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Effect of cross-flow velocity on membrane filtration performance in relation to membrane properties

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ABSTRACT

Organic fouling of membrane is one of the barriers that limit the application of membrane in water separation. Study about the effect of cross-flow velocity (CFV) on fouling rate is important to ensure that the system is operating at optimal conditions. This paper investigates the influence of CFV on the permeate flux in cross-flow configuration of nanofiltration (NF) and reverse osmosis (RO) processes by treating an organic compound in aqueous suspension. A commercial Aldrich humic acid (AHA) of 15 mg/L was used as the feed solution. The experiments were conducted at different cross-flow velocities (i.e. 0.27, 0.39, and 0.60 m/s) under a constant driving pressure of 7 bar and at an operating temperature in the range of 22–25℃. The permeate flux for both NF and RO membranes tested with AHA foulant increased slightly with time at the cross-flow velocities of 0.39 and 0.60 m/s, respectively. Despite the increase in organic rejections with the increased CFV, the rejection performance of AHA suspension was only marginally affected by the variation of CFV. Low permeate rates (flux) and strong repulsive forces were the reasons of no fouling result observed in the NF and RO membranes throughout the studied duration (i.e. 8 h). Interpretations on the chemical (contact angle and elemental composition) and physical properties (visual inspection) of membranes have further confirmed this observation. To study the fouling behavior of these membranes, it is suggested that an AHA feed with higher concentration, an addition of calcium ions, a longer filtration duration, and a higher initial flux should be used.

Keywords: Humic acid; Organic compound removal; FTIR analysis; Repulsive force; Visual inspection; Nanofiltration; Reverse osmosis

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1. Introduction

Membrane technology is widely used in water treatment due to its consistent permeate water quality, considerably high recovery rate, capabilities of handling wide fluctuations in feed, and wide areas of application [1–3]. Reverse osmosis (RO) membrane is good in rejecting dissolved ionized salts and micromolecules. If they can be claimed to have pores, the pores should not be greater than 20 Å in diameter. This is much lower than ultrafiltration (UF) which has the pore diameter of 20–100 Å [4]. *Nanofiltration* (*NF*) has similar functions as RO, but it is typically designed to reject only large *multivalent* ions. Transport mechanism in RO is solution–diffusion, while in NF, the process is governed by sieving and solution–diffusion effects because the pore sizes of NF lie between UF and RO [5,6].

Natural organic matter (NOM) is usually found in surface waters such as river water and seawater [7]. However, the concentration level of NOM in natural surface waters is low in suspension (2−15 mg/L) [8]. Although NOM is fundamentally low in concentration and harmless, it easily forms disinfection by-products, including trihalomethanes and haloacetic acids, when the source water containing NOM is chlorinated in the disinfection process [9]. The fraction of NOM consists of humic and fulvic acids [10]. Humic acid is the principal component of NOM (contains at least 70%) [8]. Despite rapid advances in the use of membrane technology in NOM removal, the issue of membrane fouling has yet to be solved.

Membrane fouling is a phenomenon where undesired substances deposit on membrane surface to form a foulant layer. The rate and extent of membrane fouling are greatly influenced by operating conditions such as the applied pressure and cross-flow velocity (CFV) [11]. Cross-flow operation is applied to the external membrane system with the aim to create a high shear condition at the surface of the membrane [12,13]. High CFV that is parallel to the membrane surface could substantially decrease resistance formed by the concentration polarization and cake layer [12,14]. This is because the increase in the feed CFV across the membrane surface will promote turbulent flow and shearing effect into the cell [15–17]. These turbulent flow and shearing effect are responsible to decrease the deposition and accumulation of foulant on the membrane surfaces and eventually may weaken the growth of the fouling layer to some extent [18–22]. Due to this effect, permeate flux increases linearly with increasing CFV [23].

Mattaraj et al. [24] conducted a study to investigate the effects of CFV on permeate flux of 10 mg/L NOM at low and high ionic strength suspensions (0.01 and 0.05 M NaCl) using thin film polyamide RO membrane from GE Osmonics. They found that the increase in CFV showed no significant effect on initial flux rate during the first 50 min of filtration time. However, the flux rate was markedly enhanced by the increase of CFV when the filtration time was extended. In a separate study carried out by Seidel and Elimelech [11], they observed that CFV caused a significant impact on the reduction of flux decline rate when the initial permeate flux was sufficiently high (i.e. 36 gfd or 61.12 LMH). They concluded that the control of proper operational parameters (initial flux and CFV) would maintain the performance of membrane and minimize the potential of membrane fouling.

Several techniques such as attenuated total reflection Fourier transform infrared (ATR-FTIR) spectroscopy, zeta potential, and contact angle have been adopted to characterize the membranes. ATR-FTIR is applied to determine the functional groups of membrane chemical properties. Zeta potential and contact angle are the indictors of membrane surface charge and hydrophobicity, which significantly influence the permeability and retention of the fluid. In general, the *surface charge* and the hydrophobicity of membranes are known to greatly affect membrane filtration performance and *membrane fouling.*

This paper presents a quantitative evaluation on the effect of CFV on the permeate flux and rejection of organic compound in NF and RO membrane processes. The study provides a detailed insight into the effect of CFV on the permeate flux and Aldrich humic acid (AHA) rejection. Interpretations on the chemical (contact angle and elemental composition) and physical properties (visual inspection) of the membranes are performed to verify the observations of no significant flux decline in these fouling tests. Verification methods that include contact angle and ATR-FTIR analysis are carried out to characterize the surface of membrane fouling.

