



The evaluation of RO membrane performance with pretreatment membrane on municipal sewage treatment

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ABSTRACT

In this research, to investigate totally the phenomenon of concentration and dilution in microfiltration membrane (MF)- reverse osmosis membrane (RO) hybrid process, it was operated for sewage with the change of variables such as temperature and TMP (trans-membrane pressure). MF modules were used directly without pretreatment and then MF permeate was sent to RO module. For evaluation of membrane fouling at a point of time, continuous process was conducted at a fixed temperature and TMP. With physical cleaning by air-stripping, permeate flux of MF module was decreased by the lapse of time. After 11 h, permeate flux in RO was decreased drastically and it implied the cleaning period of RO module. The removal efficiency of chemical oxygen demand (COD), T-N, T-P, total dissolved solid (TDS) and turbidity in RO module was observed high values over 94% regardless of the variations of temperature and TMP. It suggests that this system is very suitable for the treatment of sewage and the treated water is satisfied on environment standard. Turbidity was removed sufficiently in microfiltration modules and pH values were maintained under 6.5–8.1 range. In RO module, degree of concentration (DC) of COD, T-N, T-P and TDS was shown high values. T-N, T-P and TDS were shown very high DC values because of low removal efficiency in microfiltration modules.

Keywords: Immersed microfiltration membranes; A spiral wound reverse osmosis membrane; Sewage; Emission water quality of permeate; Environment standard

1. Introduction

Antique documents for the water pollution related closely to humans were recorded in history of the ancient Roman era's wastewater facilities. Actual sanitation had not been managed until the 1800s of industrial revolution. At that time, the industrial revolution led to the formation of slum and the abrupt outbreak of various diseases although it raised the living standard.

Such situations let advanced nations (e.g. the United Kingdom) reflect to repair sewer and construct sewage disposal plant, additionally to legislate against the environmental pollution of the sewage [1]. Meanwhile, there are many causes of water pollution such as sewage, industrial wastewater, livestock wastewater, solid waste and so on. The treatment of wastewater is indispensable to preserve the ecosystem, whose processes are characterized by various effects during operation. The treatment of wastewater enables us to

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be protected from the threat of infectious disease due to stagnant water and the flooding due to rainfall. They play important role in preserving the ecosystem and the resource of drinking water over self-purification [2]. Especially, membrane filtration as a novel alternative is expected to play an important role in treating sewage. It is more reliable of water-quality in drinking water process because of automatic operation and environmentally-friendly process. Its several merits on the process are described as follows; the easiness of scale-up, simultaneous feasibility of batch and continuous operation, the easiness of operation, and excellent removal efficiency. The other hand, its several demerits are stated as follows; economics due to the limited amount of filtration and high energy consumption in a huge plant, the outbreak of membrane fouling, the weak polymeric membrane due to the effect of pH and temperature, short life-time, the short change period of membrane, brittleness of ceramic membrane, the limited application of metallic membrane and so forth. Nevertheless, much effort to overcome them has been continuously carried out by many researchers [3–5]. Microfiltration-reverse osmosis (MF-RO) hybrid process is one of them and it has been expanded to various applications, especially for water treatment such as reuse of industrial waste-water and desalination of seawater [6–18]. As previous studies for MF-RO hybrid system had been discussed in the viewpoint of permeate (produced water), these of MF-RO hybrid system were little discussed in the viewpoint of retentate (concentrated water). In this study, both behavior of concentrated water and that of produced water were totally observed/considered to raise reuse of concentrated water as well as quality of produced water. To carry it out, hybrid process combined an immersed MF with a spiral wound RO was applied to S-sewage (influent). Behaviors of the concentrated water in retentate and the produced water in permeate were discussed by measuring chemical oxygen demand (COD), T-N, T-P, total dissolved solid (TDS) and turbidity as a function of temperature and pressure. In MF-RO hybrid process applied to sewage, optimal operating condition was investigated through running variables (e.g. Temperature, trans-membrane pressure [TMP]) to satisfy water-quality standard and achieve effective concentration. As well, optimal cleaning period was determined through the sudden change of flux as index of membrane fouling.

