Journal of Hazardous Materials B:63 (1998) 179–197

Application of cross-flow microfiltration with rapid backpulsing to wastewater treatment

José A. Ramirez, Robert H. Davis *

Department of Chemical Engineering, University of Colorado, Boulder, CO 80309-0424, USA

Received 29 January 1998; revised 8 July 1998; accepted 7 August 1998

Abstract

Cross-flow microfiltration with rapid backpulsing was investigated for the removal of suspended solids and dispersed oil from an aqueous stream. Laboratory experiments performed on clay suspensions demonstrate that rapid backpulsing can maintain the permeate flux at a level which is more than 10-fold over the long-term flux in the absence of backpulsing, without any reduction in the permeate quality. Experiments with dilute oil-in-water dispersions show that rapid backpulsing can increase the permeate flux by up to 25 times; however, the enhanced performance in this case cannot be maintained over the life of the membrane and is highly dependent on the degree of membrane use. An economic analysis shows that cross-flow microfiltration without backpulsing is not economically feasible when compared to conventional treatment methods, but that operation with backpulsing at the enhanced permeate flux levels observed for the clay experiments will result in lower costs of treated water than for conventional methods, for facilities with capacities of up to 6000 m^3 d^{-1}.

Keywords: Crossflow microfiltration; Membranes; Backpulsing; Wastewater

1. Introduction

Numerous recent studies have shown that cross-flow microfiltration is a viable treatment alternative for removing suspended contaminants from aqueous streams. Applications cited in the literature range from the removal of oil and grease from water [1–3] and the purification of nitrocellulose-manufacturing wastewater [4], to the removal

* Corresponding author. Tel.: +1 303 492 7314; fax: +1 303 492 4341; e-mail: robert.davis@colorado.edu

0304-3894/98/$ - see front matter
PII: S0304-3894(98)00204-0

of precipitated metal ions [5,6] and suspended clays [7]. The latter application is of particular relevance, owing to the increasingly widespread use of clays as adsorbing matrices for a variety of contaminants dissolved in water, including organic compounds [8] and dyes [9].

Microfiltration is generally defined as the pressure-driven flow of a suspension containing colloidal or fine particles with dimensions within the size range of 0.02 to 10 μm through a membrane or filter [10]. The microfiltration process is carried out in two types of configurations: dead-end and cross-flow. Cross-flow microfiltration is of particular interest in processes that demand continuous or semicontinuous operation; however, use in wastewater treatment applications has remained limited as a result of low long-term permeate fluxes due to membrane fouling [11].

Rapid backpulsing is a variation of the industrially established technique of back-flushing. In both processes, the transmembrane pressure is periodically reversed, with the purpose of removing foulant deposits from the surface of the membrane. In rapid backpulsing, the reverse pressure pulses are applied for very short periods of time (typically less than 1 s) at a high frequency (typically 0.1–2 Hz). The objective is to prevent or minimize the deposition of foulants on the surface of the membrane and thus to maintain high permeate fluxes throughout the operation. Experiments with biological suspensions have shown that it is possible to maintain high values of the long-term permeate flux by using rapid backpulsing, exceeding those of ordinary cross-flow microfiltration by an order of magnitude [12,13].

In this study, we have evaluated the use of crossflow microfiltration with rapid backpulsing for water treatment using two types of commercial membrane modules. Experiments were performed with suspensions of bentonite clay in water and with a dilute oil-in-water mixture. The permeate flux was maximized by varying the backpulse frequency, while the permeate was analyzed for the presence of residual particles or oil. The permeate flux values obtained were then used in a preliminary economic analysis to demonstrate the feasibility of the technique in wastewater treatment.

2. Theoretical considerations

The average permeate flux per cycle is given by the amount of permeate collected per membrane area during forward filtration between two successive backpulses, minus that lost during the short backfiltration period, divided by the cycle duration:

$$\langle J \rangle = \frac{\left(\int_0^{t_\mathrm{f}} J_\mathrm{f}(t)\,\mathrm{d}t - \int_{t_\mathrm{f}}^{t_\mathrm{f}+t_\mathrm{b}} J_\mathrm{b}(t)\,\mathrm{d}t\right)}{t_\mathrm{f}+t_\mathrm{b}}, \tag{1}$$

where J_f is the flux magnitude (volume permeate per time per membrane area) during forward filtration of duration t_f, and J_b is the flux magnitude during a backpulse of duration t_b (Fig. 1). The model of Kuberkar et al. [14] is used to provide expressions for the forward and backward fluxes. In this model, it is assumed that the membrane is not cleaned uniformly by each reverse pressure pulse. The development of preferential flow

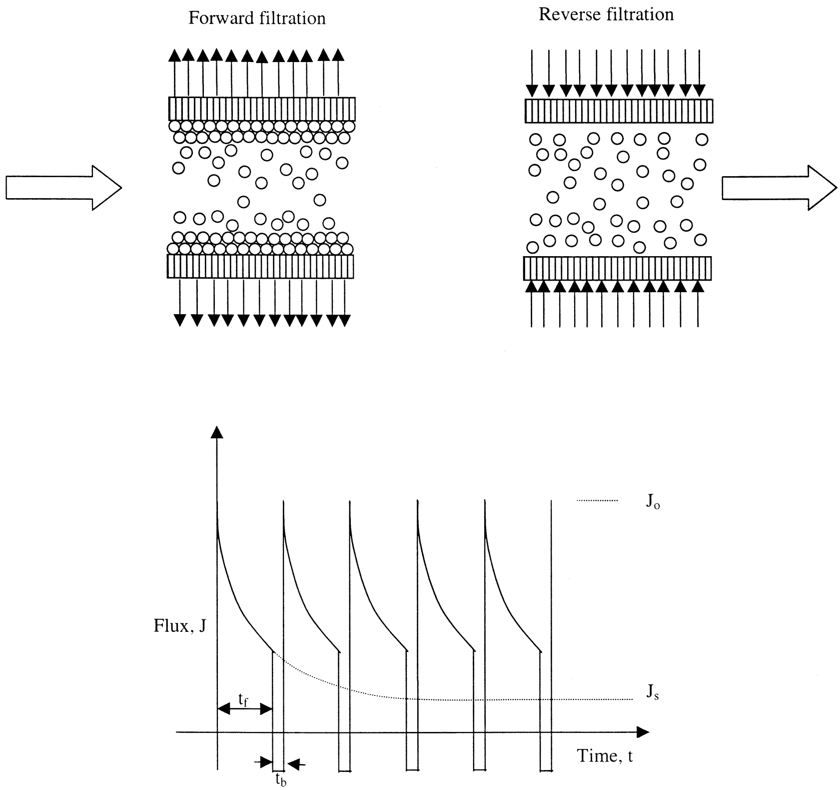

Fig. 1. Schematic of rapid backpulsing and the corresponding permeate flux during consecutive cycles of forward and reverse filtration for ideal case of complete cleaning.

paths of permeate during the reverse portion of the filtration cycle leads to the formation of cleaned patches of the membrane interspersed with uncleaned regions. The unclean regions are assumed to have a lower permeate flux equal to the long-term or steady flux, J_s, exhibited by a fouled membrane without backpulsing. On the other hand, the cleaned regions of the membrane regain the original permeability and have a higher flux.

