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Searching for ultrafiltration membrane molecular weight cut-off for water treatment in recirculating aquaculture system

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ABSTRACT

Ultrafiltration (UF) has been proposed as a promising technology in recirculating aquaculture systems (RAS) to remove both organic contaminants and other fine contaminants including viruses and pathogenic bacteria. However, fouling is still a severe problem during this application. This paper investigated the fouling behavior of three different UF membranes examined using five different aquaculture contaminants. The experiments were performed using UF membranes with molecular weight cut-off 10, 50, and 100 kDa. Humic acid, shrimp feed, *Spirulina sp.*, *Vibrio harveyi* and IHHNV were used as contaminant models. Scanning electron microscope was used to visualize the presence of foulant on the membrane surface. The results showed that fouling behavior was affected by both membrane cut-off (pore size) and foulant type. Two fouling behaviors were observed: (i) rapid flux decline at the early stage of filtration followed by relatively constant permeate flux until experiments finished, and (ii) rapid flux decline at the early stage of filtration followed by a gradual decrease in permeate flux. Due to its reliable flux value and high rejection, 100 kDa UF membrane should be considered as the most suitable UF membrane for RAS application.

1. Introduction

Aquaculture, the farming of aquatic organisms and plants in both coastal and inland areas, is very important sector in achieving world food security. Therefore, much attention has been paid for a sustainable aquaculture on the one hand. Recirculating aquaculture system (RAS), which recirculates or reuses water after undergoing treatment, is one of the developments towards sustainable aquaculture. On the other hand, a severe problem found in aquaculture system is diminishing or decreasing of animals as a result of disease or lack of oxygen. This problem is found in conventional aquaculture systems, recirculating aquaculture system (RAS) and hatchery systems. More attention should be paid for the RAS, where water is continuously reused via recycling process [1–5].

The presence of excess organic compounds in an aquaculture system lowers the dissolved oxygen (DO) content (both chemical and biological) that has an impact on the lack of oxygen supply to the animals respiratory. Organic compounds in aquaculture system can be derived from the water used itself, the residual of feed, and the metabolism of animals. It was reported that eighty percent (dry weight) of feed used in the aquaculture system is released as an animal excretion [6]. As an example, production of 100 kg catfish produced 1190 kg of dry solids, 60 kg of nitrogen compounds and 12 kg of phosphorus compounds [7]. In the more recent study [8], it was reported that 25% of the feed applied to the aquaculture system will end up as suspended solid. Therefore, the organic compounds should be reduced to the safe level for culture growth of aquatic animals such as fish and shrimp. In addition, viruses, microalgae, and pathogenic bacteria are also frequently found in aquaculture systems in which the presence of these contaminants can cause the destruction of animals.

To avoid the destruction of aquatic organisms, removal of organic compounds and other contaminants from aquaculture systems has to be conducted. This removal is usually performed by the centrifugation, clarification, gravity filtration or precipitation process, screening with different pore sizes (60–200 microns), biofilter, and biological processes [9–13]. The fundamental weakness of these techniques is the leakage of fine particles or contaminants including pathogenic bacteria and viruses [14]. In addition, biological processes have other problems such as their complexity especially for denitrification [15], high hydraulic retention time (HRT) [16], and high backwash intensity of the

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biofilter [17].

Foam fractionation has been proposed to remove those fine particles and can be combined with other processes such ozone to enhance the separation efficiency [18,19]. Nevertheless, this technology is not suitable for large and primary particles removal. Phytoremediation has also been proposed for treating aquaculture wastewater [20]. However, the use of plants in aquaculture system will block photosynthesis and consequently, the oxygen content in water will decrease. To solve these problems, application of membrane technologies in aquaculture systems has been proposed by many researchers [3,21–25]. In a recent study, it was shown that the integration of membrane filtration into RAS increased the quality of water effluent [2]. Further, several membrane technologies have been applied in small aquaculture industries [3,26]. A coupling catalytic ozonation-membrane filtration system for the recirculating aquaculture wastewater treatment using membranes coated by Ti-Mn/TiO_2 oxide was reported in a recent study by Chen et al. [27]. They claimed that their coupling system results in good permeate quality, short start-up period, and the stable operating conditions. However, special attention should be paid for the residual ozone, which is harmful to the aquatic organisms. It was reported that algae paste, decaying rotifers, and dry feed seemed to contribute to the most of membrane fouling [28].

In general, the results of both laboratory and field experiments indicated that membrane technologies deliver a promising potential for removing fine contaminants including organic compounds, viruses, and pathogenic bacteria. However, as in other applications, fouling–the decline of flux over time due to the deposition of suspended or dissolved substances on external surface, at the pore openings or within its pore [29,30]–reduces significantly the performance of the process and consequently prevents a more widespread commercial applicability in aquaculture system.

Numerous studies on the application of UF membrane in RAS have been reported. However, different results were observed among different reports. It is certainly due to different process condition, membrane property, contaminant, and feed water characteristic are involved. As a result of this situation, it is hard to determine the best molecular weight cut-off (MWCO) of UF membrane that should be used in an aquaculture system. It is important to mention that MWCO is a parameter usually used to express the retention and membrane pore size of ultrafiltration membrane. To the best of our knowledge, no systematic study has been performed to determine the most suitable UF membrane pore size that should be applied in RAS. It should be kept in our mind that the performance of a process using membrane is strongly influenced by membrane pore size or MWCO [31–33]. Systematic fouling study using seawater as the feed has been performed [34]. Nevertheless, their study was dedicated to the pretreatment of RO membrane for desalination.

As consequences of various contaminants with different characteristics exist in aquaculture systems and various commercial UF membranes with different MWCOs are available, selection of UF membrane based on their MWCO is a very important step in practical application of UF membrane in RAS. The main aim of this study is to determine the suitable MWCO of UF membrane in RAS application considering the fouling mechanism study. It is important to point out that for the practical application; the use of different MWCO membrane for the removal of different contaminants is not possible. Furthermore, the membranes used in RAS cannot be chosen only based on the manufacturer specification. The best MWCO determination is based on the efficiency and fouling behavior, which corresponds to the resulting flux. Various model contaminants of aquaculture systems, i.e. humic acid, residual shrimp feed, microalgae (*Spirulina* sp.), pathogenic bacteria (*Vibrio harveyi*) and virus (IHHNV, Infection hypodermal and hematopoietic necrosis virus) were used in this study. These five contaminants were expected to be able to represent natural organic matters (NOMs), feed residuals, microalgae, bacteria and viruses; respectively, which usually exist in aquaculture system.

