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Effect of a hydrodynamic cleaning of a cross-flow membrane system with a novel automated approach

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Abstract

The effect of different hydraulic cleaning methods was investigated in terms of flux decline and resistance using the cross-flow UF unit. The use of varied production intervals, varied ratios of periodic relaxation and the use of a periodic high-rate crossflow were investigated.

This cleaning method of relaxation followed by high-rate crossflow mainly removed the resistances of the concentration polarization and gel layer compared with weak adsorption resistance. The cleaning protocol utilizing a periodic relaxation step and/or a periodic increased cross-flow rate at a decreased pressure can lead to productivity improvements and an increase in the operational lifetime of the membrane. If the optimal frequency and duration of the cleaning step (1 h production–60 s relaxation–60 s crossflow) is used, a net productivity increase of 14.8% is achievable and a significant extension to the membrane's life results. However, a critical point of different cleaning intervals did exist. Utilizing the optimized periodic cleaning techniques developed in this study allows higher recovery rates for ultrafiltration to be achieved, without the problems of increased flux decline normally experienced when operating with high recovery rates.

Keywords: Hydraulic cleaning; Relaxation; Crossflow; Pretreatment; Ultrafiltration; Automation

1. Introduction

As a consequence of increasingly stringent standards for wastewater disposal and reuse, various new treatment technologies have emerged. Dependent upon the required effluent quality, membrane processes such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) or reverse osmosis (RO) are used. However, problems associated with continued fouling during filtration operations prevent the wide application of membrane technology.

Pretreatment has been found to effectively reduce membrane fouling, however, this produces

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additional sludge due to the addition of a coagulant or adsorbent [1,2]. Alternatively, membrane fouling can be reduced by hydrodynamic cleaning, such as higher cross-flow and long-term relaxation modes.

Operating the system at higher rates of crossflow and/or at reduced transmembrane pressures (TMP) has been found to reduce membrane fouling [3]. Increased shear created through crossflow removes the foulant layer on the membrane surface. However, these techniques result in a decreased permeate flux leading to the requirement for larger membrane systems to treat any quantity of water or wastewater and this increases the capital costs since the membrane area required will be larger.

Resistance to the cross-flow membrane system caused by concentration polarization and gel layer formation are considered to be reversible by applying clean water, while strong adsorption resistance is irreversible [4]. In a study by Cho et al. [5], flux was partially restored through the introduction of distillate water to the system followed by a period of relaxation. During this process, the absence of applied pressure to the membrane successfully reversed the resistance attributed to concentration polarization. The study also involved the increased rate of crossflow of distillate water crossflow and this was found to successfully reverse the resistance attributed to the gel layer formation.

While the results of these techniques are promising, the usage of distillate water and the single implementation of these cleaning techniques during the final stage of the filtration process minimize their potential usage in practical applications. In this study, periodic relaxation and a periodic high rate of crossflow were trialed both independently and in a combination. Effect of pretreatment associated with optimum relaxation and high-rate crossflow was also investigated.

2. Experimental

2.1. Synthetic wastewater

This study was carried out with synthetic wastewater. The composition of the synthetic wastewater is presented in Table 1. This synthetic wastewater represents effluent organic matter (EfOM) generally found in the biologically treated sewage effluent (BTSE) [6].

Table 1
Constituents of the synthetic wastewater

Compound	Concentration (mg/L)	Main molecular weight (daltons)	Fraction by DOC
Beef extract	1.8	298, 145, 65	0.065
Peptone	2.7	34,265, 128, 80	0.138
Humic acid	4.2	1543, 298	0.082
Tannic acid	4.2	6343	0.237
Sodium lignin sulfonate	2.4	12,120	0.067
Sodium lauryle sulphate	0.94	34,265	0.042
Arabic gum powder	4.7	925, 256	0.213
Arabic acid (polysaccharide)	5.0	38,935	0.156
$(NH_4)_2SO_4$	7.1		0
K_2HPO_4	7.0		0
NH_4HCO_3	19.8		0
$MgSO_4 \cdot 7H_2O$	0.71		0

