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Desalination 202 (2006) 239–246

DESALINATION

www.elsevier.com/locate/desal

Application of ultrafiltration hybrid membrane processes for reuse of secondary effluent

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Received 31 July 2005; accepted 23 December 2005

Abstract

This study evaluates the factors affecting pretreatment conditions for hybrid UF membrane processes for reuse of secondary effluent from the sewage treatment plant. The experimental results obtained from the ultrafiltration (UF) membrane process showed that the particles of the size range between 0.2 and 1.2 μm caused a significant impact on membrane fouling in all cases even with or without the coagulation process. As pretreatment of UF membrane process, the coagulation significantly improved the permeate flux. Optimal flux improvement was seen at an alum dose of 50 mg/L. In addition, it was found that the permeate flux was least declined under the coagulation condition of charge neutralization (pH 5.0). Also, the powdered activated carbon (PAC) adsorption enhanced the permeate flux. Application of the direct filtration as a pretreatment of UF membrane process was also very effective in reducing the UF membrane fouling.

Keywords: Secondary effluent; Reuse; UF membrane; Coagulation; PAC; Direct filtration

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1. Introduction

Most of the research in the past on recycling secondary effluent has been involved with activated carbon adsorption alone or in combination with ozonation or sand filtration [1]. However, in recent decades, membrane filtration has emerged as a novel technology in wastewater reclamation. The application of membrane filtration in reuse of the municipal wastewater effluent will steadily increase with more stringent discharge regulations and supply limitations [2]. Especially, the ultrafiltration (UF) membrane process is considered a cost-effective option in terms of higher permeate flux compared to nanofiltration and reverse osmosis [3]. However, the membrane

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Presented at the conference on Wastewater Reclamation and Reuse for Sustainability (WWRS2005), November 8–11, 2005, Jeju, Korea. Organized by the International Water Association (IWA) and the Gwangju Institute of Science and Technology (GIST).

doi:10.1016/j.desal.0000.00.000

can be easily fouled by the effluent organic matter present at high levels in wastewater [4]. Thus, a pretreatment process, such as coagulation, powdered activated carbon (PAC) adsorption and direct filtration processes, is necessary to decrease the fouling on the UF membrane and at the same time increase the organic removal efficiency for secondary effluent reclamation and reuse. Many studies have addressed the combined treatment of coagulation and UF in water treatment, reporting that the combined coagulation–UF membrane process tends to reduce the colloidal membrane fouling and also to improve the removal of dissolved organic matter (DOM). Also, it has been reported that PAC is more effective than coagulation in removing DOM. Although the removal of DOM is enhanced by the addition of PAC during the membrane process, the presence of PAC increases the fouling problem and the flux decline as reported by some researchers [5–7]. However, other studies found that a decrease of the membrane permeability was slower in a PAC-MF system, which may be resulted from the reduction of the organic loading to the membrane. Also Tomaszewska et al. [8] have reported that coagulation without PAC addition is less effective than the adsorption–coagulation integrated system. The aim of this study is to determine the influence of pretreatment (coagulation, PAC addition, direct filtration) on the hybrid UF membrane process for the reuse of secondary effluent from the sewage treatment plant.

2. Materials and methods

2.1. Characteristics of secondary effluent

The secondary effluent from the Busan N sewage treatment plant was collected and used as feed water. The physical and chemical characteristics of the secondary effluent are shown in Table 1. Turbidity, TOC, COD_{Cr}, coliform and color in the secondary effluent were in the ranges of 0.9–1.5 NTU, 3.3–5.2 mg/L, 25–32 mg/L, 300–700 Numbers/mL and 18–24 CU, respectively.

Table 1
Characteristics of secondary effluent from the Busan N sewage treatment plant

Parameters	Units	Values
pH	–	7.1–7.6
Turbidity	NTU	0.9–1.5
TOC	mg/L	3.3–5.2
UV_{254}	cm^{-1}	0.09–0.12
SUVA	m^{-1}/mg DOC	2.30–2.72
COD_{Cr}	mg/L	25–32
Color	CU(Pt-Co)	18–24
Coliforms	Number/mL	300–700

2.2. UF membrane process

Membrane filtration experiments were conducted in a 76 mm diameter unstirred cell (Millipore, USA) connected to a N_2-pressurized 5 L solution reservoir as shown in Fig. 1. The permeate flux was measured as a function of time at a constant pressure (fixed at 1.0 bar), and the filtrate mass was determined using a personal computer connected to an analytical electronic top-loading balance.

