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Effect of various pretreatments on the performance of nanofiltration for wastewater reuse

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ABSTRACT

In the present study, we focused on integrated processes by membrane bioreactor (MBR), SMR, and pressurized microfiltration (PMF) as a pretreatment with nanofiltration (NF) as a post-treatment for wastewater reuse. The purpose of this study is to treat wastewater through MBR, MBR with powdered activated carbon (PAC), submerged microfiltration (SMF), SMF with PAC, and PMF effluent, which is mainly used in wastewater treatment, in combination with NF as a post-treatment process. MBR with PAC was used as the main pretreatment for NF, and was compared to NF combined with other pretreatments. At constant pressure in NF, MBR-NF with PAC system has higher flux and better water quality than other systems. In order to prevent the membrane fouling by organic matters, the integrated MBR with PAC process should be employed. The integrated MBR-NF with PAC process can improve water quality for wastewater reuse. Based on the wastewater reuse standards, effluent can be available for human application. Advanced treatment of wastewater by MBR-NF system with PAC was proven to be viable. This study provides understanding on treating the wastewater through advanced technology for wastewater reuse.

Keywords: Hybrid process; Membrane bioreactor-nanofiltration; Powdered activated carbon; Pretreatment; Wastewater reuse

1. Introduction

In recent years, due to rapid industrialization and urbanization, water pollution has deteriorated. With an increase in water usage, the method of reusing treated water receives attention using membrane filtration as there is an increasing interest in the reuse of water treated as an alternative water resource, and with stricter standards of required water quality, membrane filtration technology has been in the limelight as a solution in the field of reuse. The reuse of treated water has a huge potential as such an alternative resource [1,2]. Usually, 50% of treated water is used as domestic water, while the other 50% for industry and agriculture [3,4].

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Recently, many areas chose a method of collecting sewage and wastewater for use as irrigation water and industrial water. Of them, as sewage and water reuse facilities based on reverse osmosis (RO), a plant with a capacity about 30,000 m^3/d was installed in Singapore and a facility with a capacity of about 312,000 m^3/d in Kuwait, respectively. Like this, high-level treatment of urban sewage and wastewater may be an alternative that can supply high-quality water [5].

The membrane filtration process, typically, is divided into microfiltration (MF), ultra filtration (UF), nanofiltration (NF), and RO by the operating pressure. Treatment processes such as MF and UF can overcome the limitations of the existing water treatment processes [6].

The reason why the membrane filtration process increases in the field of water treatment is that it has advantages as follows: (1) it is easier to maintain compared to the existing treatment methods; (2) it provides greater confidence in technology; (3) it can reduce the use of coagulant; (4) it uses less area compared to the existing methods; (5) it can perform subsequent separation; (6) it consumes relatively less energy; and (7) it is easy to combine with other processes [7]. Despite these advantages, the membrane filtration process has demerits as well: concentration polarization, short life expectancy and low selectivity of the membrane, and reduction of water permeation flux due to membrane fouling. Also, it has to get physical cleaning and chemical cleansing periodically due to the decrease in the water permeation flux [8].

In the membrane filtration process in the field of water purification process, MF/UF membranes have been usually used, but in the field of high-level processing and reuse, NF and low-pressure RO membranes tend to expand. In particular, since it uses raw water with very low osmotic pressure compared to the sea level, it can operate at a lower pressure than the RO membrane applied to the existing water conversion, so application expands with the merits of high flux and low energy consumption. These membrane separation characteristics are affected by a variety of operating variables such as temperature, pressure, conductivity, total dissolved solid (TDS), dissolved organic carbon, and pH and the raw water's properties [9].

The existing RO process has too high operating pressure, so it has membrane fouling and economic problem. In addition, an RO membrane has relatively more membrane fouling than a NF membrane. The NF membrane is usually used for wastewater disposal and drinking water treatment field and used as a pretreatment process step for an RO membrane separation system of seawater desalination as well as underground water and ground-surface water treatment [10]. Also, it has an ability to separate low molecular organic materials with molecular weight cut-off of about 200–500 Da, but it is difficult to separate them by the UF membrane and has an ability to separate ions (anions) with a multivalent atomic value [11]. The NF membrane can get as high flux as that of UF membrane even under an operating pressure much lower than the operating pressure of the RO membrane, usually used for ion removal, so it is economical as compared to the RO membrane [12]. However, the NF membrane can operate at a low pressure, while it is not easy to meet the water quality standard higher than the RO membrane.

