, such processes are operating efficiently by inputting electrical energy where it is needed, at the membrane surface, rather than at the more remote location of the pump.

The aim of the present article is to present an experimental study and theoretical assessment of the limiting case (regarding reduction in cross-flow velocity) of application of electric-field pulses to remove materials deposited on membrane surfaces in *dead-end* filtration. Data are presented for the dead-end ultrafiltration of silica colloids and the protein bovine serum albumin (BSA) and for the dead-end microfiltration of titania dispersions, taking as process variables the magnitude of the applied electric field, the pulse interval, the pulse duration, and the feed conditions (pH, ionic strength, concentration). A force balance model is developed to predict the filtration rate at the end of the filtration process, taking into account electrophoretic, electroosmotic, and hydrodynamic forces. It is shown that effective filtration processes for the clarification of low dispersed solutes feeds can be developed by combining electrical filter-cake release with synchronous collection of the released materials. Given the renewed international interest in dead-end membrane technology for water treatment, development of such processes could be especially timely.

Theoretical Analysis

A theoretical analysis of the result of the application of an electric-field pulse in dead-end filtration can be formulated in terms of the forces acting on a particle on the surface of the filter cake. We have previously presented such an analysis for the case of cross-flow filtration (Bowen and Sabuni, 1992). This analysis is presented here in outline form and modified for the case of dead-end filtration. The analysis is also extended to provide an analytical expression that predicts the rate of filtration expected immediately after application of an electrical pulse.

The criterion for removal of a particle from the surface of the filter cake during the application of an electric field pulse is as follows:

$$F_y + F_e < 0, \tag{1}$$

where F_y is the viscous drag on the particle due to the flow of permeate and F_e is the electrophoretic force, where forces are defined as positive if they act toward the membrane. The direction of the filtration and effects acting during the application of the electric field are shown in Figure 1.

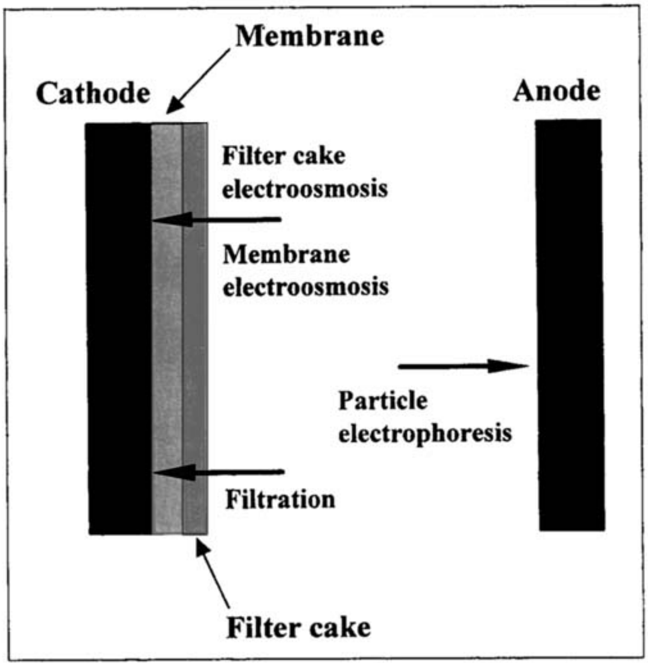

Figure 1. Effects acting during the application of electric-field pulses.

The electrophoretic force may be written

$$F_e = -fu_pE = -fu_pI/\lambda_o, \tag{2}$$

where the electric-field gradient (E) may be calculated from the current density (I) and the conductivity of the bulk solution (λ_0) (Bowen and Sabuni, 1992).

Calculation of the viscous force is more complex, as the effects of electroosmosis in the membrane and filter cake must be considered (Henry et al., 1977). The force may be calculated from

$$F_y = f\Delta P/\mu(R_m + R_c), \tag{3}$$

where it may be shown that (Bowen and Sabuni, 1992)

$$R_m = R_{om}/(1 + v_{eom}I/v_{om}) \tag{4}$$

and

$$R_c = R_{oc}/[1 + (u_pI/\lambda_o v_{oc})]. \tag{5}$$

During application of the electric field, particles are removed from the filter cake and the filtration rate increases. This removal process continues until the hydrodynamic and electrical forces become balanced. Thus, the criterion for no further removal of particles is

$$F_y + F_e = 0. \tag{6}$$

In the present article, such an analysis is taken one step further to the prediction of the filtration flux immediately after an electric pulse has been applied long enough to meet the criterion in Eq. 6. This flux (v_{end}) can be written in terms of the necessary equality:

$$v_{om} = v_{oc} = v_{\text{end}}, \tag{7}$$

where

$$v_{\text{end}} = \Delta P/\mu(R_{om} + R_{oc}). \tag{8}$$

Substituting Eqs. 2 and 3 in Eq. 6, then using Eqs. 4 and 5 to substitute for R_m and R_c, Eq. 7 to replace v_{om} and v_{oc} with v_{end}, and Eq. 8 to express R_{oc} in terms of R_{om} gives

$$v_{\text{end}} = (\mu/\Delta P)\left(u_p I^2/\lambda_0\right)\left(R_{om}u_p/\lambda_o - R_{om}v_{eom}\right) - Iv_{eom}. \tag{9}$$

All of the variables on the righthand side of Eq. 9 are readily accessible. Hence, the equation allows explicit calculation of the effect of the electric-field pulse. Such prediction will be compared with experimental data later in the article. The equation is valid as long as $v_{\text{end}} \leq \Delta P/\mu R_{om}$. That is, the clean-water flux of the membrane cannot be exceeded.

Materials and Methods

Filtration experiments were carried out in a flat-sheet dead-end filtration module constructed in our department. The overall dimensions of the process feed chamber were 11.2 cm × 3.4 cm × 0.20 cm with an effective membrane area of 23 cm^2. The membrane was supported on a stainless-steel sheet of 100-μm pore size, which was in turn supported on a stainless-steel mesh. Both sheet and mesh form the cathode. The anode was a platinized titanium sheet mounted flush with the other side of the process feed chamber.

