Desalination and Water Treatment
www.deswater.com
doi: 10.1080/19443994.2013.768769

51 (2013) 5298–5306
July

Taylor & Francis
Taylor & Francis Group

Wastewater treatment using membrane bioreactor and reverse osmosis process

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Received 15 June 2012; Accepted 25 September 2012

ABSTRACT

The objective of this study was to investigate the effect of hydraulic retention time (HRT) on removal efficiencies of organic matter, nitrogen, and phosphorus in membrane bioreactors-reverse osmosis (MBR-RO) process for treating synthetic sewage. In MBR process, turbidity was less than average 2 NTU and average removal efficiency showed more than 99% during the operational period (MBR 105 day). As a result of HRT variation, average removal efficiencies of COD_{Cr} on HRT 6, 12, 18, and 24 h were about 72.4, 84, 88.6, and 92.5%, respectively. The NH_4^+–N removal efficiency was about 60.2, 85.5, 91.3, and 92.2%, respectively. TN and TP removal efficiencies increased from 53.7 and 56.8 to 82.5 and 86.4%, respectively, as the HRT increased from 6 to 24 h. In RO process, average removal efficiencies of color and COD_{Cr} in RO permeated water were about 99.9 and 96.8%, respectively. Also, removal efficiencies of TN, NH_4^+–N, NH_4^+–N, and TP were all above average 90%.

Keywords: Membrane bioreactor; Reverse osmosis; Total phosphorous; Total nitrogen; Hydraulic retention time

1. Introduction

Membrane bioreactors (MBR) is an excellent water treatment system that replaced the separation of sludgy using the gravitational settling of existing biological treatment with a separation membrane, increasing the treatment efficiency through preservation of high-concentration microorganisms and retention of microorganisms retarded in growth rate like nitrification bacteria, etc. in the reactor, improving the quality of effluent water through combination of water treatment by membrane, representing the treated water quality as stable regardless of biological treatment conditions, and being able to remove the pathogens to some extent, too [1–3]. However, there are pointed out the problems at the final treatment stage that color and phosphorus and more should be removed by applying additional physical and chemical treatment processes including flocculation by flocculants, adsorption by granular

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7th Aseanian Membrane Society Conference (AMS7), 4–7 July 2012, Busan, Korea

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activated carbon, etc. Recently, a new concept process linking reverse osmosis (RO) to MBR process is being developed for urban wastewater reuse, and it is reported to be able to complement the disadvantages of MBR [4–6]. The integration of an MBR with RO for the treatment of wastewater produces very high water quality. Tam et al. found that the adoption of an MBR-RO system for wastewater treatment produced high water quality which satisfied the drinking water requirements of the US EPA and WHO guidelines. MBR removed organic matter, ammonia, nitrogen and suspended solids, while the subsequent application of RO improved water quality, especially with respect to the esthetical and microbial parameters [7].

Lozier and Fernandez reported that MBR-RO not only satisfied the water quality standards for secondary drinking water but also it could be possibly applied to indirect reuse of treated water, based on results of MBR-RO operation [8]. Poitel et al. used MBR-RO system for leachate water treatment. As a result, it was reported that the content of COD and BOD_5 was less than 15 and 1 mg/L, respectively, and the removal efficiency of chlorine and nitrogen was 99 and 90%, respectively [9].

In this study, we investigated experimentally the impact of MBR-RO process on process efficiency when applying it to synthetic sewage treatment. In addition, we evaluated the impact of changes in hydraulic retention time (HRT) on removal of organic compounds, nitrogen and phosphorous.

2. Experimental materials and methods

2.1. Target wastewater

The composition of synthetic sewage used in this study is shown in Table 1. In the experiment, we used dextroglucose ($C_6H_{12}O_6$) as a source of carbon supply and ammonium chloride (NH_4Cl) as a source of nitrogen supply.

Table 1
Components of synthetic sewage

Components	Concentration(mg/l)
Glucose	225
Na_2CO_3	212
NH_4Cl	59.4
$CaCl_2$	17.5
NaCl	37.5
$MgSO_4$	12.5
KH_2PO_4	90.6

2.2. Experimental method

2.2.1. Composition of MBR and RO

The membrane module of MBR used in this experiment is a submerged hollow fiber membrane (manufactured by E&E Co., Ltd), and the specifications and outline diagram of MBR-RO system are shown in Table 2 and in Fig. 1, respectively. The reactor is made of acrylic materials, and it has the effective capacity of 3 L. Through submerged placement of a submerged hollow fiber membrane inside the reactor, continuous aeration became available at the bottom of module, and to minimize the attachment of microorganisms and sludge on the surface of membrane, we installed an air diffuser.