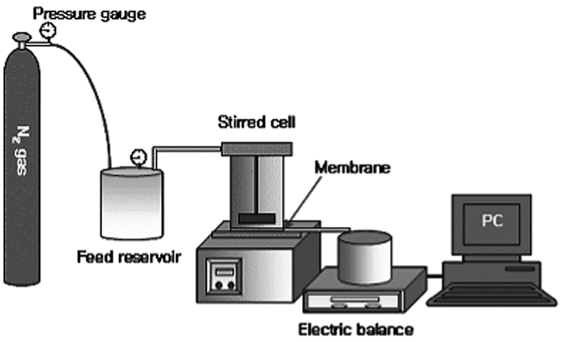

Fig. 1. Schematic of batch type UF membrane filtration assembly.

Table 2
Characteristics of powdered activated carbon (Shinki Chemical, Korea)

Iodine number (mg/g)	Moisture as packed (%)	Ash content (%)	Passing 200 mesh (%)
1090	4	1.7	96.8

2.3. Coagulant, PAC and membrane

Alum ($Al_2(SO_4)_3 \cdot 16H_2O$) was used as a coagulant. The concentration of alum stock solution was 0.25 N as alum, and the dosing solution was 10 g/L as alum which was diluted at each coagulation experiment. PAC was obtained from Shinki Chemical (Korea). The physical and chemical properties of PAC are shown in Table 2. PAC slurry was added to 2 L reactor and mixed with the feed for 30 min, then transferred into a 5 L pressure reservoir. The UF membrane used in this study is the regenerated cellulose (Millipore, USA), with a molecular weight cut-off (MWCO) of 100 kDa and a geometric area of 0.00454 m^2. The membrane material is classified as hydrophilic. The membrane surface was cleaned with deionized water, followed by soaking in 0.1 N NaOH solution for 30 min before each experiment.

2.4. DOM fractionation

An aliquot of DOM was fractionated into humic and nonhumic fractions by employing a technique based on adsorption of humic substances onto XAD-8 resin under acidic condition (pH 2). The organics adsorbed on resin were subsequently eluted with NaOH solution (pH 12) [9,10].

3. Results and discussion

3.1. Characteristics of DOC fraction of secondary effluent

Fig. 2 shows the DOC fraction and apparent molecular weight (AMW) distribution of DOC in the secondary effluent. The DOC fractions of secondary effluent used in this study were hydrophilic acid, fulvic acid and humic acid with a fraction 64, 31 and 6%, respectively. The result of AMW distribution of DOC indicates that most DOC have a molecular weight smaller than MWCO of 10 kDa, which suggests that without a signification pretreatment, most organics in secondary effluent should pass through the UF membrane having a pore size of 100 kDa MWCO.