This study aims to solve this problem by having a pretreatment process. For the treatment, membrane bioreactor (MBR), MBR with powdered activated carbon (PAC), submerged microfiltration (SMF), SMF with PAC, and pressurized microfiltration (PMF) are used, and a NF process is used at the rear of the treated outflow water to judge the possibility of the reuse of the treated wastewater.

2. Materials and methods

Characteristics of operating conditions and membranes as pretreatment, NF process was shown in Table 1.

MBR process used in this study consisted first anoxic, second anoxic, anaerobic, and aerobic reactor which was used in filtration chamber with MF membranes composed of two PVDF flat-sheet membranes [13]. Flat-sheet polyamide (PA) NF membranes with an effective area of 0.006 m^2 was used in the study. Advantages of current PA membranes are considered to be its resistance to various ranges of pH [5].

MBR, SMF, and PMF pretreatment processes were constantly fed with an influent flow rate of 0.015 m^3/d. MBR, MBR with PAC, SMF, SMF with PAC, and PMF effluent, which is mainly used in wastewater treatment, in combination with NF process. In order to compare the individual performances of various pretreatment processes in combination with NF process, same pressure was employed. The effluent of pretreatment to NF process is introduced by single-phase induction motor and separated into permeate and brine.

In the present study, the operating conditions of MBR were summarized in Table 2. The schematic diagram of pretreatment and NF process was shown in Fig. 1 [14].

Synthetic wastewater was used as feed in pretreatment with components based on wastewater treatment plant of Gyeonggi-do, South Korea [13]. Characteristics of synthetic wastewater are shown in Table 3.

Table 1
Characteristics of operating conditions and membranes

MBR				SMF			
Membrane		Conditions		Membrane		Conditions	
Type	Flat-sheet	Flow rate	0.015 m^3/d	Type	Flat-sheet	Filtration type	Dead-end
Material Pore size Membrane area	PVDF 0.08 μm 0.039 m^2	PAC Conc.	4,000 mg/L	Material Pore size Membrane area	PVDF 0.08 μm 0.039 m^2	Flow rate PAC Conc. Volume of reactor	0.015 m^3/d 4,000 mg/L 5.5 L
PMF				**NF**			
Membrane		Conditions		Membrane		Conditions	
Type	Hollow-fiber	Filtration type	Dead-end	Type	Flat-sheet	Filtration type	Cross-flow
Material Pore size Membrane area	PVDF 0.038 μm 0.039 m^2	Flow rate	0.015 m^3/d	Material Salt rejection Membrane area	PA 55% 0.006 m^2	Pressure Recovery rate	0.6 MPa 80%

Temperature of feed water was maintained at 20℃ using a water bath. The test equipment was washed with DI water for one hour when finishing each test [9]. Various parameters such as chemical oxygen demand (COD), pH, total nitrogen (T-N), total phosphorus (T-P), TDS, and turbidity were measured for feed water and permeate samples.

Equations for rejection [3], flux are as follows [16]:

$$R\,(\%) = (1 - C_p/C_0) \times 100 \tag{1}$$

$$\text{Flux}(\text{LMH}, \text{L/h/m}^2) = (Q/A) \times (\mu_T/\mu_{25}) \times (\Delta P/\text{TMP}) \tag{2}$$

where C_p is the concentration of the permeate, C_0 is the concentration of the feed, Q is the filtration flow rate, A is the effective area of the membrane, μ_T is the viscosity at actual temperature, μ_{25} is the viscosity at 25℃, and ΔP is the operating pressure.

Table 2
Operating conditions of the MBR system

Parameter	Set value
Total volume, L	12.13
Small anoxic, L	0.94
Medium anoxic, L	2.44
Large anaerobic, L	3.56
Oxic [membrane], L	5.19
Total HRT, h	16.83
Small anoxic, h	1.30
Medium anoxic, h	3.39
Large anaerobic, h	4.94
Oxic [membrane], h	7.20
Aeration intensity, L/min	3
Flow direction: Outside → In	
Return sludge (% Influent)	
Small anoxic reactor, %	100
Medium anoxic reactor, %	100
SRT, d	50
Time: On/OFF	9:1

3. Results and discussion

In Table 4, the performance of MBR process with PAC is the most efficient as compared to others, obtaining the highest effluent quality. MBR process coupled with PAC has been increasingly studied as an advanced treatment process due to the activated carbon's nature to remove soluble organic contaminants by adsorption [17]. Addition of PAC in MBR helped to increase the quality of effluent through adsorption. In adsorption technologies, PAC is the most commonly used adsorbents for dissolved organic matter removal, and the corresponding adsorption process is mature, simple, and cost-effective for wastewater reclamation [18]. The previous research showed that the addition of PAC could provide better physical