2. Materials and methods

2.1. Materials

Since the consistency of membrane properties manufactured industrially is typically better than lab-made membrane, the commercial UF membranes (supplied by Alfa Laval, Denmark) were used in this study. The membranes included GR81PP (10 kDa, polyethersulfone), GR51PP (50 kDa, polysulfone) and GR40PP (100 kDa, polysulfone). The contaminant models used were shrimp feed (SF) with the composition was (%wt) protein > 36%, crude fiber < 4%, fat > 5%, water < 12% and ash < 12% (obtained from P.T. Gold Coin, Indonesia), humic acid (HA, purchased from Sigma-Aldrich), *Spirulina platensis* (MA, obtained from Center for Bioprocess and Renewable Energy, Diponegoro University), marine bacterium *Vibrio harveyi* (VH, obtained from BBPBAP, Jepara, Indonesia), and IHHNV (obtained from BBPBAP, Jepara, Indonesia). The experiment was also performed using mixture of these contaminant models (SMH). The usage of these contaminants was based on the results of preliminary analysis of water quality from the real aquaculture system as well as previous literatures [1–5]. Real aquaculture water (RAW) with total dissolved solids ~4.1 g/L was obtained from the aquaculture pond in Jepara, Indonesia. Sodium chloride and HCl were purchased from Merck. Purified water produced from home-made RO-ion exchange system was used for all experiments. In order to apply the UF membranes in RAS, all foulant solutions were prepared by dissolving the contaminant models in 10 g/L of sodium chloride solution.

2.2. Methods

2.2.1. Investigation of fouling behavior

Study on fouling behavior was performed by investigation of membrane-solute interactions (adsorptive fouling) and membrane-solute–solute interactions (ultrafiltration fouling). The investigation of adsorptive fouling was carried out by using a dead-end stirred cell filtration system (Amicon cell model 8010 from Millipore) and followed our earlier reported work [35]. Briefly, pure water flux (J_0) was measured for each membrane sample. The pure water flux was measured at a pressure of 300 kPa for 10 kDa UF membrane and 100 kPa for both 50 and 100 kDa UF membranes. A model of contaminant solution with certain concentration (SF (1 g/L), MA (1 OD in water with 10 g/L TDS), HA (1 g/L), VH (10^8 CFU), IHHNH (10/500 mL/mL)) was added to the cell. SMH composed of 0.25 g/L SF, 0.5 OD MA and 0.25 g/L HA was also used. Thereafter, the outer membrane surface was exposed for 3 h without any flux at a stirring rate of 300 rpm and atmospheric condition. The preliminary experiments showed that 3 h of adsorption was sufficient to achieve saturation of the surface adsorption capacity for all foulant models. This experiment condition was also supported by our previous studies [30,36]. Afterward, the solution was removed, and the membrane surface was rinsed twice by filling the cell with pure water (5 mL) and shaking it for 30 s. Pure water flux (J_a) was again measured. The extent of adsorptive fouling was expressed in term of relative water flux reduction (RFR; cf. Eq (1)), which was calculated from the water fluxes at the same pressure before and after adsorptive fouling. The effect of contaminant concentration on RFR was also investigated using SF, HA and MA as contaminant models. The concentration of SF and HA was varied from 0 to 5 g/L, whereas the concentration of MA was varied from 0 to 1 OD.

$$\mathrm{RFR} = \frac{J_0 - J_a}{J_0} \quad (1)$$

A home-made laboratory scale for cross-flow filtration test was used in all ultrafiltration experiments [30]. The set-up consisted of a feed tank (3 L volume), a flow indicator, a pump, a pressure indicator connected to feed side of the membrane to determine the trans-membrane pressure and a flat–sheet membrane cell. A simplified diagram of the set-up

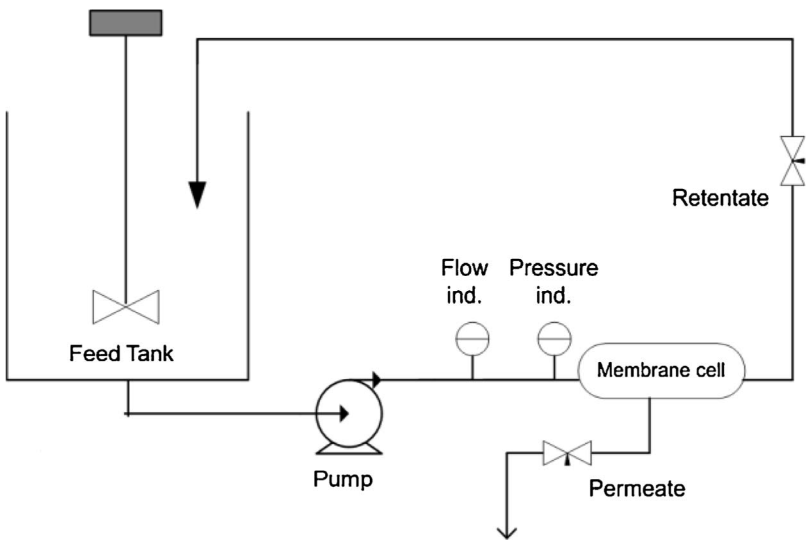


Fig. 1. The schematic cross-flow filtration equipment used for UF experiments.

is given in Fig. 1.

In each experiment, a new circular membrane disk with the effective area of 0.00125 m^2 was used. As conducted in adsorption experiments, the membrane was firstly compacted by filtering the water for at least 0.5 h at a pressure of 400 kPa. A contaminant feed model with certain concentration (SF (0.5 g/L), MA (0.1 OD in water with ~10 g/L TDS), HA (0.025 g/L), VH (10^6 CFU), IHHNH (0.5/500 mL/mL)) was added to the feed tank and pumped into membrane cell. SHM comprising of HA (0.025 g/L), SF (0.5 g/L), MA (0.1 OD), marine bacterium VH (10^6 CFU) and IHHNV (0.5/500 mL/mL) was also used as the contaminant feed model. The concentration of contaminant models was based on the characterization of actual aquaculture water. In order to maintain constant feed concentration, the retentate and permeate were returned to the feed tank. All experiments were conducted at room temperature (28 ± 2 °C) with constant trans-membrane pressure (TMP) mode (100 kPa for both 50 and 100 kDa UF membranes, and 300 kPa for 10 kDa UF membrane; the difference in pressure was due to the difference in TMP required to result in reasonable flux permeate). During UF experiments, the flux profile as a function of time was gravimetrically monitored. To investigate the effect of concentration polarization, after UF experiment the pressure was lowered to ~0 kPa for 5 min (in this way, no permeate flux was produced) then permeate flux was again measured at similar pressure with UF [30,35].