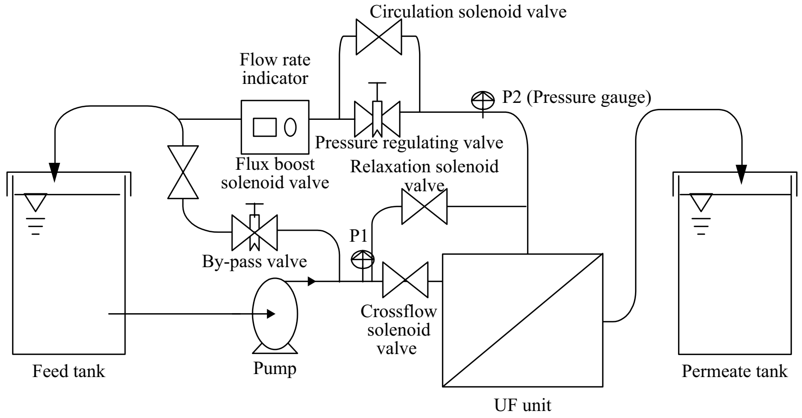

Fig. 1. Experimental set-up of the cross-flow membrane system with the inclusion of four automated solenoid valves for control of the operating modes.

Tannic acid, peptone, sodium lignin sulfonate, sodium lauryle sulfate and arabic acid represent larger molecular weight (MW) portion, while peptone, beef extract and humic acid consist of organic matters of smaller MW.

2.2. Membrane unit

Fig. 1 presents the experimental schematic of the cross-flow UF unit (Nitto Denko, Corp.) used in the study. The synthetic wastewater was pumped to the membrane module (effective membrane area 0.006 m^2; flat sheet configuration). The membrane used in this study was the NTR 7410 (Nitto Denko Corp., Japan) (Table 2).

This system allows an investigation of the effect of periodic relaxation, a periodic high-rate crossflow and a combination of both on membrane fouling and flux decline. Table 3 shows the status of the solenoid valves during each mode of operation. The automatic operations shown in Table 3 are selected through the operating mode popup screen in the SCADA system. The available operating selections include production only, periodic relaxation, periodic high-rate crossflow and a combination of the periodic relaxation followed by the periodic high-rate crossflow. The frequency and duration of each operating selection are set through the set-points popup display.

Table 2
Characteristics of ultrafiltration membrane used

Code	Material	MWCO[a] (daltons)	Contact angle(°)	Zeta potential at pH 7 (mV)	PWP[b] at 300 kPa (m^3/m^2d)	R_m (membrane resistance, $\times 10^{12}$ m^{-1})
NTR 7410	Sulfonated polysulfones	17,500	69	–98.63	1.84	14.1

[a] MWCO: molecular weight cut-off.
[b] PWP: pure water permeability.

Table 3
Status of solenoid valves during varied modes of operation

Mode	Valve position			
	Circulation	Relaxation	Crossflow	Flux boost
Production	Closed	Closed	Open	Open
Relaxation	Open	Open	Closed	Open
High-rate Crossflow	Open	Closed	Open	Closed

A key feature of this system was the control of the circulation solenoid valve situated in parallel with the pressure regulating valve. Problems associated with increased rates of crossflow creating increasing TMP were alleviated, as opening the circulation and crossflow during the high-rate cross-flow operating mode allowed simultaneous increased rates of crossflow at a decreased TMP.

2.3. Membrane resistance

A series of mathematical models has been developed to represent different membrane resistances [5]. The flux decline can be related to various resistances as given in Eq. (1):

$$J = \frac{\Delta P}{\mu(R_{\mathrm{m}} + R_{\mathrm{cp}} + R_{\mathrm{g}} + R_{\mathrm{aw}} + R_{\mathrm{as}})} \tag{1}$$

where μ is the dynamic viscosity (kPa s), R_{cp} the resistance due to concentration polarization (m^{-1}), R_{g} the resistance due to the gel layer, R_{aw} the resistance due to weak adsorption (m^{-1}) and R_{as} the resistance due to strong adsorption (m^{-1}).

The following details are the protocol that was used to investigate the resistances. The detailed experimental procedure for calculating the resistances can be found elsewhere [5].

Step 1: pure water was first filtered through the membrane until a constant flux was obtained,

Step 2: organic-containing water was introduced and the permeate rate was monitored with time,

Step 3: after the permeate rate reached a constant value, pure water was replaced with the organic-containing water and the applied pressure is released to remove concentration polarization,

Step 4: the fouled membrane was then rinsed with pure water so that the gel layer (highly concentrated organic layer) was removed from the membrane surface and pure water filtration was again performed,

Step 5: the membrane was soaked in a 0.1 M NaOH solution overnight so that weakly adsorbed organic matter on the membrane surface was desorbed, then pure water was again filtered.