2. Experimental

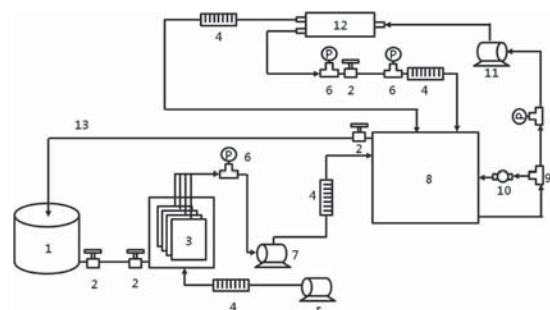
2.1. Equipments

Microfiltration modules were installed directly to feed reservoir without special pre-treatment. Feed

solution used in the experiment was collected from preliminary clarifier of environmental public corporation in “P” city and it was directly applied to microfiltration modules. The diagram of process was illustrated in Fig. 1.

2.2. Methods

Feed solution was retained in feed reservoir of MF. And it was circulated in the hybrid process with the variation of temperature and TMP. MF permeate-retaining reservoir was installed to minimize the effect of MF modules combined with the pressure of RO. Temperatures of feed solution-retaining reservoir were varied by 3 steps of 15, 25 and 35 °C. Air supplier was set up in the bottom of reservoir to minimize the difference of temperature and concentration, and to clean modules indirectly. To maintain temperature and concentration constantly in feed solution, circulation pump was installed in MF-RO hybrid process. The concentration amount in MF-permeating solution and RO-concentrating/permeating solution were directly measured and/or collected from the permeating-line after reaching at the state-state. Both collecting samples to analyze and measuring the flux were simultaneously conducted to minimize experimental error.



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|------------------|---------------|-------------------|
| 1. Feed tank | 2. Gate valve | 3. MF module |
| 4. Flow meter | 5. Air pump | 6. Pressure gauge |
| 7. Suction pump | 8. Reservoir | 9. T-type unit |
| 10. Bypass valve | 11. Pump | 12. RO module |

Fig. 1. Schematic diagram of membrane separation system.

Table 1

Experimental conditions for microfiltration and reverse osmosis membrane module

Item	Unit	Value
Pressure	MF	Pa
	RO	kPa
Aeration	MF	L/min
	Temperature	°C
		133, 200, 267, 333
		392, 490, 588, 686
		20
		15, 25, 35

Table 1 stated experimental condition for MF module and RO module. In continuous operation at constant temperature and pressure, the change of flux in permeate was checked with the period of 1 h to observe membrane fouling. The condition of operation was described in Table 2.

3. Results and discussion

3.1. Permeability

For the immersed MF, permeability of ultra-pure water was measured in permeate to observe relationship between temperature and TMP. For the experiment of ultra-pure water permeability using the immersed MF, it showed general trend that ultra-pure water permeability (J_w) increased with increasing temperature at constant pressure (permeability order: $15 < 25 < 35^\circ\text{C}$). The value of permeability at the applied pressure passed through “0” as shown in Fig. 2, indicating that membrane had no defects (e.g. large pore, water leak) and it enable to conduct the experiment (Fig. 2). For the spiral wound RO, permeability of ultra-pure water was measured in permeate to observe relationship between temperature and

TMP. It showed general trend that ultra-pure water permeability linearly increased with increasing temperature at constant pressure (Fig. 3). For the immersed MF module applied to sewage, Fig. 4 showed relationship between permeability and TMP/temperature. Fluctuation of permeability as a function of temperature/pressure was constant although the decreasing rate of permeability was 23–32% compared to that of pure-water permeability. It is inferred that primary cause of decreasing permeability is pore-blocking of membrane surface due to colloidal pollutants and secondary cause is pore-blocking of inter-pore due to adsorption of soluble pollutants. In the system, the immersed MF module played a role in controlling effectively the membrane load of RO system by blocking colloidal pollutants. Also, J/J_0 as the factor of decreasing rate in permeability was inversely proportional to operating time under continuous operation.