2.2.2. Visualization of membrane surface morphology

The top surface morphology of the membranes was visualized to observe the presence of foulant. The experiment was performed by using a scanning electron microscopy (FEI Inspect 550). The outer surface of the sample was coated with gold/palladium and sputtered for 0.5 min before analysis.

2.2.3. Analytical methods

The concentration of humic acid and residual shrimp feed was analyzed using a UV spectrophotometer (Shimadzu UV mini 1240 UV–vis Spectrophotometer). HA and SF concentrations were determined by measuring UV absorbance at 255 nm and 280 nm, respectively. MA was analyzed based on their optical density, whereas pathogenic bacteria (VH) were analyzed based on the determination total aerobic plate count (TPC). The presence of IHHNV was analyzed using Chapter 2.2-OIE 2009 Polymerase Chain Reaction (PCR) [37].

3. Results

3.1. Adsorptive fouling (membrane–solute interactions)

3.1.1. Effect of membrane cut-off

Adsorptive fouling was investigated by exposing the outer membrane surface to a definite contaminant feed solution. The relative water flux reduction (RFR) was used to identify the extent of membrane–solute interactions. The results are presented in Fig. 2.

Fig. 2 clearly shows that the RFR was influenced by both membrane pore size and contaminant type. For most of the contaminants (cf. SF, MA, HA, VH and IHHNH), GR51PP (50 kDa) showed the highest RFR followed by GR81PP (10 kDa) and GR40PP (100 kDa), respectively. Different results were observed for RAW and SMH, where the highest RFR was shown by 10 kDa UF membrane followed by 50 kDa UF and 100 kDa UF, respectively. For all contaminants used, the 100 kDa UF showed the lowest RFR. Overall, the highest RFR was shown by SMH (> 45%) followed by RAW and SF, which demonstrated comparable RFRs (32–45%). Comparable RFRs within the range 10–20% were exhibited by HA, VH, and IHHNH.

3.1.2. Effect of contaminant concentration

In order to obtain a more comprehensive information about adsorptive fouling phenomena, the effect of contaminant concentration on RFR was investigated. For most of the contaminants used, 50 kDa UF membrane showed the highest adsorptive fouling at the first adsorptive fouling examination (Section 3.1.1). As a consequent, this membrane was not involved in the study of contaminant concentration effect on RFR (Section 3.1.2.).

The results are presented in Fig. 3. It is clearly seen that the extent of adsorptive was influenced by contaminant concentration and membrane pore size. The rapid increase in RFR by increasing contaminant concentration at low concentration (less than 1 g/L for SF, less than 0.8 g/L for HA and less than 0.2 OD for MA) followed by a plateau condition beyond these concentrations were observed for both UF membranes. The RFR of 100 kDa UF membrane was lower than the RFR of 10 kDa UF membrane for all contaminants. Furthermore, 10 kDa UF membrane required lower concentration than 100 kDa UF membrane to achieve a plateau condition.

3.2. Ultrafiltration fouling with single contaminant

UF fouling behavior of aquaculture contaminant was initially investigated by ultrafiltration of humic acid (HA) solution (25 ppm). HA was used since its presence is abundant in soil and water systems [38].

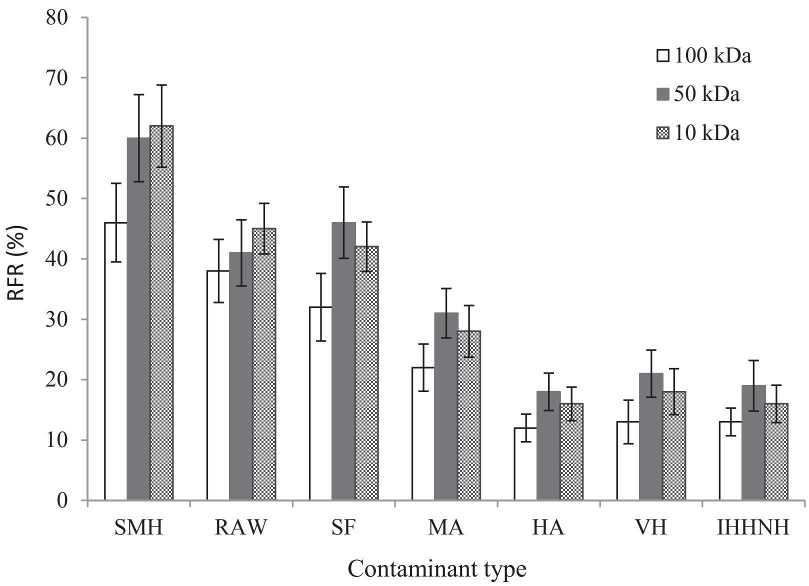
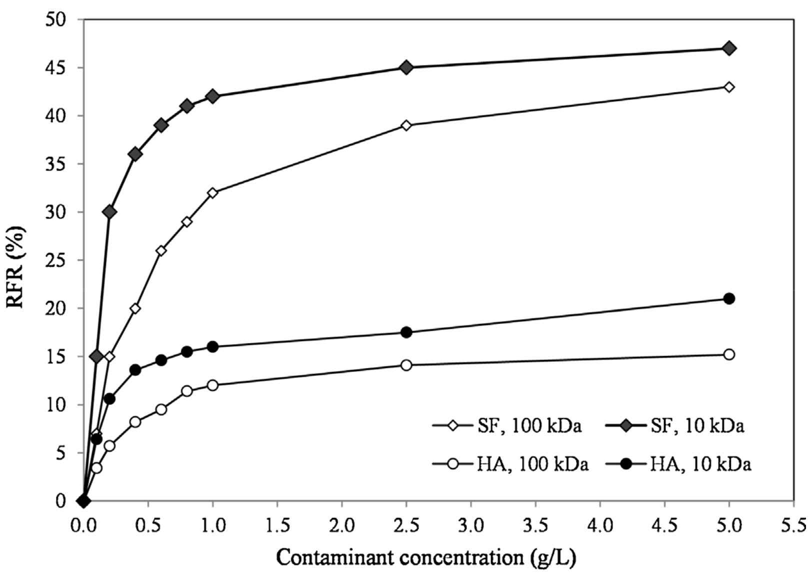


Fig. 2. Relative flux reduction (RFR) after static adsorption (3 h) to a definite contaminant feed solution (SMH (mixture of SF (0.25 g/L), MA (0.5 OD) and HA (0.25 g/L)), RAW, SF (1 g/L), MA (1 OD), HA (1 g/L), VH (10^8 CFU), IHHNH: virus (10/500 mL/mL)) of three different UF membranes (GR81PP = 10 kDa, GR51PP = 50 kDa and GR40PP = 100 kDa).