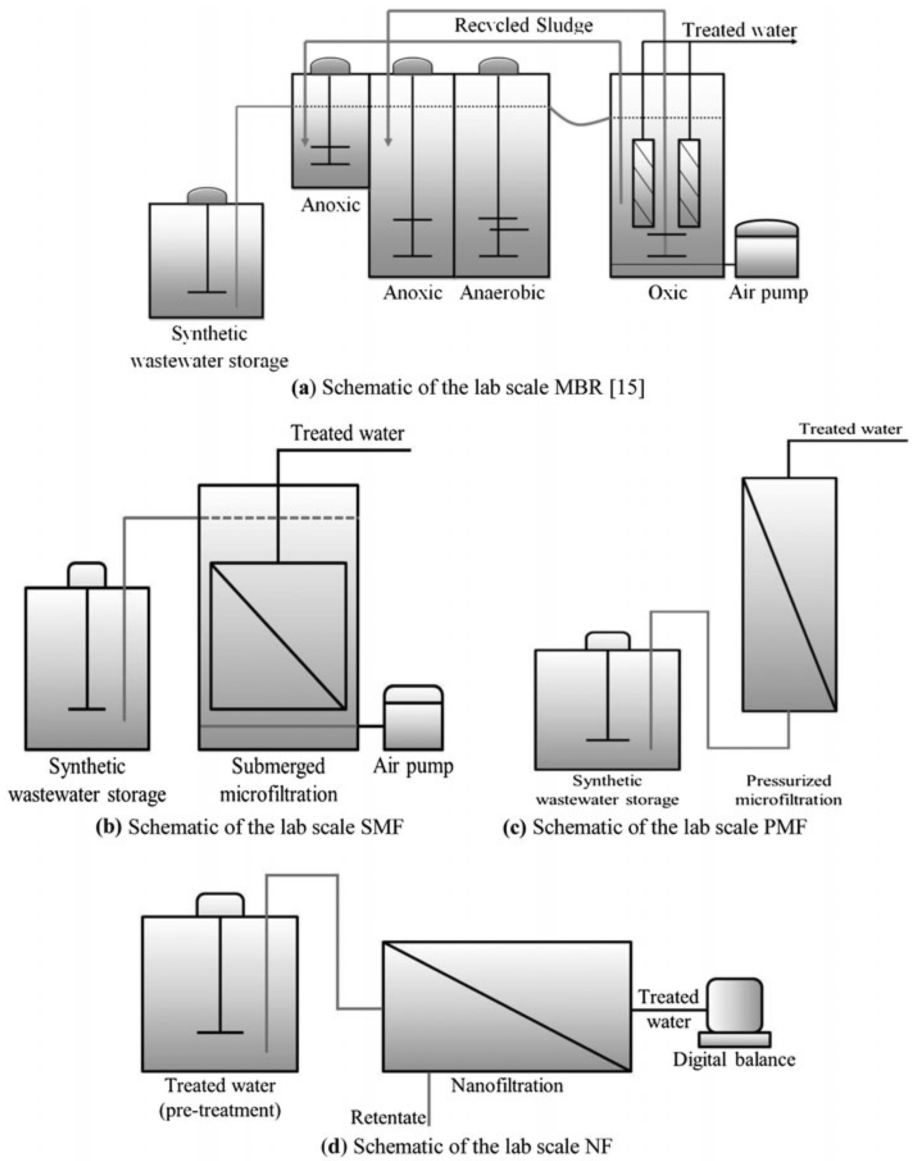

Fig. 1. Schematic diagram of lab scale: (a) MBR [15], (b) SMF, (c) PMF, and (d) NF processes.

removal of synthetic organic compounds, reduce the direct loading of dissolved organic pollutants onto the membrane and prevent membrane fouling [19]. As introducing PAC directly into the MBR process, removal of organic material occurs by PAC adsorption [20,21]. It has advantages such as increased biological activity of the micro-organisms, adsorption of soluble microbial products and increase in biodegradation [22,23].

Results showed that the effluent of MBR with PAC was stabilized by adsorption of non-readily biodegradable organic matter and adding PAC helped to improve the quality of the treated water [24].