2. Materials and methods

2.1. Membrane filtration experimental setup

All the filtration experiments were conducted using a cross-flow bench-scale membrane unit as shown in Fig. 1. The bench-scale unit was equipped with feed and collection tanks, a feed pump, a cross-flow membrane filtration cell (SEPA CF, GE Osmonics©, Minnetonka, MN), a flowmeter, pressure gauges, and an electronic scale that was connected to a computer. The cross-flow cell was designed in a plate and the

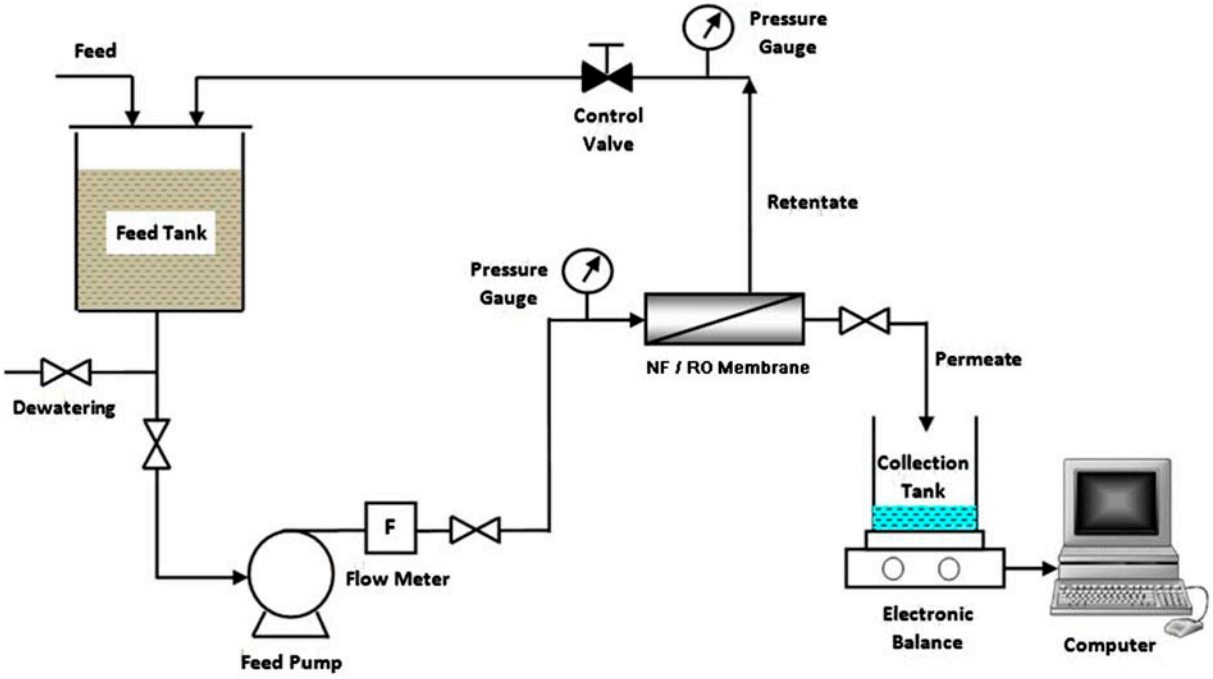

Fig. 1. Schematic diagram of bench-scale NF/RO filtration setup.

frame configuration has the thickness and width of spacer to be ~1.45 mm and 95 mm, respectively. The dimension of the cell body is 216 mm × 165 mm × 53 mm and the effective area of membrane is 0.0155 m^2, with a volume hold-up of 70 mL. The pump was driven by an adjustable/variable speed drive to control the feed inflow rate of the filtration unit. The CFVs of 0.27, 0.39, and 0.6 m/s were selected to investigate their effects on the permeate flux of membranes in this study. In addition, the constant operating pressure was maintained at ±7.0 bar. This operating pressure was adopted because it was within the typical range of NF and RO operating pressure. On low salinity applications, NF is usually working at 6–14 bar while the RO pressure is 7–20 bar [25].

The accumulative mass of permeate in the collection tank was continuously measured by an electronic balance at an interval time of 15 min. The feed inflow rate was measured and continuously logged in a data logger during the filtration process. The operating temperature of the test was monitored throughout the filtration test. It was noted that only a minimal variation (within the range of 22–25˚C) in the temperature was observed. The entire duration of the filtration process was run for 8 h. The time and mass of permeate from the cross-flow filtration cell were recorded to calculate the permeate flux, J (LMH), using Eq. (1).

$$\text{Permeate flux; } J = \frac{Q_\text{p}}{A_\text{m}} \tag{1}$$

where Q_p is the permeate flow rate, J (L/h), and A_m is the active area of membrane (m^2). The CFV (m/s), was calculated using Eq. (2) [26].

$$\text{Cross-flow velocity; CFV} = \frac{Q_\text{f}}{A_\text{c}} \tag{2}$$

where Q_f is the permeate flow rate (m^3/s) and A_c is the cross-sectional area of membrane (m^2).

2.2. Membranes characteristics

Polyethersulfone (PES) membranes which are denoted as NF1 (NF membrane) and BW30 (RO membrane) were employed in this study. Some important properties of the NF and RO membranes are given in Table 1. All the new NF and RO membrane samples were received as large sheets and were stored dry in the dark. They were cut and soaked in deionized (DI) water overnight prior to each run of experiments to remove the unwanted preservative liquids injected by manufacturers. Each of the membranes was initially filtered with ultra pure water

Table 1
Properties of the membranes used in this study

Membrane	NF	RO
Product code	NF1	BW30
Manufacturer	Amfor Inc.	Amfor Inc.
Membrane type	Flat sheet	Flat sheet
Material	Polyethersulfone (PES)	Polyethersulfone (PES)
Solute rejection (%)	98[a] ($MgSO_4$)	99.3[b] (NaCl)
Contact angle (°)	31.1 ± 1.2[c]	61.1 ± 1.0[c]
Wettability	Hydrophilic	Hydrophobic
Ionizable group (charge)	Negative[d]	Negative[d]
Water permeability, L_p (LMH/bar)	7.9[e]	3.5[e]

[a] Pressure applied at 150 psi for 2,000 mg/L of $MgSO_4$.

[b] Pressure applied at 225 psi for 2,000 mg/L of NaCl.

[c] Determined through EasyDrop contact angle measuring instrument.

[d] Given by manufacturer.

[e] Permeability test carried out with UP water at 25°C.

prior to AHA suspension in order to confirm the existence of substantial effect of AHA suspension on the flux of membranes.

2.3. Feed solution

A commercial AHA from Sigma-Aldrich was selected as the model organic foulant in this experiment. Stock solution of AHA (0.2 g/L) was prepared by dissolving the powdered form AHA in 0.05 M of sodium bicarbonate solution ($NaHCO_3$) to enhance its solubility [27]. Foulant suspension was prepared by adding DI water to the stock solution to obtain the desired concentration. In this study, concentration of the AHA tested was 15 mg/L and the dissolved AHA suspension was pH 7. The final AHA suspension was adjusted to pH 8.5 with dilute sodium hydroxide (NaOH). Table 2 shows some physical and chemical characteristics of AHA employed in the study.