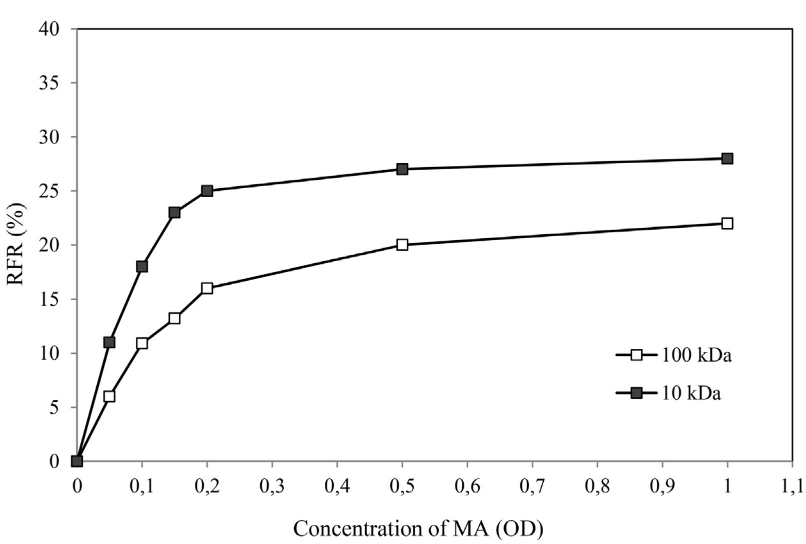


Fig. 3. Effect of contaminant concentration on RFR: SF and HA (top panel) and MA (bottom panel).

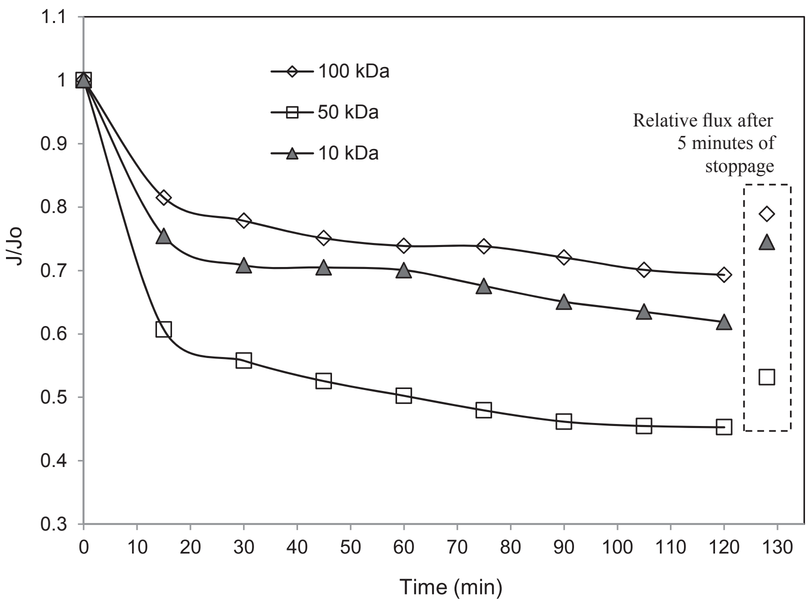


Fig. 4. Flux behavior during ultrafiltration of HA solution (0.025 g/L) using various UF membranes. The TMP was 100 kPa for 50 and 100 kDa UF membranes and 300 kPa for 10 kDa UF membrane.

The exploration of fouling mechanism of HA as the contaminant also carried out for the application of ultrafiltration in seawater [34,39]. The results are expressed in term of flux (normalized flux) as a function of filtration time, as shown in Fig. 4.

The fouling behavior using shrimp feed as a representative model for the aquaculture contaminant was also investigated. Shrimp feed was used because feed residual has been recognized as one of the most aquaculture contaminants. Fig. 5 shows the permeate flux behavior (expressed as normalized flux) for the filtration of SF solutions at a constant pressure.

Ultrafiltration of aquaculture contaminant was also examined using MA (*Spirulina* sp.). *Spirulina* sp. was selected because it can be found in many aquaculture systems and it has similar size and shape geometry with harmful algal bloom. The results are presented in Fig. 6. The performance of three UF membranes was also examined using marine bacterium (VH) and IHHNV solution. The results are presented in Figs. 7 and 8, respectively.

Ultrafiltration fouling presented in Figs. 4–8 showed that in general two different fouling behaviors were observed. The first one was rapid flux decline at the early stage of filtration and followed by relatively constant permeate flux until the filtration was stopped. This behavior was found for the filtration of SF and VH solution. The second one was rapid flux decline at the early stage of filtration followed by a gradual decrease in permeate flux approaching constant flux after a certain time of filtration. This behavior was found for the filtration of HA, MA, and IHHNV solution.

