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D. Niriella[a]; R. P. Carnahan[a]
[a] Department of Civil and Environmental Engineering, University of South Florida, Tampa, Florida, USA

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Influence of Operating Parameters on Clay Fouling of a RO Membrane

D. Niriella and R. P. Carnahan
Department of Civil and Environmental Engineering, University of South Florida, Tampa, Florida, USA

Abstract: Effect of operating parameters on permeate flux due to clay fouling on a reverse osmosis membrane in a laboratory scale plate and frame membrane module is discussed. Bentonite clay was used as the model foulant. No steady state conditions were observed even after 24 hrs of operation. The flux–time curves obtained could be generalized into three categories. In the short term, the application of higher pressure leads to higher initial permeate flux. However, this effect has a negative effect on the long term flux by the formation of a more dense and a less porous cake layer on the membrane. Particle concentration plays a more dominant role in flux reduction. The results show that an increase in particle concentration from 100 to 300 mg/l could lead to an increase in the flux decline between 15.9 to 47.7 percent. For the tested conditions, crossflow velocity did not have much of an impact on the permeate flux.

Keywords: Bentonite, clay, reverse osmosis, fouling, operating parameters

INTRODUCTION

Recently, membranes have become a very popular mode of obtaining highly purified water. However, there are many operational and maintenance problems present in membrane treatment plants that need to be solved. Of these, fouling of the reverse osmosis (RO) membrane is one of the issues that need urgent attention. Decrease in water productivity, an increase in applied pressure, membrane degradation, and a decrease in permeate quality

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Address correspondence to D. Niriella, Department of Civil and Environmental Engineering, University of South Florida, 4202, E. Fowler Avenue, Tampa, Florida 33620, USA. Tel.: 1-813-974-7226; Fax: 1-813-974-2957; E-mail: dniriell@eng.usf.edu

are effects of membrane fouling (1). Although pretreatment is carried out to remove the fouling material from feed water, 100 percent removal efficiency is impossible.

Natural water sources contain many impurities such as clay materials, colloidal silica and organic matter, oxides of iron, manganese, and aluminum, and suspended matter. Clay, which has an aluminum silicate structure, represents the vast majority of colloidal particles found in natural waters in the particle size ranging between 0.3 to 1.0 microns. Aluminium silicate clays are the product of natural weathering of the geological environment. Therefore, they are found throughout the world and are not restricted to any specific location (2). During floods and landslides, heavy sediment loads, comprising clay can be experienced in feed water sources for RO water treatment units, which could exceed the capacity of the pre-treatment units (3).

Ultrafiltration (UF) and Microfiltration (MF) fouling mechanisms cannot be applied to Reverse Osmosis (RO) membranes due to the differences in membrane pore size and permeation rates. For example, pore blocking is an important mechanism in the fouling of MF and UF membranes but not so in RO membrane fouling (4). The rapid drop in the flux observed in MF and UF membrane filtration is due to blocking of membrane pores and thereby increasing the membrane resistance. Subsequent reduction in flux is due to the formation and growth of the cake layer (5). Wang and Song, 1999 said that in the case of MF and UF, the flux decline in most instances can be generally attributed to the formation and growth of a cake layer on the membrane surface (6). Zhang and Song, 2000 showed that the permeate flux of cross-flow filtration is controlled by the dynamic process of cake formation and growth (7). Cake resistance depends on both the degree of flocculation in the feed suspension and the pressure stress that exists under the filtration conditions (8).

Several studies have been conducted on RO membrane fouling. Cohen and Probstein, 1986 investigated the cellulose acetate RO membrane fouling with ferric hydroxide (9). Although (4) investigated the RO membrane fouling by aluminium oxide colloids, (10), fouling by Silica colloids, (11), fouling by polystyrene latex, very few studies have been performed to investigate the effect of the presence of clay particles in RO feed water.

Flux decline is an important factor to be considered in membrane water treatment system designs. The affects of operating variables on performance of a membrane vary from one membrane manufacturer to another. In order to design, optimize and efficiently operate membrane systems, it is necessary to understand how the system's operating parameters effect flux-time relationship (12).

When feed water that contains small particles are filtered through a membrane, various filtration mechanisms take place. Particles separated by the membrane, in addition to the pure membrane resistance, provide additional resistance to the permeate flow. With time, as the particle deposit layer grows, this resistance also increases (13).

The objective of this paper is to systematically evaluate operating parameters, such as transmembrane pressure, crossflow velocity while exposing a low-fouling-composite 1 (LFC 1) membranes in a laboratory scale membrane unit to varying concentrations of particulate clays. The presence of other individual ions influence ionic strength of the feed solution. Ionic strength of the feed solution and the organic matter in turn influence the cake formation and fouling behavior. However, the matrix effects of these were not considered in this work. Bentonite is used as the model foulant material. Initially, the shapes of the flux-time relationship curve is discussed followed by the influence of operating parameters such as feed concentration, transmembrane pressure and cross-flow on permeate flux respectively. The last section of the paper tries to statistically model the flux decline and compare the model results with experimentally obtained values.

MATERIALS AND CHEMICALS

All chemicals used in these experiments were of ACS certified or better. Laboratory grade bentonite clay (Fisher Scientific, Fairlawn, NJ) was used in all the experimental runs. No further purification of bentonite clay particles were carried out. To induce fouling within the experimental time scale, it was decided to use a bentonite concentrations of 100, 200 and 300 mg/l respectively.