3. Results and discussion

3.1. Effect of transmembrane pressure

Fig. 2 shows the results of the flux decline when the system was operated at various TMP of 100, 300 and 500 kPa without any cleaning. When the system was operated at 100 kPa, the fouling was minimal (less than 12%). When the system was operated at 300 kPa, there was significant fouling and this increased to 38%. When the system was operated at 500 kPa, the fouling was severe and this increased to 56%. This suggests that as the pressure increases, the fouling significantly increases. Further experiments were

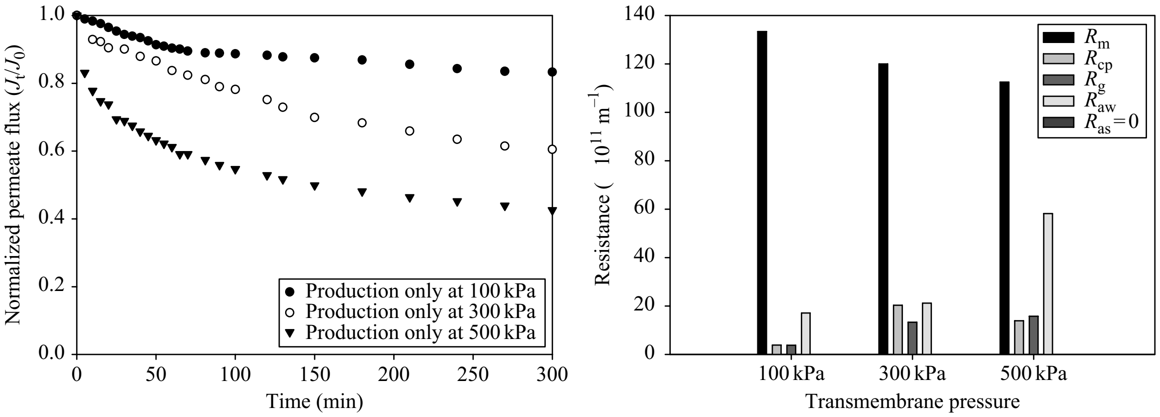

Fig. 2. Results of the flux decline versus time (left) and different membrane resistances by fouling and flux decline (right) for the three different values of transmembrane pressure used (UF membrane used = NTR 7410; MWCO of 17,500 daltons, cross-flow velocity = 0.5 m/s, transmembrane pressure = 100, 300 and 500 kPa, Reynold's number: 735.5, shear stress: 5.33 Pa).

then conducted at a TMP of 300 kPa. This allowed the relative merits of various cleaning techniques to be evaluated. Fig. 2 also presents different membrane resistances at pressures of 100, 300 and 500 kPa. The resistances of the concentration polarization and gel layer were relatively small compared with weak adsorption resistance (R_{aw}). As the TMP increased, the resistance of weak adsorption significantly increased. The recovery of membrane flux from fouled membranes after the caustic chemical cleaning was 100% for all membranes. Therefore, the dominant fouling with synthetic wastewater was caused by the weak adsorption. This fouling could be reduced through applying caustic chemical cleaning. Cho et al. [5] also reported that when there is a higher portion of fouling by weak adsorption, caustic chemical cleaning is required.

3.2. Effect of different cleaning methods

A series of experiments were then conducted to determine the effects of periodic relaxation, periodic high-rate crossflow and simultaneous periodic relaxation and periodic high-rate crossflow on the rate of flux decline. The four experiments are (i) membrane filtration with no cleaning; in this study, it is referred to as production only, (ii) production cycles of 30 min followed by 1 min of periodic relaxation, (iii) production cycles of 30 min followed by 1 min of periodic high-rate crossflow and production cycles of 30 min followed by 30 s of periodic relaxation followed by 30 s of periodic high-rate crossflow. The experiments were conducted with an operational loss of 3.2% due to production being stopped for 1 min after every 30 min of production for the periodic cleaning (relaxation, high-rate crossflow or both).