The normalized fluxes (J/Jo) at the end of HA ultrafiltration were 62%, 45% and 69% for 10, 50 and 100 kDa UF membranes, respectively. Ultrafiltration of SF solution demonstrated normalized fluxes at the end of ultrafiltration 45% for 10 kDa, 40% for 50 kDa, and 37% 100 kDa. Stable fluxes (55% for 10 kDa, 52% for 50 kDa, and 53% 100 kDa) were obtained at ultrafiltration of MA. Ultrafiltration of VH solution exhibited stable flux at 81% when 10 kDa membrane was used, 75% when 50 kDa membrane was used and 73% when 100 kDa membrane was used. Further, when IHHNV solution was used as feed, the resulted stable fluxes are 77%, 81% and 85% for 10 kDa, 50 kDa, and 100 kDa membranes, respectively. Table 1 shows rejection data of different UF membranes for different contaminants. It is shown that all UF membranes had 100% rejection for MA, VH and IHHNV. The slightly lower rejection was observed for HA, i.e., 98.5%, 98.6% and 99.8% for

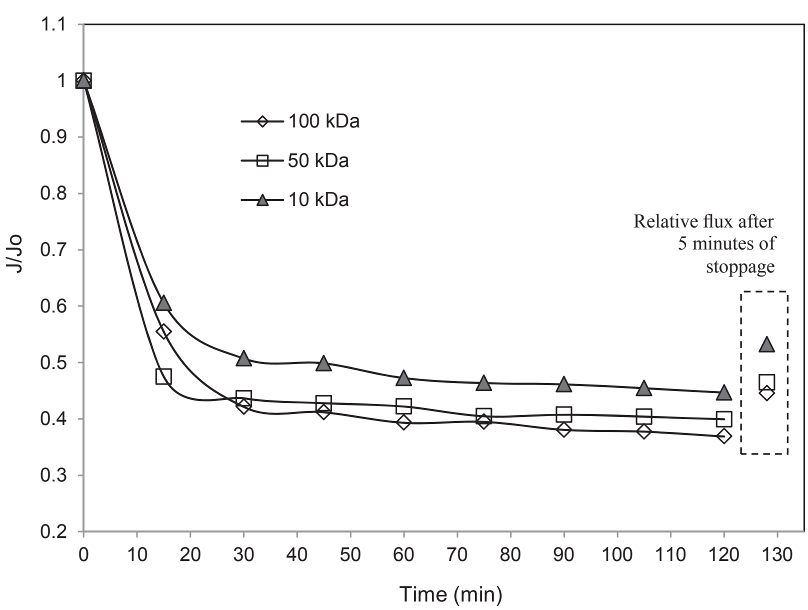


Fig. 5. Flux behavior during ultrafiltration of SF solution (0.5 g/L) using various UF membranes. The TMP was 100 kPa for 50 and 100 kDa membranes and 300 kPa for 10 kDa membrane.

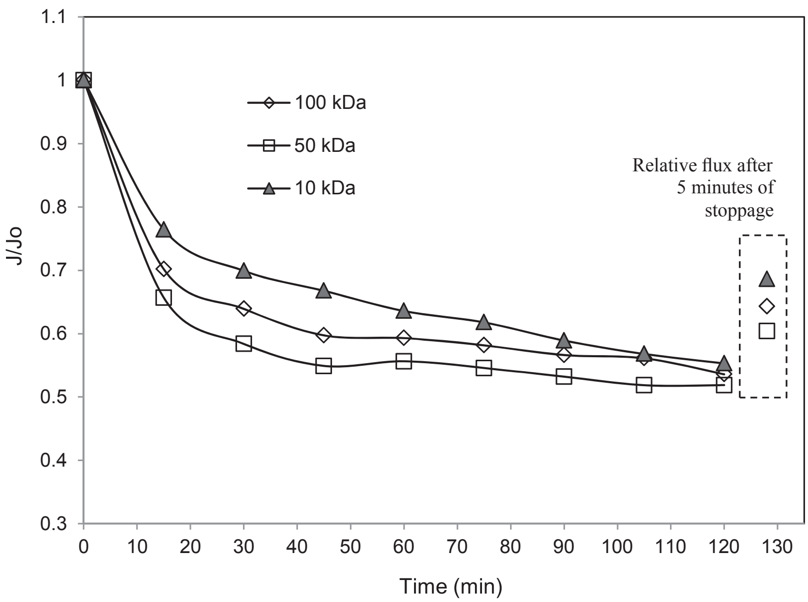


Fig. 6. Flux behavior during ultrafiltration of MA solution (0.1 D) using various UF membranes. The TMP was 100 kPa for 50 and 100 kDa membranes and 300 kPa for 10 kDa membrane.

100, 50 and 10 kDa membranes, respectively.

3.3. Ultrafiltration of mixed contaminants solution

The results of adsorption and ultrafiltration experiments using single contaminant suggest that the 100 and 10 kDa membranes showed comparable performance with respect to fouling behavior and contaminant rejection. In this part, the performances of both membranes were further evaluated. Mixed contaminants comprising of HA (0.025 g/L), SF (0.5 g/L), MA (0.1 OD), marine bacterium VH (10^6 CFU) and IHHNV (0.5/500 mL/L) were used as feed. The results are presented in Fig. 9.

Fig. 9 shows that the flux behavior observed in ultrafiltration of mixed contaminants solution was similar to the single contaminant of SF and VH, while both membranes demonstrated rapid flux decline at the early stage of filtration and followed by relatively constant permeate flux until the filtration was stopped. The stable fluxes were 20% and 37% of initial membrane flux for 10 kDa and 100 kDa membranes, respectively. The flux decline of both UF membranes was higher compared to all experiments using single contaminant. The 100 kDa membrane showed a higher normalized flux than 10 kDa membrane indicating lower flux decline. Nevertheless, both membranes showed similar rejection for all contaminants, i.e. within the range 97–100%.

In further experiments, 100 kDa UF was selected because it showed comparable normalized flux and rejection with 10 kDa membrane but it used lower pressure indicating lower energy. In addition, the permeate flux of 10 kDa membrane is relatively small for the practical application. Further examination was performed by ultrafiltration of real aquaculture water from the aquaculture system. The results are presented in Fig. 9 and Table 2.

