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Journal of Membrane Science 191 (2001) 165–178

journal of MEMBRANE SCIENCE

www.elsevier.com/locate/memsci

Fouling behavior of a pilot scale inside-out hollow fiber UF membrane during dead-end filtration of tertiary wastewater

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Received 19 January 2001; received in revised form 17 April 2001; accepted 18 April 2001

Abstract

A series of pilot-scale filtration experiments were performed systematically under various operating conditions to investigate the fouling behavior of ultrafiltration (UF) membranes to treat tertiary wastewater for resuse. All experiments were conducted using a pilot system, which consisted of six inside-out capillary polyether sulfone UF membrane modules (molecular-weight cutoff $= 150{,}000$ Da), arranged in parallel configuration. The pilot unit was operated in dead-end filtration mode and the membranes were frequently backwashed with chlorinated water. Results of this research clearly indicated that the productivity of the UF membranes, measured by the specific water flux (K_w), declined much faster as operating flux increased. This observation was attributed to enhanced solid and organic loading to the membrane surface at higher operating fluxes. Furthermore, the analysis of K_w variation against filtrate volume showed larger productivity reduction per foulant mass loading during operation at high flux rates, suggesting the formation of more compact cake layers which were not easily removed during backwashing. Pilot study results also demonstrated that increasing backwashing with chlorine addition significantly improved membrane productivity, primarily due to enhanced foulant removal by organic oxidation and biogrowth control. In addition, flux enhancement per backwashing volume increased with decreasing time between backwashing events. Ferric chloride pretreatment also markedly enhanced membrane productivity by increasing particle floc size, which led to decreased pore plugging, reduced cake layer resistance, and enhanced backwashing efficiency.

Keywords: Ultrafiltration; Water treatment; Membrane fouling; Wastewater reclamation; Dead-end filtration

1. Introduction

The application of ultrafiltration (UF) and microfiltration (MF) membrane processes to treat secondary and/or tertiary treated wastewater for reuse purposes is gaining popularity in the United States and the world today [1–4]. This is a result of the continuing depletion of fresh drinking water supplies and the development of regulations and guidelines associated with reclaimed water production [5,6]. Membrane processes provide an effective means of meeting these demands because of their ability to remove solids as well as microbial contaminants including viruses by size exclusion [7,8]. In addition, this technology requires small footprints and minimal chemical addition, in comparison with conventional wastewater reclamation processes [9].

The performance of UF/MF membrane processes can be affected by operating conditions and feed water characteristics. Several key operational conditions that control membrane performance includes operating flux [10], backwash interval (BWI) [11], and

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0376-7388/01/$ – see front matter
PII: S0376-7388(01)00455-0

pretreatment processes, such as coagulation [12–14]. Furthermore, UF/MF productivity can be significantly deteriorated by membrane fouling, which results from the mass loading of solid, organic and microbial contaminants present in the feed water [15]. The optimization of such conditions is essential to achieving the productivity and filtrate water quality necessary for water reclamation.

The main intent of this study was to assess the effect of various operating conditions on the performance of capillary fiber UF membranes during the reclamation of tertiary treated wastewater. Specifically, a series of pilot experiments were conducted using a small pilot-scale UF system to investigate the effect of operating flux, BWI, and in-line coagulation pretreatment on membrane productivity. Accordingly, membrane productivity was assessed by monitoring the temperature corrected specific flux (K_w) with respect to filtration time. In addition, feed and permeate water quality of the UF system was evaluated by measuring concentrations of solid (turbidity and particle counts), organic (UV_{254} and TOC), and microbial (total coliform and heterotrophic plate counts, HPCs) parameters under varying operating conditions. Based on pilot experimental data, fundamental fouling mechanisms during dead-end UF of tertiary wastewater were delineated for future process optimization.

2. Experimental

2.1. Source water

The source water used in this research was tertiary treated wastewater obtained from the University of Central Florida's extended aeration wastewater treatment plant located in Orlando, Florida. This facility treats wastewater generated on campus using an activated sludge system and consists of the following unit operations: bar screening, aeration, microbial decomposition, and secondary sedimentation. In addition, the wastewater undergoes sand filtration and chlorination, prior to being discharged to a nearby percolation pond. Source water for the current study was obtained immediately after sand filtration (i.e. before chlorination). Table 1 presents feed water characteristics measured during the course of this study. As shown, low turbidity and particle counts were observed primarily due to the tertiary sand filtration process. Also, the TOC and UV_{254} values measured in the feed were fairly consistent and typical of tertiary treated wastewater effluent. Finally, the microbial content of this wastewater was quite significant and offered adequate feed concentrations to assess the microbial impact on membrane fouling during this study.

Table 1
Feed water quality analysis results

Parameter	Feed water (average ± S.D.)
pH	7.0 ± 0.42
Turbidity (NTU)	1.53 ± 2.41
Particle counts (counts per ml)	2.93 ± 1.75 E + 3
UV_{254} (cm^{-1})	0.22 ± 0.02
TOC ($mg\,l^{-1}$)	8.90 ± 0.46
HPC (counts per ml)	2.01 ± 3.31 E + 6
Total coliform (counts per 100 ml)	4.79 ± 8.65 E + 5

2.2. UF pilot system

A Mini-HYDRAcap^{TM} pilot unit, developed by Hydranautics (Oceanside, CA), was used to perform all experiments pertinent to this study. The unit consists of six identical 10.2 cm (4 in.) diameter by 51.5 cm (20.3 in.) length UF membrane modules arranged in parallel. This unique membrane configuration allows for simultaneous operation of the unit at six different filtrate flux rates. Furthermore, the ability to operate each membrane at the same filtrate rate makes it possible to verify the reproducibility of membrane performance under specific operating conditions. The pilot is also designed to perform dead-end or crossflow filtration and is capable of producing filtrate flows ranging from 0.045 to 0.45 $m^3\,h^{-1}$ (0.2–2.0 gpm) per membrane module. Lastly, the temperature corrected clean water specific flux of the HYDRAcap membranes is approximately 1.48–1.97 lmh kPa^{-1} (6.0–8.0 gfd psi^{-1}).