Fig. 3 shows the results of the flux decline with time for each of the cleaning methods adopted. Relaxation was found to be slightly more effective than the high-rate crossflow. This may be because there is a slightly higher TMP during the high-rate crossflow and also that there is a crossflow continuously operating during production (although it is at a significantly lower flow rate and higher TMP). A relaxation followed by a high-rate crossflow was found to give the best results, even though both durations were halved in order to maintain

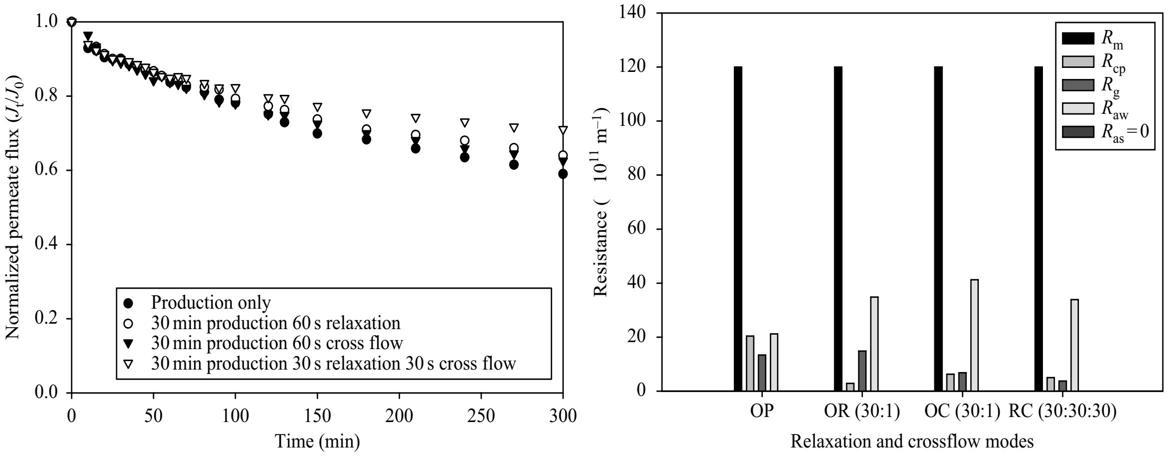

Fig. 3. Results of the flux decline versus time (left) and different membrane resistances by fouling and flux decline (right) at different cleaning techniques (UF membrane used = NTR 7410; MWCO of 17,500 daltons, cross-flow velocity = 0.5 m/s, transmembrane pressure = 300 kPa, Reynold's number: 735.5, shear stress: 5.33 Pa).

the operational losses at 3.2% for all cleaning techniques. When operated individually, relaxation and the high-rate crossflow only provided an increase in flux of only approximately 2.5%. Considering the usage of these techniques incurs an operational loss of 3.2%, there is an actual loss in productivity resulting from their usage. However, the membrane life can be extended as the rate of flux decline is slightly less. When both relaxation and the high-rate crossflow were used, there was an increase in flux of approximately 11%. This resulted in a net productivity improvement of 7.8%, after the operational losses were taken into account. Therefore, when operating under these conditions, there is both an extension to the life of the membrane and also an improvement in productivity of the system. Fig. 3 shows the variation of different resistances for the relaxation and cross-flow modes. When the relaxation and cross-flow modes were applied, the resistances associated with the concentration polarization and the gel layer significantly decreased. However, the fouling caused by weak adsorption remained high. This indicates that these modes are most likely to improve the flux decline by the fouling of the concentration polarization and the gel layer and not weak and strong adsorption.

3.3. Effect of different cleaning intervals

The above experiments showed that the best results in terms of flux decline were obtained when relaxation was followed by the high-rate cross-flow mode. Another set of experiments were conducted with increased production period. The relaxation and high-rate crossflow durations were modified accordingly. The operational losses were maintained at 3.2%. The five experiments are listed: (i) production only, (ii) production cycles of 15 min followed by 15 s of periodic relaxation followed by 15 s of periodic high-rate crossflow, (iii) production cycles of 30 min followed by 30 s of periodic relaxation followed by 30 s of periodic high-rate crossflow, (iv) production cycles of 60 min followed by 60 s of periodic relaxation followed by 60 s of periodic high-rate crossflow and (v) production cycles of 90 min followed by 90 s of periodic relaxation followed by 90 s of periodic high-rate crossflow.