EXPERIMENTAL UNIT

A schematic diagram of the laboratory scale crossflow RO test unit is shown in Fig. 1. The membrane test unit consists of membrane cell, a high pressure

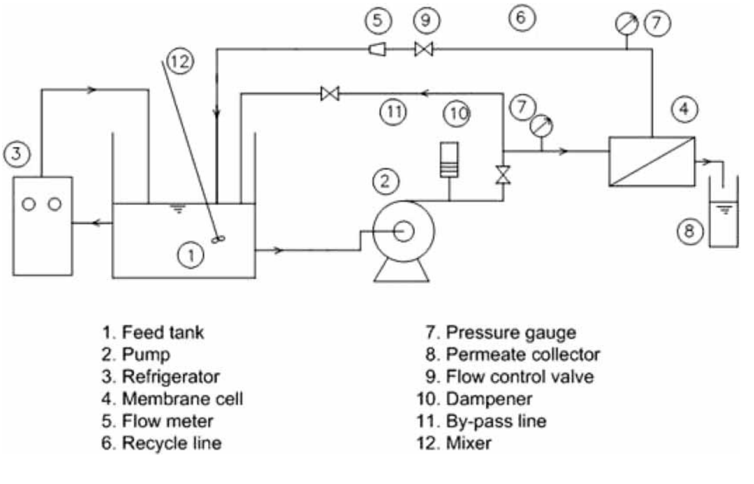

Figure 1. Schematic diagram of the experimental set-up.

positive displacement pump/motor, a feed reservoir, a mixer and a temperature control system. In this unit, the test solution is held in a 100 liter feed reservoir (1) and fed to the membrane cell by a constant flow diaphram pump (2) (Hydra-Cell, Wanner Engineering Inc. Mineapolis, Minnesota), capable of providing a maximum pressure of 1,000 psi and a maximum flow of $6.93 \times 10^{-5}\,m^3/s$ (1.1 gpm). A pulsation dampener (CAT) (10) is installed at the outlet port of the pump to eliminate vibrations created by pumping. The clay particles are held in suspension in the feed tank by continuous agitation with a mixer (12). The crossflow velocity in the membrane cell (4) is controlled by passing a portion of the test solution back to the feed tank using a by-pass valve. The desired transmembrane pressure is set by throttling the needle valve on the concentrate side of the membrane. Temperature is controlled by circulating feed water through a chiller.

All pipes and fittings on the high pressure side, which were up to the flow control valve on the concentrate side of the membrane and up to the by-pass line flow control valve were of stainless steel type 316. In all other locations, braded PVC hosing were used. For permeate collection line, PVDF pipes were used.

Membrane Cell and Holder

A stainless steel rectangular plate-and–frame membrane cell was used in the experiments. The feed water channel dimensions of the cells were 14.6 and 9.5 cm for channel length and width, respectively. The channel height is 0.2 cm.

Membrane Type and Specifications

Commercially available thin film composite Low Fouling Composite (LFC1) polyamide flat sheet membranes manufactured by Hydranuatics, Oceanside, CA were used in the experiments. The membranes operating pH range were between 3 to 10. According to manufacturer's information, its NaCl rejection capacity is 99.4% and is capable of producing 29 GFD at a test pressure of 225 psi and at a temperature of 25° C when used with 1500 ppm NaCl solution. Surface property of LFC 1 membrane has been modified during casting process to provide the membrane with low fouling characteristics. The membrane maintains a relatively neutral surface charge over both acidic and basic pH environment. Its hydrophilicity is 47° (14).

Membranes were delivered in flat sheet forms to the laboratory. The membranes were cut into coupons of size required by the membrane filtration cell, which is 7.5 × 5.5 inches and necessary holes made using a template. Each membrane coupon was washed with deionized water and then stored in fresh deionized water at 5° C, prior to using in fouling experiments. Used membrane is replaced by a new membrane at the beginning of each new experimental run.

Compaction of the Membrane

Before using the membranes in any of the experimental runs, deionized (DI) water was circulated at 500 psi (1734 kPa) for up to 12 hrs to prevent flux decline due to membrane compaction before the clay particles were added to the system.

Fouling Runs

For fouling runs with bentonite, first filtration was carried out with distilled water at the experimental pressure and cross flow rate until a steady state was reached. Once the steady state was reached, a solution containing bentonite particles were added to the system to obtain the final suspension concentration. Permeate flux was monitored with time until the steady state was reached. If the steady state is not reached by at least 8 hrs, the experiment was curtailed after 8 hrs. In all the experimental runs pH value was maintained around 7.0 and the temperature around 22 $\pm$ 1°C.

Membrane and System Cleaning After Operation

At the end of each fouling runs, the feed water in the system was replaced by tap water to flush the entire system. Thereafter, the tap water was replaced by distilled water which was used to flush the system until all the fouling particles were removed from the system.

Experimental Conditions

Transmembrane pressure (ΔP) (200, 300 and 500 psi), feed concentration (Co) (100, 200 and 300 mg/l) and crossflow velocity (V) (4.04, 8.08 and 12.9 cm/s) were selected as the variable parameters. Total mass of bentonite clay used in the system will be between 2.5 and 7.5 grams. Permeate flux which is an indication of membrane fouling taking place was considered as the dependable variable and monitored with time.

RESULTS AND DISCUSSION

Flux-Time Relationship

At the beginning, investigative experiments were carried out to determine whether a steady state condition (no variation of flux with time) could be achieved under the selected operating conditions. Surprisingly, even after 24 hrs of operation (Fig. 2), no steady state condition was reached.