Table 2
Experimental conditions of continuous operation for microfiltration and reverse osmosis module

Item		Value	Unit
Continuous operation	Pressure	MF 200 RO 588	Pa kPa
	Temperature	MF & RO	25 °C
	Aeration	MF	20 L/min

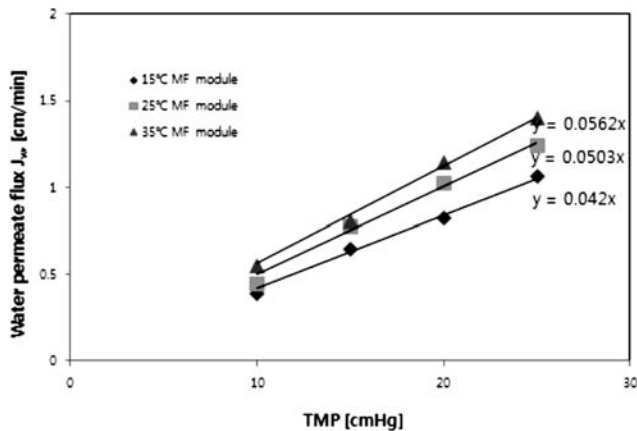


Fig. 2. Behavior of pure water permeate flux in the MF process as a function of TMP.

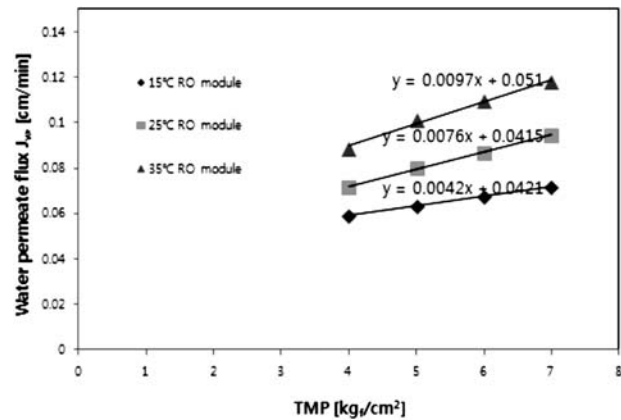


Fig. 3. Behavior of pure water permeate flux in the RO process as a function of TMP.

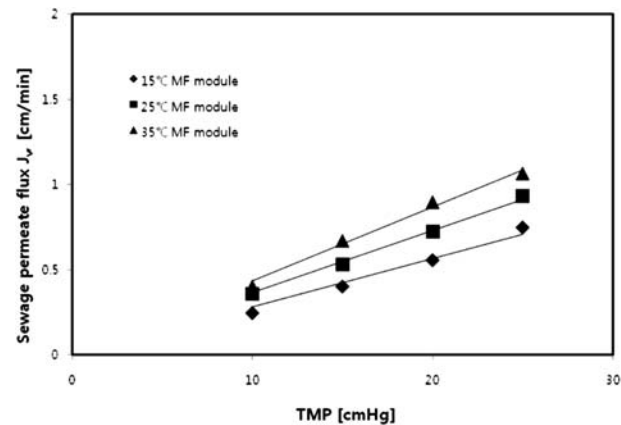


Fig. 4. Behavior of sewage permeate flux in the MF process as a function of TMP.

The results enabled to know that membrane fouling in MF module was evaluated by air-scrubbing as physical cleaning (Fig. 5). For RO module applied to sewage, Fig. 6 showed relationship between permeability and TMP/temperature. Fluctuation of permeability as a function of temperature/pressure was constant although the decreasing rate of permeability was 13–21% compared to that of pure-water permeability. The abrupt decrease of permeability appeared after 11 h of operation whereas it did not appear till 11 h of operation. It means that RO module should be cleaned around 11 h of operation (Fig. 7). It is inferred that main cause of decreasing permeability is scaling phenomenon due to laminating of soluble pollutants (e.g. virus, humic acid) unlike their cause in MF related to colloidal pollutants. It was interpreted that soluble pollutants to adsorb on RO surface resulted in the decrease of permeability.