Each membrane module contains approximately 2000 capillary polyether sulfone (PES) fibers that provide a total membrane surface area of approximately 1.9 m^2 (20 ft^2). Furthermore the membrane modules are designed with an inside-out flow configuration; that is, feed water enters the center of the capillary tubes, filters through the wall, and is collected outside of the fibers. The capillary fibers have an inside diameter of approximately 0.8 mm (0.0031 in.) and an average molecular-weight cutoff (MWCO) of 150,000 Da. In addition, these membranes have several characteristics necessary for the treatment secondary/tertiary wastewater including the ability to withstand a wide pH range (2–13) and a high tolerance to free chlorine exposure (100 ppm). Such features allow for versatility in cleaning and backwashing procedures.

The pilot unit is also equipped with an automated membrane backwashing system and is capable of performing a semi-automated membrane integrity test. The backwashing system acts to remove foulants accumulated on the membrane surface during filtration. The system initiates a series of steps during which backwash water is pumped under pressure (207–241.5 kPa) through the filtrate side and across the inside of the capillary membranes. Furthermore, this system is designed to inject chlorine into the backwash water to promote chemical breakdown of foulants present on the membrane surface. The membrane integrity test system allows the operator to identify any breaches in integrity of the membrane fibers. Specifically, the test is a pressure-hold method in which air pressure (103.4–137.9 kPa) is applied to the inside of the membrane fibers. Final assessment of membrane integrity is completed by visual inspection of the filtrate flow meters. The presence of air bubbles in the flow meters indicates a degradation of membrane fibers.

2.3. Backwashing

Backwashing of the membrane modules was accomplished through an automatic sequence of steps controlled by a programmable logic controller, equipped on the pilot unit. The automated unit operations, which occurred during backwashing, included forward flush, bottom backwash, top backwash, soak cycle, and final rinse. Accordingly, backwashing was initiated by a forward flush of the modules in which feed water was driven across the inside of the capillary fibers to shear any foulants accumulated on the surface. Next, filtrate water was pumped to the filtrate side of the membrane modules by the backwash pump and forced to exit the bottom end of the membrane modules. This step was immediately followed by a similar step in which the backwash water was removed through the top end of the modules. It should be noted the pressure during backwashing was between 207 and 241.5 kPa (30–35 psi). In addition, during these steps, the chlorine pump was initiated and remained active until the soak cycle, at which time all pumps were shut off to promote chlorine degradation of contaminants present on the membrane surface. Lastly, the backwashing cycle was completed by a final rinse of the membranes. Accordingly, backwash water was pumped to the filtrate side of the membrane and exited to drain through the feed and concentrate lines. Table 2 presents experimental sequence times of the backwashing operations used in this study. As shown, each backwashing event lasted a total of 65 s, which included 24 s of NaOCl dosing at a rate of approximately 40 ml min^{-1}. This dosing rate provided a 25 ppm NaOCl concentration in the backwash water during bottom and top backwashing steps. The filtration time between backwashing events, defined as the BWI, ranged from 15 to 30 min.

Table 2
Backwashing sequencing times

Unit operation	Sequence time
Forward flush	9 s
Bottom backwash[a]	12 s
Top backwash[a]	12 s
Soak[a]	20 s
Final rinse	12 s
Total backwash	65 s
Backwash interval	Every 15–30 min

[a] These steps were conducted with filtrate water containing 25 ppm NaOCl.

2.4. Ferric chloride pretreatment

To study the effect of coagulant pretreatment on UF membrane performance, several experiments were conducted in which the feed water was dosed with ferric chloride prior to membrane filtration. The coagulant was continuously injected into the source water at the beginning of the 5.1 cm (2 in.) PVC pipeline leading from the feed storage tank to the feed pump. This allowed for a mixing length of approximately 4.6 m (15 ft) prior to UF membrane filtration. Experimental doses included 7 and 14 ppm as $FeCl_3$. Dosing was applied using an LMI Milton

Roy (Acton, MA) chemical metering pump with a maximum flow capacity of 45.4 l per day (12 gpd). The ferric chloride used in dosing was 40% by weight and was purchased from Archer Daniels Midland Co. (Decatur, IL) at the onset of the project.

2.5. Pilot operation

The general sequence of steps followed during pilot operation included membrane cleaning, clean water flux analysis, membrane integrity testing, and filtration experiments. Prior to each experiment, the membranes were thoroughly cleaned using both acidic and basic solutions. Specifically, the membranes were cleaned by filtering and recycling a citric acid solution (pH 2–2.5) for 60 min in the crossflow mode. This process was then repeated using a sodium hydroxide solution of pH 12–12.5.

To assess whether a given cleaning procedure adequately restored the specific flux capacity of the membranes, a clean water flux profile was developed for each individual membrane module. The profiles were generated by measuring the transmembrane pressure (TMP) of each membrane module during dead-end filtration of potable water at filtrate flow rates ranging from 0.09 to 0.27 $m^3 h^{-1}$ (0.4–1.2 gpm). This information was then used to determine the K_w associated with each clean membrane module. These values were compared to the specific flux values determined at the onset of the project (i.e. new membranes) to determine the percent recovery of the initial productivity. If the recovery was less than 75%, the cleaning procedure was repeated.