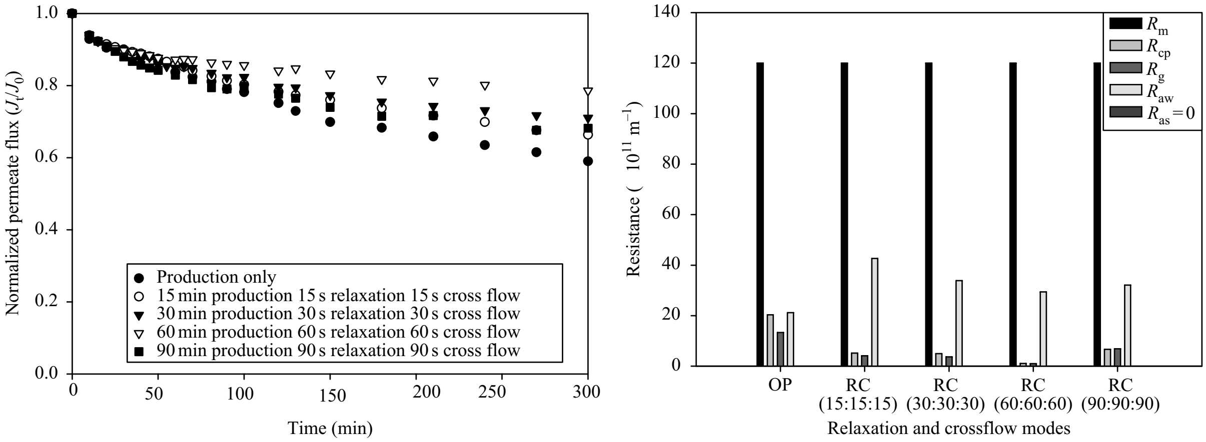

Fig. 4. Results of the flux decline versus time (left) and different membrane resistances by fouling (right) at different production periods (UF membrane used = NTR 7410; MWCO of 17,500 daltons, cross-flow velocity = 0.5 m/s, trans-membrane pressure = 300 kPa, Reynold's number: 735.5, shear stress: 5.33 Pa).

Fig. 4 shows the results of the flux decline with time for each of the experiments. Table 4 indicates that the effectiveness of the cleaning interval (relaxation and crossflow) diminishes with increased durations past the 60 s relaxation and 60 s cross-flow point. This is indicated by an additional flux recovery of only 0.12% for the additional 1 min of cleaning in the case of the 90 s relaxation and the 90 s crossflow. Prior to this additional interval, the rate of flux recovery was approximately 1.2% for each 30 s of cleaning. Fig. 4 also presents the resistance for different production periods. The fouling from only weak adsorption was significant when a 60-s relaxation and 60-s cross-flow cleaning was adopted. In the case of the short-term experiments using the production periods of 15 and 30 min, the flux declined to approximately 2 and 4%, respectively. The flux decline continued after 90 min of production and it reached almost 5%, suggesting that the utilization of the cleaning method resulting in a 3.2% operational loss is eventually justified. It is therefore evident that a critical point exists where the application of a periodic cleaning technique is optimal.

Table 4
Flux recovered for each varied production interval

Experimental condition	Total flux recovered (%)
15 min prod,15 s rel, 15 s cross	0.625
30 min prod, 30 s rel, 30 s cross	1.22
60 min prod, 60 s rel, 60 s cross	2.88
90 min prod, 90 s rel, 90 s cross	3

3.4. Effect of cleaning time ratio

Fig. 5 shows the experimental results obtained when the optimal production duration of 60 min was used with different ratios of relaxation and a relatively high-rate cross-flow velocity. The experimental conditions are: (i) production only without any cleaning, (ii) production cycles of 60 min followed by 20 s of periodic relaxation followed by 100 s of periodic high-rate crossflow, (iii) production cycles of 60 min followed by 60 s of periodic relaxation followed by 60 s of periodic high-rate crossflow, (iv) production