We represented the operating conditions of MBR and RO processes in Tables 3 and 4, respectively. Using the air diffuser installed at the bottom of module, we operated the volume of aeration as aeration intensity of 5 L/min taking into consideration, the supply of dissolved oxygen content of more than 2 mg/L and reduction of membrane pollution. To maintain the mixed liquor suspended solids (MLSS) concentration before MBR operation as 3,000 ± 200 mg/L, we regulated the sludge retention time (SRT) as 10 days by removing a proper volume of sludge in the reactor and conducted the experiment

Table 2
Membrane module specifications

System type	Submerged
Material	PVDF
Membrane type	Hollow fiber
Pore size	0.4 μm
Total membrane surface area	$0.11 m^2$

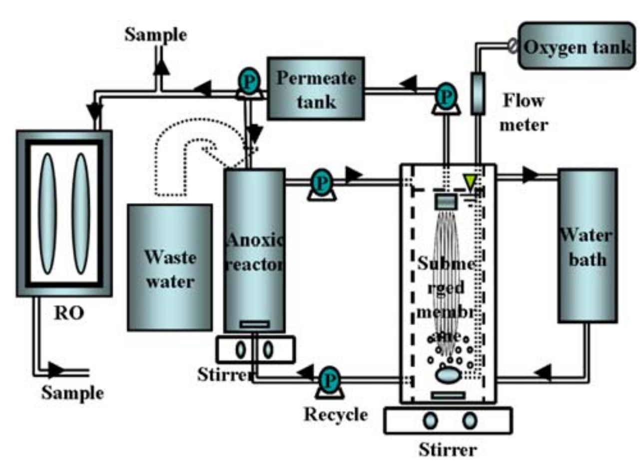

Fig. 1. Schematic diagram of MBR-RO system.

Table 3
MBR-operating conditions

Operating conditions				
Working volume	Anoxic reactor	5 l		
	MBR reactor	3.5 l		
Feed volume	10 l			
Operating pressure	>10 kPa			
SRT	10 day			
HRT	6hour	12hour	18hour	24hour
Flux(l/m^2·h)	4.43	2.16	1.4	1.02
Aeration intensity	5 l/min			
Dissolved oxygen	2–3 mg/l			
Temperature	25 ± 1 ℃			
pH	7.0 ± 0.5			

Table 4
Operating conditions and configuration of an RO system

Operating pressure	200psi
Capacity	200 l/h
RO pump	DS-960(Speck pump)
Vessel	AFV-4,040
Membrane	AG-4040F

by regulating HRT as 6, 12, 18, and 24 h corresponding to operation period of 0–30, 30–60, and 60–90 days, respectively.

To maintain the activation of microorganisms, we maintained the temperature at 25℃, using a water bath, and regulated pH near the neutral point using sodium bicarbonate. We maintained the MLSS concentration of sludge used in an aeration tank through periodic addition of sludge and discharge of it from MBR reaction tank to anoxic tank to maintain the MLSS concentration as 3,000 ± 200 mg/L while adapting the sludge returned from sewage treatment facilities to the synthetic sewage. In addition, we arranged some of the MBR permeated water in a permeated water tank leaving a certain volume for sample analysis flowing again in MBR reactor to maintain the concentration in MBR reactor. In addition, we evaluated the treatment characteristics of RO process by using the MBR effluent water of HRT 24 h as RO influent water.

3. Results and discussion

During the experimental period, we conducted the experiments under the operating conditions in reactor regulated for MLSS concentration as 3,000 ± 200 mg/L, F/M ratio as 0.15 (kg-COD/kg-MLSS.day), C/N ratio as 2.5, aeration intensity as 5 L/min, SRT as 10 days, HRT as 6, 12, 18, and 24 h corresponding to 30, 60, 90, and 105 days of operation period, respectively. To fix SRT to 10 days, we regulated it through periodic return and disposal of sludge in an aeration tank. In addition, we maintained the return ratio of anoxic tank to MBR reactor as 0.37, F/M ratio as 0.4 (kg-COD/kg-MLSS.day), and C/N ratio as 2.5.

The data shown in Fig. 2 represents the experimental results of analyzing the turbidity of permeated water in an aeration tank during operation period targeting synthetic sewage using MBR. The turbidity of treated water coming through the hollow fiber membrane in an aeration tank was maintained at a concentration of less than average 3NTU during operation period, showing the removal efficiency more than 99%. This is because the solid–liquid separation, one of membrane characteristics was realized effectively by the removal of suspended matters through the hollow fiber membrane with nominal diameter of 0.4 μm. However, the color of effluent water takes on a light yellow, which is the common results that appear at the time of biological treatment due to soluble microbial products, matters created by a soluble or colloidal microbial metabolism, showing the limits of MBR directly.