Next, each membrane was tested for integrity using the semi-automated pressure hold test equipped on the unit. The test was initiated by introducing approximately 137.9 kPa (20 psi) of air pressure to the feed side of each membrane. A damaged membrane would allow the air to escape through the membrane and therefore can be detected by the presence of air bubbles in the filtrate valve.

Following integrity testing, membrane filtration experiments were performed in dead-end filtration mode under various operating conditions. A total of fifteen pilot experiments were performed during the course of the study. During each experiment, the source water was gravity-fed to the pilot unit from the nearby wastewater treatment facility. The feed water was collected in a 208 l (55 gal) storage tank equipped with a float valve to prevent overflow. Feed water was then pumped from the storage tank and passed through a 300 μm pre-filter, prior to entering the bottom of the membrane modules. The operating flux was set between 34 and 102 $l\,m^{-2}\,h$ (lmh) for each membrane module by adjusting the feed pressure and filtrate flow valves equipped on the unit. In addition, ferric chloride (0–14 ppm) was injected continuously at a point located just after the feed storage tank. Throughout the course of a given experiment the membrane modules were frequently backwashed (15–30 min) by chlorinated water as describe above. The filtrate produced was stored in 322 l (85 gal) storage tank and partially used during backwashing. The duration of each experiment ranged from 60 to 240 h depending on operating conditions and source water supply.

2.6. Membrane productivity assessment

The productivity of each membrane module was monitored under various experimental conditions by evaluating the decline of the specific flux (K_w) with respect to operation time. The K_w of each membrane was evaluated three times a day as presented in the following equation.

$$K_w = \frac{F_{w20}}{\Delta P} \tag{1}$$

where K_w is the specific flux of the membrane, ΔP the pressure gradient across the membrane, and F_{w20} the water flux normalized to 20°C. The pressure gradient, often described as TMP, was determined by the average pressure difference across the membrane, that is

$$\Delta P = \tfrac{1}{2}(P_f + P_c) - P_p \tag{2}$$

where P_f, P_c, and P_p are feed, concentrate, and filtrate stream pressures, respectively. Because water flux across a membrane changes with temperature, the flux was normalized at a temperature of 20°C for direct comparison as follows [8].

$$F_{w20} = \frac{F_w}{2.72^{0.019(T-20^\circ C)}} \tag{3}$$

where F_w is water flux and T temperature (°C). It should be noted the temperature correction factor of 0.019 presented in Eq. (3) was used throughout the

Table 3
Water quality analysis methods

Category	Parameters	Methods
General	pH	Electrometry (SM[a] 4500-H^+ B)
Solid	Turbidity	Nephelometry (SM 2130 B)
	Particle counting	Electrical sensing (SM 2560 B)
Organic	UV_{254}	Ultraviolet absorption (SM 5910 B)
	NPDOC	Sodium persulfate/UV oxidation (SM 5310 C)
Microbial	Total coliform	Membrane filtration (SM 9222 B)
	Heterotrophic plate counts	Pour plate (SM 9215 B)

[a] SM: standard methods for the examination of water and wastewater [16].

course of the project to correct K_w for feed water temperature variations. This value was provided by Hydranautics and verified during the course of this work using pilot operational data. The values used to determine K_w in this calculation (Eq. (1)) were obtained by reading and logging the run hour, feed water temperature, feed pressure, concentrate pressure, filtrate pressure, and filtrate flows from the gages and meters present on the pilot unit. This data was collected 5 min after a given backwashing event to ensure stable readings. Because the unit was operated with a constant feed pressure, it was often necessary to readjust filtrate flow meters to their experimental values before recording the feed and filtrate pressures.

2.7. Water quality analysis

A complete water quality analysis was also conducted during each filtration experiment to investigate the performance of the UF membranes at retaining solid, organic and microbial foulants present in the feed water. Two water quality parameters were measured to assess each foulant group as follows: turbidity and particle counts for solids, UV_{254} and TOC for organics, and total coliform and HPC for microorganisms. Each of the water quality parameters were measured in accordance to specific methods outlined in the “standard methods for the examination of water and wastewater” [16] as identified in Table 3. The measurement frequency of the above water quality parameters was established for a 10-day continuous filtration period. During this time, pH, turbidity and particle counts were measured daily; UV_{254}, and TOC were measured on days 1, 5 and 10; and total coliforms and HPC were measured on days 1 and 10.

All samples, with the exception of those used to assess microbial parameters, were collected in 500 ml amber bottles. Sampling bottles were rinsed with acid (1:1 HCl) and deionized (DI) water before each sampling event. To prevent contamination of foulants removed from backwashing, all samples were collected in a timely manner after 5 min of filtration time following a backwash event. In addition, each sampling port valve was opened for 30 s prior to sampling to flush any contaminates present in the sampling lines. Upon collection, samples were stored in a cooler and immediately transported to a nearby location for water quality analysis. Microbial samples were collected in 50 ml sealed plastic containers in accordance to US EPA sampling procedures for this particular container type.