Fig. 9 shows that rapid flux decline followed by a gradual flux decrease was observed. Although the filtration has already been performed 2 h, a gradual flux decline was still observed. The normalized flux was 56% of initial membrane flux after two hours' ultrafiltration. Compared to UF of mixed contaminants solution, ultrafiltration of real aquaculture water resulted in lower flux decline. Table 2 shows the rejection of some important parameters for aquaculture system. It is seen that the membrane had 100% rejection for IHHNV, VH and suspended solid. In addition, the rejection of turbidity, ammonia, and COD were 99.5%, 97.6%, and 86.4%, respectively. The lowest rejection was

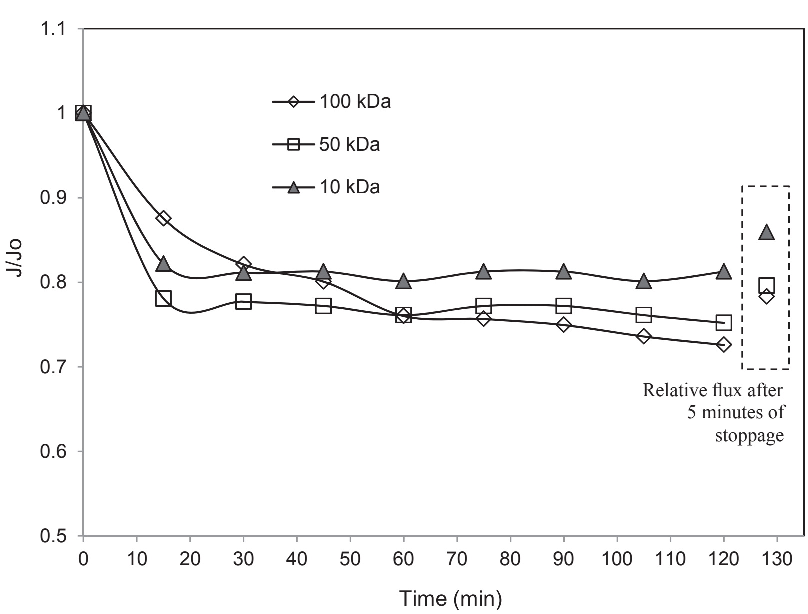


Fig. 7. Flux behavior during ultrafiltration of VH solution (10^6 CFU) using various UF membranes. The TMP 100 kPa for 50 and 100 kDa membranes and 300 kPa for 10 kDa membrane.

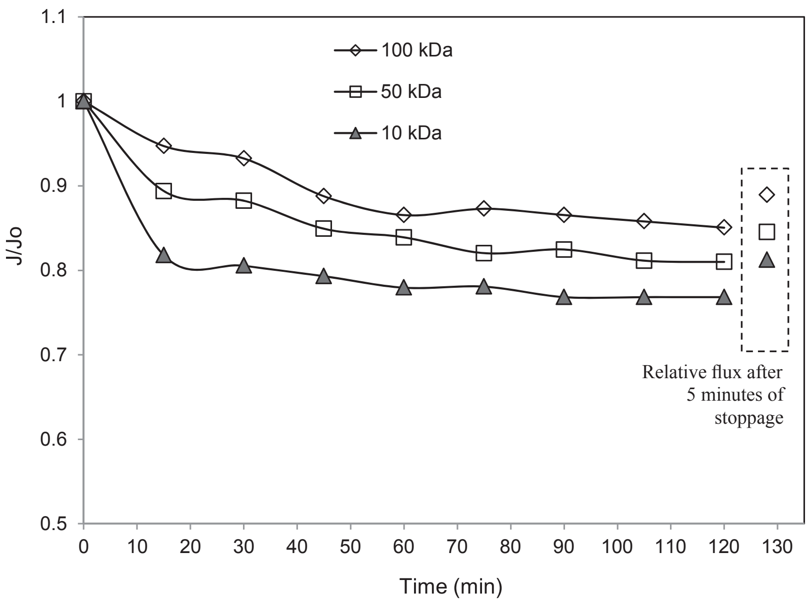


Fig. 8. Flux behavior during ultrafiltration of IHHNV solution (0.5/500 mL/mL) using various UF membranes. The TMP was 100 kPa for 50 and 100 kDa membranes and 300 kPa for 10 kDa membrane.

Table 1
Rejection of different contaminants by different UF membranes.

Nr	Contaminant	Rejection (%)		
		10 kDa	50 kDa	100 kDa
1	Humic acid (HA)	98.2 ± 2.5	92.6 ± 3.3	94.5 ± 2.4
2	Shrimp feed (SF)	100	98.3 ± 2.1	99.3 ± 1.8
3	Microalgae (*Spirulina* sp.)	100	100	100
4	*Vibrio harveyi* (VH)	100	100	100
5	IHHNV	100	100	100

observed for total phosphate, i.e. 11.1%.

3.4. Membrane characterization

Visualization of membrane surface was performed to support previous explanations. It is clearly seen that all foulants were significantly deposited on membrane surface (Fig. 10). In addition, foulant segregation on the membrane surface was also observed for all foulants.

Table 2
Feed and permeate quality of ultrafiltration of pond water using 100 kDa membrane.

No	Parameter	Unit	Real aquaculture water	Permeate	Rejection (%)
1	Turbidity	JTU	156.8	0.78	99.5
2	Ammonia	mg/l	0.042	0.001	97.6
3	Nitrite	mg/l	< 0.001	< 0.001	–
4	Nitrate	mg/l	< 0.001	< 0.001	–
5	COD	mg/l	175.6	23.9	86.4
6	*Total Phosphate*	mg/l	0.054	0.048	11.1
7	*Total Suspended Solid*	mg/l	238	1.73	~100
8	*Vibrio harveyi* (VH)	CFU/ml	1300	0	100
9	IHHNV	–	+	–	100

4. Discussion

RFR was used as an indicator to evaluate adsorptive fouling caused by the contaminant models. As can be seen from Fig. 2, the static

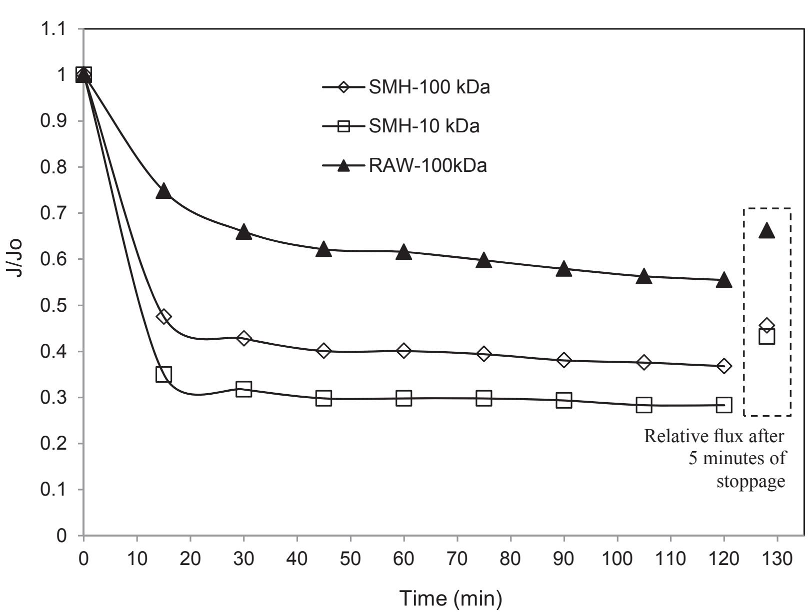


Fig. 9. Flux behavior during ultrafiltration of mixed contaminants solution and real pond water. The TMP was 100 kPa for 100 kDa and 300 kPa for 10 kDa.