Experiments were controlled and monitored by a PC fitted with an IEEE interface. The power supply used for applying the electric field (Farnell Instruments Ltd. AP100-30) was linked directly to this interface. This power supply uses switching techniques that result in a fast rise time—0.6 s under the experimental conditions—and a fall time of 1.0 s. A pH probe, temperature probe, and pressure transducers were linked via an A/D converter. Filtration rates were determined for the ultrafiltration experiments by using an electronic balance (Salter Ltd. EK-1200A) linked via a RS232 interface. Filtration rates for microfiltration were determined using an automated level sensing device (Bowen et al., 1989). The system reservoir held up to 3 L of feed. For ultrafiltration experiments the system was pressurized with nitrogen gas, and for microfiltration experiments the filtration was driven by a centrifugal pump (Micropump, 445), in both cases at a pressure of 100 $\text{kN}\cdot\text{m}^{-2}$.

In the operation of the filtration cell, a valve on the feed side of the membrane module opened automatically after the application of the electric-field pulse to allow collection of the released cake. Pressurization was maintained during this process, and the volume discharged was equivalent to the volume of the feed chamber.

The membranes used for ultrafiltration were Nadir (polyethersulphone, Hoechst AG) with a molecular-weight cutoff (MWCO) of 30,000. The membranes used for microfiltration had a specified pore size of 0.2 μm (polysulphone, Dow Danmark). The electrokinetic properties of the membranes were determined using a computerized electroosmotic technique (Bowen and Clarke, 1984). The ultrafiltration experiments were carried out with Syton W30 colloidal silica (SiO_2) (Monsanto) or bovine serum albumin fraction V (BSA) (Sigma Chemical Co.). The microfiltration experiments were carried out with technical-grade titania (TiO_2) (BDH). Electrophoretic mobilities of all three test systems were determined using a Malvern Zetamaster (Malvern Instruments). The mean size of the silica and titania were determined by photon correlation spectroscopy using a Malvern Instruments Lo-C with a 75-mW Ar laser. The equivalent spherical hydrodynamic radius of BSA was calculated from the specific volume (Cantor and Schimmel, 1980) to be 3.2 nm (Bowen and Williams, 1996). For filtration experiments, silica and titania were dispersed in KNO_3 in a range of ionic strengths and pH (adjusted using HCl or KOH). Silica and titania concentrations of samples taken during filtration experiments were determined by evaporation to dryness. BSA filtration experiments were carried out in phosphate buffer, and protein concentrations were determined by UV adsorption at 280 nm.

Experimental Results and Discussion

Characterization of the membranes and colloids

The interpretation of the results and the use of Eq. 9 requires a knowledge of the specific electroosmotic flow rate (v_{eom}) of the membranes. This can be obtained by direct measurement of the rate of electroosmosis at the membranes in appropriate electrolytes (Bowen and Clarke, 1984) but in the absence of particles. Such data are presented in Table 1. For the ultrafiltration membrane the data are presented for both KNO_3 solutions (as used for silica colloids) and phosphate buffer (as used for BSA). In both cases the specific electroosmotic flow rates are negative and increase in magnitude as the ionic strength is decreased or the pH is lowered (except for pH 3 in KNO_3). The variation with ionic strength is much greater for the phosphate buffer, but the variation with pH is less in that case. Similar trends are also found for the microfiltration membrane.

Equation 9 also requires a knowledge of the electrophoretic mobility of the particles (u_p), which may be determined using commercial equipment. Data are given in Table 2. For the two inorganic systems studied, the magnitude of the electrophoretic mobility is maximum in a 0.01-M solution, decreasing at higher and lower ionic strengths. The mobility of BSA increases in magnitude as the ionic concentration is lowered. For all three systems, the magnitude of the mobility increases with increasing pH. Also shown in the table are the mean diameters of the particles obtained by photon correlation spectroscopy. Although the particle size is not required by Eq. 9, a knowledge of particle size is useful, as significant aggregation would influence the filtration behavior. The data show that no aggregation of SiO_2 occurred under the conditions studied. However, there is evidence of aggregation of TiO_2 at the highest ionic strength and to a lesser degree at the lowest pH used. The size of BSA puts it at the lower limit of the instrument available, so direct size measurements have not been quoted. However, no BSA aggregates (which would have been within the size range of the instrument) were detected for any of the conditions studied.

Table 1. Electroosmotic Properties of Membranes for the Various Solution Conditions Used for the Filtration Experiments

(a) Ultrafiltration Membrane

At Constant pH = 8 in KNO_3		At I = 0.01 M in KNO_3	
I	v_{eom} $(\mu m \cdot s^{-1}/A \cdot m^{-2}) \times 10^2$	pH	v_{eom} $(\mu m \cdot s^{-1}/A \cdot m^{-2}) \times 10^2$
0.1	−0.32	3	−1.92
0.01	−0.90	4	−2.99
0.001	−2.59	6	−2.56
0.0001	−5.16	8	−0.90

At Constant pH = 8 in Phosphate Buffer		At Constant Phosphate Buffer Concentration (0.01 M)	
Concentration (M)	v_{eom} $(\mu m \cdot s^{-1}/A \cdot m^{-2}) \times 10^2$	pH	v_{eom} $(\mu m \cdot s^{-1}/A \cdot m^{-2}) \times 10^2$
0.1	−0.33	5	−3.17
0.01	−2.10	6	−2.70
0.001	−5.11	7	−2.25
0.0001	−12.5	8	−2.10

(b) Microfiltration membrane

At Constant pH = 8 in KNO_3		At I = 0.01 M in KNO_3	
I	v_{eom} $(\mu m \cdot s^{-1}/A \cdot m^{-2}) \times 10^2$	pH	v_{eom} $(\mu m \cdot s^{-1}/A \cdot m^{-2}) \times 10^2$
0.1	−0.20	4	−1.13
0.01	−0.53	6	−0.94
0.001	−4.89	8	−0.53
0.0001	−20.6		

v_{eom} = specific electroosmotic flow rate.