3. Results and discussion

3.1. Feed and filtrate water quality

Fouling of size exclusion membranes such as UF and MF is attributed to the mass loading of solid, organic and microbial contaminant present in the feed water to the membrane surface. Fig. 1(a) presents average feed and average filtrate values of turbidity during experiments conducted at operating fluxes ranging from 34 to 102 lmh (20–60 gfd). As shown, the average filtrate turbidity concentrations measured during operation at the above experimental flux rates were all below 0.25 NTU. Similarly, average filtrate turbidity

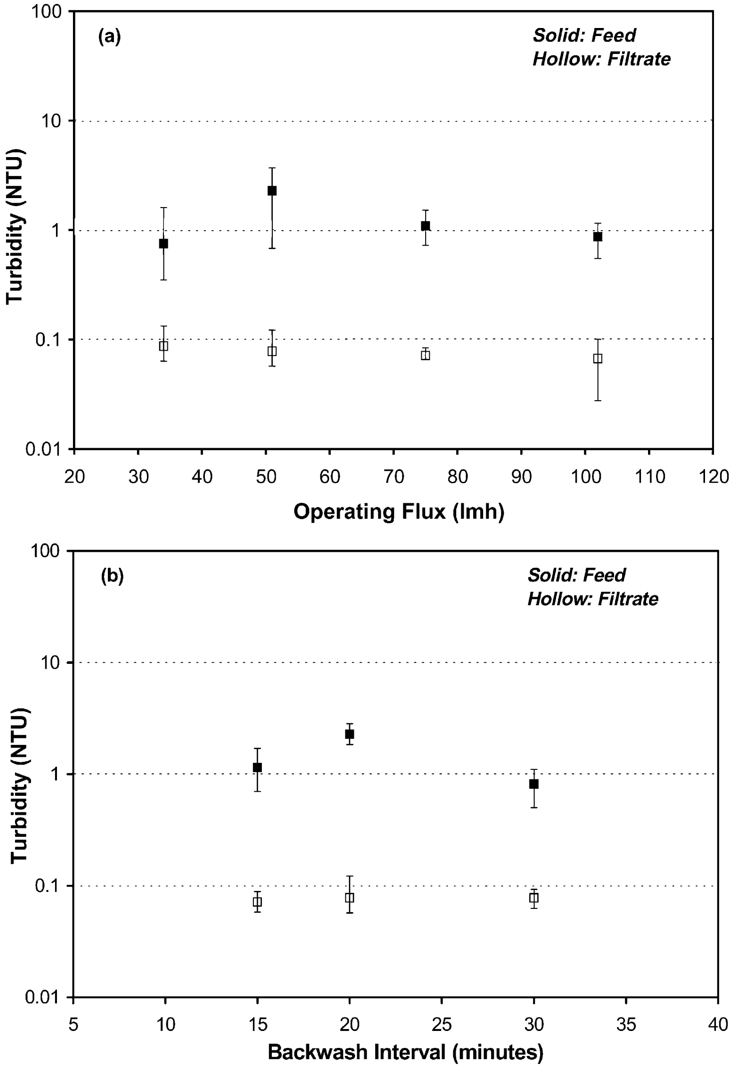

Fig. 1. Effect of operating conditions on solid retention. (a) Feed and filtrate turbidity vs. operating flux and (b) feed and filtrate turbidity vs. BWI. BWI was set to 20 min for the experiment (a), while the operating flux was maintained at 51 lmh (30 gfd) during the experiment (b). Filtrate turbidities are average values of the six UF membranes tested.

values measured during experiments conducted at BWIs ranging from 15 to 30 min were also below 0.1 NTU as shown in Fig. 1(b). These observations indicated that UF membrane was very effective at rejecting turbidity, independent of operating flux and backwashing interval. Consequently, the retained particles would impose significant fouling potential on the system.

In addition to particle loading, the importance of organic matter in UF/MF membrane fouling has also been pointed out by several investigators [18–20]. The average measurements of UV_{254} and TOC in the feed and filtrate water during the course of this project indicated low rejection of organic matter (i.e. UV_{254} rejection $\approx$ 4.1% and TOC rejection $\approx$ 5.6%), independent of operating conditions. Although significant portions of organic matter passed through the UF membranes during this study, organic substances seem to play an important role in membrane fouling during wastewater reclamation. The presence of organic constituents in source water can cause significant productivity loss by plugging membrane pores, adsorbing to the internal matrix of the membrane, and forming a cohesive gel on the cake layer [8]. In addition, the organic matter can also enhance biological activities in the membrane system, which may increase biofouling.

Lastly, microbial constituents present in the feed water can cause severe reduction of membrane productivity by forming a biofilm layer on the membrane surface. Based on the feed microbial concentrations measured during the present study, the log removal value (LRV) of total coliform ranged from 1.2 to 7.0 with 33% of the filtrate samples containing counts less than the limit of the assay (0.1 CFU/100 ml), indicating infinite rejection. Similar results were shown by Hong et al. [21], whom reported LRVs of total coliform ranging from 5.4 to 8.9 during MF pilot studies. The authors also illustrated that LRV can be limited by the feed water concentration, which may explain the slightly lower LRVs of total coliform observed during this study.

3.2. Operating flux

The effect of operating flux on UF membrane productivity was systematically investigated by conducting experiments at 34, 51, 75 and 102 lmh. Each experiment was performed in dead-end filtration with a BWI of 20 min. The results are presented in Fig. 2(a) as a function of operating time. As indicated the specific flux values (K_w) used in this analysis were based on average values obtained from four of six membranes tested after excluding the two most deviant. In order to account for variations in initial specific flux (K_{w0}), the K_w values were normalized with respect to the K_{w0} determined by linear regression. In a pilot- or full-scale operation, the productivity declines at slower rate compared to typical bench-scale experiments, and is often observed to drop linearly with respect to filtration time [12,15]. As shown, the declining slopes clearly indicate that K_w decreased with time for each operating flux tested over a given time period and set of experimental conditions. In addition, the magnitude of the decline rates steadily increased as operating flux increased; suggesting that the operation at higher fluxes caused a significant increase in membrane fouling.