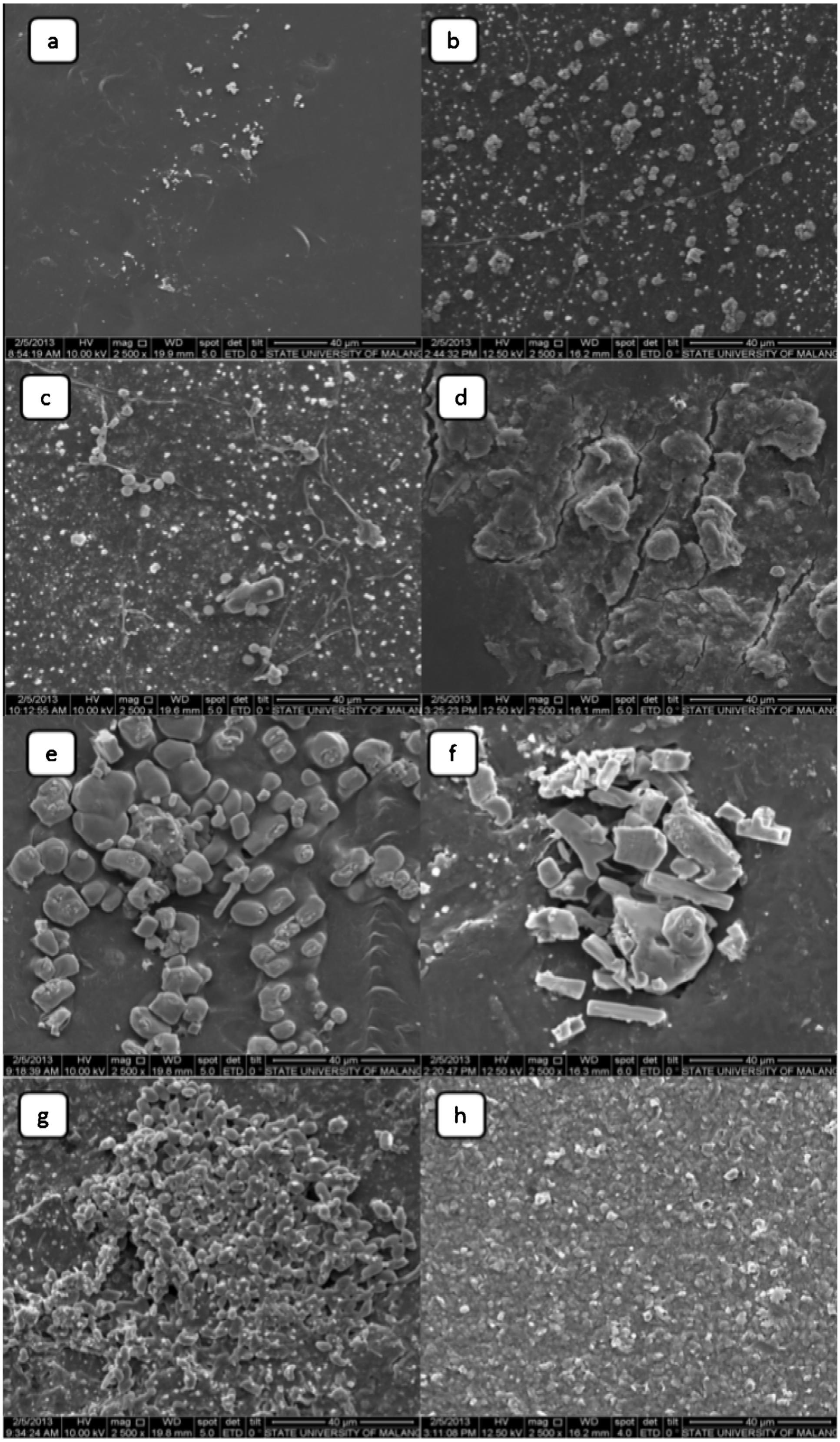


Fig. 10. Scanning electron micrographs of the upper surface of PSH 100 kDa UF membranes: (a) before used, (b) used to filter HA, (c) used to filter SF, (d) used to filter MA, (e) used to filter IHHNV, (f) used to filter VH, (g) used to filter all contaminant mixture, and (h) used to filter RAW.

adsorption of all contaminants used in this study caused membrane fouling as indicated by their RFR (the higher RFR means the higher adsorptive fouling). All contaminant models (except RAW and SMH) showed a systematic trend of adsorptive fouling, where the 100 kDa UF membrane had the lowest RFR followed by the 10 kDa and 50 kDa UF membranes, respectively. Different behavior was exhibited by a more complex feed (RAW and SMH), where the increase in pore size decreased the RFR indicating lower adsorptive fouling. The highest RFR was observed at the experiments using RAW as feed for all membranes. These results suggest that SMH was the strongest foulant in term of adsorptive fouling. Furthermore, for the single contaminant, SF was identified as the strongest foulant. The results obtained from adsorptive fouling are in agreement with the result obtained from ultrafiltration experiment, where SMH and SF are the two foulant models that generated highest flux decline.

It is clearly seen that the extent of adsorptive fouling was influenced by foulant concentration and membrane pore size. Increasing foulant concentration would rise RFR which shows more severe adsorptive fouling has occurred at higher concentration. This phenomenon was well explained in our previous publication [36]. The influence of pore size (expressed as MWCO) on adsorptive fouling can be explained by blocking model theory. If the size of foulant is smaller than the membrane pore size, the foulant can access the membrane pores causing adsorption in the membrane pores followed by a pore size constriction. When the foulant size is increased, the possible pore blocking will be higher. Complete blocking will occur when the membrane pore size is equal to the foulant size. Nevertheless, if the foulant size is further increased the possibility of pore blocking becomes lower, and a cake layer formation will be larger. It is understood that the pore blocking mechanism will result in a larger flux reduction than pore narrowing.