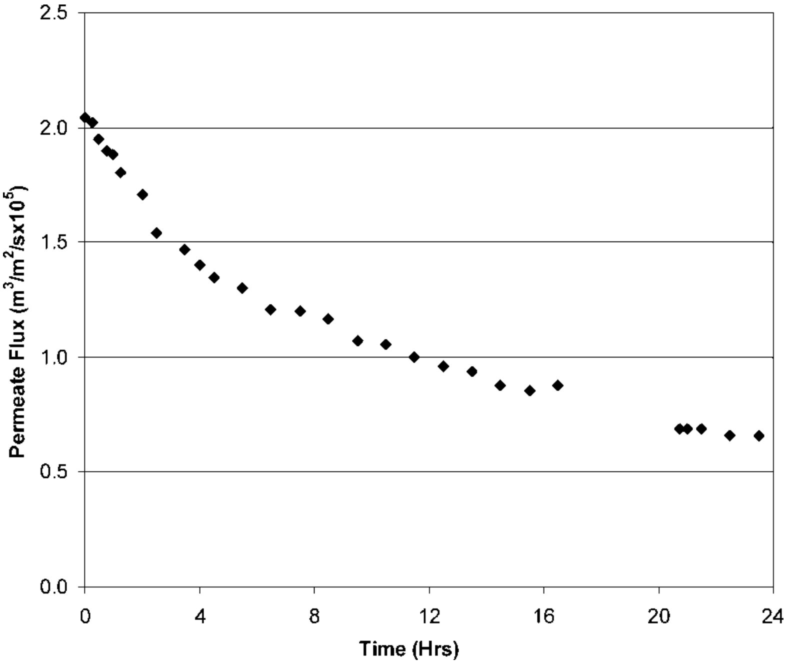

Figure 2. Flux vs time for applied pressure (ΔP) = 300 psi, Ben. concentration (Co) = 100 mg/l and cross flow velocity (V) = 12.9 cm/s.

Flux-time curves obtained could be generalizes into three categories:

Flux exhibits an initial decrease, which could be a rapid exponential type or near linear (Figs. 3 and 4) followed by a slower almost linear decrease with time or almost linear (Fig. 5) until the run is curtailed.

The rapid exponential flux decline followed by a slower linear behavior was observed during the application of operating pressures of 500 psi and 300 psi at bentonite concentrations of 300 mg/l. The closer examination of the flux time relationships show that an initial rapid linear drop followed by a slower linear portion for runs showed that they all came from runs, with operating pressures of 500 psi, at feed concentrations of 100 mg/l and at higher concentrations for operating pressures of 300 psi. A constant linear relationship between flux vs time was displayed in all experiments with operating pressures at 200 psi and for lower feed concentrations.

It can be argued that the rapid exponential or linear decrease is dependent on both the applied pressure and feed concentration. Lowering the pressure and concentration results in an initial flux decline that becomes linear and less steep. Pillay and Buckley, 1988, found that in the case of crossflow microfiltration (CFMF) of a limestone suspension, the initial rapid decrease in flux was due to the rapid formation of the cake layer, which could be the

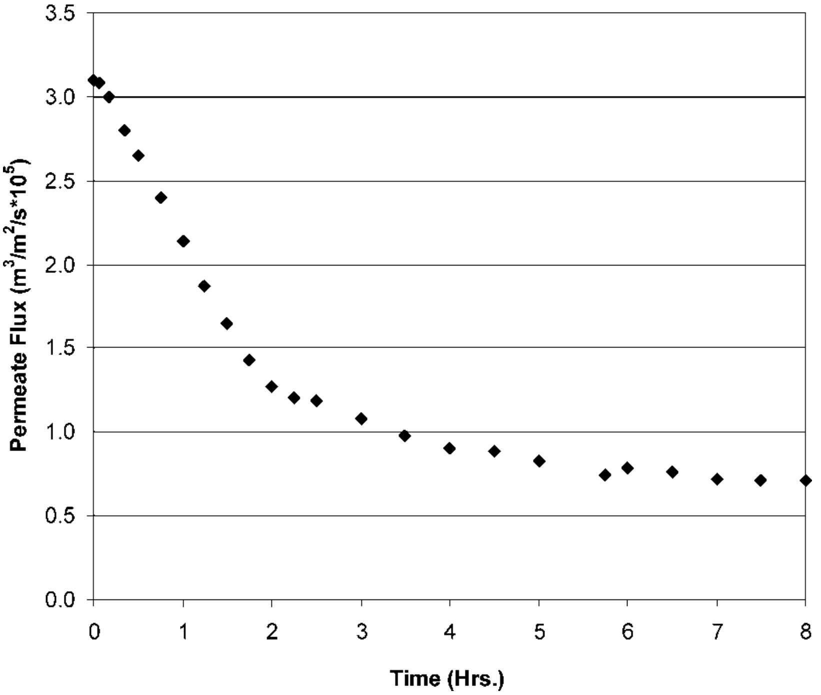

Figure 3. Flux vs time for applied pressure (ΔP) = 500 psi, Ben. concentration (Co) = 200 mg/l and cross flow velocity (V) = 8.08 cm/s.

same for bentonite. The application of higher pressures (more than 200 psi) and the compressible nature of clay particles (13) could result in compression of it. This may not occur in low pressure systems such as MF systems. Thereafter, the cake thickness stays a fixed value while changing its characteristics (eg. Porosity), which is the reason for the slow decrease in the flux. In the case of the bentonite, it could be due to the slow transport of finer particles into the cake leading to the reduction of cake permeability, together with, cake compression due to higher applied pressure.