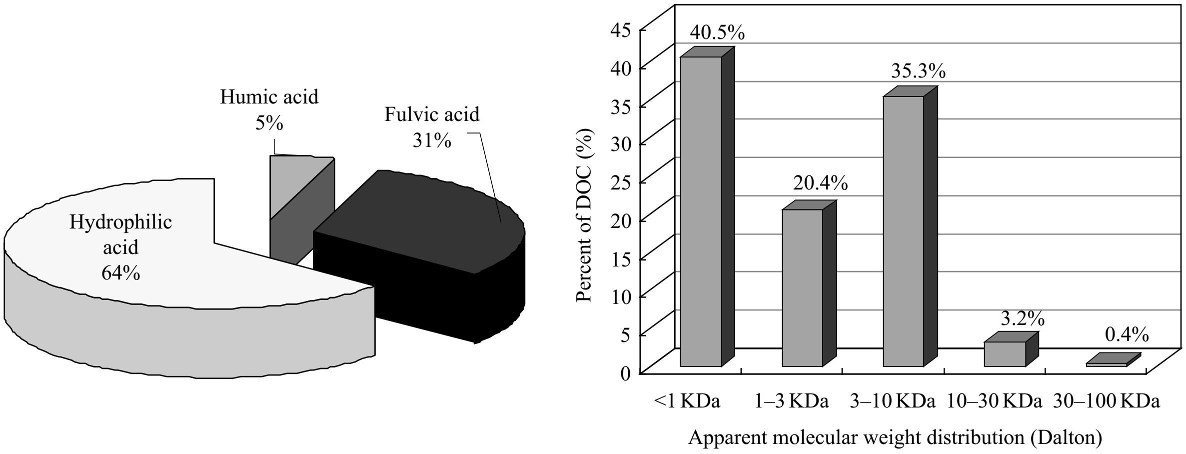

Fig. 2. DOC fraction and AMW distribution of DOC in secondary effluent.

3.2. Effect of particulate matter on membrane fouling

Secondary effluents contain a wide variety of suspended and colloidal particles that cause changes in color and turbidity [11]. Therefore, we tried to find out the effect of organic floc and colloidal matter on the membrane fouling by examining the effect of the prefiltration and coagulation process on the changes in permeate flux. Fig. 3 shows the membrane filtration result of either secondary effluent alone or secondary effluent after filtration with 1.2 and 0.2 μm filter. As shown in the figure, the difference of the permeate flux barely appeared in the case of filtration with or without the 1.2 μm filter. However, in the condition of filtration with 0.2 μm filter, the permeate flux difference appeared. This result indicates that the relatively large particulate matters sized over 1.2 μm do not cause the fouling of the UF membrane used.

Fig. 4 shows the membrane filtration result with either secondary effluent after rapid mixing (50 mg/L as alum), or secondary effluent after filtration with 0.2 μm filter followed by rapid mixing. As shown in the figure, the permeate flux improved under various pretreatment conditions: 13.0% when pretreatment was made with 0.2 μm filter, 24.4% when an alum dosage of 50 mg/L was applied for rapid mixing and 40% when an alum dosage of 50 mg/L was applied for rapid mixing after filtration with 0.2 μm filter. These results indicate that the particles over the size of 0.2 μm caused a significant impact on membrane fouling in all cases even with or without the coagulation. In addition, as shown in Table 3, when the UF membrane process was applied with prefiltered effluent, the UV_{254} and DOC were removed only 1.0% compared to the application of UF alone, while the permeate flux increased by 13.0%. Besides, the permeate flux for the prefiltered and coagulated water increased by 40.0% compared to the application of UF alone. Lahoussine-Turcaud et al. [12] suggested

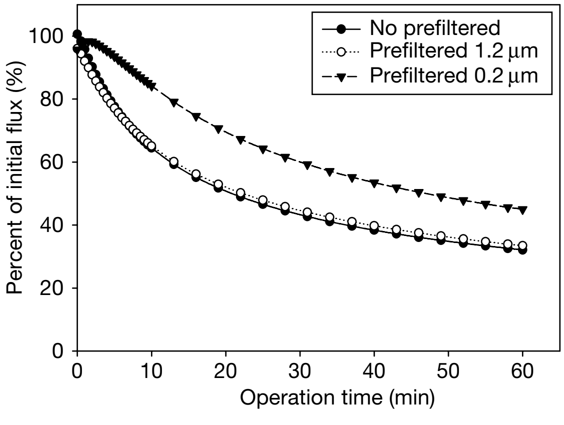

Fig. 3. Flux decline of secondary effluent after prefiltering through 1.2 and 0.2 μm filter.

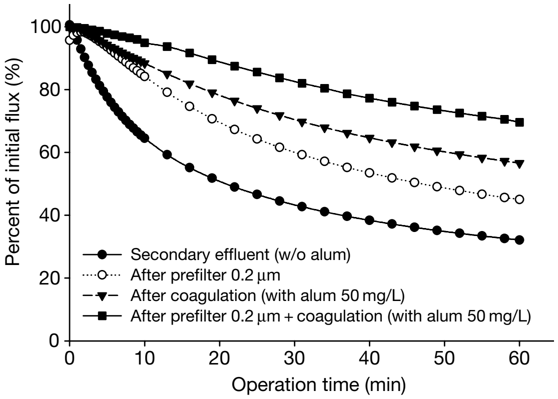

Fig. 4. Changes in the flux of UF membrane under various pretreatment conditions.