2.4. Analytical methods

The rejection parameter of AHA was characterized based on the concentration of permeate. The AHA concentrations of feed and permeate samples were determined by measuring the wavelength of UV absorbance at 254 nm using UV/Vis spectrophotometer (Perkin Elmer Lambda 35, Wellesley, USA). Solute rejection, R can be defined by Eq. (3).

$$\text{Rejection}, R\% = 1 - \frac{C_p}{C_f} \times 100\% \tag{3}$$

where C_p is the concentration of permeate and C_f is the concentration of feed.

The hydrophobicity behavior of the fresh/filtered membranes was examined using an EasyDrop contact angle measuring instrument (KRÜSS GmbH, Hamburg, Germany), based on the standard sessile drop method. DI water was used as the reference solvent in the

Table 2
Physical and chemical characteristics of AHA

Appearance	Powder form
Manufacturer	Sigma-Aldrich
Color	Dark brown
pH	pH 6−7 in suspension
Solubility	Soluble in water and relatively better solubility in $NaHCO_3$ [27]
Molecular weight (g/mol)	4,170 [28]
Hydrophobicity	AHA contains 85% of hydrophobic and 15% hydrophilic fractions. Thus, it is relatively hydrophobic due to greater hydrophobic fraction [29]

hydrophobicity measurement. The membrane samples were cut into the desired size and soaked into DI water overnight to remove any remaining preservative on the surface of the membrane. They were dried in the air overnight and then mounted on a support prior to the measurement on the next day. Approximately 3.0 μL droplet of DI water was dropped on the membrane specimen and the contact angle was measured immediately after the drop placement. At least five droplets were placed on five different spots for each membrane sample measured and the value of contact angle was determined by *averaging the readings taken.* The dry membranes (fresh and filtered membranes) were analyzed using an ATR-FTIR spectroscopy (Nicolet 6700, Thermo Scientific, Waltham, MA) to qualitatively identify the fouling occurrence and to quantitatively determine the foulant functional groups on the surface of the membranes. Spectra for the fresh and filtered membranes were compared with the characteristic changes on the functional chemistry of unknown material on the membrane surfaces. ATR-FTIR spectroscopy can provide insight into the chemical nature of deposited solutes on membrane surface based on the peak spectrum. The adsorbed compounds were examined by the ATR-FTIR in the transmittance mode.

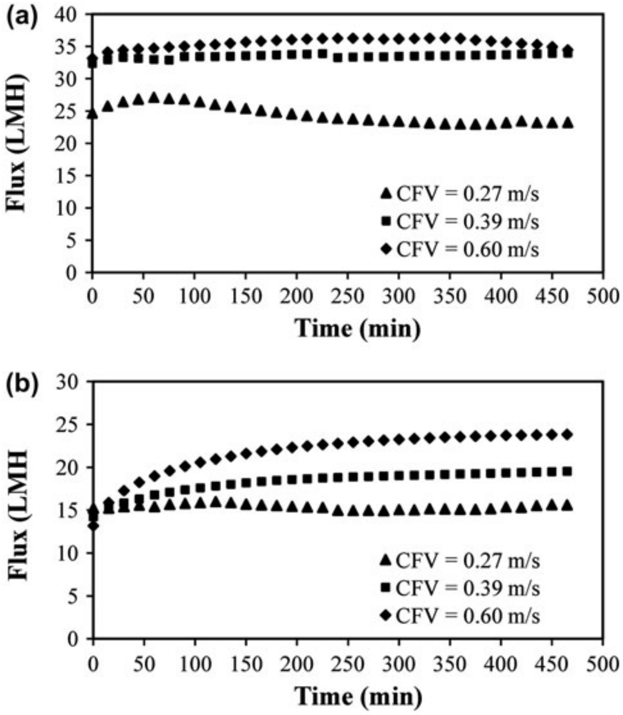

Fig. 2. Permeate flux as a function of filtration time at different cross-flow velocities. Driving pressure = 7 bar; organic compound concentration = 15 mg/L AHA. (a) NF membrane. (b) RO membrane.

3. Results and discussion

3.1. Effect of CFV on flux performance

The effect of CFV on the permeate flux of NF and RO membranes treating AHA suspension is shown in Fig. 2(a) and (b). The permeate flux of both membranes was less sensitive to the CFV at the beginning of the filtration process. The flux differences were only noticeable after 50 min of the filtration time. By comparing the fluxes generated at different cross-flow velocities, higher flux was achieved when higher CFV was imposed for both membranes [30]. This has proven that CFV is an important operating parameter in enhancing the flux of membranes while treating the AHA suspension. However, this phenomenon has not been observed at a low CFV (i.e. 0.27 m/s). The permeate flux of the RO membrane during the filtration of AHA suspension at a low CFV was almost constant over the entire filtration time (Fig. 2(b)). This is probably attributed to the pores of membrane, which are unable to open completely at the low CFV. It is believed that the CFV could help to promote membrane pores opening and enlargement [31]. Issues related to pore opening and enlargement will be further discussed in the next paragraph. Meanwhile, the reduction of flux over filtration time for the NF membrane at the low CFV (i.e. 0.27 m/s) was probably caused by the unsteady flow of NF filtration process rather than the flux decline. This could be due to the fact that the permeate flux was taken when the equilibrium state of filtration process has yet been achieved. The NF flux variation between the initial and final fluxes in the end of filtration period was noticeably insignificant, i.e. 24.7 and 23.2 LMH (flux decline ratio = 5.9%), respectively. Therefore, this condition could not be categorized as a flux decline. Studies conducted by Chilukuri et al. [32] demonstrated that severe fouling tends to occur at fluxes above 200 LMH. They added that at fluxes below 50 LMH, only a relatively insignificant flux decline and low fouling had occurred. The results imply that severe fouling does not commonly occur and it greatly depends on the initial permeate flux. Similar observation was found in the membranes with low permeation rates (i.e. NF45 and YM3) employed by Cho et al. [33]. It should also be noted that the NF1 and BW30 membranes employed in this study are at lower permeate rates than the NF45 and YM3. The low permeation rates (flux) could be the reason of the two membranes to experience a negligible concentration polarization and adsorption [33]. Furthermore, Jiang et al. [34] also revealed that blocking behavior of membrane filtration at a low flux (i.e. 18 LMH) could hardly be observed. They claimed that this phenomenon was attributed to the fact that the chance of particles, colloids, and macromolecules to deposit on membrane was very low, and consequently resulted in a low fouling rate.