Effects of Operating Variables on Flux

Applied Pressure

Figures 6 and 7 show the typical effect of applied pressure on flux decline. The results show that for the same bentonite concentration and flow velocity, application of a higher pressure could yield higher permeate flux at the beginning, it could also lead to a greater flux decline.

This feature shows the importance the applied pressure force plays in filtration. Higher initial permeate flux brings more clay particles towards the

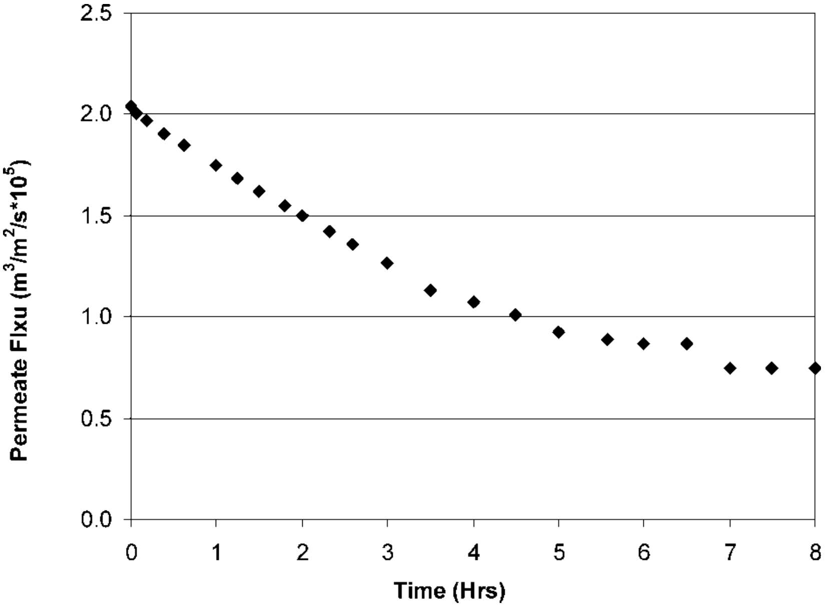

Figure 4. Flux vs time for applied pressure (ΔP) = 300 psi, Ben. concentration (Co) = 200 mg/l and cross flow velocity (V) = 8.08 cm/s.

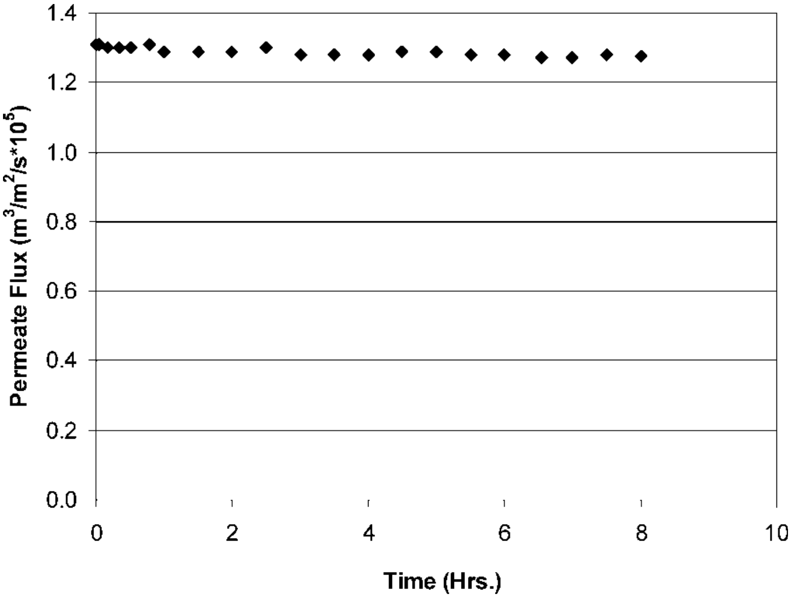

Figure 5. Flux vs time for applied pressure (ΔP) = 200 psi, Ben. concentration (Co) = 100 mg/l and cross flow velocity (V) = 4.04 cm/s.

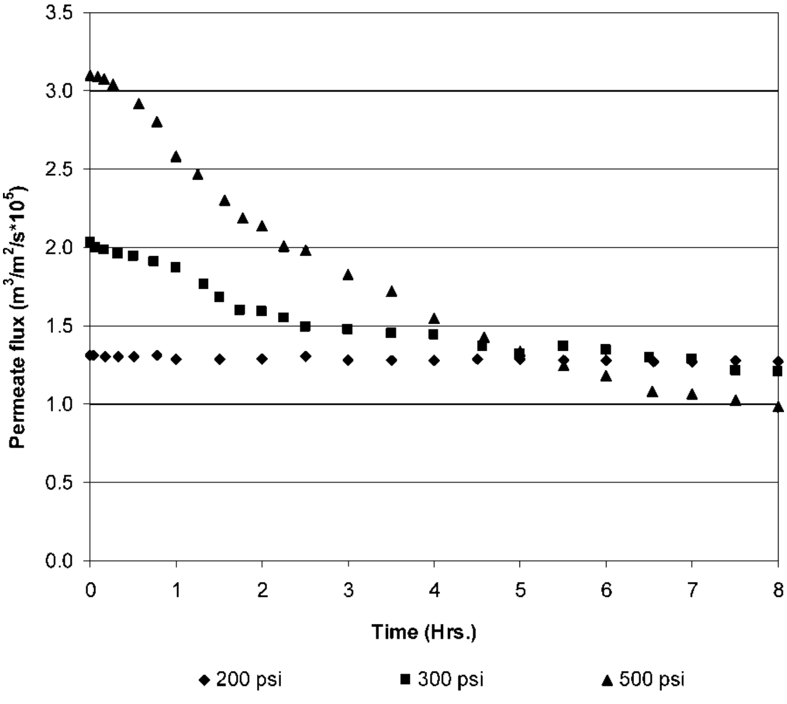

Figure 6. Flux vs time applied pressure, Ben. concentration (Co) = 100 mg/l and cross flow velocity (V) = 4.04 cm/s.

membrane for a cake layer to be formed, and the pressure force also compact the particles and the end results are the formation of a less porous well compacted cake layer on the membrane surface leading to a lower permeate flux.