As shown in Fig. 2, MBR and MBR with PAC were able to remove organic matter and T-N. Effective removal of COD was achieved with an average removal efficiency of 96.52% by MBR with PAC. Wastewater was contained a large amount of the organic matter [25]. Normally, the MBR is good for organic matter and nitrogen removal [13]. In order to efficiently remove the organic matter, biological response is very important. SMF and PMF to use physical methods as compared to the MBR biological reaction, COD removal ratio is very low.

T-N, T-P, TDS, and turbidity were removed by approximately 83.16, 50, 48.44, and 83.06%, respec-

Table 3
Characteristics of synthetic wastewater

Parameter	Concentration (mg/L)
Glucose	500
$NaHCO_3$	300
NH_4HCO_3	50
KH_2PO_4	22.5
$MgSO_4 \cdot 7H_2O$	50
$MnSO_4 \cdot H_2O$	0.03
$ZnSO_4 \cdot 7H_2O$	0.04
$CaCl_2 \cdot 2H_2O$	10
$FeCl_2 \cdot 4H_2O$	0.32
Yeast extract	50

tively. It can be seen that the MBR process has the highest rate of removal in T-N removal, which is judged to be caused by nitrification and denitrification. Nitrification is performed by autotrophic micro-organism, and ammoniac nitrogen is oxidized into nitrite nitrogen and nitrate nitrogen, and the oxidization of ammoniac nitrogen occurs through two steps [26]. In step 1, ammoniac nitrogen is oxidized into nitrite nitrogen, which is usually carried out by micro-organisms in the genus of *nitrosomonas* or *nitrosococcus*. In step 2, nitrite nitrogen is oxidized into nitrate nitrogen, and this reaction is performed by micro-organisms classified into the genus of *nitrobacter* or *nitrosocystis*. The following is a formula showing the processes of biological nitrification [Eqs. (3) and (5)] and denitrification [Eq. (6)] [27].

$$\text{Organic N} \rightarrow \text{Decomposition} \rightarrow NH_4^+ \tag{3}$$

$$\begin{aligned} NH_4^+ + 1.5\,O_2 &\rightarrow \text{Nitrosomonas} \\ &\rightarrow NO_2^- + H_2O + 2H^+ + 240 \sim 350\ \text{kJ} \end{aligned} \tag{4}$$

$$NO_2^- + 0.5O_2 \rightarrow \text{Nitrobacter} \rightarrow NO_3^- + 65 \sim 90\ \text{kJ} \tag{5}$$

$$NO_3^- \rightarrow NO_2^- \rightarrow NO \rightarrow N_2O \rightarrow N_2 \tag{6}$$

The energy generated by these processes has a very close relationship with the growth of nitrification micro-organisms itself since the nitrification micro-organisms use it to synthesize organic materials necessary for them. In denitrification, while there is sufficient dissolved oxygen, the micro-organisms get energy using the dissolved oxygen as an electron

Table 4
Characteristics of synthetic wastewater and various pretreatment effluents

Parameter	Average value					
	Synthetic wastewater as a feed	MBR (PAC)	MBR	SMF (PAC)	SMF	PMF
COD, mg/L	500	17.4(±7.55)	18.7(±7.51)	136(±26.26)	185(±22.56)	197(±25.62)
pH	8.2	7.79(±0.08)	7.83(±0.08)	7.89(±0.11)	7.93(±0.12)	7.93(±0.11)
T-N, mg/L	37.4	6.3(±0.60)	7.2(±0.62)	24(±0.63)	25(±0.81)	27(±0.94)
T-P, mg/L	5.0	2.5(±0.41)	2.3(±0.39)	4.2(±0.93)	3.9(±0.42)	4(±0.95)
TDS, mg/L	256	132(±25.74)	143(±25.68)	159(±34.39)	181(±27.85)	196(±35.82)
Turbidity, NTU	5	0.847(±0.06)	0.924(±0.06)	0.936(±0.11)	0.933(±0.07)	0.947(±0.08)