Overall, there are obviously no significant sign of flux decline in the NF and RO membranes filtering AHA suspension at different cross-flow velocities (Fig. 2). This observation indicated that fouling has not occurred in the NF and RO membranes throughout the entire duration of the experimental runs. In contrast, some results demonstrated that the flux increased proportionally with the filtration time, e.g. RO membrane at 0.39 and 0.60 m/s. This observation is more pronounced at a high CFV than a low one. This is probably caused by the phenomenon of active and inactive pores of membrane as well as pore size enlargement of membranes. First, it was suspected that the inactive pores of membrane have been activated/transformed to an active condition after the filtration process. The pores which were in the active condition are capable of allowing more solutes to flow through and eventually increase the flux. In addition, it is believed that the original pores of membrane are narrow before the filtration processes. Once the dense membrane is flowed through with feed water under a high driving flowrate, the pores of membrane tend to be enlarged [31].

According to the study conducted by Košutić et al. [35], the pores of membrane can be formed in various patterns and structures. The common patterns of membrane pores are in the shape of circular and non-circular, open or closed, and may have a continuous (porous membrane) or non-continuous network (dense membrane) [35]. It has also been agreed that the driving flowrate (force) plays a significant role in the transport mechanisms of NF and RO membranes because they are formed by tight pores. In addition, previous study has reported that the pore sizes of PES-based membrane can be enlarged to 100% in an alkaline environment, in contrast, a low pH can potentially reduce the pores by 10–25% [36]. Therefore, it is suspected that the pore size of these PES-based NF and RO membranes had experienced enlargement as the AHA suspension in an alkaline environment (i.e. pH 8.5). Furthermore, a high CFV would help to enhance permeate flux due to increased back-transport of solute to the bulk solution [24], and hence decrease the solute accumulation on the surface of membrane [37]. To study the fouling behavior of these membranes, it is suggested that a greater AHA feed concentration with an addition of calcium ions [38], a longer filtration duration [28,39], and a higher initial flux [32] should be implemented due to their relatively high resistances to organic fouling.

When comparing the flux between NF and RO membranes, as expected, the flux of NF membrane was higher than that of RO membrane over the filtration time at all cross-flow velocities applied. This is because of the NF1 (NF) membrane has a relatively higher permeability than the BW30 (RO) membrane (Table 1). Besides, it is learnt from literature that hydrophobic membranes tend to produce lesser flux and are more sensitive to fouling than hydrophilic membranes [40,41]. Thus, the hydrophilic characteristic of the NF membrane could be the reason to yield a higher flux than the hydrophobic RO membrane in this study.

Fig. 3 presents the comparison of final permeate flux and AHA rejection for NF and RO membranes as a function of different cross-flow velocities. From Fig. 3(a), it is clearly seen that the permeate flux increased linearly with the increased CFV because the mass transfer increased at a high cross-flow velocity as discussed earlier [42]. However, the trend of flux increase for the NF and RO was not proportional. A higher CFV was more effective to increase the flux and reduce fouling in the RO membrane than the NF

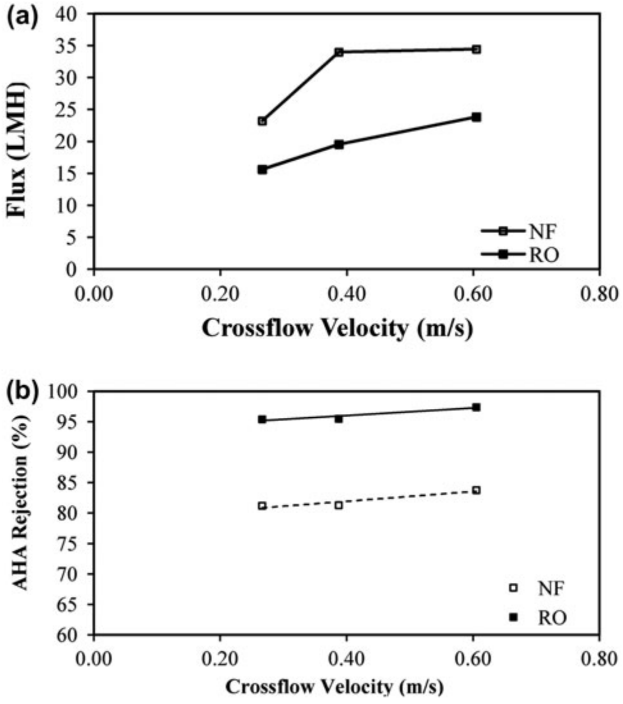

Fig. 3. Variation of permeate flux and AHA rejection with CFV for NF and RO membranes. Driving pressure = 7 bar; organic compound concentration = 15 mg/L AHA. (a) permeate flux and (b) AHA rejection (%).

membrane. The finding was supported by Choi et al. [43] who found that the permeate flux varied linearly with the CFV in MF and UF filtration processes. A high CFV increases the permeate flux due to the increase in solute back-transport to the bulk solution [13,24]. In addition, a high CFV would also decrease the flux decline and solute accumulation by sweeping away the solute from the membrane surface [24,44]. Even though the permeate flux increased linearly with an increase in the surface shear force as a result of the increase in CFV, the permeate flux plateaued once a certain shear force level was reached [12]. This phenomenon happened in the NF membrane. Increasing the CFV above 0.39 m/s did not result in a further increment in the permeate flux for the NF membrane. As the CFV increased from 0.27 to 0.39 m/s, the permeate flux of NF membrane increased rapidly from 23.2 to 34 LMH (flux decline ratio = −46.6%), while the flux of the NF membrane increased marginally from 34 to 34.4 LMH (flux decline ratio = −1.2%) at a high CFV region. The results indicated that the maximum flux of NF membrane was almost achieved when the CFV was 0.39 m/s. Liu et al. [45] revealed that an increase in the CFV would lead to an increase in the equilibrium permeate flux for the NF and RO membranes. The shearing effect at lower CFV had given a larger effect on reducing the extent of flux decline than that at higher CFVs. They added that the increase in the stabilized permeate flux at a high CFV is likely caused by the reduced concentration polarization and foulants adsorption on membrane surface. However, the rate of permeate flux increase as a function of CFV for the NF membrane was less than that of RO membrane at high CFV. The achievable permeate flux of NF membrane could not be further increased once the limiting flux had been reached.

3.2. Effect of CFV on rejection performance

The rejection performance of NF and RO membranes was evaluated in terms of AHA rejection and permeate quality.