Table 2. Electrophoretic Properties and Size (Mean Diameter) of Particles for the Various Solution Conditions Used for the Filtration Experiments

(a) SiO_2

At Constant pH = 8 in KNO_3			At I = 0.01 M in KNO_3		
I	Size (nm)	Mobility	pH	Size (nm)	Mobility
0.1	67.0	−3.26	3	69.1	−0.39
0.01	68.4	−4.90	4	69.1	−1.11
0.001	70.1	−4.69	5	68.9	−1.91
0.0001	70.9	−4.52	6	68.9	−3.15
			8	68.4	−4.90

(b) BSA

At Constant pH = 8 in Phosphate Buffer			At Constant Phosphate Buffer Concentration		
Conc.	Size (nm)	Mobility	pH	Size (nm)	Mobility
0.1		−0.01	5		−0.70
0.01		−1.95	6		−1.11
0.001		−1.89	7		−1.83
0.0001		−2.07	8		−1.95

(c) TiO_2

At Constant pH = 8 in KNO_3			At I = 0.01 M in KNO_3		
I	Size (nm)	Mobility	pH	Size (nm)	Mobility
0.1	398	−2.70	4	195	−1.51
0.01	153	−3.79	6	166	−3.96
0.001	143	−3.64	8	153	−3.79
0.0001	143	−3.26			

*Units of mobility are $(\mu m\ s^{-1}/V\ m^{-1}) \times 10^2$.

Ultrafiltration of SiO_2

Typical results showing the effects of applied electric-field pulses on filtration flux in the dead-end ultrafiltration of silica colloids are shown in Figure 2. Figure 2A shows the effects of increasing the applied voltage for 10-s pulses at 20-min intervals for 5 g L^{-1} SiO_2 at pH 8 in a 10^{-2}-M KNO_3 solution. With an applied voltage of 12.5 V there is little effect, since such a voltage is below the threshold for removal of particles from the filter cake. However, increasing the voltage to 50 V or 100 V gives substantial increases in flux immediately after the application of each pulse, showing that the filter cake has been released from the surface of the membrane. This cake is then collected by discharge from the feed compartment. The flux subsequently declines with time as a new cake builds up. Figure 2B shows the effects on the filtration flux of increasing the interval between pulses of 10 s at 100 V under identical solution conditions. As the interval is increased from 10 to 40 and then 60 minutes, so the filter cake builds up to a greater extent and the flux declines further. Even so, application of an electric-field pulse gives a substantial increase in flux as the cake is released from the membrane surface.

Table 3 contains more information on the overall process performance for selected operating (Table 3a) and feed (Table 3b) conditions. The average filtration rate increases as the applied voltage is increased, as the pulse interval is decreased, and generally as the pulse duration is decreased, though with an indication of an optimum duration of 7.5 s. The average (for all pulses) recovery of deposited particles successfully collected in the discharge increases in a parallel fashion and reaches 100% for the highest applied voltages and shortest pulse intervals. (Small errors inherent in the analysis procedure are responsible for the percentages slightly in excess of 100%.) The highest concentration factors (concentration in collected discharged cake/feed concentration) are obtained at the highest voltages, longest pulse intervals, and shorter pulse durations.

With variation of feed conditions, the greatest average filtration rates are obtained at the highest pH values and the lowest ionic strengths, trends that generally follow the electrophoretic mobility data in Table 2. As expected, the average filtration rates increase as the feed concentration decreases. The average cake recovery reaches 100% for the highest pH values and the highest feed concentrations. The latter suggests that a small fraction of particles may be irreversibly adhering to the membrane, perhaps an initial layer, this being most noticeable at lower feed concentrations. This suggestion is supported by the increase in percentage of cake recovery measured for successive pulses at low feed concentrations—recovery for the first two pulses may be less than half that of subsequent pulses. There is an indication in the data that the cake recovery is highest in a 0.01-M solution, though the values at lower ionic strength are also high. The cake concentration factor increases with increased pH, decreased ionic strength and decreased feed concentration.

Ultrafiltration of BSA

The success of pulsed filter-cake removal in the ultrafiltration of silica particles as described in the preceding section is