During the backpulse part of the cycle, it is assumed in the model [14] that a fraction of the membrane area is instantly and completely cleaned, so that the reverse flux during the backpulse is simply a weighted average of the clean membrane flux, J_o, and the fouled membrane flux, J_s:

$$J_b = \beta J_o + (1-\beta) J_s, \tag{2}$$

This equation is valid when the transmembrane pressure or driving force is simply reversed during backpulsing (as is the case in the present work); otherwise, the right-hand-side should be multiplied by the ratio of the magnitudes of the backpulse and forward transmembrane pressures.

A slightly more complicated expression is required for the forward flux, since it equals J_o for the cleaned fraction of the membrane only at the start of the forward part of the cycle and then declines due to the deposition of foulants on the membrane. Fortunately, the flux decline for short periods of crossflow microfiltration may be approximated by dead-end filtration theory [10], and so accounting for both the cleaned and uncleaned regions yields [14]

$$J_f(t) = \frac{\beta J_o}{(1 + t/\tau)^{1/2}} + (1 - \beta) J_s, \tag{3}$$

where τ is the time constant for growth of the fouling deposit on the membrane surface [13].

When Eqs. (2) and (3) are substituted into Eq. (1), the theory predicts an optimum backpulsing frequency (where the frequency is $1/(t_f + t_b)$) for a fixed backpulse duration (t_b). At frequencies lower than the optimum, the average or net flux is reduced by significant fouling during the relatively long periods of forward filtration. At frequencies greater than the optimum, the net flux is reduced because of the significant loss of permeate during the backpulses separated by relatively short periods of forward filtration.

The clean membrane permeate flux, J_o, the long-term fouled membrane flux, J_s, and the foulant growth time constant, τ, can all be easily measured from a forward filtration experiment without backpulsing [13]. The backpulsing cleaning efficiency, β, is then treated as a fitted parameter which may be determined from measurement of the net permeate collected at different backpulsing frequencies.

3. Materials and methods

In this work, cross-flow microfiltration experiments with and without backpulsing were performed for aqueous clay suspensions of different concentrations, as well as for a dilute crude oil-in-water dispersion. The backpulse duration and frequency were varied for fixed values of the transmembrane pressure and cross-flow velocity which are typical of medium-scale and large-scale applications in wastewater treatment [15,16]. The objective of the experimental study was to determine the maximum permeate fluxes when using rapid backpulsing applied at different frequencies.

3.1. Foulants and membranes

The model clay-in-water suspension was prepared by combining the desired amount of bentonite clay (Aldrich Cat. No. 28,533-4) with 2 l of tap water at 25°C and agitating vigorously in a capped conical beaker. The resulting suspension exhibited a number-averaged particle diameter of 4.6 μm (suggesting the presence of clustered aggregates) and a standard deviation of 1.7 μm, as determined by analysis with a Coulter Multisizer. The oil-in-water dispersion was prepared by adding a heavy crude oil (API 12 weight, density 0.972 g cm^{-3}, supplied by UNOCAL from the Hueneme field in California) to tap water at a concentration of 50 mg l^{-1}. A blender (Osterizer Model 890-28M) was used to mix the oil and water at a high setting for approximately 2 min. Analysis with a

Coulter Multisizer gave a number-averaged diameter of the oil droplets of approximately 2 μm, with most droplets being in the 1 to 10 μm range.

Two types of membrane filters were employed in the experiments. The first was a tubular α-alumina ceramic membrane manufactured by the Membralox division of US Filter, while the second was an unmodified polysulfone hollow fiber cartridge manufactured by AG Technology. The tubular ceramic membranes had a porosity of 35%, an internal diameter of 0.7 cm, a useful length of 20.6 cm, a surface area of 45.3 cm^2, and either 0.2 or 0.8 μm nominal pore diameter. The hollow fiber cartridge was composed of numerous small lumens of internal diameter 0.1 cm, giving the cartridge a total surface area of 420 cm^2, and the pore size employed was either 0.2 or 0.45 μm.

3.2. Apparatus

A schematic of the experimental apparatus is provided in Fig. 2. The experiments were operated in a diafiltration or dialysis mode; the retentate was returned to the feed reservoir while the feed vessel liquid level was maintained constant by replenishment with clean water. In addition, the feed solution was changed at 30-min intervals during the experiment in order to avoid concentration variations due to particle or droplet adhesion to the vessel walls, or due to settling or creaming. The feed water of controlled temperature was pumped (P-1, Lobbee pump, 6LOE) through the interchangeable membrane module (MF, US Filter, 1T170 or AG Technology CFP-2-E-4A/CFP-4-E-4A) at a fixed flow rate (Q_f) and pressure. The reverse pressure pulses were generated through a computer-controlled, three-way valve connected to a pressurized water reservoir (T-1, Gellman, 15203). The mass of permeate collected throughout the

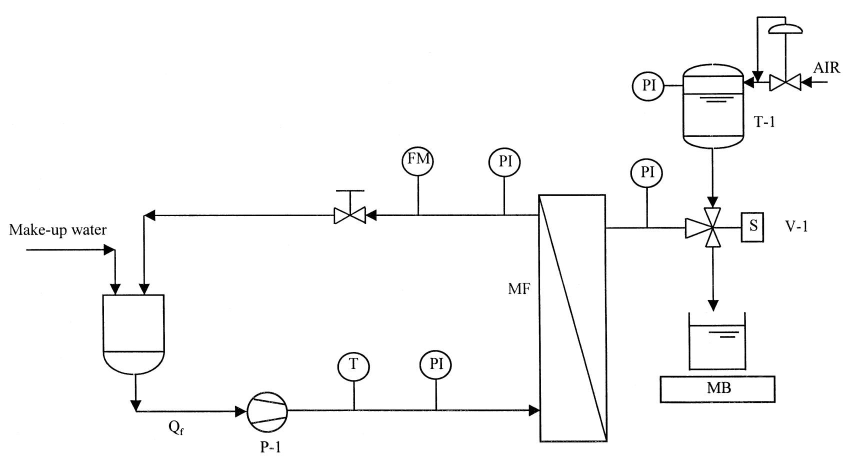

Fig. 2. Experimental apparatus for rapid backpulsing experiments. P1—gear pump of variable rpm, MF—microfiltration test module, V-1—three-way solenoid valve, T-1—pressurized backpulse reservoir, MB—electronic mass balance. Pressures were measured by pressure gauges (PI), while a paddle wheel flow-meter (FM) was used to monitor the cross-flow velocity.

experiment was recorded by an electronic balance (MB, Mettler, PM4000), while the total permeate lost due to backpulsing was calculated from the difference between the initial and final volumes of water in the pressurized reservoir, T-1. The average net rate of permeate flow was determined by the difference between the mass of permeate collected and the mass of permeate lost due to backpulsing, per unit time; the net permeate flux was then calculated by dividing this quantity by the membrane surface area.

3.3. Experimental method

Most experiments were repeated from 2 to 4 times for each set of conditions. Each run began by filtering clean tap water through the clean membrane until the permeate flux was stable (usually after a period of 40 min). At this point, a switch was made to the bentonite (or oil-in-water) suspension, with simultaneous start-up of the backpulsing mechanism. The experiment was then run until the net permeate flux stabilized (typically after a period of 1.5 h), after which the net average permeate flux for the particular run was recorded. The values of the key operating parameters are given in Table 1.