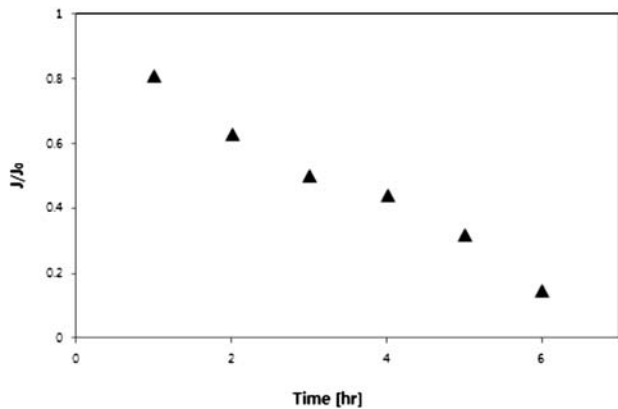


Fig. 5. Behavior of sewage permeate flux ratio J/J_0 in the MF process as a function of operating time.

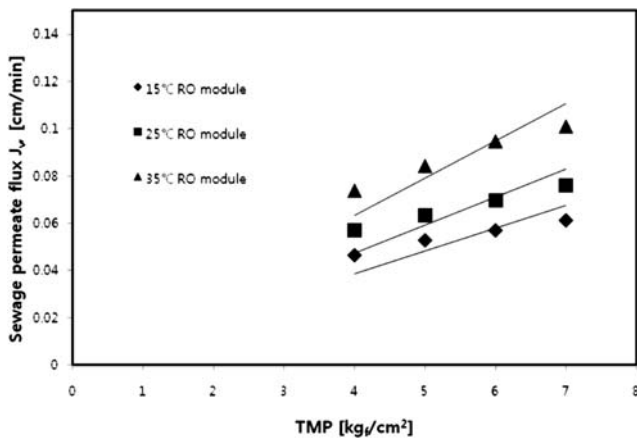


Fig. 6. Behavior of sewage permeate flux in the RO process as a function of TMP.

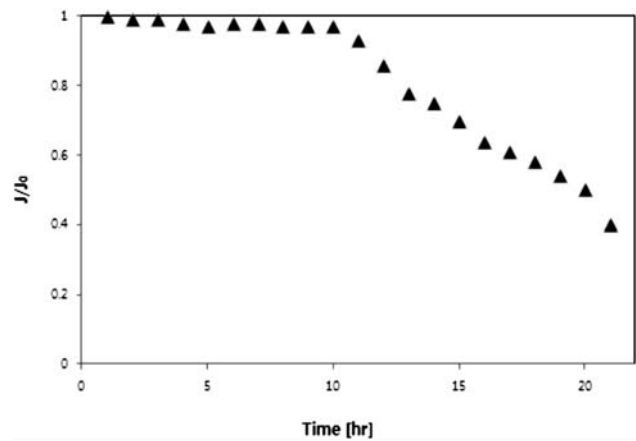


Fig. 7. Behavior of sewage permeate flux ratio J/J_0 in the RO process as a function of operating time.

3.2. COD, T-N, T-P

The system was sequentially constituted of the immersed MF module and RO module. To observe the efficiency of treatment, permeate in each process was measured by various analytical items (e.g. COD, T-N, T-P). Removal efficiencies of CODs were 65–70% in the immersed MF module and 95% in RO module (Fig. 8). For permeate of MF and retentate of RO, respectively, the degree of concentration for COD was calculated. As shown in Fig. 9, the degree of COD concentration was 0.07–0.09 as a function of temperature. The results were interpreted in the viewpoint of colloidal pollutants and soluble pollutants. In other word, wastewater supplied to the immersed MF module gave the load for MF membrane with colloidal pollutants and soluble pollutants. The efficiency of COD removal due to the reason described above was

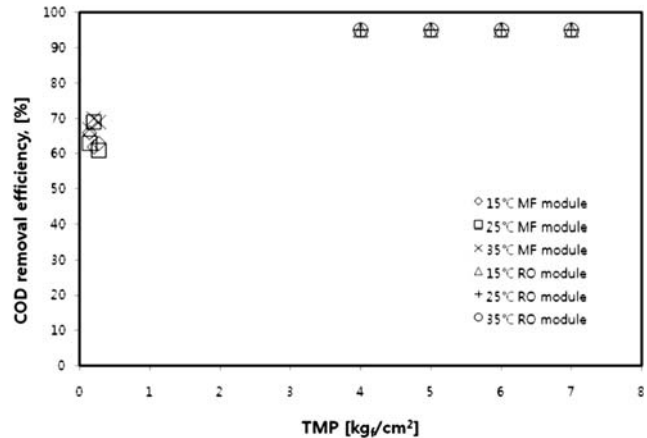


Fig. 8. Behavior of COD removal efficiency in the process as a function of TMP.