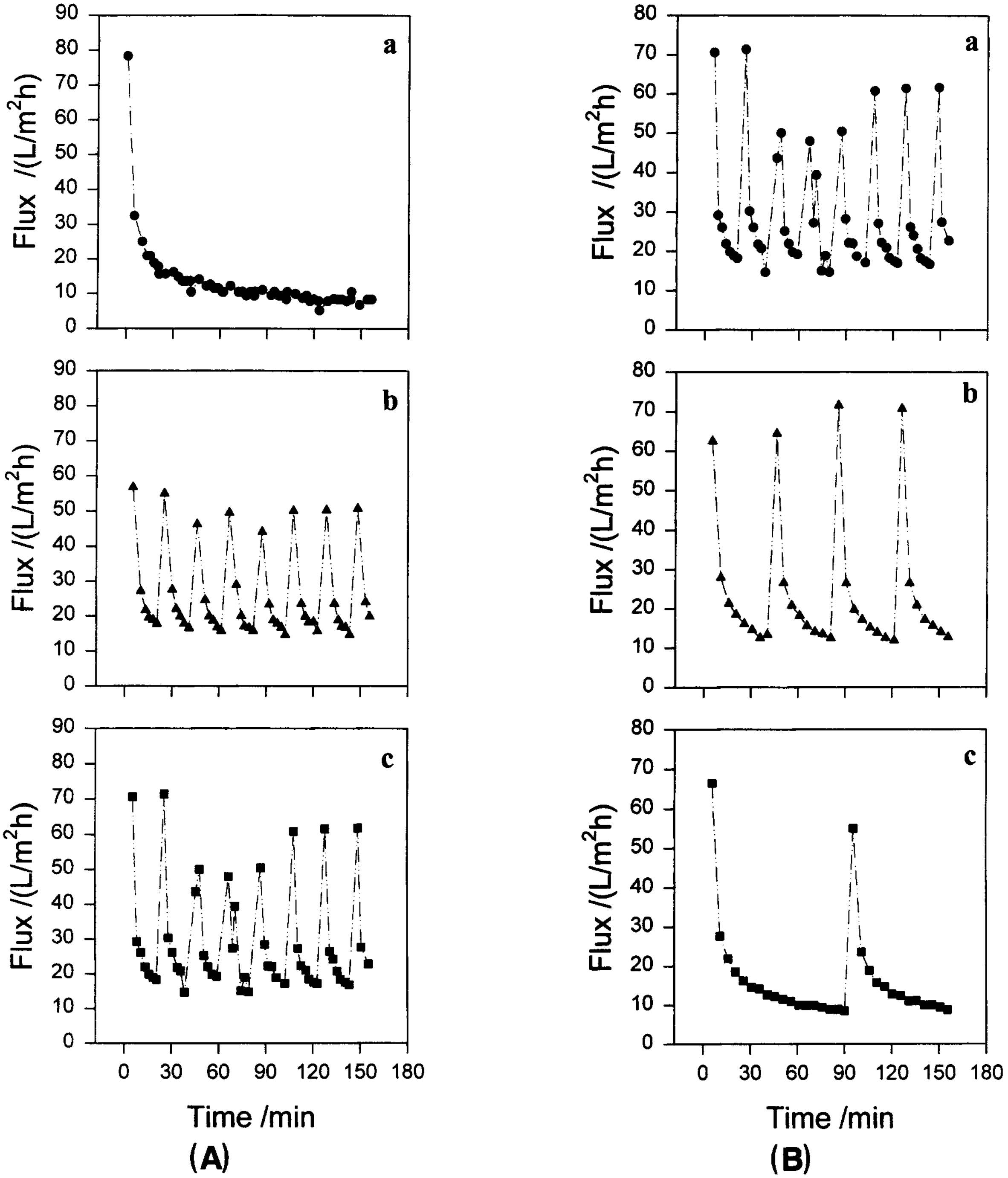

Figure 2. Filtration flux as a function of time in the pulsed electrophoretic dead-end ultrafiltration of 5 g L^{-1} SiO_2; pH 8.0; 0.01-M KNO_3; 10-s pulses.

(A) Effect of variation of applied voltage, 20-min interval. (a) 12.5 V; (b) 50 V; (c) 100 V. (B) Effect of variation of pulse interval, at 100 V. (a) 20 min; (b) 40 min; (c) 90 min.

encouraging. However, many industrial ultrafiltration processes require the separation of softer materials, in particular biological materials such as proteins. It is therefore of interest to test the applicability of the pulsed field process with such materials, and BSA has been chosen as a typical example protein that has been widely used for filtration studies.

Figure 3A shows typical results of increasing the applied voltage for 7.5-s pulses at 20-min intervals for 1 g L^{-1} BSA at pH 8.0 in a 0.01-M phosphate buffer. Even at an applied voltage of 12.5 V there is a significant increase in flux, indicating filter-cake release. As the applied voltage is increased, the increase in flux after each pulse becomes more marked. Figure 3B shows the effects on the filtration flux of increasing the interval between pulses for 7.5 s of 100 V under identical solution conditions. As was observed for silica, as the interval is increased, the filter cake builds up to a greater

Table 3a. Variation of Operating Conditions in the Filtration of SiO_2*

Voltage (V)	Interval (min)	Duration (s)	Avg. Filt. Flux ($L \cdot m^{-2} \cdot h^{-1}$)	Avg. Cake Recovery (%)	Cake Conc. Factor
(a) *Variation of applied voltage*					
12.5	20	10	11.0	73	1.1
25	20	10	12.3	74	1.3
50	20	10	29.3	97	2.9
100	20	10	33.4	101	3.0
(b) *Variation of pulse interval*					
100	10	10	45.9	102	2.5
100	20	10	33.4	101	3.0
100	40	10	24.9	91	3.4
100	90	10	16.6	69	3.6
(c) *Variation of pulse duration*					
100	20	3	18.1	78	3.1
100	20	7.5	19.3	77	3.3
100	20	15	16.1	72	2.6
100	20	40	14.5	68	2.3

*5 g/L, SiO_2; 0.01 M, KNO_3; pH 8.0

extent and the flux declines further, but application of an electric-field pulse substantially increases the flux.

Table 4 contains more information on the overall process performance for selected operating (Table 4a) and feed (Table 4b) conditions for the filtration of BSA. The results broadly parallel those found for silica. Thus, the average filtration rate increases as the applied voltage is increased, as the pulse interval is decreased, and generally as the pulse duration is decreased, again with an optimal duration of 7.5 s. Such an interval can be interpreted as representing a balance between the minimum time required for effective movement of the cake when an electric field is applied and the loss of filtration time for long pulses coupled with possible changes to the contents of the filtration cell caused by the release of electrolysis products. The average recovery of deposited protein successfully collected in the discharge increases in a similar manner. The maximum reached is 96%, suggesting that irreversible deposition of BSA occurs to a greater extent than a similar deposition of silica. Cake concentration factors increase with increased applied voltage, increased pulse interval, and show a decrease for the longest and shortest pulse durations.

The average filtration rates are greatest at the highest pH, paralleling the mobility data of Table 2. Variation in the average filtration rates with ionic strength is more complex, with a maximum at 0.001 M. Average cake recoveries are again high, though less than 100%, except for the lowest feed concentration studied. In contrast to the case of silica, average cake recovery decreases as the concentration increases. This may reflect some irreversible aggregation on the membrane surface at the highest concentration used. The cake concentration factor increases with increased pH and decreased feed concentration, with a more complex pattern as the ionic strength varies.

Microfiltration of TiO_2

The membrane microfiltration of dilute dispersions of particles is also an important industrial requirement that would benefit from improved operation in the dead-end mode. Ex-

Table 3b. Variation of Feed Conditions in the Filtration of SiO_2*

pH	I (M)	Conc. (g/L)	Avg. Filt. Flux ($L \cdot m^{-2} \cdot h^{-1}$)	Avg. Cake Recovery (%)	Cake Conc. Factor
(a) *Variation of pH*					
3	0.01	5	19.7	91	2.0
4	0.01	5	26.9	94	2.6
6	0.01	5	29.9	96	2.7
8	0.01	5	33.4	101	3.0
(b) *Variation of ionic strength*					
8	0.1	5	19.0	76	1.6
8	0.01	5	33.4	101	3.0
8	0.001	5	37.6	95	3.1
8	0.0001	5	39.8	97	3.3
(c) *Variation of feed concentration*					
8	0.01	0.1	126.0	75	8.6
8	0.01	1.0	65.8	95	4.8
8	0.01	10.0	24.3	101	2.4

*Applied voltage, 100 V; pulse interval, 20 min; pulse duration, 10 s.