Membranes were reused after each experiment, following a fairly elaborate cleaning procedure. After each experiment, 5 l of a 1% w/v solution of caustic detergent (Alconox, VWR Scientific Products) were forced through the membrane in the reverse direction of permeate flow. After this, the ceramic membrane was removed from its housing and manually scrubbed with clean detergent solution (this step does not apply to the hollow fiber cartridge). The membrane was rinsed in clean water and then immersed in a 0.25% Alconox detergent solution for storage. For membranes used on oil-in-water

Table 1
Summary of parameter values used for the rapid backpulsing experiments

Membrane characteristics
Tubular ceramic, I.D. 7 mm, $L = 21$ cm, Nom. pore size: 0.2, 0.8 μm
Hollow fiber (polysulfone), I.D. 1 mm, $L = 36.2$ cm, Nom. Pore size: 0.2, 0.45 μm
Feed conditions
Bentonite clay in water
Concentrations: 0.04 g l^{-1}, 0.2 g l^{-1}, 2.0 g l^{-1}
$T = 25°C$; $\Delta P = 130$ kPa (20 psi)
Crude oil in water
Concentration: 50 ppm
$T = 40°C$; $\Delta P = 130$ kPa (20 psi)
Process parameters
Forward filtration transmembrane pressure: 130 kPa (20 psi)
Backward filtration transmembrane pressure: 130 kPa (20 psi)
Cross-flow velocity: 2.6 m s^{-1} (clay), 3.5 m s^{-1} (oil)
Backpulse duration: 0.2, 0.5 s
Backpulsing intervals: 0.5, 1, 3, 5, 10, 20, 50 s

filtration, the same procedure was employed; however, manual scrubbing with clean pure ethyl alcohol (Sigma Aldrich, HPLC grade) and subsequent rinsing in water were performed prior to membrane storage. The apparatus was cleaned by circulating warm tap water (40°C), followed by 0.2 wt.% sodium hydroxide, and a final rinsing cycle with warm tap water (40°C).

The permeate quality was assessed by visual inspection with a microscope (Nikon, Labophot) and by turbidity measurements on a nephelometric turbidimeter (Monitek, TA1), for the experiments with bentonite clay. A Horiba (Model OCMA-220) oil-content analyzer was used to determine the total oil and grease content of both feed and permeate, for the experiments with crude oil. The analyzer utilizes tetrachloroethylene to extract the hydrocarbon phase from the water, and then measures the absorbance at a wavelength of 2930 cm^{-1} [17].

4. Experimental results and discussion

4.1. Experiments without backpulsing

A typical permeate flux curve for the filtration of a dilute model mixture of bentonite of concentration 0.2 g l^{-1} in water under nonbackpulsed conditions at a cross-flow

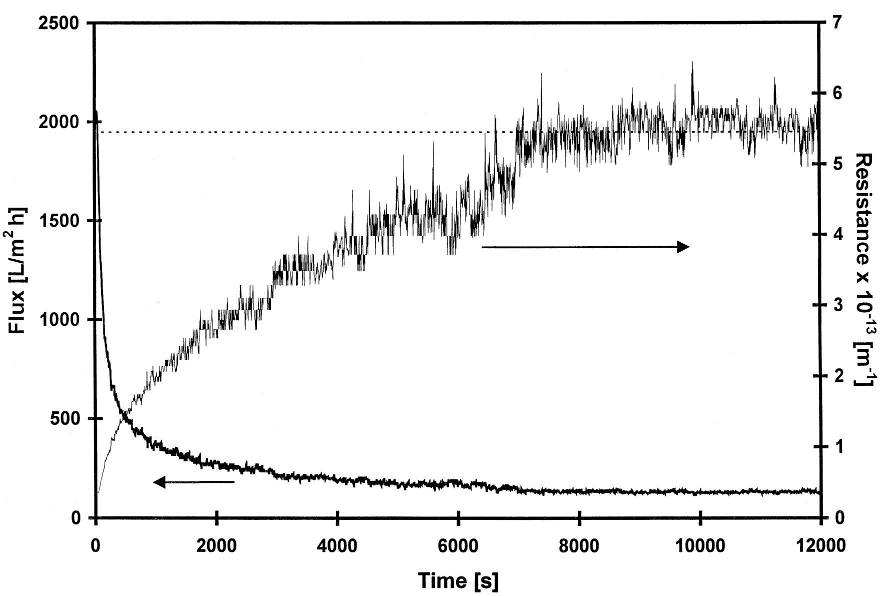

Fig. 3. Typical experimental flux and resistance curves for a 0.2 g l^{-1} bentonite suspension under ordinary cross-flow filtration without rapid backpulsing. The membrane used was a 0.8 μm nominal pore diameter, tubular α-alumina filter. The cross-flow velocity was 2.6 m s^{-1}, the water temperature was 25°C, and the transmembrane pressure was 20 psi (130 kPa). The dotted line is the water flux for the clean membrane.

velocity of 2.6 m s^{-1} and a transmembrane pressure of 130 kPa (20 psi) is shown in Fig. 3, for a ceramic membrane with 0.8 μm nominal pore diameter. The dramatic flux decline from an initial value of 1950 l m^{-2} h^{-1} to a final value after 3 h or 125 l m^{-2} h^{-1} (a reduction of 94%) is readily apparent. This decline in flux is a result of the deposition of particles on the membrane surface. This external fouling is the dominant contribution to membrane flux degradation when the linear dimensions of the majority of the suspended particles are larger than the membrane pores. The dominating fouling mechanism was identified through a technique described by Tracey and Davis [18]. The method involves examining the total resistance (which is inversely proportional to the permeate flux) as a function of time (see Fig. 3). A concave-up curve indicates a predominance of internal fouling, while a concave-down curve (as in Fig. 3) is indicative of external fouling.

Fig. 4 shows the results of a similar experiment with a 50 ppm crude oil-in-water dispersion and a 0.2 μm ceramic membrane. Once again, a very low value of the long-term permeate flux results (the long-term flux for these conditions stabilized at a value of 30 l m^{-2} h^{-1} after 2 h). From the concavity of the resistance curve at early times, the dominant fouling mechanism in this case is initially internal (due to blockage or constriction of the membrane pores); after a brief time period, the dominant resistance to permeate flux switches to an external oil layer which forms on the membrane surface. Similar findings have been reported by Mueller et al. [19], for fouling experiments with different oil-in-water concentrations and membranes. Based on the analysis of Nazzal

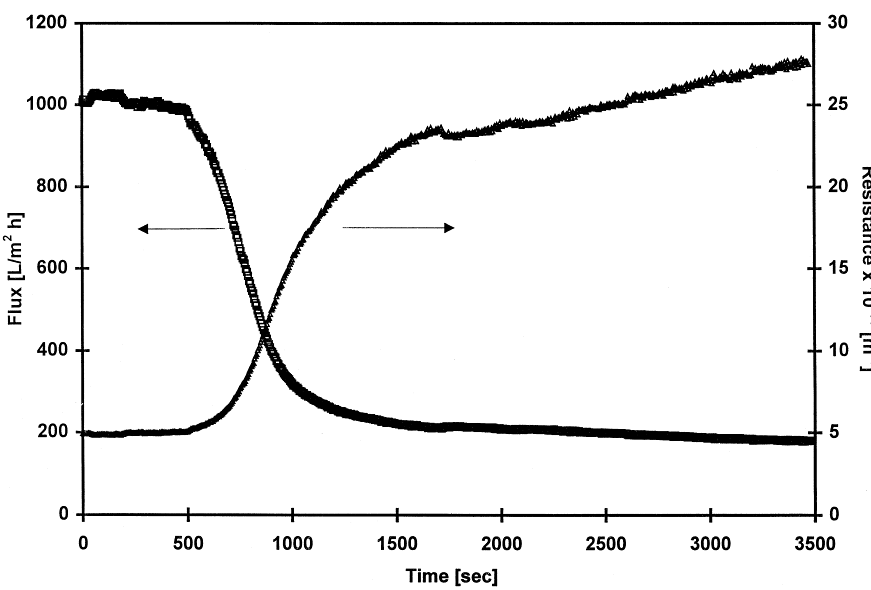

Fig. 4. Typical experimental flux and resistance curves for a 50 ppm crude oil-in-water dispersion under ordinary cross-flow filtration without rapid backpulsing. The membrane used was a 0.2 μm nominal pore diameter, tubular α-alumina filter. The cross-flow velocity was 3.5 m s^{-1}, the water temperature was 40°C, and the transmembrane pressure was 20 psi (130 kPa).