Thus, to resolve these problems, it is required to add another process, and in this study, we intended to resolve the afore-said problems by applying RO process. In the experimental results shown in Fig. 3, the concentration of COD_{Cr} represents that of $SCOD_{Cr}$, and looking through the results of its removal movement, the average concentration of COD_{Cr} for influent water is represented as 219 mg/L, while the average

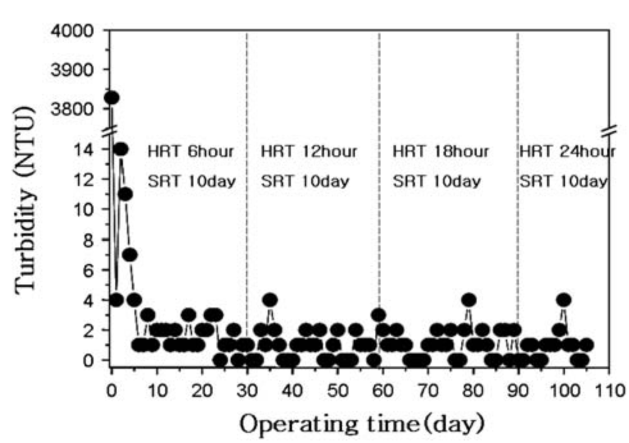

Fig. 2. Changes in turbidity according to HRT and operating time.

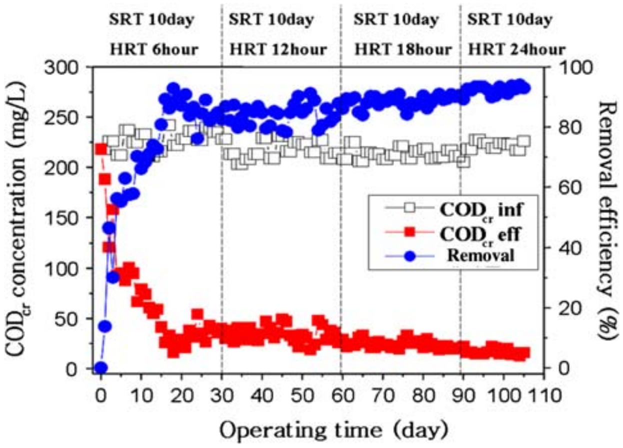

Fig. 3. Changes in concentration of COD_{cr} depending on HRT and operating time.

concentration of COD_{Cr} for effluent water after 15 days of MBR operation is represented as 29.5 mg/L, showing a stable quality of treated water, respectively. However, the concentration of COD_{Cr} is represented as decreasing before 15 days, showing an unstable quality of treated water, which is considered as due to the fact that microorganisms require a period of adaptation in environment to decompose the organic matters in wastewater.

In general, in order for microorganisms to adapt to the environment, an enzyme activity in microbial organisms is carried out and the microorganisms decompose the organic matters after being adapted to the environment through the secretion of the enzyme. In addition, in the removal of organic pollutants using the microorganisms, it is considered that stable treatment efficiency for organic matters can be achieved through retention of proper concentrations of wastewater and microorganisms that can be treated in MBR reactor and retention of optimized activation state of microorganisms as well.

The concentration of COD_{cr} after HRT 12 h (after 30 days of MBR process) was reduced in a stabilized manner, showing a high removal efficiency of 88.7% on an average, which is higher than conventional removal efficiency of organic matters for activated sludge process [10]. This is because suspended solids and microorganisms that cannot be removed by separation of sediments are completely excluded through combination of membrane process and biological treatment process.

Therefore, it is considered that during the process of solid–liquid separation by membrane, the decomposition efficiency of organic matters is increased by reaction of concentrated microorganisms in reactor.

Fig. 4 shows the experimental results representing the changes in concentration of NH_4^+–N, TN and NO_3^-–N corresponding to operating time in MBR process. The average concentrations of influent water for NH_4^+–N, TN were 42.3 and 46.4 mg/L, respectively. NH_4^+–N, TN showed a stable quality of treated water after 20 days of MBR process, and the average concentrations of NH_4^+–N, TN after HRT 18 h (after 60 days of MBR process) were represented as 3.7 and 10.3 mg/L, respectively. TN showed relatively low removal efficiency compared with NH_4^+–N and also showed the scope of change smaller than that of NH_4^+–N. This is considered due to the fact that although NH_4^+–N was removed effectively through nitrification, the removal efficiency of TN is lower than that of NH_4^+–N due to increased concentration of NO_3^-–N created relatively as shown in Fig. 4(c). C/N ratio of influent water affects the biggest impact on effective removal efficiency of nitrogen at the time of biological treatment, and in this study, we obtained approximately 90% removal efficiency of NH_4^+–N and TN on an average as a result of 105 day operation by maintaining the C/N ratio as approximately 2.5, showing the results similar to the study results that represented 90% removal efficiency of nitrogen in total in an operation while maintaining the C/N ratio as approximately 3 [11].

Fig. 5 shows the experimental results representing the changes in concentration of TP corresponding to operating time in MBR process. The average concentration of TP for influent water flowing in MBR was 21.3 mg/L. The results showed a stable quality of treated water after 15 days of operating time, and the average concentration of TP for MBR effluent water after HRT 18 h (after 60 days of MBR process) was represented as 2.9 mg/L, showing a removal efficiency of 86%, which is higher than those of HRT 6 and 12 h. The removal of TP was achieved generally in a stabilized manner, which was considered due to the fact that the phosphorus accumulated in sludge was also removed because there was a disposal of sludge through periodic regulation of SRT and MLSS.