Particle Concentration

The typical effect of bentonite clay particle concentration on the fouling rate is shown in Figs. 8–10. It appears that particle concentration plays a dominant role in the permeate flux decline. The results show that greater flux decline is achieved at higher feed concentrations. The large decline of permeate flux at higher feed particle concentration is due to increased particle transport. As the particle transport rate to the membrane increases (product of permeate flux and particle concentration), the overall rate of clay deposition on the membrane increases resulting in higher resistance to water flow through the membrane.

Based on the flux—time results, quantitative estimation of the flux decline at the end of 8 hour runs, for each operating condition, was carried out. The results are presented in Table 1. The results show that an increase in particle concentration from 100 to 300 mg/l could lead to an increase in the flux decline between 15.9 to 47.7 percent. The higher value reported at

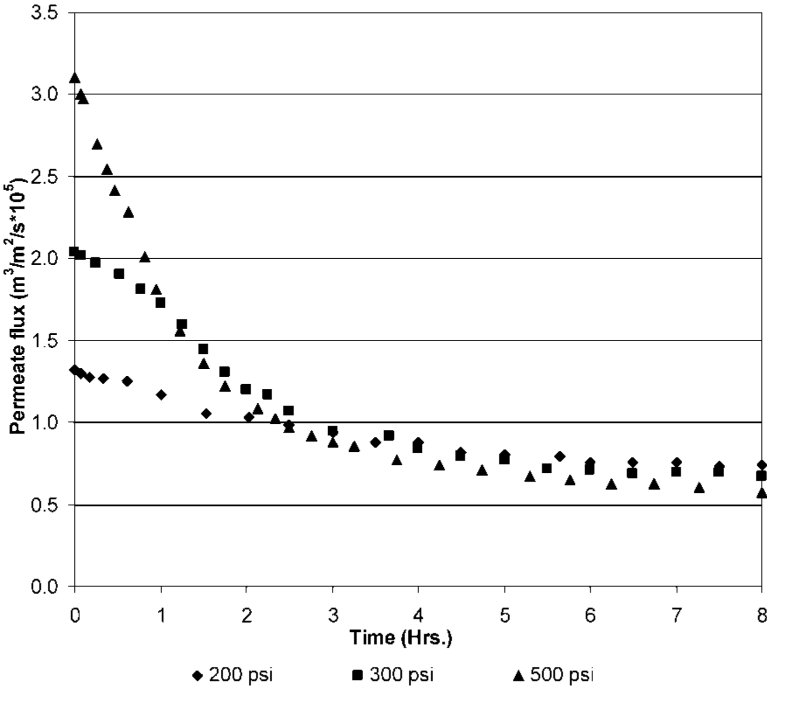

Figure 7. Flux vs time for applied pressure, Ben. concentration (Co) = 300 mg/l and cross flow velocity (V) = 12.9 cm/s.

lower pressures and vice versa. As the pressure increases, the effects of particle concentration on flux decline reduces.

Crossflow Velocity

The typical effect of crossflow on bentonite fouling is presented in Figs. 11 and 12. The results for the flux vs time curves for 300 and 500 psi pressures and at 100 and 300 mg/l bentonite concentration show almost a similar pattern. This shows that for the tested conditions, crossflow velocities do not have much of an impact on the flux. In order to determine the flow regimen, Reynolds number was calculate and the values obtained were 85, and 270 for the velocities of 4.04 and 12.9 cm/s, respectively. The calculated Reynolds numbers were within the laminar flow regions for the rectangular channel of the used membrane cell. The shear stress generated within the above Reynolds number range may not be adequate enough to show the differences in bentonite particle removal mechanism from the membrane for the used module. This may be the reason for cross flow velocity for displaying a poor relationship to permeate flux.