Table 2
Summary of experimental results showing the long-term flux for clean water filtration (J_o), long-term flux for clay or oil suspension without backpulsing (J_s) the maximum observed average permeate flux with rapid backpulsing ($\langle J \rangle_{max}$) at the backpulse duration t_b, the time constant for cake growth (τ) and the best-fit cleaning efficiency (β)

Membrane	Feed	t_b (s)	τ (s)	J_o ($l\ m^{-2}\ h^{-1}$)	J_s ($l\ m^{-2}\ h^{-1}$)	$\langle J_{max} \rangle$ ($l\ m^{-2}\ h^{-1}$)	β (−)
Ceramic, 0.8 μm	Clay, 0.04 g l^{-1}	0.5	380	1950 ± 290	270 ± 19	2020 ± 270	1.0
Ceramic, 0.8 μm	Clay, 0.2 g l^{-1}	0.5	82	1950 ± 290	125 ± 8	1300 ± 100	0.82
Ceramic, 0.8 μm	Clay, 0.2 g l^{-1}	0.2	82	1950 ± 290	125 ± 8	1300 ± 130	0.83
Ceramic, 0.8 μm	Clay, 2.0 g l^{-1}	0.5	9	1950 ± 290	89 ± 8	410 ± 110	0.24
Polysulfone, 0.45 μm	Clay, 2.0 g l^{-1}	0.5	10	2070 ± 20	200 ± 13	610 ± 20	0.26
Ceramic, 0.2 μm	Oil, 50 ppm	0.5	n/a	$1950^a \pm 290$	30 ± 6	748^a; 184^b	n/a
Polysulfone, 0.2 μm	Oil, 50 ppm	0.5	n/a	$1740^a \pm 10$	80 ± 7	405^a; 92^b	n/a

[a]Unused membrane.
[b]After 200 h (ceramic) or 20 h (polysulfone) of use.

and Wiesner [2] for a similar suspension and membrane, it may be argued that, since the transmembrane pressure exceeds the critical pressure necessary for forcing the smaller oil droplets into the larger pores in the membrane (considering that the membrane

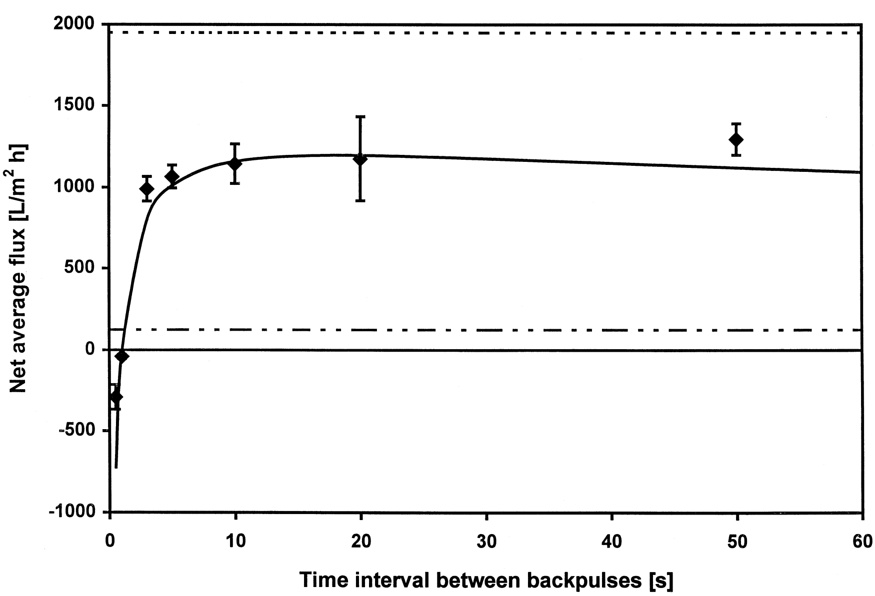

Fig. 5. Average net permeate flux vs. the duration of forward filtration between backpulses for a bentonite suspension of concentration 0.2 g l^{-1} with backpulsing at the same conditions of Fig. 3, and with a backpulse duration of 0.5 s and a reverse transmembrane pressure of 20 psi (130 kPa). The solid curve represents the best fit of the data based on the model with $\beta = 0.82$. Error bars represent 90% confidence intervals on four repeats. The dotted line is the clean membrane water flux and the dashed-dotted line is the long-term flux without backpulsing.

exhibits a distribution of pore sizes about the nominal pore size of 0.2 μm), the internal fouling mechanisms of pore blockage or constriction are dominant at early times. After the larger pores have been blocked (or constricted), the oil droplets cannot penetrate the smaller pores due to the fact that the required critical pressure is higher than the available transmembrane pressure (this critical pressure decreases with increasing membrane pore size and contact angle, but increases with increasing droplet size [2]). At this point, deposition of an oil layer on the surface of the membrane becomes the dominant resistance to permeate flow.

4.2. Experiments with backpulsing

Rapid backpulsing is particularly effective in minimizing external fouling, since the external fouling layer is lifted off the membrane by the reverse flow and swept out of the filter by the cross flow. The emphasis of the backpulsing experiments was on maximizing the permeate flux by varying the frequency with which the reverse pulses were applied, so that the effectiveness of the technique relative to operation without backpulsing could be evaluated. The reverse pressure pulses were applied at time intervals of 0.5, 1.0, 3.0, 5.0, 10, 20 and 50 s, with a fixed backpulse duration of either 0.2 or 0.5 s. A summary of the experimental conditions employed is given in Table 1, while a summary of the main experimental results pertaining to the permeate flux is given in Table 2.

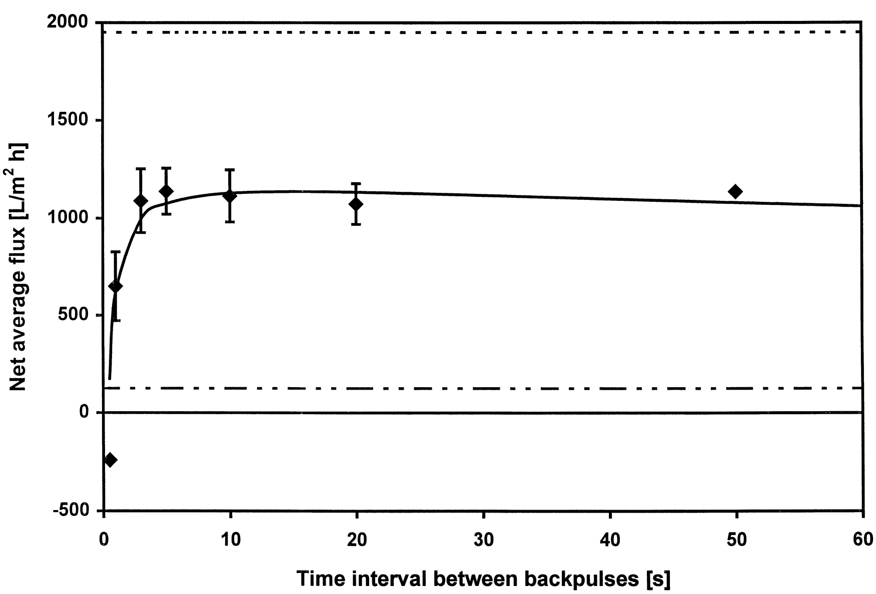

Fig. 6. Average net permeate flux vs. the duration of forward filtration between backpulses for a bentonite suspension of concentration 0.2 g l^{-1} with backpulsing at a lower backpulse duration of 0.2 s. All other experimental conditions are the same as those for Fig. 5. The solid line represents the best fit of the data based on the model with $\beta = 0.83$. Error bars represent 90% confidence intervals on four repeats.