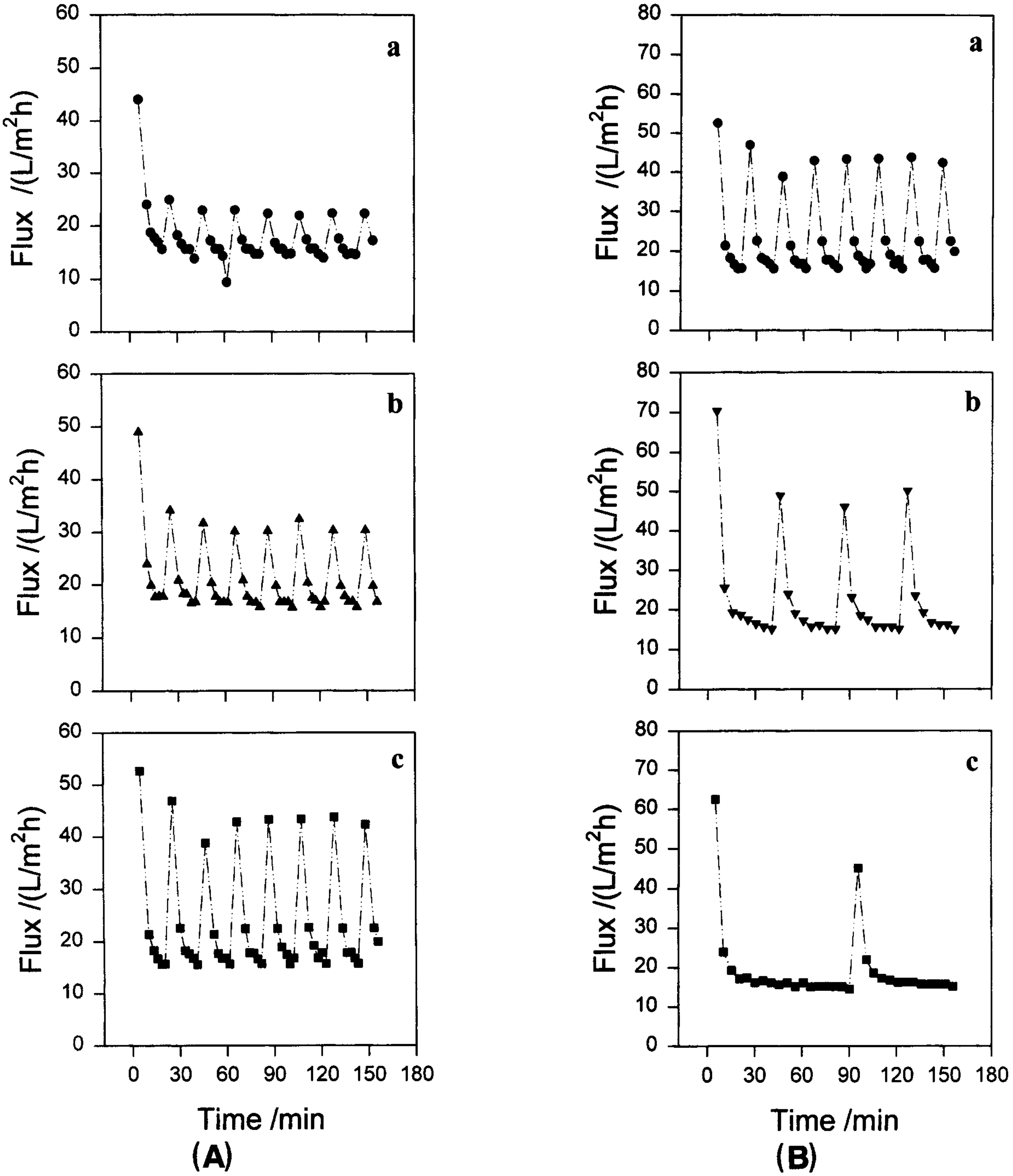

Figure 3. Filtration flux as a function of time in the pulsed electrophoretic dead-end ultrafiltration of 1 g L^{-1} BSA; pH 8.0; 0.01-M phosphate buffer; 7.5-s pulses.

(A) Effect of variation of applied voltage, 20-min interval. (a) 12.5 V; (b) 50 V; (c) 100 V. (B) Effect of variation of pulse interval, at 100 V. (a) 20 min; (b) 40 min; (c) 90 min.

amples of the effects of the application of electric-field pulses on filtration flux in the dead-end microfiltration of titania dispersions are shown in Figure 4A. Figure 4A shows the effects of increasing the applied voltage for 10-s pulses at 20-min intervals for 1-g L^{-1} TiO_2 at pH 8 in 0.01-M KNO_3 solution. 12.5 V is below the threshold voltage for removing particles from the filter cake, but increasing the voltage to 50 V or 100 V gives very substantial increases in the flux immediately after the application of each pulse. Figure 4B shows the effects of increasing the interval between pulses. Again, substantial increases in flux are obtained after applying electric-field pulses, even when the cake is allowed to build up for long periods. As was the case for the ultrafiltration experiments, there were no detectable particles present in the permeate.

Tables 5a and 5b contain more selected data for the overall process performance in the filtration of titania. The trends

Table 4a. Variation of Operating Conditions in the Filtration of BSA*

Voltage (V)	Interval (min)	Duration (s)	Avg. Filt. Flux ($L \cdot m^{-2} \cdot h^{-1}$)	Avg. Cake Recovery (%)	Cake Conc. Factor
(a) *Variation of applied voltage*					
12.5	20	10	19.2	92	1.9
25	20	10	20.0	93	2.0
50	20	10	23.0	94	2.2
100	20	10	26.7	96	2.5
(b) *Variation of pulse interval*					
100	10	10	39.2	94	2.1
100	20	10	29.4	94	2.5
100	40	10	23.7	79	2.9
100	90	10	20.1	60	3.4
(c) *Variation of pulse duration*					
100	20	3	22.5	92	2.2
100	20	7.5	26.7	96	2.5
100	20	15	25.1	90	2.5
100	20	40	22.5	68	1.6

*1 g/L, BSA; 0.01 M, phosphate buffer; pH 8.0

of the average filtration rate vary with applied voltage, pulse interval, and pulse duration, and follow those observed for silica and BSA, though the filtration rates are much greater due to lower hydraulic resistance of both the microfiltration membrane and the deposited particles. The average percent of cake recovery increases with increased applied voltage and decreased pulse duration. The data show that the optimum pulse duration for maximum cake recovery has increased to 20 s for this system. This may indicate some in-pore particle deposition. The cake concentration factors are much higher than for ultrafiltration, as the rate of filtration is much higher while the interval between pulses has remained constant.