The sharp decrease in productivity observed during operation at high operating flux rates is primarily attributed to the increase of foulant mass loading, which can result in irreversible fouling [15]. As presented in Eq. (4), the cumulative foulant loading of a given contaminant to the membrane during dead-end filtration can expressed as a function of the operating flux (F_w), filtration time (t), and the bulk concentration of the respective contaminate (C_b).

$$\text{Mass loading per unit membrane surface area} = F_w C_b t \tag{4}$$

The loading of each foulant present in the feed water can be determined by using various water aggregate quality parameters: for example, turbidity or particle counts for solid loading. Therefore, when assessing the influence of operating flux on membrane productivity, it is imperative to examine the feed water quality. In particular, several investigators have reported that particle feed water characteristics played a significant role in productivity decline during UF/MF pilot studies [1,17].

To further study the significance of operating flux on productivity decline, K_w/K_{w0} was also plotted against total filtrate volume, as presented in Fig. 2(b). Assuming steady feed water quality, identical or similar slopes were expected for all of experimental runs in this analysis. However, as shown in Fig. 2(b), the decline in normalized specific flux associated with the production of a given filtrate volume slightly increased with increasing operating flux. This finding is attributed to the structural characteristics of the fouling layers formed during operation at high flux rates. Specifically, the cake layers may become more compressed as a result of the stronger drag force induced by the high filtrate flows. This can cause the fouling layer to be more compact and lead to larger hydraulic resistance (per a given particle loading) to filtrate flow. In addition, it is expected the densely packed nature of the foulants causes removal by backwashing to be more difficult.

The effect of operating flux on productivity decline was further analyzed by calculating the runtime associated with each of the studied flux rates

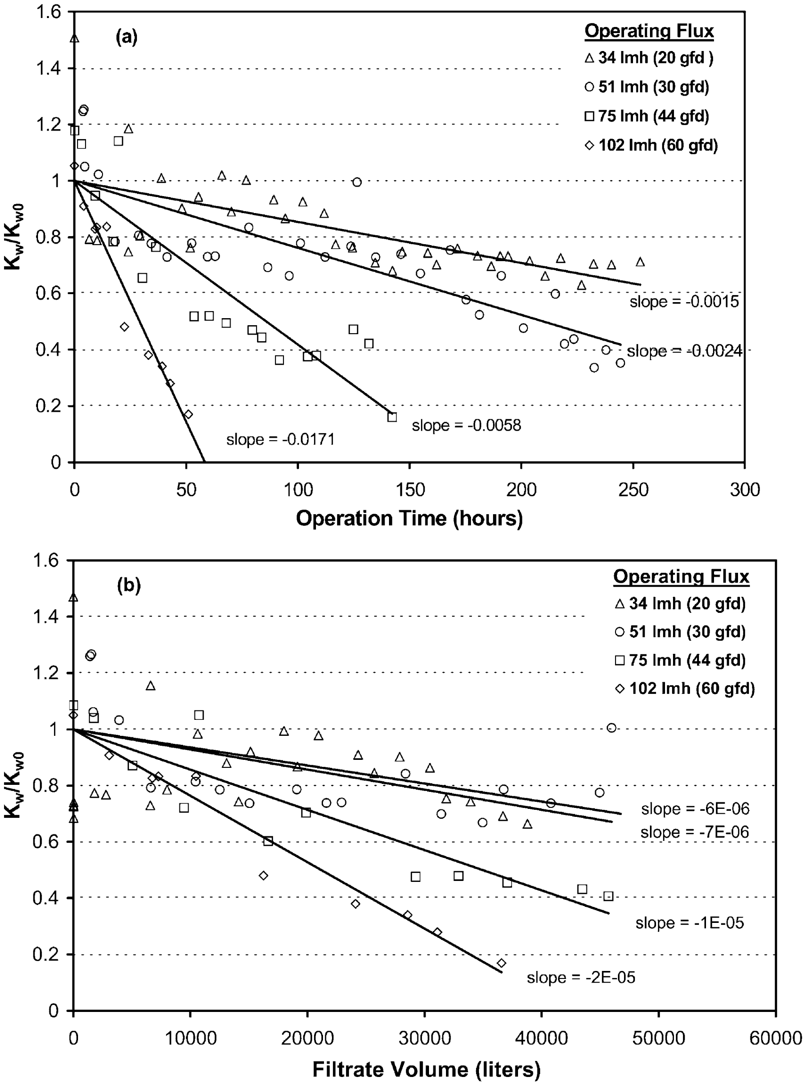

Fig. 2. Effect of operating flux on UF membrane productivity. (a) Normalized K_w (K_w/K_{w0}) vs. operation time and (b) normalized K_w (K_w/K_{w0}) vs. filtrate volume. BWI was set to 20 min. Filtrate volume shown (b) represents the total filtrate produced by four membrane modules containing a combined membrane area of 7.4 m^2 (4 m × 1.85 m). Regression lines are based on linear least squares fit and are presented as solid lines.

(34–102 lmh). As indicated in Fig. 3, the runtime is defined as the filtration time between cleaning events and represents the time at which the maximum operation TMP of the membrane is achieved. Accordingly, the runtimes shown were predicted using the linear normalized specific flux decline rates associated with each value of flux as presented in Eq. (5). As shown, the runtime (days) was calculated by subtracting the initial (K_{wi}) and final (K_{wf}) normalized values of K_w and dividing by the corresponding normalized specific flux decline rate. The K_{wf} value was determined by dividing the operating flux by the maximum TMP (120.7 kPa) specific to the membranes investigated. In addition, K_{wi} and the normalized specific flux decline rate, $\Delta(K_w/K_{wi})/\Delta t$, were obtained from the linear regression. As expected, the achievable runtime