Table 3
Percent of initial flux and removal of UV_{254} and DOC after each pretreatment

Pretreatment conditions	Percent of initial flux (%)	Removal (%)	
		UV_{254}	DOC
UF alone	32.1	9.3	6.67
0.2 μm prefilter + UF	45	10.4	7.1
Alum 50 mg/L + UF	56.5	33.7	22.2
0.2 μm prefilter + alum 50 mg/L + UF	70	38.3	28.8

that the flux characteristics of coagulated water can be explained in terms of the back-transport velocity of particle; particles near 0.2 µm in diameter produce rapid fouling, while particles greater than 3.0 µm in size have little effect on flux.

3.3. Effect of coagulant dose and coagulation pH on membrane fouling

Fig. 5 shows the effects of the coagulant dose on the changes in the permeate flux. The permeate flux was measured with the rapid mixed sample under the mixing intensity of $G = 550\ s^{-1}$ for 1 min. As shown in the figure, the coagulation pretreatment significantly improved the permeate flux. Optimal flux improvement is seen at an alum dose of 50 mg/L. Higher alum dose (70 mg/L), however, have a detrimental effect on the optimum permeate flux. Some researchers also concluded that higher coagulant doses produced sticky aluminum micro-floc, which formed a cake layer on the membrane [13].

Fig. 6 shows the effect of coagulation pH on the changes in permeate flux. The coagulation pH was adjusted to 5.0 and 7.0 which correspond to an optimum charge neutralization and optimum sweep floc coagulation condition, respectively. As shown in the figure, the permeate flux was least declined under the condition of charge neutralization (pH 5.0). However, the effect of coagulation pH appears to have a less dramatic effect on flux than coagulant dose. Lee et al. [14] have also reported that when deadend, constant-pressure, decline-flux filtration was performed, fouling was reduced (compared to raw water) under coagulation conditions of charge neutralization and fouling increased under the sweep-floc conditions.

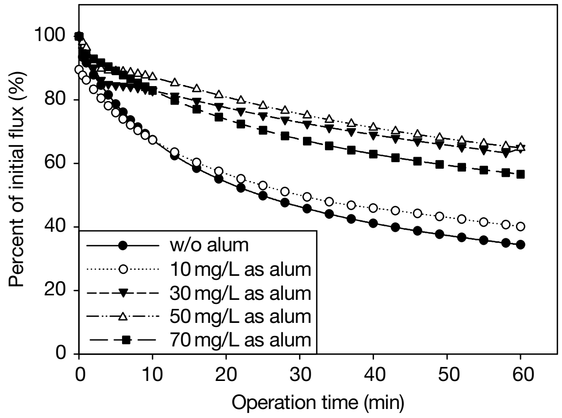

Fig. 5. Changes in flux of UF membrane under various coagulant doses.

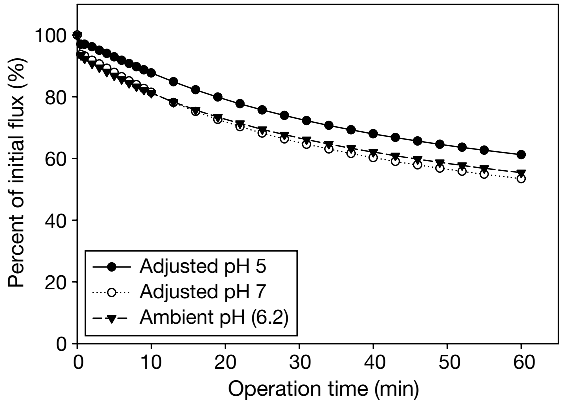

Fig. 6. Effect of coagulant pH on the changes in flux of UF membrane (alum dose: 50 mg/L).