4.2.1. Bentonite suspensions

Fig. 5 shows the measured net flux plotted against different values of the forward filtration time for a suspension of bentonite clay in water of concentration 0.2 g l^{-1}. Each point on the chart represents the average value of the permeate flux obtained from 2 to 4 repeats of experiments. In all cases, the duration of the backpulse was maintained at $t_b = 0.5$ s. It is clear that rapid backpulsed operation is highly effective in restoring the permeate flux. The maximum flux with backpulsing of $\langle J \rangle_{max} = 1300$ l m^{-2} h^{-1} is two-thirds of the clean water flux and more than 10-fold greater than the long-term flux in the absence of backpulsing. The solid line in Fig. 5 represents the model prediction for the net flux based on the model of Eqs. (1)–(3) and a best-fit value of $\beta = 0.82$ for the cleaning efficiency. In contrast, we found that the flux recovery after a long forward filtration experiment followed by a 10-min period of reverse filtration (or backflush) is only 56% of the original clean water flux. This suggests that rapid backpulsing operation reduces the degree of irreversible fouling on the surface of the membrane.

Fig. 6 shows the measured average net flux vs. the forward filtration time for the same 0.2 g l^{-1} bentonite suspension of Fig. 5, but at a shorter backpulse duration of $t_b = 0.2$ s. The results are very similar to those with $t_b = 0.5$ s; the best-fit value for the cleaning efficiency is $\beta = 0.83$, and the maximum net flux is again $\langle J \rangle_{max} = 1300$ l m^{-2} h^{-1}. Apparently, the shorter backpulse provides essentially the same cleaning that

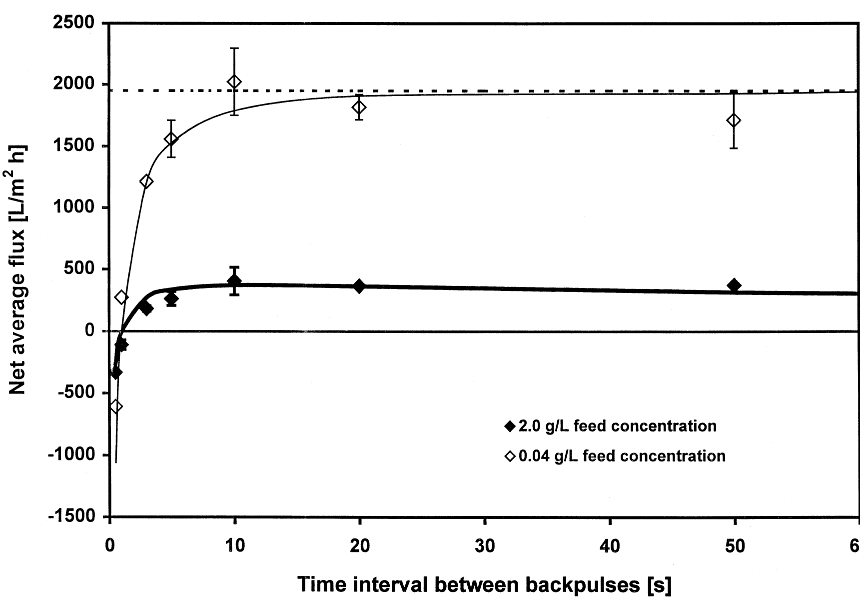

Fig. 7. Average net permeate flux vs. the duration of forward filtration between backpulses for bentonite suspensions of two different concentrations. Experiments were run at the same conditions as those of Fig. 5. The solid curves represent the best fit of the data based on the model with $\beta = 1.0$ for 0.04 g l^{-1} feed and $\beta = 0.24$ for 2.0 g l^{-1} feed. The lower curve is for a feed concentration of 2.0 g l^{-1} (thickest line), while the upper curve is for a feed concentration of 0.04 g l^{-1} (thinnest line). Error bars represent 90% confidence intervals on four repeats. The dotted line is the clean membrane water flux.

the longer one does for this system, but the fluid lost during backpulsing is small in either case compared to the fluid gained during forward filtration. The high-frequency case of $t_f = 1.0$ s is an exception, since then the fluid lost during backpulsing is significant for the longer backpulse of duration $t_b = 0.5$ s (in fact, leading to a negative net flux).

Fig. 7 shows the measured net flux plotted against different values of the forward filtration time for two additional concentrations of bentonite clay in water. The utilization of rapid backpulsing is most effective at low concentrations, since high concentrations lead to rapid fouling during forward filtration, and thicker foulant layers which are not as easily removed. The best-fit cleaning indices are $\beta = 1.0$ and 0.24 for $c = 0.04$ g l^{-1} and 2.0 g l^{-1}, respectively. For the most dilute suspension (0.04 g l^{-1}), the net permeate fluxes achieved with backpulsing are essentially equal to the clean water flux. Since the time constant for cake growth is relatively large (Table 2) for this dilute suspension, backflushes are also expected to be effective, provided that they are no more than a few minutes apart.

Fig. 8 shows the results of backpulsing experiments with a hollow fiber unmodified polysulfone membrane with nominal pore size 0.45 μm and a feed suspension of bentonite of concentration 2.0 g l^{-1}. For this case, the maximum flux improvement over the long-term flux is on the order of 3-fold, as compared to the observed flux improvement of 4.5-fold for the ceramic membrane at the same operating conditions.

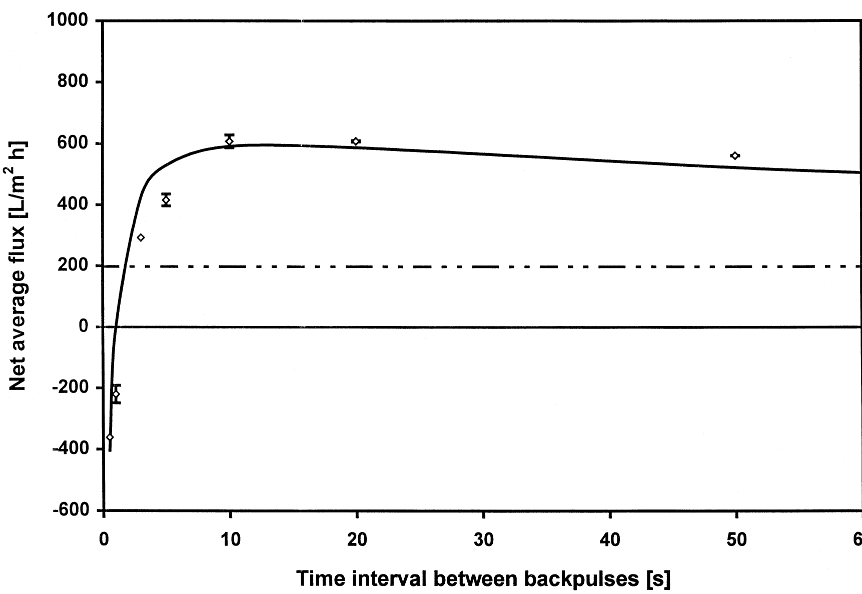

Fig. 8. Average net permeate flux vs. the duration of forward filtration between backpulses for a bentonite suspension of concentration 2.0 g l^{-1} with backpulsing of duration 0.5 s for a 0.45 μm nominal pore diameter, hollow fiber polysulfone cartridge. The solid curve is the best fit of the data with the model using $\beta = 0.26$. All other experimental conditions are the same as those for Fig. 5. Error bars represent 90% confidence intervals on two to four repeats. The dashed–dotted line is the long-term flux without backpulsing.