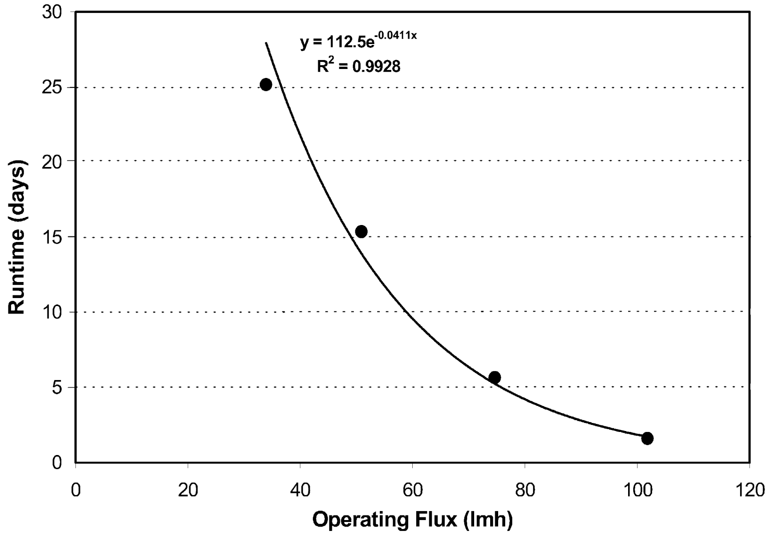

Fig. 3. Runtime vs. operating flux. BWI was set to 20 min. Runtimes shown represent the filtration time before reaching a TMP of 120.7 kPa (17.5 psi). Runtimes were calculated using the normalized K_w decline rates determined from regression analysis.

decreased steadily as flux increased as follows: 25.1 days at 34 lmh (20 gfd); 15.4 days at 51 lmh (30 gfd); 5.7 days at 75 lmh (44 gfd) and 1.6 days at 102 lmh (60 gfd). Such results are indicative of the increased fouling occurring at high operating flux rates, which as described above, is attributed to an increase in mass loading and alteration of the cake layer characteristics.

$$\text{Runtime} = \frac{(K_{wf}/K_{wi}) - (K_{wi}/K_{wi})}{\Delta(K_w/K_{wi})/\Delta t} \times \text{CF} \quad (5)$$

where K_{wf} is final value of specific flux (lmh kPa^{-1}), K_{wi} is initial value of specific flux (lmh kPa^{-1}), $\Delta(K_w/K_{wi})/\Delta t$ is normalized specific flux decline rate (h^{-1}), and CF is conversion factor (day h^{-1}).

3.3. Backwash interval

As previously stated, the BWI is one of the primary factors affecting UF/MF membrane performance. Particularly, in dead-end filtration, backwashing by air and/or water is the only means to remove foulants from the system. The BWI was defined in this study to be the continuous filtration time between backwashing events. Accordingly, operating with a 30 min BWI would result in two backwashing events per hour. The effect of BWI on UF productivity was studied by conducting experiments at various BWIs including 15, 20, and 30 min. Each experiment was conducted at a flux of 51 lmh (30 gfd) in dead-end filtration mode. A comparison of the normalized temperature corrected values of specific flux (K_w/K_{w0}) decline as function of operation time is provided in Fig. 4. The trend lines, developed from linear regression, clearly indicated that K_w declined much faster as the BWI increased. This observation suggested that decreasing the time in between backwashing significantly improved UF membrane productivity. The elimination of UF fouling observed with operation at the 15 min BWI is consistent with a previous study [15] in which the author reported significant increase in runtime for backwash frequencies of 15 min or less.

The effect of BWI on the productivity was further analyzed by comparing the decrease of specific flux to cumulative backwash volume. This comparison revealed that after backwashing with approximately 11,000 l of filtrate during operation at BWI of 15, 20 and 30 min, the respective initial normalized specific flux values dropped by 63, 75 and 88%, respectively. Such results clearly indicated that the flux enhancement per unit backwashing water volume decreased with increasing BWI. This result demonstrated that the efficiency of backwashing was dependent on the BWI. It is expected that increasing time between

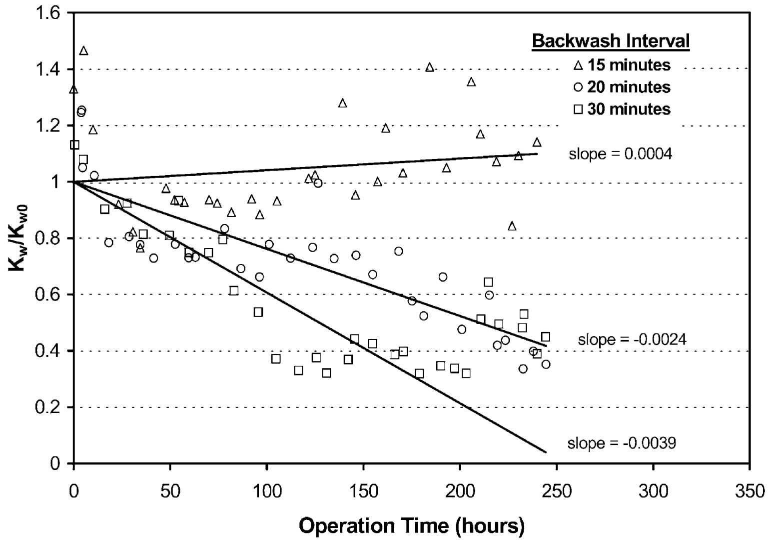

Fig. 4. Effect of BWI on UF membrane productivity: normalized K_w (K_w/K_{w0}) vs. operation time. Operating flux was set to 51 lmh (30 gfd). Backwashing was performed for 65 s, which included 24 s using backwash water with 25 ppm NaOCl. Regression lines are based on linear least squares fit and are presented as solid lines.

backwashing events allows the formation of a thicker fouling layer that is not easily removed, and thus leads to irreversible fouling. In addition, during extended times between backwashing with chlorine, organic matter can build up in the pores and on the surface of the membrane leading to irreversible fouling.