Fig. 6 shows the results of experiment that analyzed the quality of treated water for color and turbidity in RO permeated water by introducing RO process in post-treatment experiment for the water permeated by membrane in MBR process after HRT 24 h (after 105 days of MBR process). After setting the permeation time under operation conditions of RO system as 5, 10, 15, 20, and 25 min, respectively, and also regulating the operation pressure as 200 psi, we conducted the permeation experiment using RO membrane of AG-4040F. Using DS-960 pump (Speck

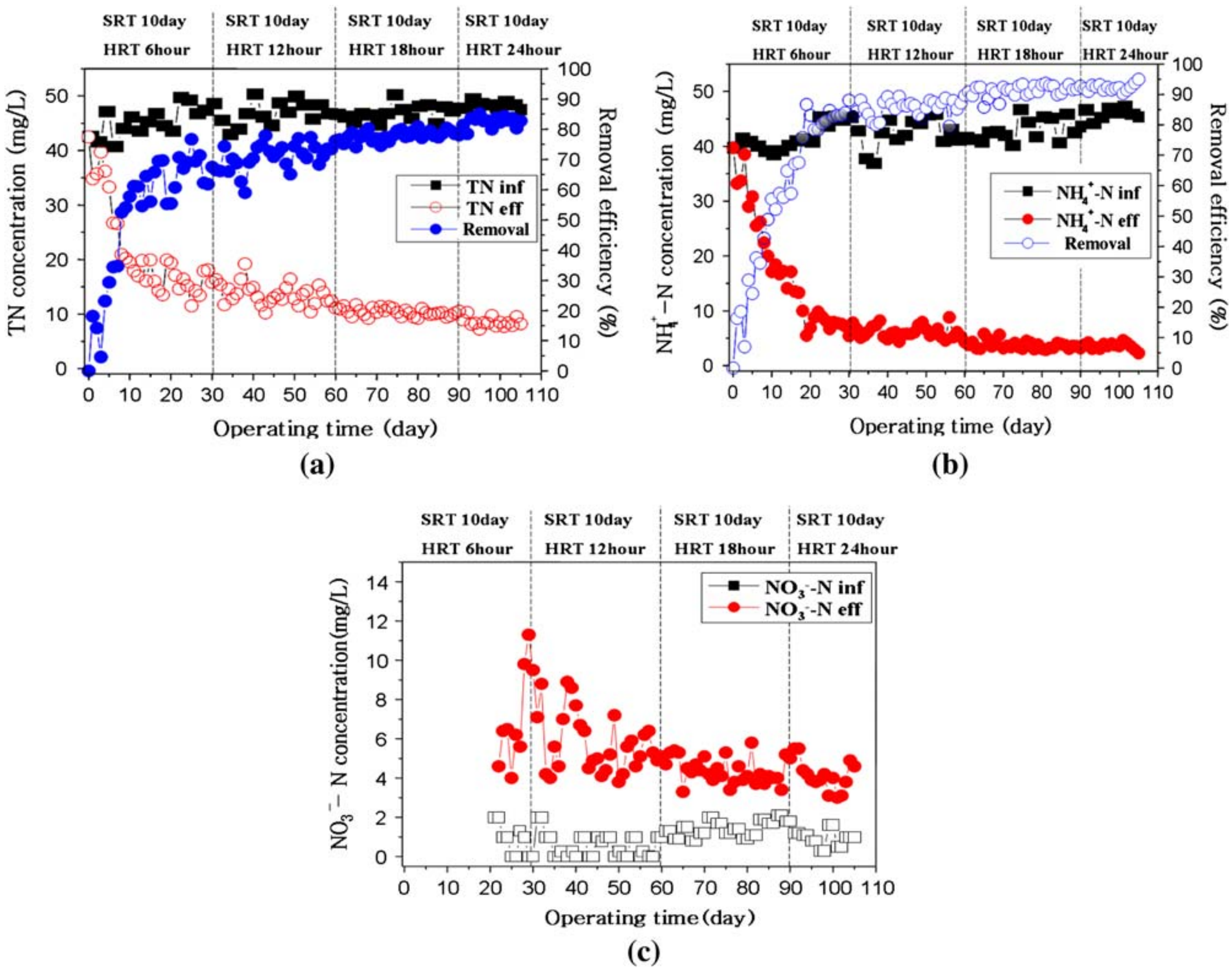

Fig. 4. Changes in concentration of TN (a), NH_4^+–N (b) and NO_3^-–N (c) according to HRT and operating time.

pump), we conducted the experiment by operating RO system at 200L/H speed. Fig. 6 represents the removal efficiency of more than 90% at average color for 1 PtCo in RO-permeated water when using RO permeating process targeting MBR effluent water, showing the concentration lower than Seoul tap water quality standards (5 PtCo). In addition, for removing turbidity, the experiment showed the results of removal efficiency of 90% on an average, representing a stable removal efficiency of turbidity.