3.2.1. AHA rejection

Solutes rejection is another key factor for the performance evaluation of membrane processes. Fig. 3(b) shows the AHA rejection as a function of CFV for the NF and RO membranes. Surprisingly, the effectiveness of AHA rejection increased with the increased CFV. As the CFV increased from 0.27 to 0.39 m/s, the AHA rejection for the NF membrane increased from 81.2 to 83.8%. Similar trend of AHA rejection with the increase in CFV was also observed for the RO membrane. The AHA rejection rates at the cross-flow velocities of 0.27, 0.39, and 0.60 m/s for the RO membrane were 95.4, 95.6, and 97.7%, respectively. The results were supported by Chang et al. [19], who conducted a systematic experimental study to investigate the effect of operating parameters on the NOM rejection for a lake water in an NF filtration. The performance of NF membrane on NOM rejection was assessed by the measurement of DOC. They claimed that the foulant on the membrane surface was removed more effectively at a high CFV [19]. However, they reported that the effect of CFV on the DOC rejection ratio was not significant for an operating pressure below 7.6 bar. Their results showed good comparisons with the observations obtained from the present study, as the filtration system was performed at a constant driving pressure of 7 bar. Besides, Liu et al. [45] also investigated the effect of CFV on the COD removal of textile wastewater treated by NF and RO membranes. They elucidated that the better rejections of COD for both the NF and RO membranes at a high CFV can be attributed to the reduction of concentration polarization and membrane fouling. The extent of concentration polarization and membrane fouling is associated with the loss of membrane rejection capability. It is learnt that an increase in the CFV would increase the mass transfer coefficient and subsequently minimize the effect of concentration polarization, causing higher solutes rejection by the membrane [46]. Despite the organic rejections increased with the increased CFV, the rejection performance of AHA suspension was only marginally affected by the variation of CFV. This observation was in good agreement with studies reported by Öner et al. [47], whereby they discovered that the increase in flow rate did not affect salt, boron, and silica rejections significantly.

3.2.2. Permeate quality

Fig. 4 illustrates the permeate quality of NF and RO effluents as a function of CFV in the filtration of AHA foluant. The results demonstrated that the permeate effluent of NF and RO membranes in rejecting AHA was reasonably consistent. The concentration of AHA in the permeate was lower than 3.0 mg/L for both the NF and RO membranes, with the RO membrane in particular showed slightly better AHA rejection than the NF membrane. The AHA concentration in the RO permeate was as low as 0.4 mg/L, while the higher value of permeate concentration was achieved in the NF membrane (2.5 mg/L). The results indicated that the RO membrane has successfully

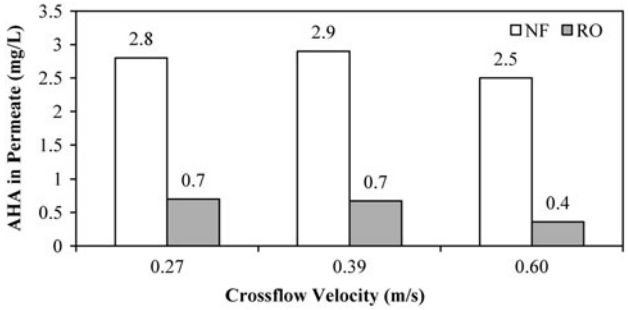

Fig. 4. Permeate quality in AHA concentration (mg/L) for NF and RO membranes. Driving pressure = 7 bar; organic compound concentration = 15 mg/L AHA.

reduced the organic content (i.e. in AHA form) to a greater extent than the NF membrane tested. The effectivenesses of the NF and RO membranes in organic rejection can be explained in terms of their intrinsic properties [48]. Typically, RO membrane is capable of rejecting all solutes constituent including monovalent ions in the solution, while the smallest solute that can be rejected by the NF membrane is a divalent ion. This can be traced from the comparatively high passage of the AHA species in the NF membrane than that of RO membrane. Even though the final permeate quality for both the NF and RO membranes was quite consistent under the three different cross-flow velocities tested, it was found that a slightly better quality of permeate was generated with a high CFV. Literatures have proved that fouling layers on the RO membrane surfaces result in a high tendency for the passage of species, and hence produce a poor quality of permeate due to a high diffusion mechanism [48]. In addition, a low rejection efficiency was also caused by combined effect of reduced surface charge and thickness of the top selective layer [49]. Table 3 summarizes the performance of membranes in this AHA filtration test. The effects of CFV on both the flux and rejection efficiency have already been discussed previously.

3.3. Impacts of membrane surface properties upon fouling tests

The absence of fouling by AHA suspension in these filtration systems cannot be explained only in terms of flux decline and rejection performances. The chemical properties (contact angle and elemental composition) and physical properties (visual inspection) of the membranes were tested: (1) to investigate the effect of CFV on the NF and RO membranes, and (2) to verify the results of no significant flux decline in the NF and RO filtration experiments obtained for the AHA suspension.

3.3.1. Hydrophobicity analysis—contact angle

The hydrophobicity of a surface is defined as the tendency of the surface to absorb water. A hydrophilic surface has a water-loving characteristic and it tends to absorb water, while hydrophobic surface posses a water-hating characteristic to repel water. The contact angle measurement is commonly used to describe the hydrophobicity behavior of a membrane surface. The contact angle θ is an angle between water and surface of membrane. In general, the contact angle in the range of $0 < \theta < 90^\circ$ corresponds to hydrophilic surfaces, while the contact angle in the range of $90 < \theta < 180^\circ$ represents the hydrophobic membrane surfaces [40]. Generally, the fouled membrane would encounter a significant reduction in the degree of contact angle compared with the fresh membrane [50,51]. Her et al. [51] revealed that the fouled membrane which experienced a severe flux decline had contact angle of about 7°–10° lower than the fresh membrane.

Table 3
Summary of bench-scale membrane performance in AHA filtration tests

	NF						RO					
	Permeate flux (LMH)		J^a (%)	AHA (mg/L)		R^b (%)	Permeate flux (LMH)		J^a (%)	AHA (mg/L)		R^b (%)
Cross-flow velocity (m/s)	J_i	J_v		In	Out		J_i	J_v		In	Out	
0.27	24.7	23.2	5.9	14.9	2.8	81.2	15.2	15.6	−2.3	15.2	0.7	95.4
0.39	32.4	34.0	−4.9	15.5	2.9	81.3	14.2	19.5	−37.2	15.5	0.7	95.5
0.60	33.2	34.4	−3.8	15.4	2.5	83.8	13.2	23.8	−80.9	15.4	0.4	97.4

[a] Flux decline ratio % = (initial flux, J_i − final flux, J_v)/initial flux, J_i × 100 (%).