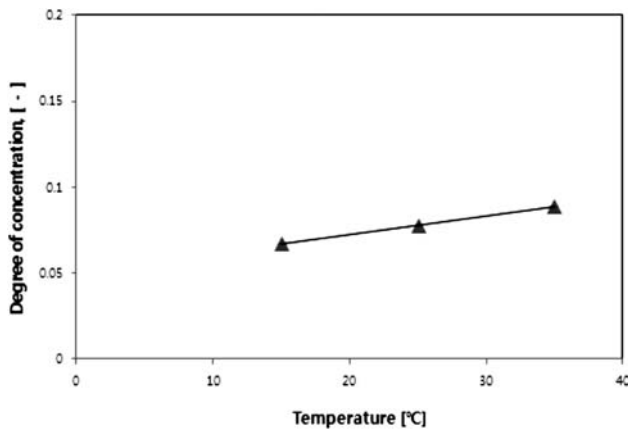


Fig. 9. Behavior of degree of COD concentration in the RO process as a function of temperature.

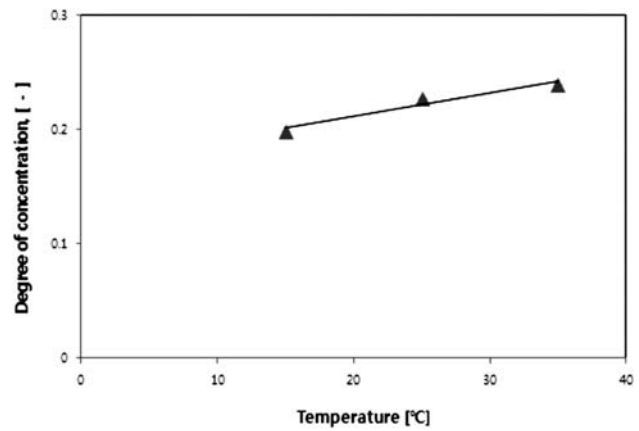


Fig. 11. Behavior of degree of T-N concentration in the RO process as a function of temperature.

65–70%. The soluble pollutants can't be removed without RO-connected process because substances treated by MF process are mostly soluble. It led to COD removal efficiency of 95%, indicating the RO module was effectively connected to the overall system. Removal efficiencies of T-N were 30–35% in the immersed MF module and 96% in RO module, respectively (Fig. 10). As shown in Fig. 11, concentration degree of T-N some increased linearly with increasing temperature while the fluctuation of concentration degree was not observed. The concentration degree of T-N increased from 0.20 to 0.24 with increasing temperature. The removal efficiency of T-N was 96% in RO module whereas that in MF module was very low. Nitrogen-induced compounds result in the eutrophication of water and they are soluble in water, indicating that the MF module is not suitable to remove nitrogen-induced compound. As a result, the

removal efficiency of T-N was low 30–35% in the MF module while that in RO module was high 95%. The system effectively controlled the water-quality without the process based on biological treatment (see Fig. 12). The concentration degree of T-P in MF module was not even expected as the removal efficiency of T-P was very low. By contrary, the concentration degree of T-P in RO module was expected as the removal efficiency of T-P was over 98%. The removal efficiency of T-P increased from 0.22 to 0.39 with increasing temperature at the applied pressure (Fig. 13). Like the result of T-N, the results of T-P can also be interpreted in the viewpoint of the features of pollutants. Namely, phosphorus-induced compounds result in the eutrophication of water and they are soluble in water, indicating that the MF module is not suitable to remove phosphorus-induced compound. Therefore, RO-connected system is indispensable to remove