Fig. 7 shows the results of experiment that analyzed the quality of treated water for COD_{Cr} in RO permeated water by introducing RO process in post-treatment experiment for the water permeated by membrane in MBR process. Looking through the experimental results shown in Fig. 7, the average concentration of COD_{Cr} after 5 min of RO filtration process was 6.9 mg/L, representing the removal efficiency of 75% on an average. These results show that the quality of treated water through RO filtration process is available for recycled water instead of the water quality not having satisfied the recycled water in MBR process.

Fig. 8 shows the results of experiment that analyzed the quality of treated water for NH_4^+–N, NO_3^-–N, TN in RO-permeated water by introducing RO filtration process in post-treatment experiment for the permeated water permeated by membrane in MBR process. The average concentrations of initial NH_4^+–N, NO_3^-–N, and TN in influent water (MBR-permeated water) of RO filtration process were 6.3, 7.4, and 16.2 mg/L, respectively. However, after five minute application of RO filtration process, the removal efficiencies for NH_4^+–N and NO_3^-–N were represented as high ratio of more than 70% in all cases, and in

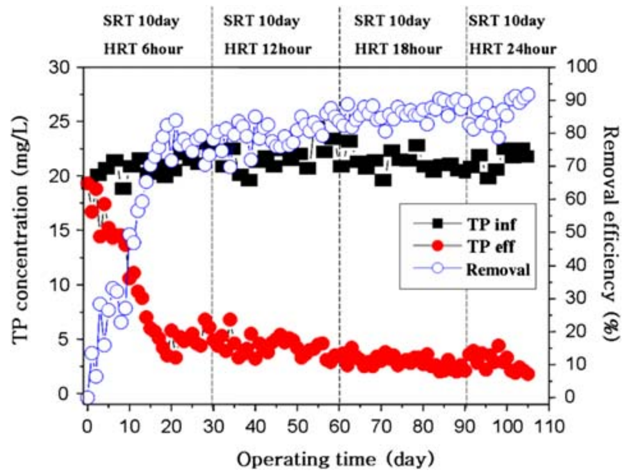

Fig. 5. Changes in concentration of TP according to HRT and operating time.

case of TN, the average concentration after 5 min of initial operation was represented as 7.5 mg/L with removal efficiency of 53.7% on an average. The concentration of NH_4^+–N and NO_3^-–N after 25 min of RO filtration process was represented as 1.04 and 0.6 mg/L, respectively, and their removal efficiencies were represented as high ratio of 84 and 92%, respectively. And in case of TN, it represented 84.6%, good results for removal characteristics as 2.5 mg/L according to results of 20 min performance of RO filtration process.

Fig. 9 shows the experimental results that analyzed the quality of treated water for TP in RO-permeated water. Looking through the experimental results shown in Fig. 9, the average concentration of initial TP in MBR effluent water was 6.5 mg/L. As the results of RO filtration process performed targeting MBR effluent water, the average concentration after 5 min of initial operation was represented as 0.09 mg/L, showing the removal efficiency of 98.6%, and as the results of RO filtration process for 20 min, it was represented as 0.02 mg/L, showing the removal efficiency of 99.7% on an average. This can be evaluated as showing a stable quality of treated water during entire operation period.

3.1. Material balance of MBR-RO process

Figs. 10–12 show the material balance diagram of COD_{Cr}, TN and TP in MBR-RO process and represented the removal efficiency through each analytic method after collecting a certain volume of samples in effluent water of each process and MBR reactor.

Fig. 10 represents the material balance diagram for COD_{Cr} in MBR-RO process. 87.7% of COD_{Cr} among initial 100% was removed while passing the MBR process and the remaining 12.3% was processed by application of RO filtration process, but only 3.2% of COD_{Cr} was not removed in MBR process.

Fig. 11 represents the material balance diagram for TN in MBR-RO process. Looking through the material balance diagram, the average concentration of TN in influent water in MBR process was 47.2 mg/L, but average 70.8% was removed while passing the MBR process. However, among 29.2% of TN not removed, NO_3^-–N that hardly existed in influent wastewater occupied approximately 50% of TN in MBR-treated water after being created through nitrification of NH_4^+–N. In addition, through application of RO filtration process to remaining 29.2% of TN in treated water not removed in MBR process, only 8.9% of TN in total was not removed.