[b] Rejection, R % = (feed concentration, C_f − permeate concentration, C_p)/feed concentration, C_f × 100.

For relatively less severe fouling membranes, the decrease in contact angle value was only about 5˚. In a separate study carried out by Ruohomäki et al. [50], they reported that severe fouling only occurred when there is a marked decrease in the contact angle (i.e. 60˚–22˚). Even though the contact angle recedes from nearly 60˚ to 45–50˚ after filtration with humic acid, the flux decline was not obvious. The contact angles of the NF and RO membranes were measured and tabulated in Table 4. It is found that the fresh NF membrane exhibits hydrophilic-like behavior as the contact angle is 31.1˚. The results of contact angle revealed that the NF membrane was not fouled by the AHA foulant, as the contact angle of NF membrane increased significantly regardless of the CFV applied in comparison with the fresh NF membrane. In fact, the hydrophilic membranes are well known to be less susceptible to adsorption fouling by organic particles due to organic matters which are less favorably to be adsorbed onto hydrophilic membrane than the hydrophobic one [3,40]. Thus, it can be concluded that no fouling sign in terms of chemical membrane properties (i.e. contact angle) was observed in the NF filtration test.

Polyethersulfone material should exhibit a hydrophobic characteristic when the value of contact angle is about 60˚ [52]. Thus, the fresh PES-based RO membrane used in this study has categorized as hydrophobic with the contact angle of 61.1˚. However, the RO membrane may have changed from hydrophobic to hydrophilic after filtration with AHA suspension as the contact angle for all the tested membranes decreased to less than 60˚. The results of contact angle revealed that the RO membrane was probably fouled by the AHA foulant. The reduction in contact angle of the RO membrane indicated that the adsorption of humic acid had probably occurred. However, it should also be noted that highest reduction in the contact angle was only 6.1˚. A number of researchers, Her et al. [51] and Ruohomäki et al. [50], suggested that this marginal reduction was insufficient to prove that the membrane has been fouled. The decrease in contact angle after filtration with AHA suspension is probably caused by the surface interactions of membrane with AHA suspension. The exposure of membranes to reactive radicals from $NaHCO_3$ and NaOH in AHA suspension may probably change the surface tension of membranes.

3.3.2. FTIR analysis

Transmittances of ATR-FTIR of NF and RO membranes were compared to identify the chemical composition differences between the fresh and filtered membranes. Figs. 5 and 6 show the spectra of ATR-FTIR obtained for the fresh and filtered membrane with AHA suspension of 15 mg/L at different cross-flow velocities for the NF and RO membranes, respectively. In comparison with the FTIR spectrum of fresh membrane, insignificant transmittance was observed on both the membranes after expose to AHA filtration. It is apparent that no changes in the functional groups of fresh membranes have occurred after filtration with AHA suspension. Despite being exposed to different cross-flow velocities, all the filtered membranes yielded almost an identical spectral pattern with slight differences in intensity. Dominant chemical functional groups of AHA suspension were shown in the spectra of AHA suspension (Figs. 5 and 6). Three dominant peaks in the spectrum were identified as –OH stretching, C=O stretching, and C–H bending at wavenumber of 3,396, 1,646, and 685 cm^{-1}, respectively. The broad band of peaks at wavenumber of 3,396 cm^{-1} indicated the presence of large amounts of –OH groups in the AHA suspension. The ATR-FTIR peaks detected for the fresh and filtered NF and RO membranes are tabulated in Table 5. The variations on the peaks of spectra of the filtered membranes are expected to be observed at the three dominant peaks of AHA suspension, if the membrane fouling and adsorption persist. To be more specific, the FTIR peaks of the fresh membrane are expected to decrease in transmittance intensity for a

Table 4
Contact angle of the studied NF and RO membranes

Conditions	Contact angle (˚) NF	Contact angle (˚) RO
Fresh membrane	31.1 ± 1.2	61.1 ± 1.0
Membrane filtered with AHA suspension at CFV = 0.27 m/s	43.8 ± 8.9	56.5 ± 3.6
Membrane filtered with AHA suspension at CFV = 0.39 m/s	57.0 ± 1.1	55.0 ± 3.1
Membrane filtered with AHA suspension at CFV = 0.60 m/s	59.8 ± 0.2	56.1 ± 3.4

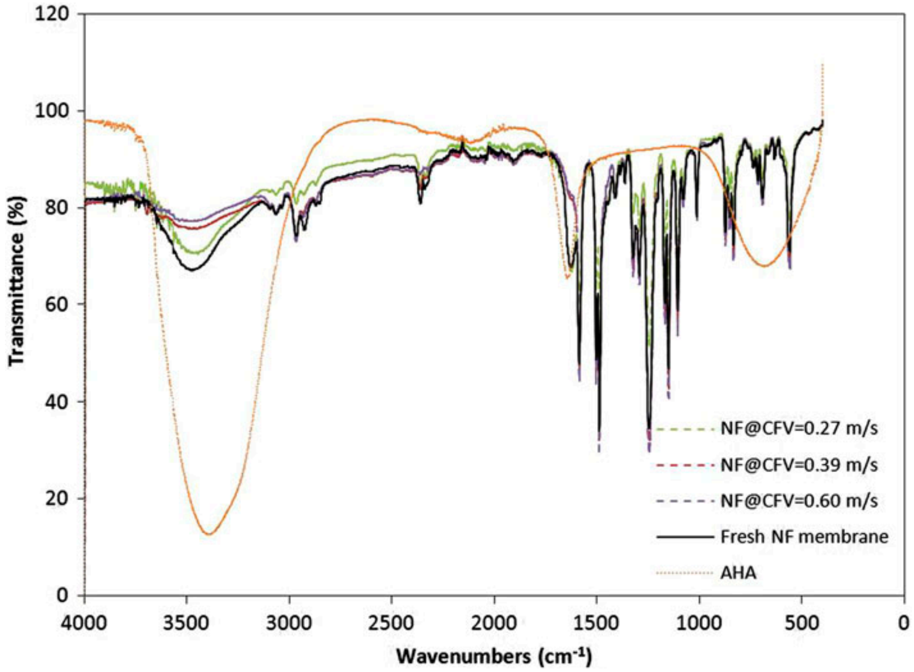

Fig. 5. ATR-FTIR spectra comparison of NF membrane at different cross-flow velocities. Driving pressure = 7 bar; organic compound concentration = 15 mg/L AHA.