As the feed conditions vary, it is found that the average filtration rates are greatest at the highest ionic strengths and lowest pHs, in contrast to the findings for ultrafiltration. This is in part due to the variation of particle size with solution conditions (Table 2). Larger particles give filter cakes of a lower specific resistance. However, the average percentage of cake recovery is highest at the highest pH, which gives the highest particle electrophoretic mobility. Again there is optimal cake recovery in a 0.01-M solution. The cake concentration factors obtained follow a pattern that reflects their dependence on both the average filtration rate and the average cake recovery.

Comparison of theory and experiment

A predictive description of flux–time curves, such as those shown in Figures 2–4, has two main requirements. The first is a description of the time dependence of dead-end filtration in the absence of the electric field. Such a description has recently been provided for charged inorganic colloids (Bowen and Jenner, 1995) and proteins (Bowen and Williams, 1996), taking electrostatic, dispersion, entropic, and hydration interactions into account. The second requirement for a complete description of the process is the prediction of the filtration flux immediately after the electric-field pulse has been applied. This is the main concern of the present section.

Table 4b. Variation of Feed Conditions in the Filtration of BSA**

pH	I (M)	Conc. (g/L)	Avg. Filt. Flux ($L \cdot m^{-2} \cdot h^{-1}$)	Avg. Cake Recovery (%)	Cake Conc. Factor
(a) *Variation of pH*					
5	0.01	1	18.6	86	2.1
6	0.01	1	23.8	90	2.3
7	0.01	1	28.1	90	2.3
8	0.01	1	28.3	96	2.6
(b) *Variation of ionic strength*					
8	0.1	1	14.7	76	1.7
8	0.01	1	28.3	96	2.6
8	0.001	1	29.9	94	2.6
8	0.0001	1	22.2	89	2.3
(c) *Variation of feed concentration*					
8	0.01	0.1	63.1	104	4.7
8	0.01	1.0	26.7	96	2.5
8	0.01	10.0	12.5	73	1.2

**Applied voltage, 100 V; pulse interval, 20 min; pulse duration, 10 s.

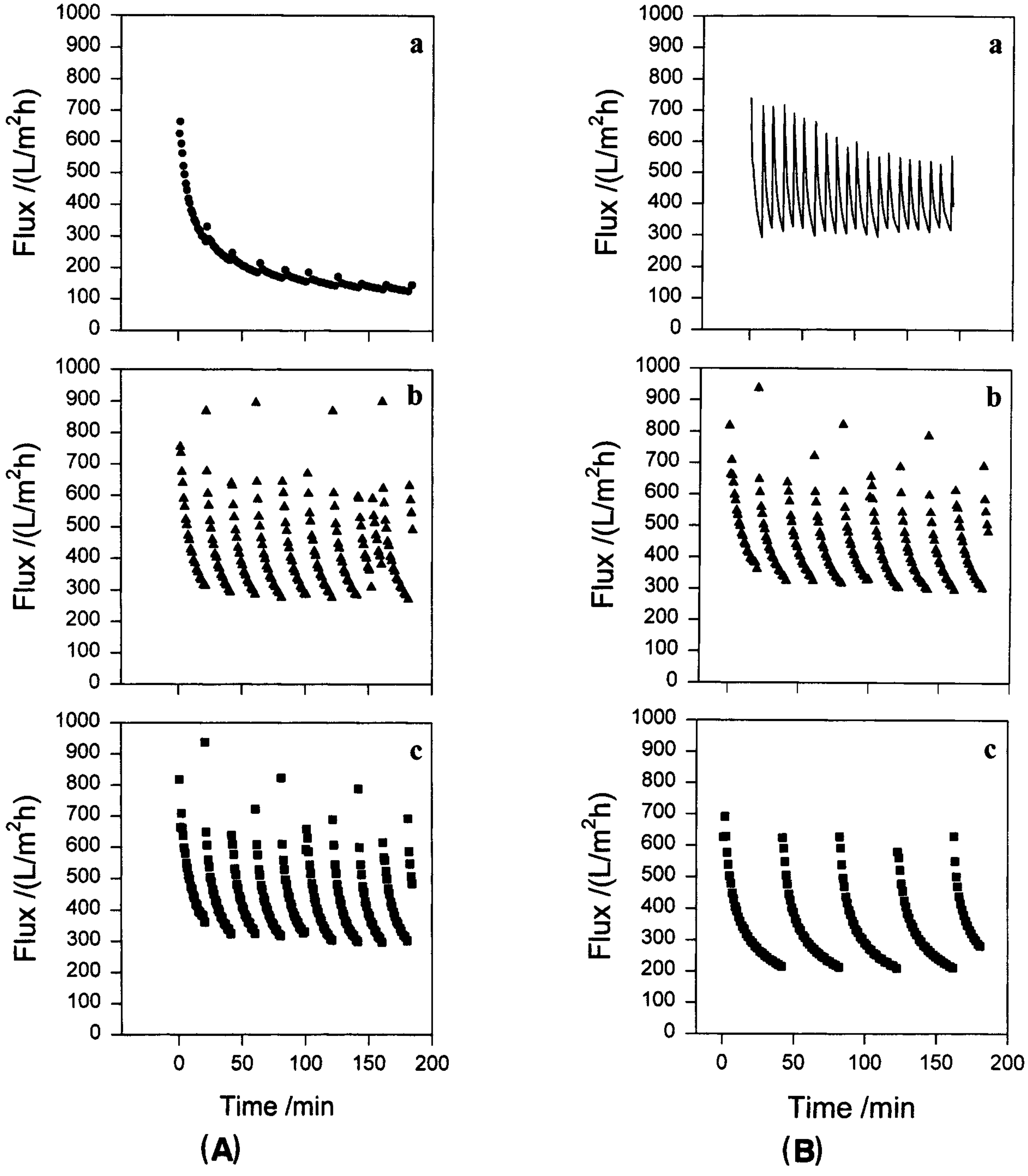

Figure 4. Filtration flux as a function of time in the pulsed electrophoretic dead-end microfiltration of 1 g L^{-1} TiO_2; pH 8.0; 0.01-M KNO_3; 10-s pulses.