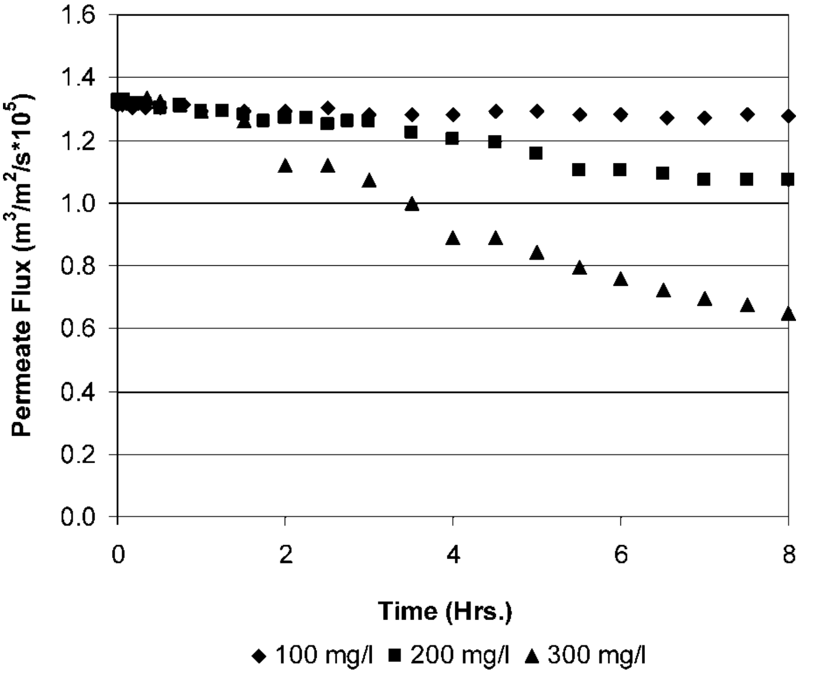

Figure 8. Flux vs time for Bentonite concentration, applied pressure(ΔP) = 200 psi and cross flow velocity (V) = 4.04 cm/s

Occurrence of Critical Flux

The critical flux is defined as the lowest flux that creates a fouling layer on the membrane surface (15). In our case, no such conditions were observed. However, the results of the 8 hour runs show that the membrane, if operated around the 200 psi region, displays very low fouling behavior.

Mass Deposited vs Flux Decline Relationship

Figure 13 gives the mass deposited vs flux decline relationship. The plot gives a reasonably good match ($R^2 = 0.8692$) between the two parameters. The result confirms that the main reason for the flux decline in the runs may be the formation of a cake layer on the membrane surface.

Statistical Model

Based on the experimental data, an empirical model was derived, using the general linear model function of the SPSS software (SPSS Inc., Chicago, Illinois), to predict flux decline at an end of 8 hour runs for the unit for a

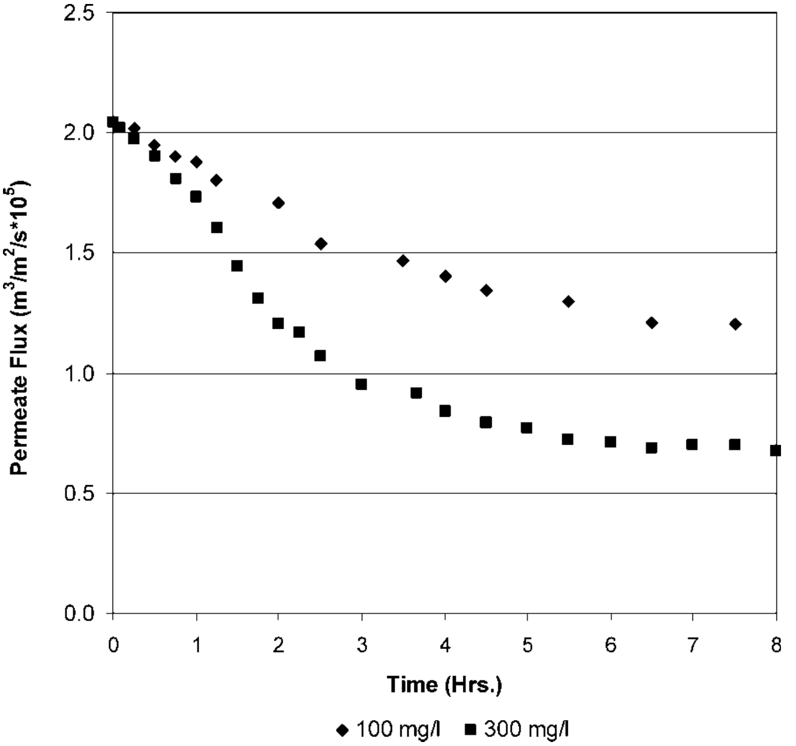

Figure 9. Flux vs time for Bentonite concentration, applied pressure (ΔP) = 300 psi and cross flow velocity (V) = 12.9 cm/s.

given transmembrane pressure, particle concentration and crossflow velocity. The expression for the flux decline (FD) was of the form.

$$
\begin{aligned}
FD = {} & 88.865 - 28.129\,LnP - 20.795\,LnC + 2.757(LnP)^2 - 0.085\,LnP * U \\
& + 0.785 * (LnC)^2 + 0.046 * U^2 - 0.025 * U * (LnC)^2 - 0.009 * LnP * U^2 \\
& + 0.044 LnP * LnC * U - 0.464(LnP)^2 * LnC + 4.988 LnP * LnC
\end{aligned}
$$

In applying the above equation, one must be quite aware of the units. The applicable units for Flux decline (FD), pressure (P), concentration (C) and cross flow velocity (U) are $m^3/m^2/s \times 10^5$, psi, mg/l and cm/s, respectively.

The relationship between the experimentally derived values and the model values provides a good fit with a $R^2 = 0.956$ and is given in Fig. 14. The model is based on the results of three level, three factor experimental