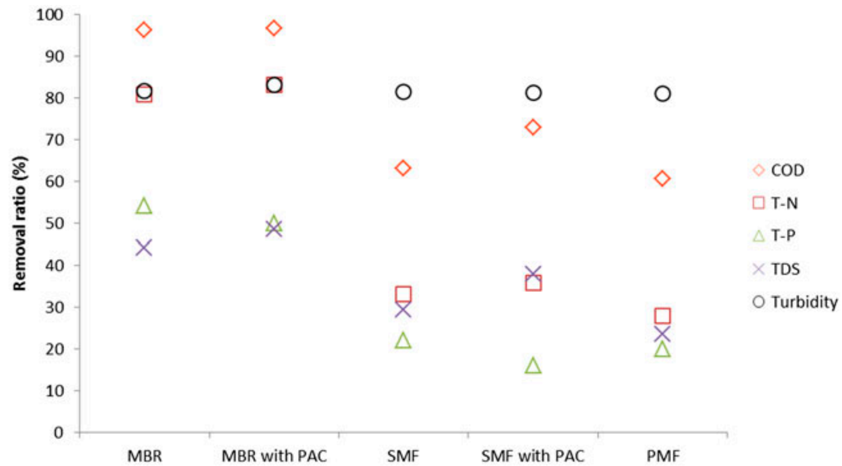

Fig. 2. Removal ratio of various pretreatments.

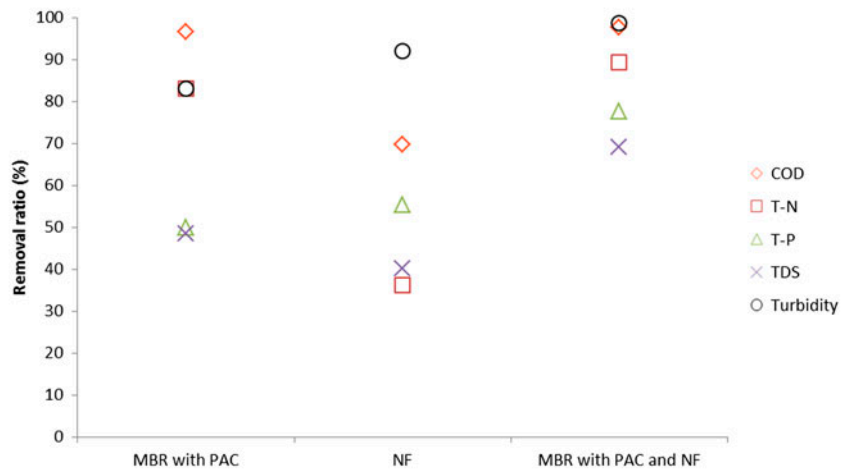

Fig. 3. Removal ratio of MBR with PAC, NF, and MBR with PAC and NF.

acceptor, but if there is free oxygen in the form like NO_3^- or NO_2^-, while there is insufficient dissolved oxygen, they use this as an electron acceptor, and nitrogen removal in water becomes possible by flying the nitrate nitrogen generated through the reaction of biological nitrification in the form of nitrogen gas through the reduction process into the air [28].

However, it is found that the rate of removal of T-P is as low as 50.00%. The removal principle of phosphorus in MBR process is based on the release of phosphorus in anaerobic condition followed by significant uptake or accumulation of phosphorus in aerobic condition [29]. Since phosphorus cannot be emitted into the air, it should be pulled out in the sludge to remove, it is judged that due to high MLSS concentration, SRT becomes longer, so the rate of removal of phosphorus becomes lower [30].

Fig. 3 shows the rate of removal of the MBR with PAC, that in the NF single filtration and that of a mixing process. When NF is filtered alone, the rates of removal of COD, T-P, and turbidity were 69.82, 55.35, and 92.10%, respectively, so they were, respectively, 5.35% and 9.04% higher than MBR with PAC, but the rates of removal of other items were lower. By the characteristics of NF membrane, it can remove high molecule organic matters, but it can hardly remove low molecular organic matters, and through this, it is found that the reason for the low rate of removal is that most extraneous water properties consist of glucose (180.156 Da) [11]. Therefore, it is judged that mutual supplementation is possible through linking to pretreatment.

The result of the change in the NF flux through a continuous experiment on lab-scale MBR with PAC