3.4. Effect of PAC dose on permeate flux

Fig. 7 shows the changes in permeate flux obtained during the UF filtration of PAC suspension at doses of 50, 100 and 150 mg/L, and it also shows the changes in the permeate flux after backwashing in different PAC doses. It can be seen that PAC addition leads to less reduction in the permeate flux. In the case of filtering the secondary effluent without the addition of PAC, the permeate flux was reduced more than 50% of initial flux at 30 min filtration time, and the PAC dose of 100 mg/L showed the lowest flux reduction, followed by 50 and 150 mg/L. The flux reduction was smaller at 100 mg/L than the dose of PAC 50 mg/L because the DOM that cause the membrane fouling was removed to a greater extent by absorption on PAC. At a PAC dose of

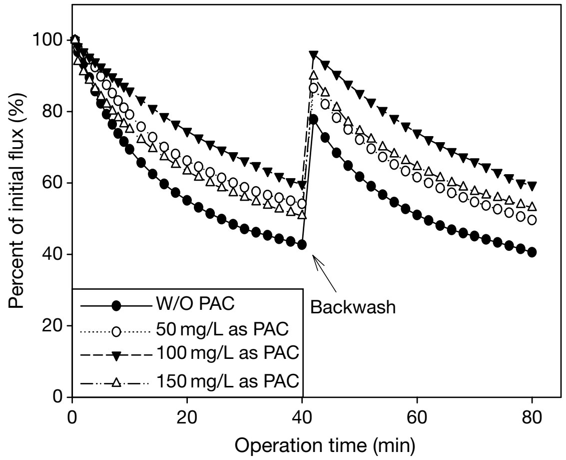

Fig. 7. Flux decline and recovery after physical backwashing on hybrid UF process.

150 mg/L, the thicker PAC cake layer formed on the membrane surface may result in greater filtration resistance and then result in a decrease in the permeate flux. In addition, as shown in Fig. 7, the flux recovery following the physical backwashing was around 80% for the UF alone process, but the recovery over 90% was obtained with the addition of PAC.

Fig. 8 indicates the percentage of initial permeate flux after 30 min of UF membrane filtration under various pretreatment conditions. Overall, the reduction in permeate flux varied between 25 and 55% depending on pretreatment conditions. It can be seen that the final flux obtained from coagulation pretreatment was relatively higher than those from PAC pretreatment. That is, the DOM causing membrane fouling was preferentially removed by coagulation rather than PAC adsorption.

3.5. Effect of direct filtration on permeate flux

Fig. 9 describes the change of permeate flux following the addition of PAC after direct filtration. The direct filtration process was implemented with the rapid mixing of the secondary effluent with 30 mg/L of alum, followed by filtration through the anthracite–sand dual media filter at a filtration rate of 180 m/day. Fig. 9 shows that the direct filtration leads to a much higher permeate flux, regardless of PAC dose. Thus, the addition of PAC after direct filtration does not contribute to more reduction of UF fouling. This illustrates that the colloidal and organic matters mainly causing membrane fouling were substantially removed through the direct filtration process. In addition, the change in flux decline

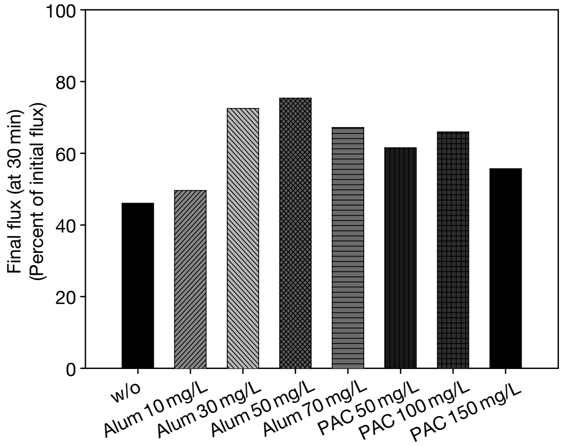

Fig. 8. Final flux for various pretreatment conditions with hybrid UF processes.