However, both the long-term flux without backpulsing and the maximum net flux with backpulsing are higher for the polysulfone membrane. The cleaning efficiency is essentially the same for the two membranes at this high concentration.

In all of the experiments discussed above, both backpulsed and nonbackpulsed, the permeate streams contained no visible particles and were very clear. For example, the nephelometric turbidity of the permeate for the 0.2 g l^{-1} experiments was always less than 1.4 NTU (nephelometric turbidity units), whereas the 'clean' tap water had 1.0 NTU and the feed suspension had 50 NTU. It is concluded that rapid backpulsed operation does not significantly affect the high quality of permeate delivered, at least for the suspensions considered herein.

4.2.2. Oil-in-water dispersions

Initially, backpulsing with both ceramic and polysulfone membranes yielded large increases in the net permeate flux for the oil-in-water dispersions. However, it was later found that the degree of flux increase gradually degraded with each subsequent use of membranes, despite the cleaning procedures employed between experiments. Fig. 9 shows such effects for both the ceramic (α-alumina) tubular membrane and the unmodified polysulfone hollow fiber module. The data were not fit to the model in this case, since the oil-in-water dispersions exhibit internal fouling during the early stages of

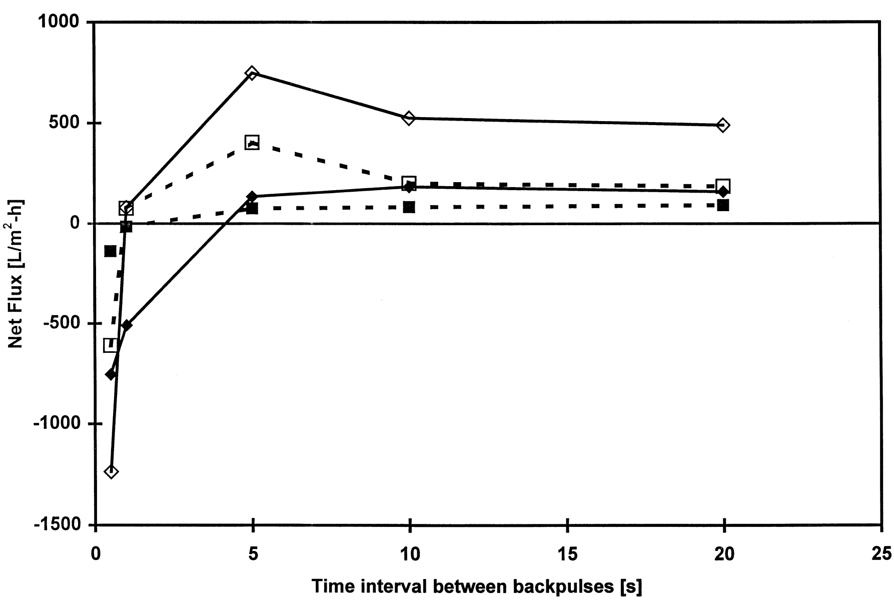

Fig. 9. Net flux vs. forward filtration time between backpulses for oil-in-water backpulsing experiments with a backpulse duration of 0.5 s done with new and used (after approximately 20 h of experimental use) 0.2 μm unmodified polysulfone hollow-fiber membrane (upper and lower dashed curves, respectively) and new and used (after approximately 200 h of use) 0.2 μm α-alumina tubular membrane (upper and lower solid curves, respectively). The oil content in the feed was 50 ppm, the cross-flow velocity was 3.5 m s^{-1}, and the feed temperature was 40°C.

forward filtration, whereas the model is based on external fouling. It is clear that, even though rapid backpulsing was effective in enhancing the permeate flux early in the life of the membranes (up to 25-fold for the ceramic membrane, and 5-fold for the polysulfone membrane), the performance severely declined with membrane use. This is consistent with what has been observed in field trials using cross-flow microfiltration of oil field waters with low-frequency backflushing [15,20], where the net permeate flux eventually declines to that of long-term nonbackpulsed operation. This decline in performance was not seen for the experiments with clay suspensions described earlier.

As shown in Table 2, the ratio $\langle J \rangle_{max}/J_s$ (maximum net flux with backpulsing divided by long-term flux without backpulsing) declines from 25 to 6 with 200 h of use of the ceramic membrane, and from 5 to 1.1 with 20 h of use of the polysulfone membrane. It is reasonable that the performance of the ceramic membrane declined more slowly, since the highly hydrophilic nature of the α-alumina layer has a stronger tendency to reject the organic phase. The more hydrophobic unmodified polysulfone presents a chemically more favorable surface for deposition of the suspended oil droplets, and for penetration of the oil into the membrane pores (due to the smaller contact angles between the oil droplet and surface), resulting in more severe irreversible and internal fouling and a relatively short useful operating life. This combination of irreversible and internal fouling is most likely the reason behind the poor performance of the polysulfone membrane after a relatively short period of use. In all cases, both backpulsed and nonbackpulsed, the permeate was very clear and contained only 1–6 ppm total hydrocarbons.

5. Economic analysis

In order to determine whether rapid backpulsing is of benefit in practice, a preliminary economic analysis was performed. We examine here the typical case of a system operated in a feed-and-bleed mode as appropriate for medium-scale and large-scale continuous wastewater treatment processes (see, for example, Ref. [21]). In these systems, a large part of the retentate stream is recirculated back to the filtration module in order to achieve the desired cross-flow velocity within the membrane. The reverse pressure pulses can be generated by a pressurized (or elevated) reservoir which is periodically replenished with clean permeate water. In this study, we consider a filtration system based on the use of ceramic modules, as they provide the best resistance to the typically harsh conditions found in wastewater treatment. A more complete description of the proposed system can be found in Murkes and Carlsson [22].

The cost of the wastewater treatment membrane plant can be apportioned into capital and operating costs. Capital costs represent the investment required to provide a given capacity for treated water production. This investment includes costs such as land, engineering and construction, and installations (civil, electrical, mechanical, etc.). Common practice for calculating the investment required per unit volume of treated water ($ m^{-3} of treated water) involves annualizing the initial cost incurred in erecting the facility and dividing this by its intended annual capacity. The operating costs comprise those expenses associated with plant operation and maintenance, such as energy consumption, membrane replacement, labor, preventive and corrective maintenance, and waste disposal.