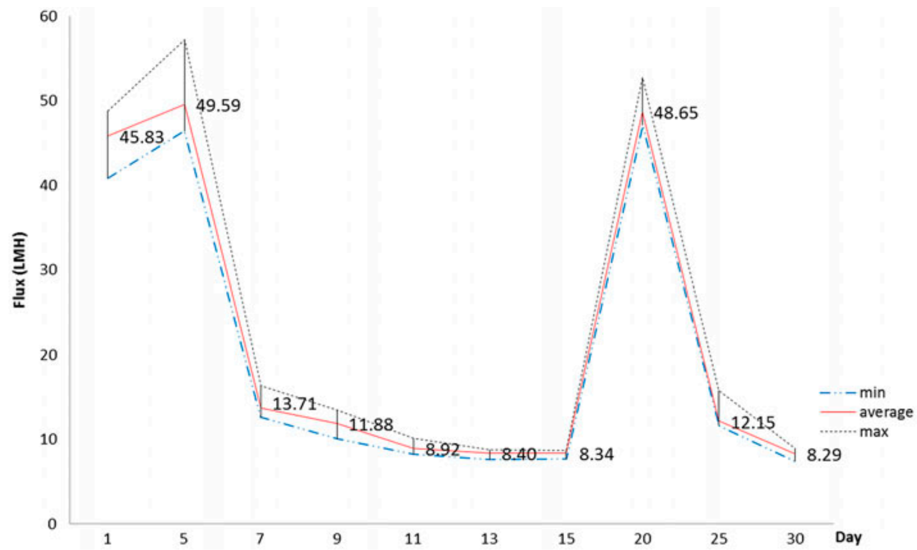

Fig. 4. Flux of MBR with PAC and NF.

Table 5
Comparison of feed, permeate, and water standard

	Average value				
Parameter	Synthetic wastewater as a feed	MBR (PAC)	NF	MBR with PAC and NF	Human application water standard[a]
COD, mg/L	500	17.4(±7.55)	150.9(±6.37)	5.36(±1.73)	≤20
pH	8.2	7.79(±0.08)	7.93(±0.20)	7.34(±0.17)	5.8–8.5
T-N, mg/L	37.4	6.3(±0.60)	23.89(±0.82)	4.02(±0.17)	≤10
T-P, mg/L	5.0	2.5(±0.41)	2.23(±0.14)	0.8(±0.06)	≤1
TDS, mg/L	256	132(±25.74)	153.42(±10.12)	79.10(±8.36)	NS[b]
Turbidity, NTU	5	0.847(±0.06)	0.395(±0.07)	0.064(±0.05)	NS[b]

[a]Ref. [34].
[b]No standards.

and NF for a month is shown in Fig. 4. The average value of the initial flux is 45.83 LMH, relatively lower than that on day 5, and it is judged that the initial MBR is not stabilized, so the quality of the discharged water decreases, which makes the flux decreased. After day 5, the average flux increases to 49.59 LMH and at the same time, the flux later decreased sharply, showing an average of 13.71 LMH on day 7, and it decreased to 8.34 LMH on day 15. These symptoms are due to membrane fouling. At this point, it was judged that as membrane chemical cleaning would be necessary, chemical cleaning was carried out with 0.1% NaOCl and 0.1% nitric acid [31,32].

As a result of the chemical cleaning, the flux was recovered up to 48.65 LMH, but on day 25, the flux was 12.15 LMH, faster than the degree of the decrease in the initial flux. It is judged that since organic materials and divalent ions such as Ca^{2+} or Fe^{2+} contained in extraneous water were not completely removed by the chemical cleaning, they remained in NF membrane, which increased the pollution speed, and divalent ions form a gel layer severely adherent to membrane surface cause membrane fouling [33].

According to the guidelines on the domestic reuse of treated wastewater as given in Table 5, it is found that for the discharged water quality of the MBR with PAC, T-P concentration was 2.5 ± 0.41 mg/L, which did not meet the criteria, and for NF, COD, T-N, and T-P were 150.9 ± 6.37, 23.89 ± 0.82, and 2.23 ± 0.14 mg/L, respectively, which also did not satisfy the criteria. However, as shown in Fig. 3, for the rates of removal of the six items listed above through the mixing process, COD was

97.73%; T-N, 89.24%; T-P, 77.68%; TDS, 69.10%; and turbidity, 98.73%.

Based on the wastewater reuse standards, effluent can be available for human application and industrial water.

4. Conclusions

In the present study, we have successfully operated integrated processes by MBR, SMR, and PMF with NF. The MBR with PAC process have been evaluated and compared with different pretreatment processes for enhancing water qualities. This study reveals the following points:

(1) The integrated MBR with PAC and NF process was able to enhance water quality for wastewater reuse.
(2) The advantage of nitrification and denitrification of MBR is important as a pretreatment for wastewater reuse.
(3) Flux of the MBR with PAC and NF integrated process was decreased by divalent ions as a main foulant.
(4) It is possible to provide understanding on treating wastewater via advanced technology.
(5) The MBR with PAC and NF integrated process has better water qualities than the water standard. Effluent can be available for human application.
(6) This study has operated at laboratory scale, so it will increase the size of the device and the number of filtration conditions.

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