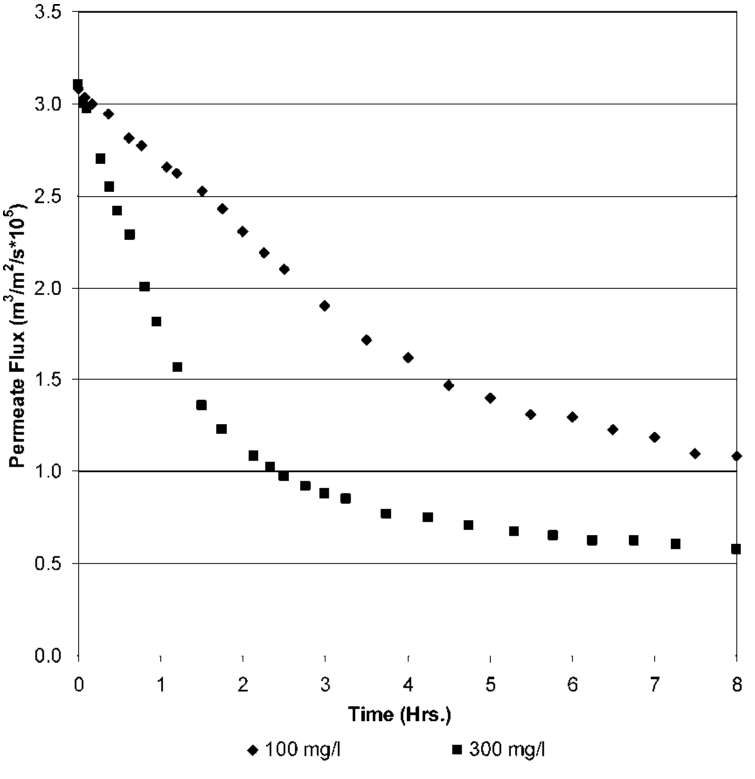

Figure 10. Flux vs time for Bentonite concentration, applied pressure (ΔP) = 500 psi and cross flow velocity (V) = 12.9 cm/s.

design which was carried out in the study and is calibrated only for the laboratory membrane unit that was used. However, at an industrial scale, a similar arrangement and model could be generated easily by selecting appropriate level and factors.

Table 1. Percentage flux decline with concentration

Pressure (psi)	Crossflow velocity (cm/s)	Bentonite concentration (mg/l) 100	300	Percentage increase in flux decline
200	4.04	3.1	50.8	47.7
	12.9	18.3	43.2	24.9
300	4.04	40.9	71.4	30.5
	12.9	41.7	67.2	25.5
500	4.04	68.1	84.0	15.9
	12.9	64.9	81.3	16.4

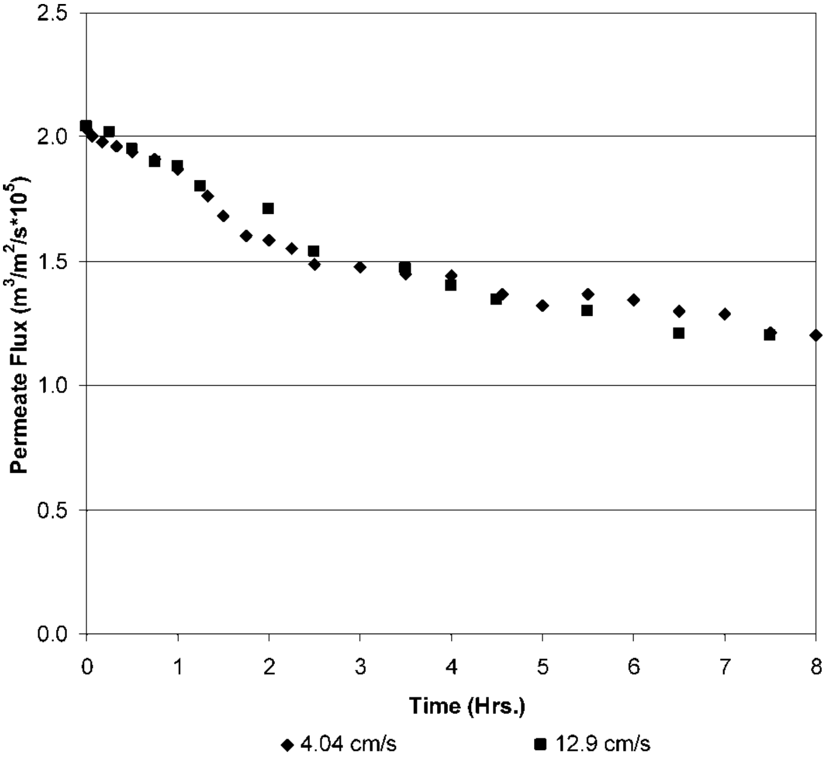

Figure 11. Flux vs time for cross flow velocity, applied pressure (ΔP) = 300 psi and Bentonite concentration (Co) = 100 mg/l.

CONCLUSIONS

Effect of operating parameters on permeate flux due to clay fouling on a LFC 1 membrane installed in a laboratory test cell is discussed. Bentonite was used as the model foulant. No steady state conditions were observed even after 24 hrs of operation. The flux–time curve could be generalized into three categories. Flux exhibits an initial decrease, which could be rapid exponential type or near linear followed by a slower almost linear decrease with time or almost linear until the run is curtailed. In the short term, the application of higher pressure leads to higher initial permeate flux. However, this effects has a negative effect on the long term flux by the formation of more dense and less porous cake layer on the membrane. Particles concentration plays a more dominant role in flux reduction. With the increase in particle concentration, the rate of particle transport towards the membrane increases and the overall rate of clay deposition on to the membrane increases resulting in higher resistance to water flow through the membrane thereby, decreasing the flux further.