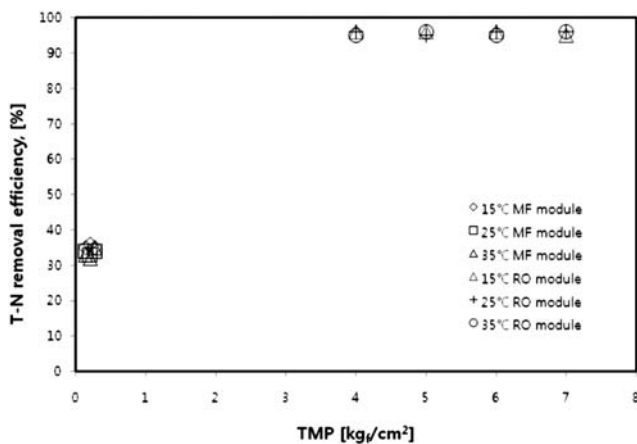


Fig. 10. Behavior of T-N removal efficiency in the process as a function of TMP.

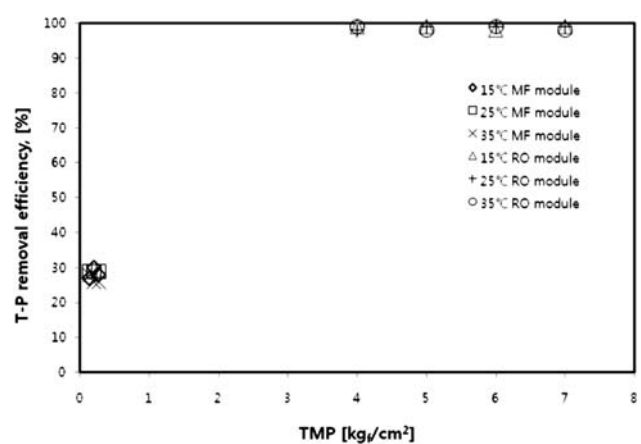


Fig. 12. Behavior of T-P removal efficiency in the process as a function of TMP.

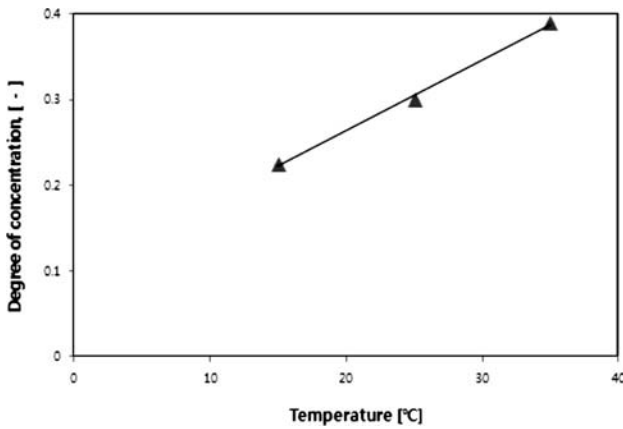


Fig. 13. Behavior of degree of T-P concentration in the RO process as a function of temperature.

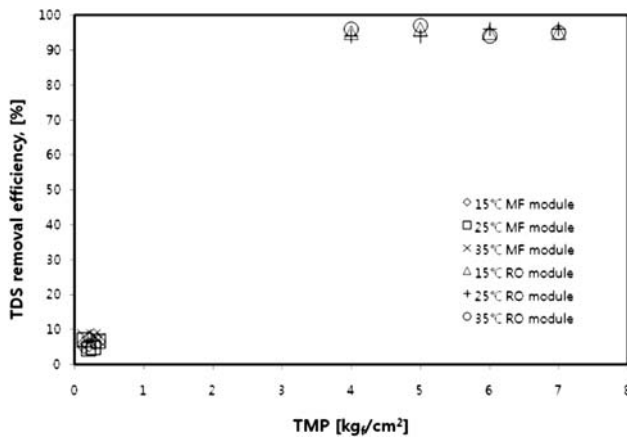


Fig. 14. Behavior of TDS removal efficiency in the process as a function of TMP.