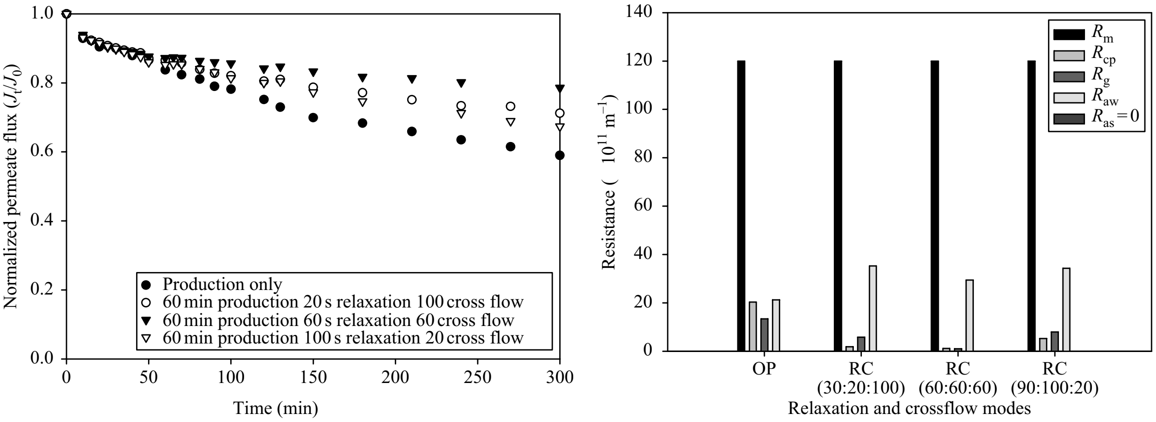

Fig. 5. Results of the flux decline versus time (left) and different membrane resistances by fouling and flux decline (right) at the three different cleaning ratios (UF membrane used = NTR 7410; MWCO of 17,500 daltons, cross-flow velocity = 0.5 m/s, transmembrane pressure = 300 kPa, Reynold’s number: 735.5, shear stress: 5.33 Pa).

cycles of 60 min followed by 100 s of periodic relaxation followed by 20 s of periodic high-rate crossflow.

Although the production and cleaning periods were maintained to keep the operational loss of 3.2%, it was found that the best results in terms of flux maximization occurred when there was an equal time allocated for the relaxation period and the high-rate cross-flow period. Interestingly, there was a higher rate of flux recovery when the period of relaxation was extended, however, these gains were eroded quickly once production started with an increased rate of flux decline. Possibly, during the extended relaxation period, the foulant layer reduction that helped with the initial flux recovery did not have sufficient time to be completely removed due to the shortened cross-flow duration. Thus, when production started again, there was the higher rate of flux decline.

3.5. *Effect of flocculation with optimum cleaning conditions*

Fig. 6 presents the effect of pretreatment of flocculation associated with the optimum cleaning conditions using 1 h production–60 s relaxation–60 s crossflow. Here, the supernatant of flocculation underwent UF without any prefilters. The permeate flux after flocculation was not improved with the hydraulic cleaning method and it may be due to the reduction of initial concentration and/or large molecular weight (MW) of the feed [7]. As the initial concentration and/or large MW of the feed after flocculation decreased, the cleaning roles of relaxation and crossflow were marginal. This phenomenon can be considered in more detail in terms of the variation of resistances (Fig. 6). All the resistances (R_{cp}, R_g and R_{aw}) after flocculation at 21 mg-Fe/L $FeCl_3$ were minimized. This suggests that there is not enough fouling to be removed by the cleaning of the relaxation and crossflow.

3.6. *Effect of adsorption with optimum cleaning conditions*

Fig. 7 shows the effect of pretreatment of PAC adsorption combined with the cleaning conditions. The supernatant after PAC adsorption was used for the UF experiments. Here, it should be noted that a small amount of PAC