The annualized total capital costs per unit treated volume, which include total initial membrane and nonmembrane costs annualized over the design life of the plant, are calculated by the following expression due to Wiesner et al. [23]:

$$\mathrm{CC} = \frac{\left(c_{\mathrm{mod}} \cdot N_{\mathrm{mod}} + \$1.50 \times 10^{5}\, N_{\mathrm{mod}}^{0.74}\right)(\mathrm{AF})}{Q}, \tag{4}$$

where c_{mod} is the cost ($) of a single membrane module (including both membrane and housing), $N_{\mathrm{mod}} = \mathrm{Integer}(A_{\mathrm{reqd}}/A_{\mathrm{mod}} + 0.5)$ is the number of required modules, A_{mod} is the total surface area of the membrane module in question, A_{reqd} is the required total membrane area, and AF is the amortization factor (0.10 yr^{-1} for the present application). The nonmembrane cost factor ($\$1.50 \times 10^{5}$) includes pumps, valves, and piping for periodic membrane backflushing and so includes the primary capital costs for a backpulsing system. The required membrane area is a function of the plant capacity or volumetric treatment rate, Q, and the attainable net permeate flux, $\langle J \rangle$:

$$A_{\mathrm{reqd}} = \frac{Q}{\langle J \rangle}. \tag{5}$$

The main operating costs considered herein are energy (for pumping feed, recycling concentrate, and pumping backpulse permeate to attain the required backpulsing pressure), membrane replacement, maintenance and labor. The model used in these calculations is largely based on the model for operating costs of Pickering and Wiesner [24]. The costs of pumping were determined by knowing the required pressure head, pumping flow rate and pump efficiency. The cost of pumping the permeate for backpulsing was adjusted by the ratio of backpulse duration to total cycle duration, $t_{\mathrm{b}}/(t_{\mathrm{b}} + t_{\mathrm{f}})$, in order to account for cyclic pump operation. The cost of retentate recycling is given by the cost of supplying the energy required to maintain a specified cross-flow velocity in the membrane module. The membrane replacement costs were considered as a variable (or operating) cost as opposed to a periodic investment of capital. This cost contribution was considered as a constant operating cost by assuming that the membranes are replaced at a fixed interval per the recommendations of the manufacturer; for example, the typical replacement interval for the ceramic membranes is 8 years [24]. The required labor was calculated from standard data for man-hour requirements for fluid processing plants [25], while the cost of maintenance (materials and labor) was taken as an annualized 1.5% of the nonmembrane cost [16]. The cost of waste disposal is highly dependent on the nature of the feed and on plant location and should be carefully considered for each specific plant. For the general case considered herein, the contributions of these costs for the cases examined (conventional treatment and nonbackpulsed and backpulsed microfiltration) were assumed to be equal.

The costs for a conventional treatment plant have been previously calculated elsewhere [16,23]. These include capital and operating costs for the standard operations of rapid-mixing and flocculation, gravity settling, and multimedia and cartridge filtration. Table 3 summarizes the cost of treated water for three different wastewater treatment plant capacities. The nonbackpulsed microfiltration calculations are based on a permeate

Table 3
Cost comparison between the conventional approach to water treatment, cross-flow microfiltration (CFMF) without backpulsing, and cross-flow microfiltration with rapid backpulsing

Plant capacity (m^3 d^{-1})	Cost of treatment (\$ m^{-3})						
	Conventional	Nonbackpulsed CFMF			Rapid backpulsed CFMF		
	Total	Capital	Operating	Total	Capital	Operating	Total
500	0.88	1.25	0.55	1.80	0.36	0.08	0.44
2000	0.49	0.93	0.54	1.47	0.28	0.08	0.36
6000	0.32	0.76	0.53	1.29	0.23	0.08	0.31

Calculations are based on a system employing ceramic membrane modules with 90 l m^{-2} h^{-1} and 410 l m^{-2} h^{-1} flux values for nonbackpulsed and backpulsed operation, respectively.

flux of 90 l m^{-2} h^{-1}, while the calculations for operation with rapid backpulsing are based on an operating flux of 410 l m^{-2} h^{-1} (as determined from our experiments with the ceramic membrane and the most concentrated bentonite suspension). It is seen that nonbackpulsed operation cannot compete with the conventional process within the range of plant capacities examined; however, operation at the higher flux resulting from rapid backpulsing results in cross-flow microfiltration becoming a viable alternative in plants with capacities of up to 6000 m^3 d^{-1}. The capital costs, including the membrane modules, are generally higher than the operating costs.

The higher permeate flux levels that result from rapid backpulsing have a direct effect on both capital (membrane and nonmembrane) and operating costs. Lower membrane

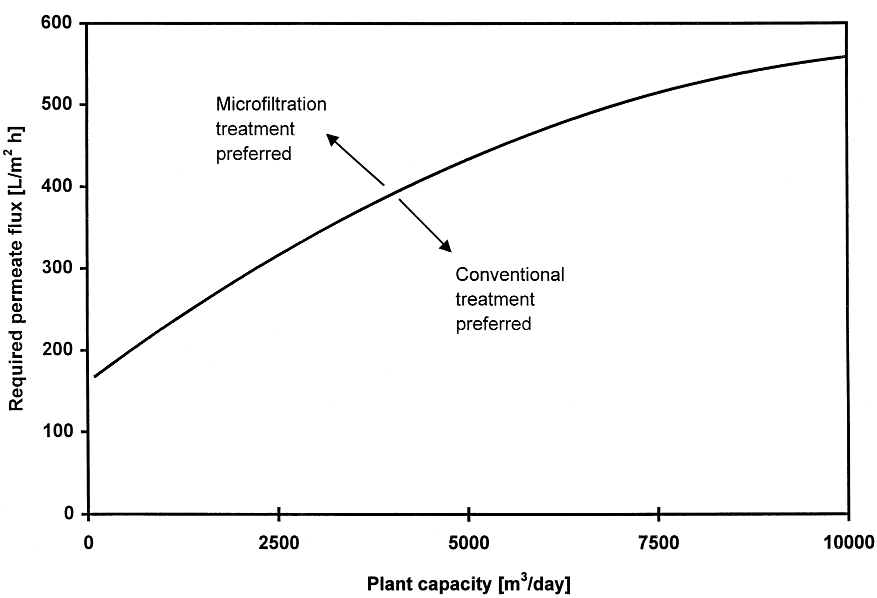

Fig. 10. Required permeate flux for membrane filtration of wastewater to be competitive with conventional treatment as a function of capacity of the treatment plant.

capital costs result from the diminished membrane area requirements with rapid backpulsing. The reduction in nonmembrane capital costs is a direct consequence of the more compact system (smaller area and volume requirements, thus less costly civil, electrical and mechanical installations). Finally, the lower operating costs result mainly from the reduction in the costs of membrane replacement (which again traces back to the lower membrane area associated with higher net flux) and retentate recycling (a higher rate of recycle is needed with more membrane modules in order to maintain the specified cross-flow velocity).

A chart showing the minimum required permeate flux for rapid backpulsed microfiltration to be economically competitive with conventional pretreatment is presented in Fig. 10. The chart shows which treatment option results in lower costs of treated water for a particular design capacity, if one knows a priori the value of the attainable permeate flux for the feed to be treated. Backpulsing experiments such as those described in this study can be performed with actual samples of the wastewater feed in order to determine the maximum attainable permeate flux, and thus to make the final decision on which route of treatment to pursue. For the chart in Fig. 10, 2.6 m s^{-1} cross-flow velocity and 20 psi transmembrane pressure were used for all plant capacities; these values are in the middle of the optional ranges [15,16], which are nearly independent of plant capacity [16].