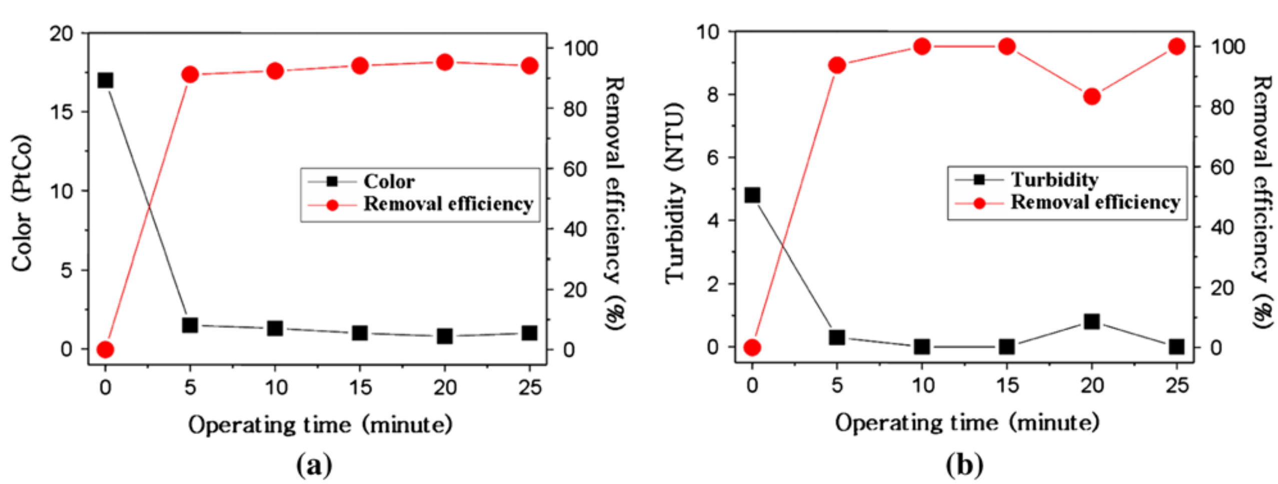

Fig. 6. Changes in color (a) and turbidity (b) at the time of RO filtration targeting MBR effluent water.

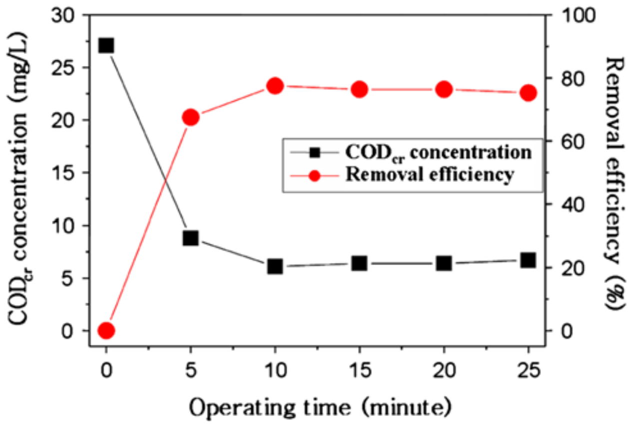

Fig. 7. Changes in concentration of COD_{cr} at the time of RO filtration targeting MBR effluent water.

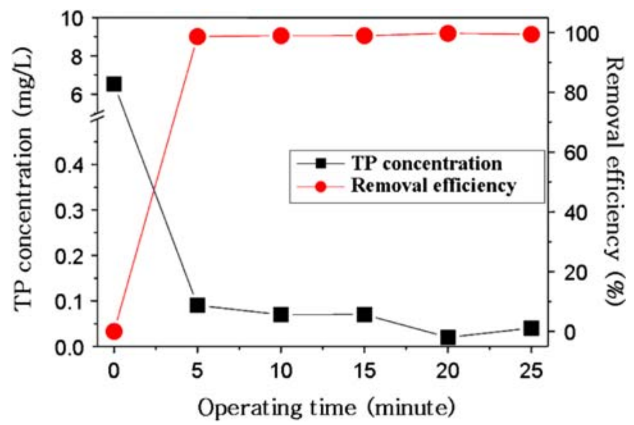

Fig. 9. Changes in concentration of TP at the time of RO filtration targeting MBR effluent water.

Fig. 12 represents the material balance diagram for TP in MBR-RO process. In case of TP, 76.7% was removed through treatment in MBR process and the remaining 23.0% of TP was removed by application of RO filtration process for post-treatment, reaching the removal efficiency of 99.7%, but only 0.3% was not removed in MBR process.