Difference in adsorptive fouling behavior was shown by the 10 and 50 kDa membranes, where the 50 kDa membrane showed higher adsorptive fouling than the 10 kDa membrane for all individual foulants. The 10 kDa membrane showed higher adsorptive fouling than 50 kDa membrane for mixed foulants (SMH and RAW). The possible reason for this phenomenon is foulant–foulant interactions. The interaction between or among foulants in the feed can alter the fouling behavior caused by the individual foulant.

Ultrafiltration data suggest that fouling behavior and steady permeate fluxes were influenced by both membrane cut off (membrane pore size) and contaminant type. This means the ratio of contaminant size to membrane pore size is more relevant to be used for explaining fouling phenomena rather than pore or foulant size itself. All filtration experiments showed a rapid flux decline at the beginning of filtration followed by either gradual decrease for longer filtration time and finally tended to level off after relatively long time filtration or by relatively constant permeate flux until the filtration has been finished. This difference in fouling behavior is certainly due to the difference fouling phenomenon.

The rapid flux decline at the beginning of filtration was caused by concentration polarization. In addition, fouling could also contribute to this rapid decline. Concentration polarization is the filtration phenomenon that cannot be avoided and occurs due to the build-up of foulant concentration on the membrane surface as consequence of filtration process. In order to prove the contribution of fouling, we stopped the filtration just after 5 min filtration for 10 s. The filtration was then started again and the flux could not be restored to the initial value (the flux was higher than the flux at 5th min filtration but lower than initial flux). The increase in flux after stopping the filtration suggests that concentration polarization contributed to the flux decline. Nevertheless, because their flux increases were only within the range 5–20%, fouling was the dominant reason for the flux decline.

For the case, where rapid flux decline at the early stage of filtration followed by relatively constant permeate flux, fouling by pore blocking (tends to complete blocking) should be another reason for the sharp flux decline beside concentration polarization. It is important to point out that the adsorption should not be the reason for this rapid decline at the early stage because adsorption fouling requires a certain time to occur and relatively high concentration (Fig. 3). Our experiments showed that it takes at least 30 min for adsorption (data not shown). Thereafter, both adsorption and foulant deposition on the membrane surface leading to cake layer formation took place. This cake layer formation is evidenced by the results of membrane surface visualization using SEM (Fig. 10). Nevertheless, it is believed that this cake layer did not influence the resulted permeate flux as indicated by constant flux after rapid flux decline. In this case, porous cake layers should be formed.

For the case where rapid flux decline at the early stage of filtration followed by a gradual decrease in permeate flux, fouling by pore narrowing should be the reason for this rapid declining beside concentration polarization. Then, adsorption fouling and cake layer formation occurred. In this case, adsorption fouling and cake layer influenced the resulted permeate flux as indicated by a gradual flux decrease. Furthermore, it is reasonable to explain that a stable external membrane fouling seems to be the most dominant fouling. Deposit of multilayers foulant on the membrane was also clearly observed by SEM (Fig. 10). A similar phenomenon was explained earlier by Saidi et al. [40].

Experiments using single foulant solution demonstrated that 50 kDa membrane showed the highest fouling as indicated by its smallest normalized flux (J/Jo). Beside the pore morphology, the higher initial flux of 50 kDa membrane could cause higher CP that will promote a higher possibility of irreversible fouling formation. Furthermore, for the same UF membrane used, SF and MA were identified as two strongest contaminants leading to highest flux decline. By contrast, VH and IHHNV were identified as the two weakest contaminants leading to lowest flux decline. For the same contaminant, 50 kDa UF membrane generally showed the highest flux reduction among other types of used membranes. The 100 and 10 kDa membranes showed comparable flux decline. In addition, PSH 100 kDa was effective in removing contaminants in aquaculture systems to meet the standard in aquaculture systems. For the same contaminant, the membrane with larger pores (PES 50 kDa) is prone to internal fouling than the membrane with smaller pores (10 kDa). Definitely, this explanation is not proper for the membrane with very large pore size compared to contaminant size. This phenomenon is in agreement with the study by Rickman et al. [41].

Considering the permeate flux and rejection data obtained from the experiments using single contaminant, 100 kDa and 10 kDa membranes were further examined for ultrafiltration of mixed contaminants. Both 100 and 10 kDa membranes demonstrated fouling behavior, where rapid flux decline at the early stage of filtration followed by relatively constant permeate flux. The reason used to explain fouling behavior for single foulant solution is still valid to be used here. The results also indicated that 100 kDa membrane was less prone to fouling than 10 kDa membrane as indicated by its higher J/Jo (37% vs 28%). Further examination using real aquaculture water was conducted using 100 kDa membrane. Rapid flux decline at the early stage of filtration followed by a gradual decrease in permeate flux was observed. The 100 kDa UF membrane exhibited very high rejection for all components in real aquaculture water except total phosphate (only 11.1%). However, the concentration of total phosphate in permeate is still lower compared to aquaculture standard.

5. Conclusions

Study of the effects of membrane pore size (expressed as MWCO) and foulant type on fouling behavior and foulant rejection has been performed using three different commercial UF membranes and five different foulant models. The experimental results showed that fouling behavior was influenced by both membrane cut off (pore size) and foulant type. Two fouling behaviors were observed, i.e. (i) rapid flux decline at the early stage of filtration followed by relatively constant permeate flux until the experiment finished, and (ii) rapid flux decline at the early stage of filtration followed by a gradual decrease in permeate flux. Concentration polarization and fouling by pore blocking are believed to be the reason for the sharp flux decline for fouling behavior (i). Furthermore, both adsorption and foulant deposition on the membrane surface leading to cake layer formation occurred. Nevertheless, it is supposed this cake layer did not influence the resulted permeate flux. Concentration polarization and fouling by pore narrowing were the reason for the sharp flux decline of fouling behavior (ii). Further, adsorption fouling and foulant deposition on the both membrane surface and the membrane pore surface leading to cake layer formation occurred, then finally decreases permeate flux. Thus, it is reasonable to state that a stable external membrane fouling seems to be the most dominant fouling mechanism for fouling behavior (ii). In general, 100 kDa membrane showed the lowest fouling among all membranes examined. It is worth noting that all membranes had similar rejections for all contaminant models. Therefore, based on the basis of permeate flux behavior and rejection data, the 100 kDa UF membrane appeared to be the most promising ones among other membranes examined for RAS application.

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