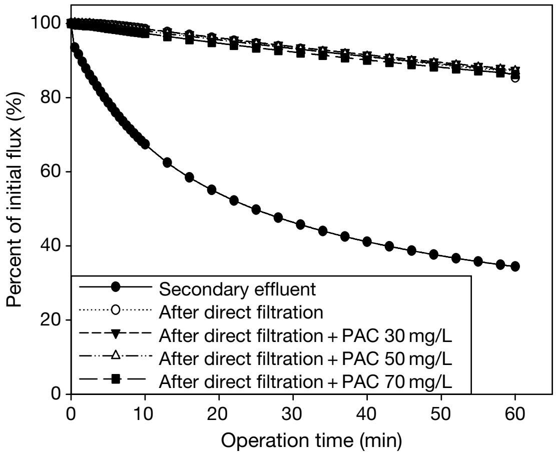

Fig. 9. Effect of PAC dose on permeate flux after direct filtration (alum dose: 30 mg/L).

between raw and pretreated water represents a change in fouling potential, where the fouling potential is reduced if less flux decline is observed. To compare the flux decline from different operation conditions, the *Reduction in Fouling Potential* can be calculated with the following equation [15]:

$$\Phi = \frac{(J_C - J_R)}{(100 - J_R)} \times 100 \qquad (1)$$

where Φ = reduction in fouling potential (percent); J_R = final flux with raw water (percent of initial permeability); J_C = final flux with pretreated water (percent of initial permeability).

Fig. 10 presents the reduction of fouling potential by coagulation, PAC addition and direct filtration calculated from Eq. (1). Significant differences in the performance of UF hybrid membrane processes can be observed between each pretreatment method. It can be seen that the reduction in fouling potential obtained from direct filtration was greatly higher than those from either coagulation or PAC addition alone as a pretreatment. Thus, the application of direct filtration as a pretreatment of the UF membrane process was also very effective in reducing the UF membrane fouling.

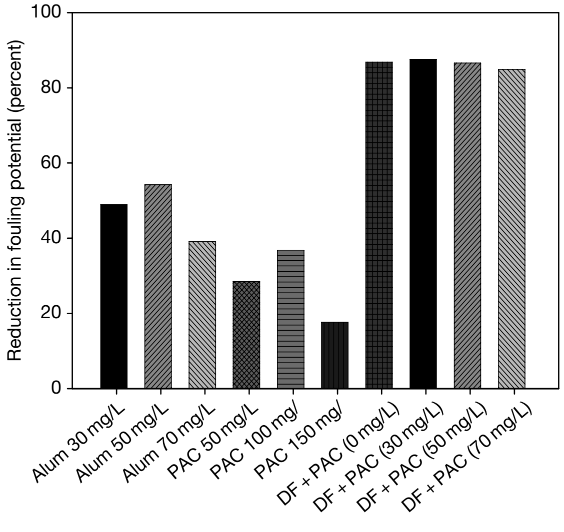

Fig. 10. Reduction in fouling potential under various pretreatment conditions with hybrid UF processes.

4. Conclusion

In this study, we found that the particles of the size range of 0.2–1.2 μm caused a significant impact on membrane fouling in all cases with or without the coagulation. When coagulation pretreatment was used as a UF hybrid membrane process, an alum dose of 50 mg/L presented the least reduction in membrane fouling. The result of the permeate flux on coagulation pH indicated that pH 5.0 showed a flux decline lower than other pH conditions. These results led us to conclude that the permeate flux was least declined under the condition of charge neutralization (pH 5.0). When PAC addition as a pretreatment was used as a UF hybrid membrane process, the PAC dose of 100 mg/L showed the lowest flux decline, followed by 50 and 150 mg/L.

In addition, applying the direct filtration as a pretreatment of UF membrane process was very effective in reducing the UF membrane fouling. Thus, the coagulation process alone or the direct filtration can be retrofitted to a pretreatment of UF membrane process. Therefore, these hybrid UF membrane processes can be applied for direct reuse of secondary effluent as a type of urban reuse, agricultural reuse, landscape and industrial reuse or as a pretreatment before a second step of a reverse osmosis membrane process.

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