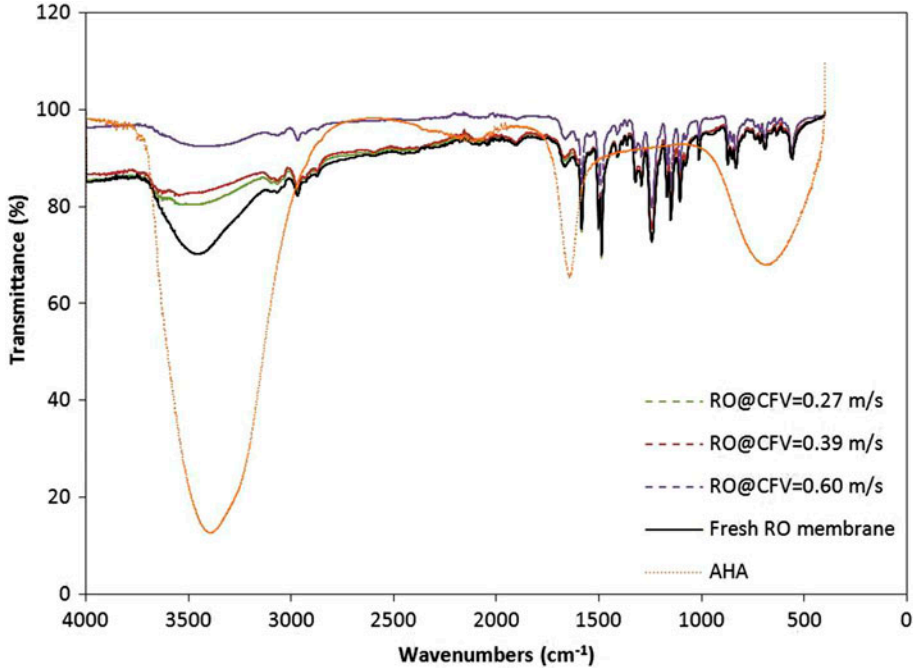

Fig. 6. ATR-FTIR spectra comparison of RO membrane at different cross-flow velocities. Driving pressure = 7 bar; organic compound concentration = 15 mg/L AHA.

fouled membrane. The decrease in transmittance intensity indicated that the original clean surface of membrane has been coated by the functional groups of foulants [53]. However, there were no significant differences in the spectra ranging from 500 to 4,000 cm^{-1} between the spectra of fresh and filtered membranes.

Table 5
ATR-FTIR peaks detected for the fresh and tested NF and RO membranes

Wavenumber (cm^{-1})	Functional groups
3,400–3,600	−OH stretching
2,960–3,095	C–H stretching
1,550–1,610	C=O stretching
1,445–1,485	$-CH_2$ bending
1,430–1,470	$-CH_3$ bending

These spectra seem to be less affected by the three dominant peaks of AHA suspension. These results are consistent with the results of flux performance, whereby an increase in the flux is observed (Fig. 2).

3.4. Further explanation on the correlation of membrane and AHA characteristics with no fouling results

Nghiem and Hawkes [38] reported that a decrease in solution pH of humic acid could result in a low charge density of these organics. The charge density of a particular species is dependent on its isoelectric point (IEP). The IEP of a molecule is the pH at which the molecule has a net electrical charge of zero. For instance, if the solution pH is above the IEP, the charge will be negative, while below the IEP, the charge will be positive. As has been reported elsewhere [50], the IEP of AHA is around 3; therefore, the net charge of AHA would be negative since the pH of AHA suspension is 8.5 (above the IEP). Due to the usage of negatively charged membranes in this study, a strong repulsive force was created when these negatively charged membranes reacted with the negatively charged AHA suspension [54]. Fig. 7 illustrates the repulsive force due to both negatively charged surfaces. Besides, the adsorption of humic acid was found to be the strongest at pH 3 (close to the IEP of humic acid) and independent on the material of membrane [50]. At high pH, the organic adsorption and foulant–membrane adhesion were less severe due to the electrostatic repulsive force of negatively charged humic acid with the negatively charged membranes. However, the suspension in a high ionic strength will cause a reduction of this negative charges by compressing the electric double layers, neutralizing, and screening the charge of humic acid [38]. In addition, the organic fouling becomes more severe with the presence of Ca^{2+} ions. Part of the negative charges was used to neutralize the Ca^{2+} ions. Both conditions (i.e. low pH and high ionic strength of suspension) have significantly contributed to the reduction of the electrostatic repulsive forces between humic acid molecules in suspension and subsequently the increase of foulant−membrane adhesion. In contrast, high charge density (negative charge) of the AHA suspension at pH 8.5 with the absence of ionic strength and Ca^{2+} ions in this study had created a strong electrostatic repulsive force between foulant-membranes, and eventually caused a reduction in the adhesion forces. Ruohomäki et al. [50] extended their research to verify the result of obtaining no fouling with the measurement of contact angle. They found that severe fouling phenomenon was not detected in the UF and NF membranes when the suspension was at a pH range of 7−10. The results are in good agreement with the fact that the flux is relatively higher for a suspension under a basic and neutral pH, than the acidic conditions [42]. They claimed that the sign of humic acid adsorption was only detected when the contact angle decreased from 60° to 22° at pH 3. From the foregoing discussion, it can be concluded that no fouling results was obtained in this study due to the insignificant reduction in contact angle as observed in Section 3.3.1. It is believed that the AHA suspension with pH 8.5 is the reason of pore enlargement and flux enhancement [36]. Therefore, it is safe to claim that the NF and RO membranes employed in this study possess a good resistance against organic fouling.

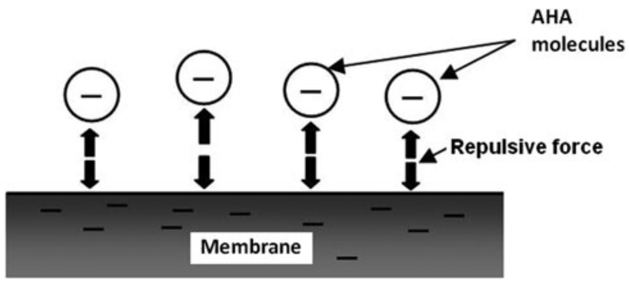

Fig. 7. Repulsive force due to both negatively charged surfaces.