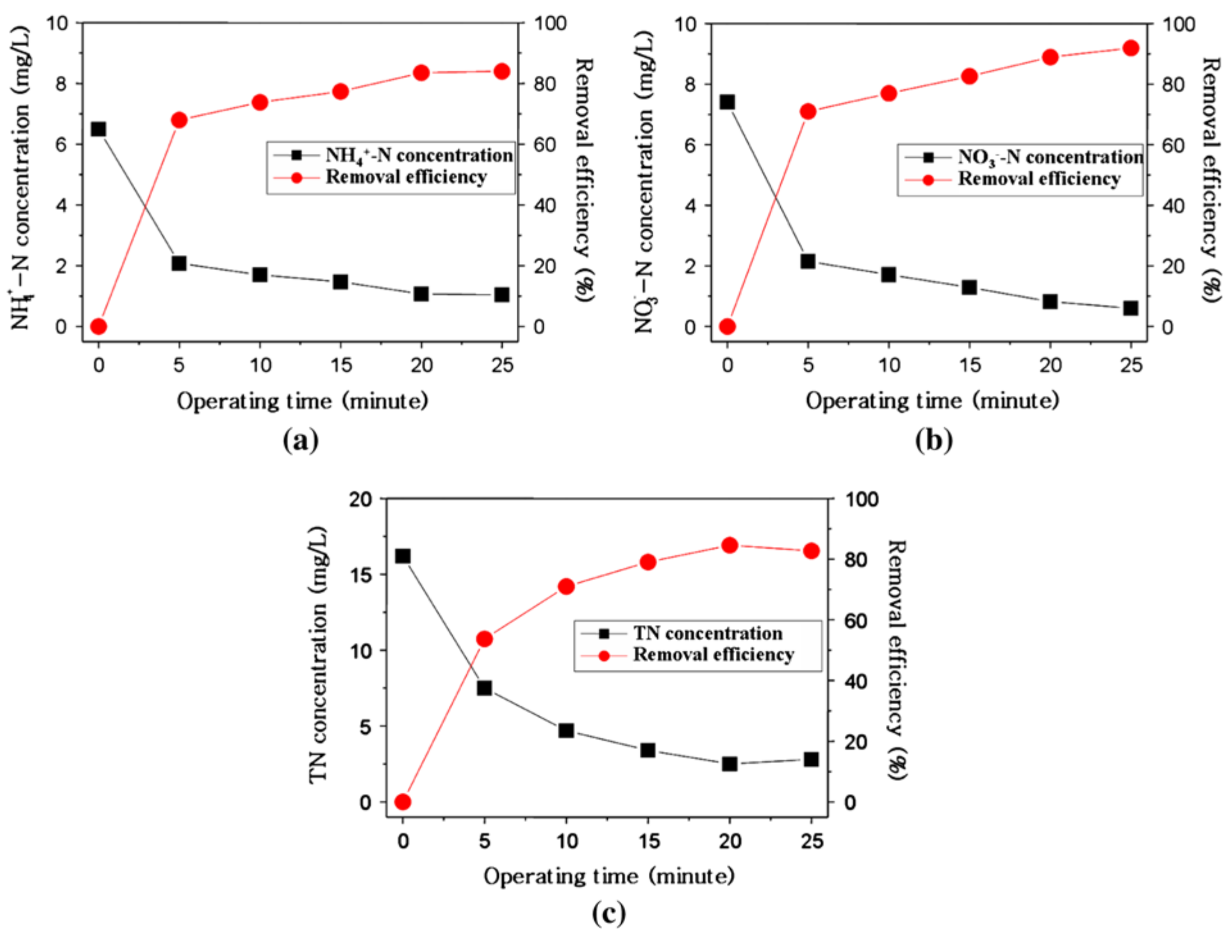

Fig. 8. Changes in concentration of NH_4^+–N (a), NO_3^-–N (b), TN (c) at the time of RO filtration targeting MBR effluent water.

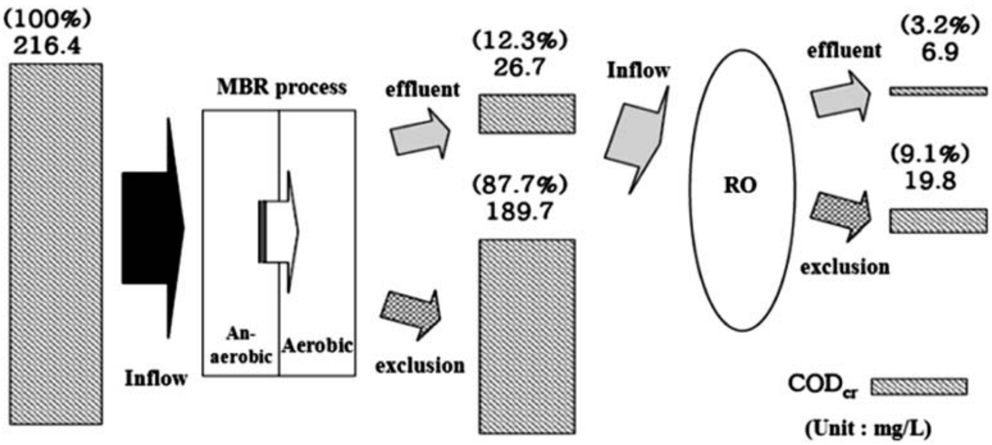

Fig. 10. The material balance for COD_{Cr} in MBR-RO process.