(A) Effect of variation of applied voltage, 20-min interval. (a) 12.5 V; (b) 50 V; (c) 100 V. (B) Effect of variation of pulse interval, at 100 V. (a) 10 min; (b) 20 min; (c) 40 min.

The basis for prediction of the filtration flux following an electric pulse (v_{end}) is Eq. 9. This equation requires a knowledge of v_{eom}, the specific electroosmotic flow rate (Table 1); u_p, the particle electrophoretic mobility (Table 2); R_{om}, the membrane resistance in the absence of the electric field, obtained from the clean electrolyte permeation rate; λ_o, the feed electrolyte conductivity; μ the feed electrolyte viscosity; I, the applied current density; and ΔP, the applied pressure. Thus, all of the required parameters are available.

One of the best tests of the agreement between theory and experiment is a comparison of v_{end} and v_{max} (the experimental flux immediately after each pulse, averaged for an experimental run) as the applied current is increased. Such a comparison is shown in Figure 5 for the case of silica dispersions. The overall agreement between theory and experiment is excellent. The main deviation is a slight underestimation of the experimental flux at the lowest current, as some of the deposited filter cake is removed by the liquid flow through the

Table 5a. Variation of Operating Conditions in the Filtration of TiO_2*

Voltage (V)	Interval (min)	Duration (s)	Avg. Filt. Flux ($L \cdot m^{-2} \cdot h^{-1}$)	Avg. Cake Recovery (%)	Cake Conc. Factor
(a) *Variation of applied voltage*					
12.5	20	10	225	20	1.3
25	20	10	251	52	4.9
50	20	10	325	69	8.5
100	20	10	329	94	10.9
(b) *Variation of pulse interval*					
100	5	10	482	102	4.4
100	10	10	406	95	7.0
100	20	10	329	94	10.9
100	60	10	295	83	16.2
(c) *Variation of pulse duration*					
100	20	5	321	65	9.1
100	20	10	329	94	10.9
100	20	20	362	97	9.9
100	20	40	386	96	9.9

*1 g/L, TiO_2; 0.01 M, KNO_3; pH 8.0.

feed compartment when the valve is opened for cake collection. Figure 6 shows a comparison for BSA. The overall agreement is reasonable, with the trends in the experimental data being well reproduced. As for silica, there is an underestimation of the experimental filtration flux at the lowest current. There is also an overestimation at the highest currents. Nevertheless, the overall agreement between theory and experiment, with no adjustable parameters, for the ultrafiltration of both an inorganic and biological process feed is very encouraging.

Figure 7 compares theory and experiment for titania. Again the theory predicts the overall pattern of the experiments. It has already been shown experimentally that for titania the average percentage of cake recovery when the applied voltage is varied are substantially lower than those for silica or BSA (compare Tables 3–5). A major difference between the systems is that the Hamaker constant for titantia ($A_H = 26 \times 10^{-20}$ J) (Israelachvili, 1992) is substantially greater than that of either silica ($A_H = 0.795 \times 10^{-20}$ J) (Bowen and Jenner, 1995) or BSA ($A_H = 0.753 \times 10^{-20}$ J) (Bowen and Williams, 1996). During the filtration process the particles in the filter cake are forced together by the hydraulic drag in the cake (Bowen and Jenner, 1995). Particles with higher Hamaker constants are more likely to aggregate under these circumstances, leading to a higher filtration flux, but they may also be more difficult to remove from the membrane surface, leading to a lower flux recovery when the electric field is applied. Clearly the overall filtration behavior of titania is more complex than that of either silica or BSA.

Conclusions

This article has presented an experimental and theoretical study of the use of pulsed electric fields to remove filter cakes from membranes in equipment that allows subsequent recovery of the displaced cake. The effectiveness of such a process has been experimentally quantified for two process feeds in ultrafiltration (silica and BSA) and for one process feed (titania) in microfiltration. Varying the key operating parameters of the magnitude of the applied field, the pulse duration, and the pulse interval, coupled with variation of the important feed conditions (pH, ionic strength, and particle concentration) has identified the conditions under which the pulsed electric fields can efficiently release filter cakes for collection. These conditions allow filtration in dead-end mode at substantially improved fluxes. It is worth noting that the fluxes achieved for both ultrafiltration and microfiltration are comparable to or better than those frequently reported for cross-flow filtration, but without the mechanical complexity or the higher energy costs of the latter process (Cheryan, 1986).

Table 5b. Variation of Feed Conditions in the Filtration of TiO_2*

pH	I (M)	Conc. (g/L)	Avg. Filt. Flux ($L \cdot m^{-2} \cdot h^{-1}$)	Avg. Cake Recovery (%)	Cake Conc. Factor
(a) *Variation of pH*					
4	0.01	1	706	67	7.7
6	0.01	1	532	93	8.4
8	0.01	1	406	95	7.0
(b) *Variation of Ionic Strength*					
8	0.1	1	492	75	2.4
8	0.01	1	482	102	4.4
8	0.001	1	439	90	3.1
8	0.0001	1	373	79	4.4

*Applied voltage, 100 V; pulse interval, 10 min for (a) and 5 min for (b); pulse duration, 10s.