6. Conclusions

The effectiveness of rapid backpulsing in enhancing the permeate flux in cross-flow microfiltration has been demonstrated. Experiments show that rapid backpulsing is most effective for the treatment of feed streams containing relatively nonadhesive suspended particulates, and that its effectiveness declines with increasing feed concentration. For backpulse durations of $t_b = 0.2$ and 0.5 s, it was observed that extremely high backpulsing frequencies (greater than approximately 1 Hz) are counterproductive due to the increased permeate loss during the frequent reverse pulses, while much lower backpulsing frequencies (less than 0.01–0.1 Hz) tend to result in lower permeate fluxes due to fouling during the prolonged operation in the forward filtration part of the cycle.

Permeate flux improvements from 3-fold to over 10-fold were observed for suspensions of bentonite clay of various concentrations in water. The degree of flux enhancement observed was comparable for the two membrane modules tested, while the permeate quality was always very high. Our preliminary results with oily wastewaters (particularly the results of experiments early in the life of the membranes) suggest that operation with rapid backpulsing could prove to be useful if coupled with an additional means of combating foulant deposition. For example, for the case where a ceramic membrane is employed, a periodic cleaning procedure consisting of direct firing of the membrane could be implemented, and applied whenever the permeate flux declined to levels below those required for economical operation. On the other hand, the alternative of utilizing less expensive, more lightweight polymeric membranes should be investigated further; perhaps modifying the chemical nature of the surface of the membrane and operation in the rapid backpulsing mode would permit prolonged backpulsed operation between off-line membrane cleaning.

An economic analysis on a cross-flow microfiltration system based on the data for clay suspensions and ceramic membrane modules reveals that operation without backpulsing (at a permeate flux of approximately 100 $l\ m^{-2}\ h^{-1}$) results in prohibitively high water treatment costs. Rapid backpulsed operation (at a permeate flux of approximately 400 $l\ m^{-2}\ h^{-1}$) results in lower treatment costs than conventional treatment for plant capacities under 6000 $m^3\ d^{-1}$.

The degree of flux enhancement will depend on the particular feed water to treat. In practice, pilot membrane fouling and rapid backpulsed testing should be performed with the actual plant feed wastewater, in order to realistically estimate the maximum attainable net permeate flux and the best values of the key backpulsing parameters. Based on the current work, it is expected that rapid backpulsed microfiltration will be an effective treatment alternative for wastewaters containing dilute concentrations of suspended particulates and droplets.

Acknowledgements

The authors acknowledge the United States Department of the Interior, Bureau of Reclamation, the Center for Separations Using Thin Films at the University of Colorado, and the United States Environmental Protection Agency for their financial support of this project. Appreciation is also extended to Lisa Henthorne and Frank Leitz of the Bureau of Reclamation for their comments and suggestions, to Harlan Polishook of AG Technology for valuable discussions on the AGT membranes, and to Cliff Juengst of UNOCAL for providing the oil samples. The authors also thank Jill Fletcher and Marc Kroll for assisting with the experimental work.

References

[1] S.H. Hyun, G.T. Kim, Synthesis of ceramic microfiltration membranes for oil/water separation, Sep. Sci. Tech. 32 (1997) 2927.
[2] F.F. Nazzal, M.R. Wiesner, Microfiltration of oil-in-water emulsions, Water Environ. Res. 68 (1996) 1187.
[3] A.B. Koltuniewicz, R.W. Field, T.C. Arnot, Cross-flow and dead-end microfiltration of oily-water emulsion: Part I. Experimental study and analysis of flux decline, J. Membr. Sci. 102 (1995) 193.
[4] X. Shen, J.K. Park, B.J. Kim, Separation of nitrocellulose-manufacturing wastewater by bench-scale flat-sheet crossflow microfiltration units, Sep. Sci. Tech. 29 (1994) 333.
[5] D.J. Chang, S.J. Hwang, removal of metals ions from liquid solutions by crossflow microfiltration, Sep. Sci. Tech. 31 (1996) 1831.
[6] S.M. Hasany, Najamuddin, M. Ikram, Uptake of traces of selenite by manganese dioxide from aqueous solutions, Sep. Sci. Tech. 32 (1997) 1945.
[7] C.F. Vasilieff, T.A. Doneva, L.G. Ljutov, Cross-flow microfiltration of bentonite-in-water dispersions: Initial transient effects at low concentration, J. Membr. Sci. 119 (1996) 65.
[8] J.H. Lee, D.I. Song, Y.W. Jeon, Adsorption of organic phenols onto dual organic cation montmorillonite from water, Sep. Sci. Tech. 32 (1997) 1975.
[9] Y.C. Hsu, C.C. Chiang, M.F. Yu, Adsorption behavior of basic dyes on activated clay, Sep. Sci. Tech. 32 (1997) 2513.
[10] R.H. Davis, Theory of crossflow microfiltration, in: W.S.W. Ho, K.K. Sirkar (Eds.), Membrane Handbook, Van Nostrand- Reinhold, New York, 1992, p. 457.

[11] S.D.N. Freeman, O.J. Morin, Recent developments in membrane water reuse projects, Desalination 103 (1995) 19.
[12] V.G.J. Rodgers, R.E. Sparks, Effect of solution properties on polarization redevelopment and flux in pressure pulsed ultrafiltration, J. Membr. Sci. 78 (1993) 163.
[13] S.G. Redkar, R.H. Davis, Cross-flow microfiltration with high frequency reverse filtration, AIChE J. 41 (1995) 501.
[14] V. Kuberkar, P. Czekaj, R.H. Davis, Flux enhancement for membrane filtration of bacterial suspensions using high-frequency backpulsing, Biotech. Bioeng., under review.
[15] A. Zaidi, K. Simms, S. Kok, The use of micro/ultrafiltration for the removal of oil and suspended solids from oilfield brines, Water Sci. Technol. 25 (1992) 163.
[16] G. Owen, M. Bandi, J.A. Howell, S.J. Churchouse, Economic assessment of membrane processes for water and wastewater treatment, J. Membr. Sci. 102 (1995) 77.
[17] Horiba OCMA 220 analyzer, Users manual, Tokyo, Japan.
[18] E.M. Tracey, R.H. Davis, Protein fouling of track-etched polycarbonate microfiltration membranes, J. Colloid Interface Sci. 167 (1994) 104.
[19] J. Mueller, Y. Cen, R.H. Davis, Crossflow microfiltration of oily water, J. Membr. Sci. 129 (1997) 221.
[20] A. Zaidi, K. Simms, S. Kok, R. Nelson, in: J.P. Ray, F.R. Engelhart (Eds.), Produced Water, Plenum, New York, 1992, p. 489.
[21] L.J. Zeman, A.L. Zydney, Microfiltration and Ultrafiltration, Marcel Dekker, New York, 1996, p. 388.
[22] J. Murkes, C.G. Carlsson, Crossflow Microfiltration, Wiley, Chichester, UK, 1988, p. 58.
[23] M.R. Wiesner, J. Hackney, S. Sethi, J.G. Jacangelo, J.M. Laine, Cost estimates for membrane filtration and conventional treatment, J. AWWA 85 (1994) 33.
[24] K.D. Pickering, M.R. Wiesner, Cost model for low-pressure membrane filtration, J. Environ. Eng. 119 (1993) 772.
[25] M.S. Peters, K.D. Timmerhaus, Plant Design and Economics for Chemical Engineers, 4th edn., McGraw-Hill, New York, 1991, p. 198.