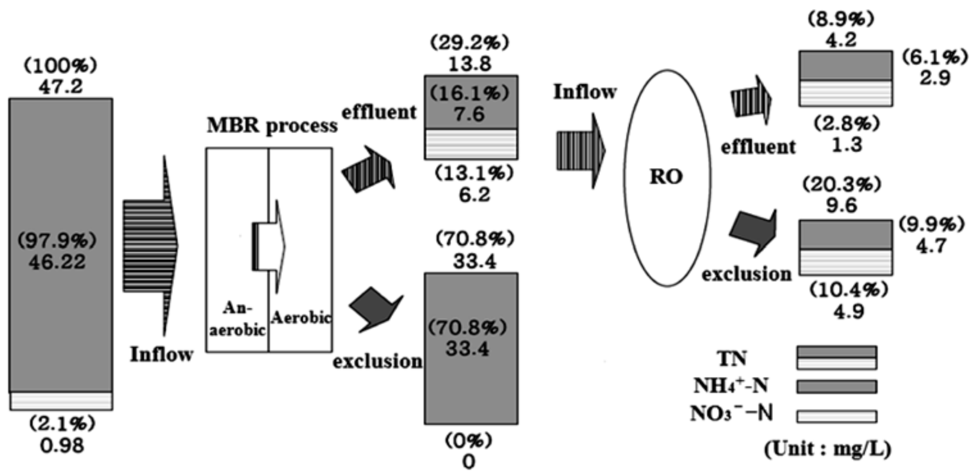

Fig. 11. The material balance for TN in MBR-RO process.

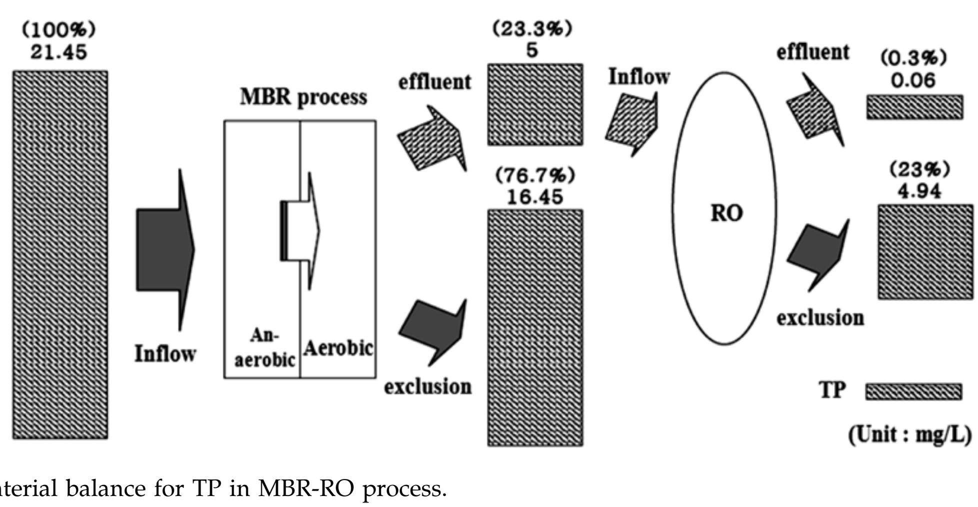

Fig. 12. The material balance for TP in MBR-RO process.

4. Conclusion

As a result of treatment for synthetic sewage using MBR-RO filtration process, we obtained the conclusion as follows.

(1) As a result of application of MBR process, the turbidity of effluent water was maintained at concentration less than average 2NTU during operation period, showing the removal efficiency of more than 99%, and in case of COD_{Cr}, the concentration was reduced in a stabilized manner after HRT 12 h (after 30 days of MBR operation).
(2) After 60 days operation of MBR process, the concentrations of NH_4^+–N, NO_3^-–N, TN were 3.7, 4.3, 10.3 mg/L, respectively, and the removal efficiency of TN was represented as relatively low compared with NH_4^+–N and the scope of change was also represented as smaller than that of NH_4^+–N. This is considered due to the fact that although NH_4^+–N was removed effectively through nitrification, the removal efficiency of TN is lower than that of NH_4^+–N due to increased concentration of NO_3^-–N created relatively.
(3) The average concentration of TP for MBR effluent water after HRT 18 h (after 60 days of MBR operation) was represented as 2.9 mg/L, showing a removal efficiency of 86%, which is higher than those of HRT 6 and 12 h. This is because the phosphorus accumulated in sludge was also removed due to disposal of sludge through periodic regulation of SRT and MLSS.
(4) As the results of application of RO filtration process to MBR effluent water, the average color in RO-permeated water represented the removal efficiency of more than 90% at 1 PtCo and for the removal of turbidity in RO permeated water, it showed the removal efficiency of 90% on an average, representing a stable removal efficiency of turbidity.
(5) As the results of application of RO filtration process to MBR effluent water as target wastewater, the removal efficiencies of COD_{Cr}, NH_4^+–N, TN, and TP were represented as more than 90%, showing the possibility of recycled water.

Acknowledgments

This manuscript was supported by Basic Science Research Program through National Research Foundation of Korea (NRF 2012-0005227) funded by the Ministry of Education, Science and Technology and we hereby express our thanks.

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