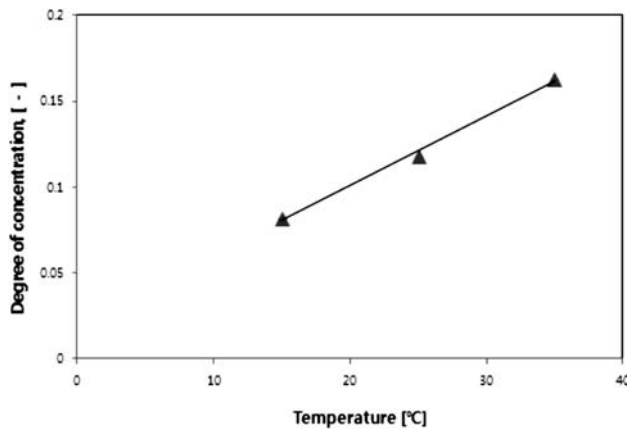


Fig. 15. Behavior of degree of TDS concentration in the RO process as a function of temperature.

phosphorus-induced compound. The removal efficiency of T-P was low 25–30% in the MF module while that of RO module was high 98%. The system was effectively controlled the water-quality without the process based on biological treatment.

3.3. TDS, turbidity, pH

The removal efficiency of TDS was high 94% in RO module whereas that in MF module was very low 10% (Fig. 14). For permeate of MF and retentate of RO, respectively, the degree of concentration for TDS was calculated. The degree of TDS concentration was not expected as the removal efficiency of TDS was low 6–8% in MF module. However, that of RO module was expected as the removal efficiency of TDS is very high 94%. The removal efficiency of TDS increased from 0.08 to 0.16 with increasing temperature, independent of the applied pressure (Fig. 15). Like the result of T-N and T-P, the results of TDS can also be interpreted in the viewpoint of the features of pollutants. As soluble chemicals are soluble in water, the MF module is not suitable to remove soluble substances. Therefore, RO-connected system is indispensable to remove soluble substances. With the operation of the system, the removal efficiency of turbidity was very high 99.7% in RO module while that in MF module was 99% (Fig. 16). Meanwhile, the removal efficiency of turbidity in RO module was independent of temperature/applied pressure. In the condition of constant temperature, the pH value of permeate in MF module was 7.9–8.0 (increasing) whereas that in RO module was 6.5–6.7 (decreasing) (Fig. 17). They were satisfied with the range of management and effluent standard of environmental regulation.

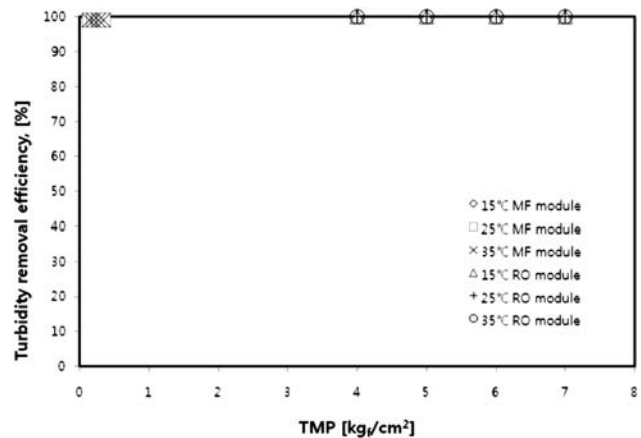


Fig. 16. Behavior of turbidity removal efficiency in the process as a function of TMP.

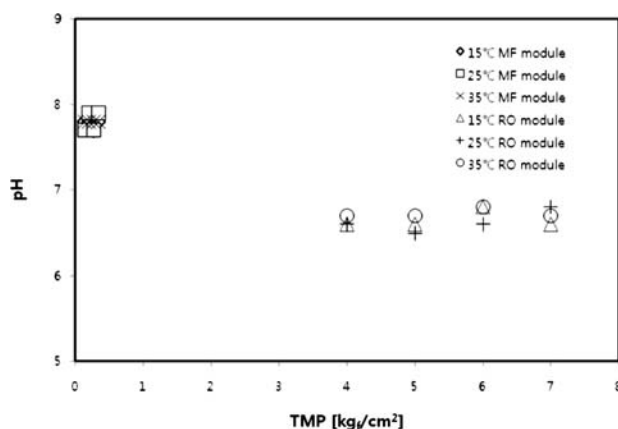


Fig. 17. Behavior of pH in the process as a function of TMP.