The enhancement of productivity can be explained by several flux restoration mechanisms associated with backwashing. These mechanisms include convection, oxidation, and biofouling control [22]. Solid foulants, which block the membrane pores and/or accumulate on the surface of the membrane, are removed by shear, which results from the reverse flow of filtrate water through membrane. Furthermore, chlorine present in the backwash water can oxidize or destroy organic matter adsorbed or accumulated on the membrane surface. In addition, chlorine is capable of repressing microbial growth and thus prevents the formation of biofilm on the membrane surface. The degree to which backwashing enhances membrane productivity is dependent on both the feed water characteristics and the frequency at which it is performed.

The importance of chemical-enhanced backwashing has been well documented in previous studies involving UF/MF treatment of secondary/tertiary wastewater and low quality surface waters. Laine et al. [10] reported the termination of chlorine injection in backwash water after 20 days of runtime resulted in severe fouling of the membranes within 5 days. Van Houtte et al. [14] reported it was necessary to backwash every 5–20 min, depending on feed water quality, during MF filtration of secondary treated wastewater. Van der Graff et al. [2] reported that backwashing every 15 min with the addition of sodium hypochlorite every 120 min resulted in steady and dependable UF pilot operation during wastewater filtration experiments. Hofman et al. [11] determined the optimal backwashing regime to consist of backwashing every 20 min for duration of 30 s with chemical enhanced backflushing and soaking every 3 h during UF filtration of surface water. The authors also indicated it was necessary to decrease the time between backwashing and operate at lower flux rates during periods of poor feed water quality to prevent irreversible fouling.

3.4. Ferric chloride pretreatment

The introduction of a coagulant to the feed water prior to membrane filtration is a common practice to control fouling [8]. However, direct addition of coagulants to the membrane system (i.e. in-line coagulation) has not been explored thoroughly in the

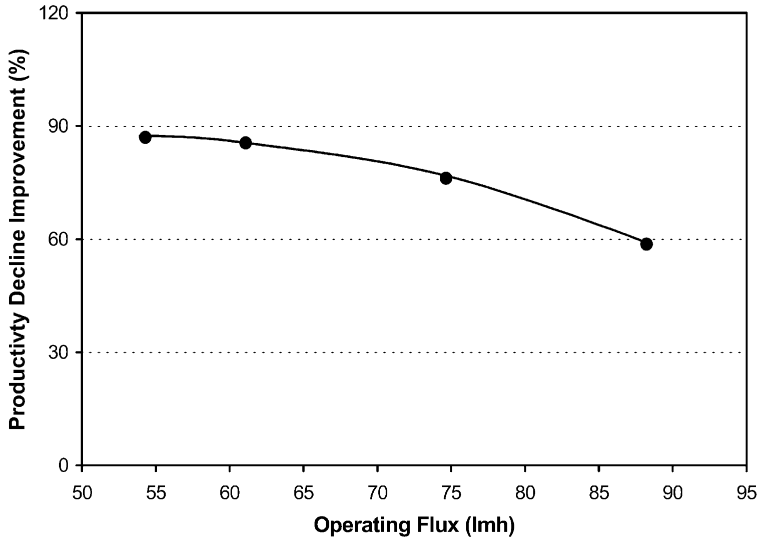

Fig. 5. Productivity decline improvement vs. operating flux. Productivity decline improvement was determined by comparing the normalized productivity decline rates measured during experiments conducted using 0 and 7 ppm ferric chloride pretreatment. BWI was set to 20 min for each experiment.

literature. Accordingly, the effect of in-line coagulation on UF productivity was studied by conducting experiments with 0 and 7 ppm ferric chloride pretreatment. Fig. 5 presents the productivity decline improvement realized by the addition of 7 ppm ferric chloride for operating flux rates ranging from 54 to 88 lmh (32–52 gfd). The productivity decline improvement was determined by calculating the difference between the normalized productivity decline rates measured during operation at 0 and 7 ppm ferric chloride pretreatment. Fig. 5 clearly demonstrated that the addition of 7 ppm ferric chloride prior to UF filtration significantly improved productivity decline for each operating flux investigated. Further examination of the data also indicated a clear trend of decreasing improvement with increasing operating flux; that is, 87% at 54 lmh (32 gfd), 85% at 61 lmh (36 gfd), 76% at 75 lmh (44 gfd), and 59% at 88 lmh (52 gfd). This trend is consistent with the results presented above in which the decline in productivity at higher operating flux rates was attributed to alteration of the cake layer due to increased drag force.

As a result of coagulation by the addition of ferric chloride, average feed turbidity and particle counts increased from approximately 1.1 to 4.7 NTU and from 4500 to 6500 counts per ml, respectively. An interesting observation was that the increase in particulate matter resulting from the addition of ferric chloride did not result in membrane fouling. To further investigate the effect of particle loading on productivity, K_w/K_{w0} was plotted against cumulative mass loading based on particle counts (>2 μm), as presented in Fig. 6. The plot clearly indicated that pretreatment by ferric chloride (14 ppm) caused much less hydraulic resistance to filtrate flow per given particle mass loading. This observation indicates that ferric chloride pretreatment was successful at destabilizing colloidal particles ($\leq$1 μm) present in the feed water which resulted in the formation of large particles that were readily retained during UF filtration. The relationship of particle size to UF membrane performance during the filtration of treated wastewater was also observed by Tchobanoglous et al. [1] who reported a disproportionate impact on performance from feed water containing different particle sizes.