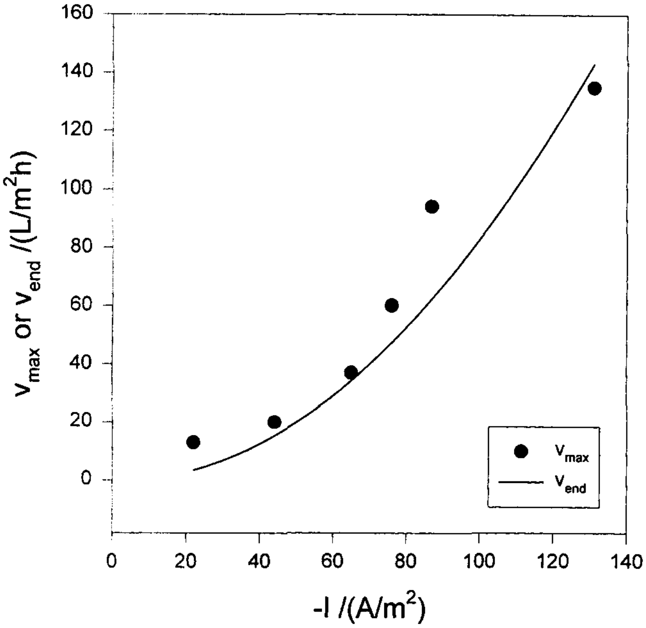

Figure 5. Predicted (v_{end}) vs. experimental (v_{max}) filtration fluxes immediately after an electric-field pulse as a function of the applied current.

5 g L^{-1} SiO_2; pH 8.0; 0.01-M KNO_3; 10-s pulse duration; 20-min pulse interval. Voltage range 12.5–75.0 V.

Currently, industry is very interested in the use of dead-end membrane processes, especially for water treatment. The use of electrically enhanced dead-end membrane filtration could be facilitated if there were methods available for predicting the performance of such processes. Methods already exist for predicting the rate of dead-end filtration of colloids in the absence of electric fields. In addition the pulsed-field process requires the prediction of the rate of filtration immediately after the application of the pulse. This article has presented such a theoretical analysis and has shown that the predictions show excellent agreement for the case of ultrafiltration and reasonable agreement for the case of microfiltration.

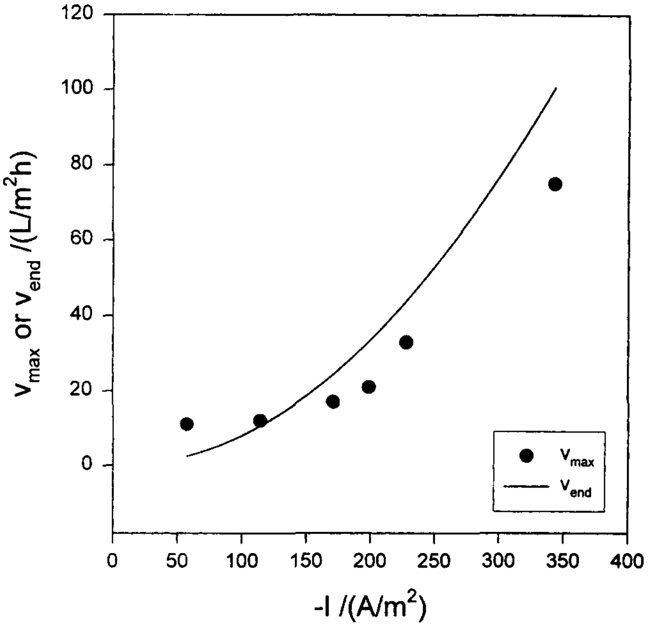

Figure 6. Predicted (v_{end}) vs. experimental (v_{max}) filtration fluxes immediately after an electric-field pulse as a function of the applied current.

1 g L^{-1} BSA; pH 8.0; 0.01-M phosphate buffer; 7.5-s pulse duration; 20-min pulse interval. Voltage range 12.5–75.0 V.

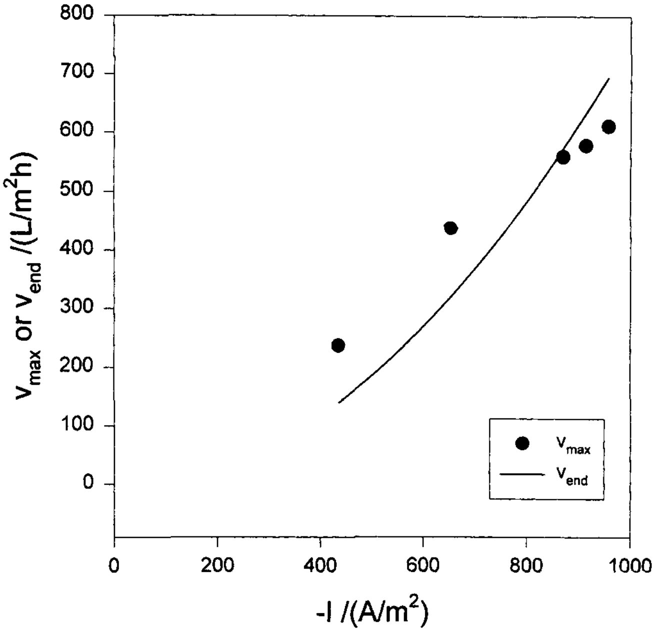

Figure 7. Predicted (v_{end}) vs. experimental (v_{max}) filtration fluxes immediately after an electric-field pulse as a function of the applied current.

1 g L^{-1} TiO_2; pH 8.0; 0.01-M KNO_3; 10-s pulse duration; 20-min pulse interval. Voltage range 12.5–75.0 V.

The pulsed-field dead-end membrane filtration process requires only low levels of electrical power input. It is mechanically much simpler than filter-cake removal by pressure backflushing, and unlike the latter does not stress the membrane in a direction in which it is not supported. It is a very promising technique worthy of further assessment through scale-up.

Acknowledgment

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Notation

f = friction factor, N·s·m^{-1}
I = ionic strength, M, in the tables
R_c = hydraulic resistance of the filter cake, m^{-1}
R_m = hydraulic resistance of the membrane, m^{-1}
R_{oc} = hydraulic resistance of the filter cake in the absence of an electric field, m^{-1}
v_{oc} = flux through the cake in the absence of an electric field, m·s^{-1}
v_{om} = flux through the membrane in the absence of an electric field, m·s^{-1}
μ = electrolyte viscosity, N·s·m^{-2}

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