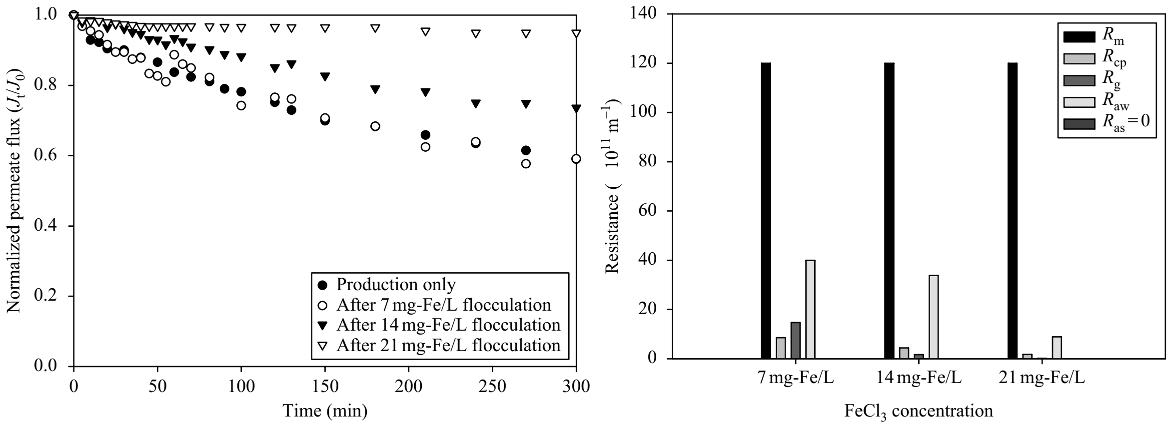

Fig. 6. Results of the flux decline versus time (left) and different membrane resistances by fouling and flux decline (right) after $FeCl_3$ flocculation for the optimal cleaning conditions (UF membrane used = NTR 7410; MWCO of 17,500 daltons, cross-flow velocity = 0.5 m/s, transmembrane pressure = 300 kPa, Reynold's number: 735.5, shear stress: 5.33 Pa).

particle still remained in the pretreated water. The flux after PAC adsorption with the automated cleaning could not increase with 0.1 and 1 g/L PAC adsorption and may be due to the coated PAC particle on the membrane surface. Thiruvenkatachari et al. [8] reported that the coated membrane with PAC can effectively stop the fouling agents in the wastewater reaching the membrane pores and thereby limit membrane fouling. They also found that, without any pretreatment or addition of PAC in the tank, the PAC-coated membrane also had the ability to retain organic materials. The resistances after PAC adsorption are shown in Fig. 7.

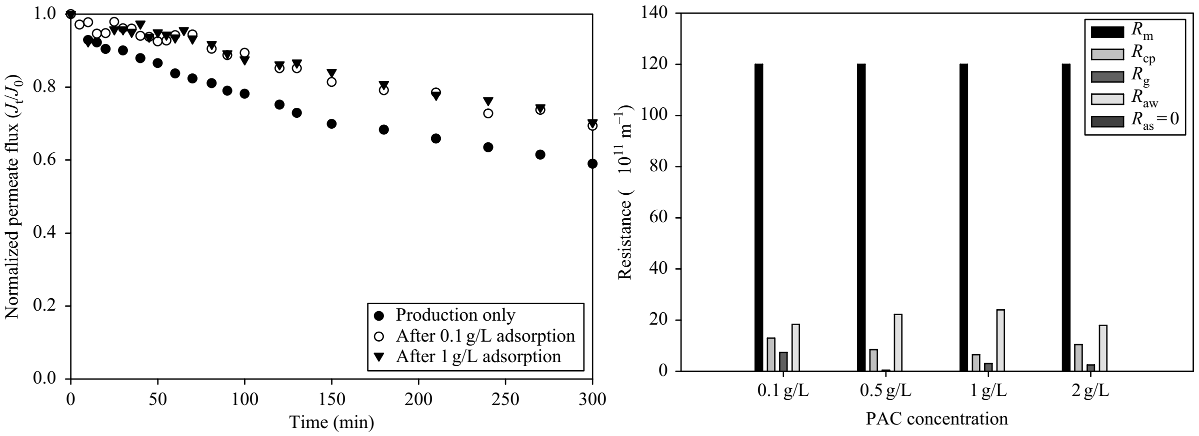

Fig. 7. Results of the flux decline versus time and different membrane resistances by fouling and flux decline after adsorption for the optimal cleaning conditions (UF membrane used = NTR 7410; MWCO of 17,500 daltons, cross-flow velocity = 0.5 m/s, transmembrane pressure = 300 kPa, Reynold's number: 735.5, shear stress: 5.33 Pa).

4. Conclusions

In this study, the effect of different hydraulic cleaning methods was investigated with the cross-flow UF unit. The cleaning methods in the SCADA system consisted of relaxation, high-rate crossflow and relaxation and high-rate crossflow. The optimum cleaning conditions were associated with different pretreatment (flocculation and adsorption). The results led to the following conclusions:

(1) Membrane fouling in a cross-flow UF unit can be minimized by the periodic relaxation and high-rate cross-flow cleaning method. This cleaning method mostly removed the resistances of the concentration polarization and gel layer compared with weak adsorption resistance (R_{aw}).

(2) A cleaning protocol utilizing a periodic relaxation step and/or a periodic increased cross-flow rate at a decreased pressure can lead to productivity improvements and an increase in the operational lifetime of the membrane. If the optimal frequency and duration of the cleaning step is used, a net productivity increase of 14.8% is achievable and a significant extension to the membrane's life results.

(3) In this study, the cleaning interval of 1 h production–60 s relaxation–60 s crossflow indicated the best result. However, a critical point of different cleaning intervals did exist.

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