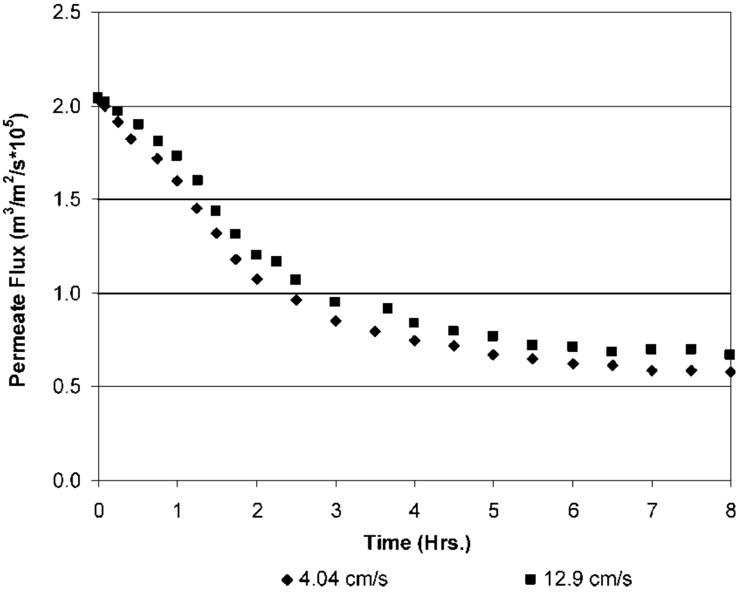

Figure 12. Flux vs time for cross flow velocity, applied pressure (ΔP) = 500 psi and Bentonite concentration (Co) = 300 mg/l.

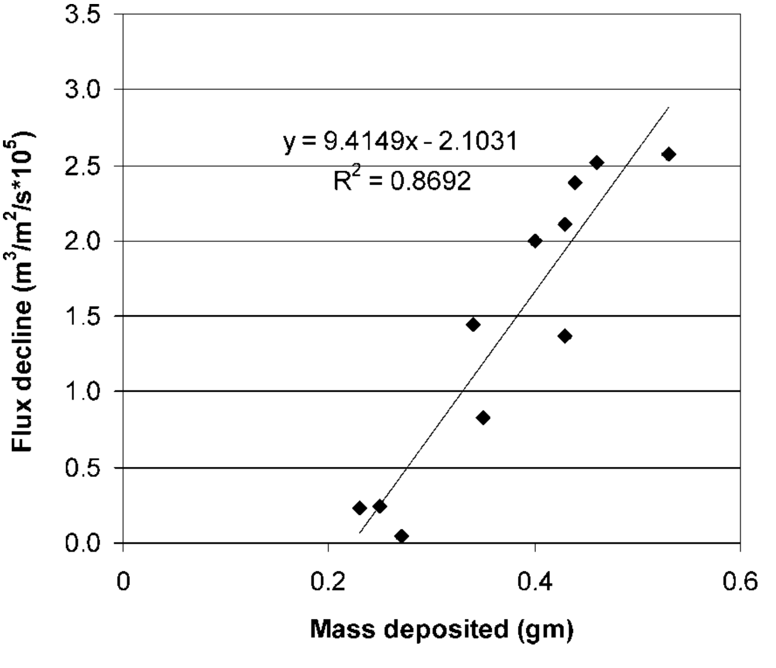

Figure 13. Flux decline vs mass deposited.

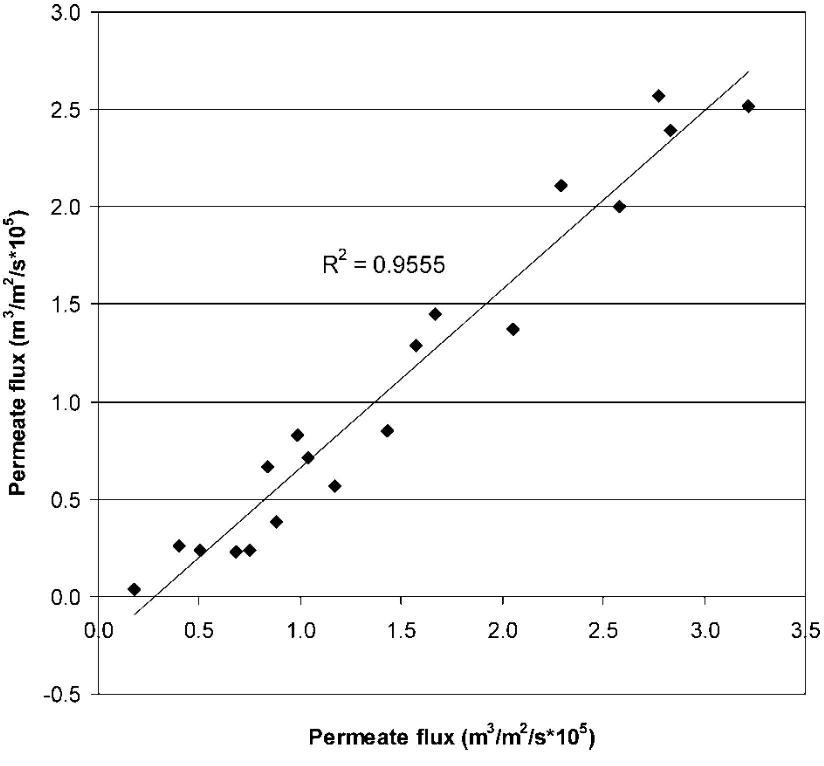

Figure 14. Model predicted vs experimental values.

The results show that an increase in particle concentration from 100 to 300 mg/l could lead to an increase in the flux decline between 15.9 to 47.7 percent. However, this effect tends to get somewhat reduced as the transmembrane pressure increases. For the tested conditions, crossflow velocity did not have much of an impact on the flux. The calculated Reynolds numbers 85 and 270 were within the laminar flow range for a rectangular channel and the shear stress generated within the Reynolds number range may not be adequate enough to show a marked difference in the bentonite particle removal for the used module. The poor relationship displayed by the crossflow velocity to permeate flux could be the result of the above.

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