The mechanisms responsible for enhanced membrane productivity resulting from coagulation pretreatment include reduction in the amount of foulants entering the pores, conditioning of the cake layer, and an increase in back transport [8]. As a result of charge neutralization and possibly sweep flocculation by the precipitated metal hydroxide complexes,

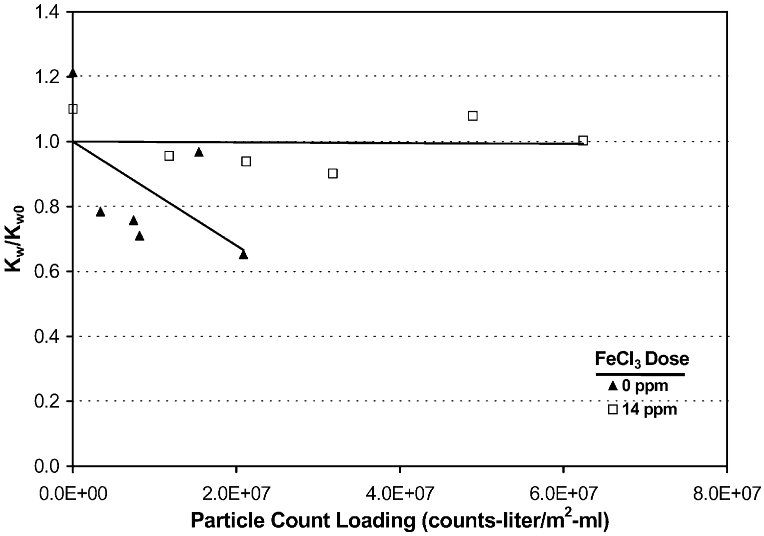

Fig. 6. Normalized K_w (K_w/K_{w0}) vs. particle count loading. Particle counts measured during experiments conducted with 0 and 14 ppm ferric chloride pretreatment. Experimental conditions: flux = 51 lmh (30 gfd) and backwash interval = 20 min. Particle mass loading is estimated by particle counts (counts per ml) × filtrate volume per unit membrane surface area ($\mathrm{l\,m^{-2}}$). Regression lines are based on linear least squares fit and are presented as solid lines.

colloidal particles present in the feed water form large aggregates and are prevented from entering the membrane pores. Furthermore, the specific resistance of the cake layer would be reduced with larger particles allowing more flow to pass through the membrane. Lastly, as particle size increases due to aggregation, the effect of inertial lift and shear force become more pronounced and more particles are back transported from the membrane surface minimizing concentration polarization. However, it should be noted that this mechanism is not applicable to our study because experiments presented in Fig. 5 were performed at a dead-end filtration mode and there was no crossflow causing inertial lift and shear.

In addition to particle aggregation, ferric chloride pretreatment could increase organic foulant removal. However, the water quality analysis performed in this study did not indicate any significant enhancement of organic matter rejection by the experimental ferric dose concentrations (i.e. 7–14 ppm). It is expected that a much higher dose would be required to reduce organics in this source water, probably due to its low SUVA value (i.e. $UV_{254}/TOC \approx 2.5\,\mathrm{m^{-1}\,l\,mg^{-1}}$), which indicates the feed water had a low hydrophobic fraction.

Several recent studies [13,14,17,23,24] have also shown the addition of a coagulant prior to UF membrane filtration resulted in stable membrane productivity. Lahoussine-Turcaud et al. [23] demonstrated that coagulant addition during UF filtration of river water shifted the membrane fouling from chemical reversible to hydrodynamic reversible. Taylor and Kothari [17] reported that coagulation with ferric sulfate greatly reduced fouling during MF/UF filtration but that the choice of coagulant was membrane specific. Braghetta et al. [24] also observed an increase of 25–50% in operating specific flux, when filtering water that had undergone coagulant pretreatment.

4. Conclusions

Primary inferences from this research are summarized as follows.

- Membrane fouling increased significantly as operating flux increased. The increased fouling rate at high operating fluxes was primarily attributed to an increase in mass loading of solid, organic and microbial contaminants. In addition, a comparison of K_w decline versus filtrate volume for various values of operating flux suggests that enhanced hydraulic resistance of the fouling layer induced by filtrate flow also contributed to increased fouling observed at high operating flux. As a result of increased fouling, the runtime associated with the maximum tolerable TMP of the membranes, decreased sharply with increasing operating flux.

- The K_w decline rate increased with increasing BWI. This increase in productivity decline can be explained by the occurrence of irreversible fouling resulting from the longer filtration time between backwashing events. It was also demonstrated that flux enhancement per unit chlorinated backwashing water volume decreased with increasing BWI, indicating that the effectiveness of backwashing was a function of BWI. This observation may be explained by the formation of a thicker fouling layer, less organic removal by oxidation, and enhanced microbial growth associated with an increased time interval between backwash events.
- The continuous in-line addition of ferric chloride prior to UF filtration significantly reduced the rate of K_w decline for experimental flux values ranging from 54 to 88 lmh (32–52 gfd). However, the improvement in productivity decline was observed to decrease as the operating flux increased. This observation is consistent with the results from the assessment of flux on productivity in which it was shown higher flux rates caused greater membrane fouling.

Acknowledgements

This research was conducted as a project of the US Bureau of Reclamation. The UF pilot unit was manufactured and provided by Hydranautics (Oceanside, CA). Dr. Mark Wilf and Steve Alt of Hydranautics provided technical support throughout the course of the project. Microbial analysis was performed in the Department of Molecular Biology and Microbiology at the University of Central Florida (UCF) by Claudia Romero under the direction of Dr. Saleh A. Naser. The authors would also like to recognize Dr. Charles Norris and Maria Pia Real of the Environmental Systems Engineering Institute (UCF) for their assistance in the TOC analysis. Lastly, the authors would like to thank Larry Eflin (plant operator), and the entire staff of the UCF's wastewater treatment plant for their cooperation and support during this project.

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