3.4.1. Visual inspection of membrane surfaces

Visual inspection of the membrane include assessing the color, odor, particle deposits, and faults of the elements [55]. Color variation of the membrane surfaces is a major visual inspection criterion in this study. The color intensity represents the amount of mass covered on the surface of membranes [56]. Both the NF and RO membranes tested at CFV of 0.60 m/s were relatively "clean" and "clear" when observed with naked eyes as shown in Fig. 8(c) and (f), respectively. On the contrary, light brown color was visually detected for the NF and RO membranes tested at a low CFV i.e. 0.27 m/s (Fig. 8(a) and (d)). These observations implied that these membranes were more

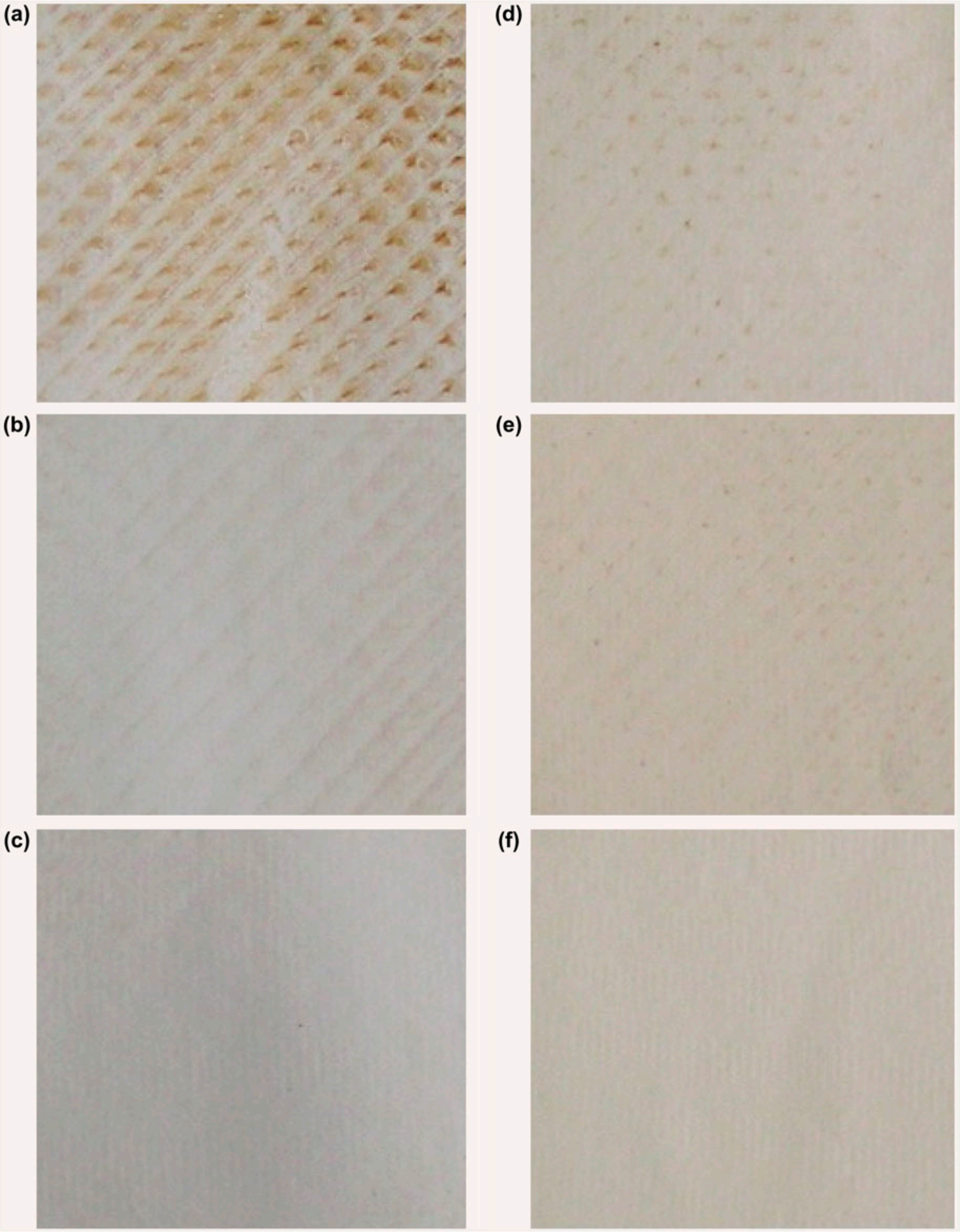

Fig. 8. Visual inspection of the membranes after filtration with AHA suspension under different cross-flow velocities: (a) NF@0.27 m/s. (b) NF@0.39 m/s. (c) NF@0.60 m/s. (d) RO@0.27 m/s. (e) RO@0.39 m/s. (f) RO@0.60 m/s.

"dirty" than the membranes that tested at higher cross-flow velocities (0.39 and 0.60 m/s). However, no serious and visible flaws, cracks, patches of scales, or damages have been observed on the membrane surfaces filtered with the AHA suspension. Visible appearance of brown patches on the samples of membrane surfaces indicated the presence of a severe fouling layer [57]. These visual inspection results have further confirmed that severe membrane fouling has not occurred in this study.

4. Conclusion

A cross-flow bench-scale membrane unit was employed to study the effect of CFV on the permeate flux and AHA rejection in NF and RO membranes. The following conclusions can be drawn from this study:

1. Despite an increase in the organic rejections with the increased CFV, the rejection performance of AHA suspension was only marginally affected by the variation of CFV.
2. Fouling phenomena have not been observed in the NF and RO membranes throughout the studied duration (i.e. 8 h). The low permeation rates (flux) and strong repulsive force could be the reasons of the two membranes to experience a negligible concentration polarization and adsorption.
3. The interpretations on the chemical (contact angle and elemental composition) and physical properties (visual inspection) of membranes are performed to verify the observations of no significant flux decline in these fouling tests. The results of contact angle revealed that the NF membrane was not fouled by the AHA foulant as the contact angle of NF membrane increased significantly regardless of the CFV applied in comparison with the fresh NF membrane. Besides that, it is apparent that no changes in the functional groups of fresh membranes have occurred after filtration with AHA suspension from the FTIR spectrum. No serious and visible flaws, cracks, patches of scales, or damages have been observed on the membrane surfaces filtered with the AHA suspension.

It is safe to claim that the NF and RO membranes employed in this study possess a good resistance against organic fouling. To study the fouling behavior of these membranes, it is suggested that an AHA feed with higher concentration, an addition of calcium ions, a longer filtration duration, and a higher initial flux should be used.

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