4. Conclusions

Performances of RO–MF hybrid process was evaluated through filtration feature, retentate concentration, and permeate dilution of sewage as follows:

- (1) In the case of sewage, the decreasing rate of permeability in MF was 23–32% of pure-water permeability and that of RO was 13–21% of pure-water permeability. Both permeabilities were linearly proportional to temperature and TMP.
- (2) Under air-scrubbing of MF, permeability of MF for sewage linearly decreased with increasing time
- (3) The abrupt decrease in permeability in RO module appeared around 11 h of operation, implying that the system should be cleaned after 11 h of operation.
- (4) Removal efficiency for COD, T-N, T-P, TDS, and turbidity was over 94% in MF-RO hybrid process, implying that RO module was greatly contributed to hybrid system. The water quality of RO permeate was not restricted in environmental standard for sewage.
- (5) In the case of applying to RO module with controlling temperature and TMP, the applied pressure was independent of the system although permeability linearly increased with increasing temperature.
- (6) The low removal efficiency for T-N, T-P and TDS in MF module led to the high-concentration degree for RO module.

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References

- [1] W.M. Edmunds, Geochemistry's vital contribution to solving water resource problems, *Appl. Geochem.* 24 (2009) 1058–1059.
- [2] C.S. Ok, et al., *Water Quality and Management on Water Resources*, Daehakseorim, Seoul, 1999, pp. 14–15.
- [3] K. Hwang, Filtration characteristics and membrane fouling in cross-flow microfiltration of BSA/dextran binary suspension, *J. Membr. Sci.* 347 (2010) 75–76.
- [4] M. Abkov, Recovery of vanillin from lignin/vanillin mixture by using tubular ceramic ultrafiltration membranes, *J. Membr. Sci.* 301 (2007) 221–222.
- [5] J. Seo, Membrane choice for wastewater treatment using external cross flow tubular membrane filtration, *Desalination* 249 (2009) 197–198.
- [6] A. Rahimpour, Development of high performance nano-porous polyethersulfone ultrafiltration membranes with hydrophilic surface and superior antifouling properties, *Appl. Surf. Sci.* 255 (2009) 9166–9168.
- [7] M. Ulbricht, Advanced functional polymer membranes, *Polymer* 47 (2006) 2218–2219.
- [8] M. Sairam, Spiral-wound polyaniline membrane modules for organic solvent nanofiltration (OSN), *J. Membr. Sci.* 349 (2010) 124–125.
- [9] A. Nordin, Influence of module configuration on total economics during ultrafiltration at high concentration, *Chem. Eng. Res. Des.* 88 (2010) 1555–1556.
- [10] J.R. Park, Operation and Cleaning Characteristics of Submerged Membrane Bio-Reactor for Advanced Wastewater Treatment, Ph.D. dissertation of Hongik University, 2005, pp. 17–19.
- [11] S. Mondal, Generalized criteria for identification of fouling mechanism under steady state membrane filtration, *J. Membr. Sci.* 344 (2009) 6–7.
- [12] M. Mota, Influence of cell-shape on the cake resistance in dead-end and cross-flow filtrations, *Sep. Purif. Technol.* 27 (2002) 138–139.
- [13] D. Wirth, Water desalination using membrane distillation: Comparison between inside/out and outside/in permeation, *Desalination* 147 (2002) 139–140.
- [14] K.R. Ha, Comparison of Submerged/Pressurized Type Membrane System by Long Term Results in MF Drinking Water Treatment, Thesis of Sungkyunkwan University, 2007, pp. 17–26.
- [15] F. Meng, Recent advances in membrane bioreactors (MBRs): Membranefouling and membrane material, *Water Res.* 43 (2009) 1491–1492.
- [16] K.H. Lee, H.S. Choi, Sewage treatment using immersed MF and RO membrane, *J. Korean Soc. Environ. Technol.* 12(3) (2011) 189–194.
- [17] T.R. Jang, Study of Effects of Various Pretreatments on Membrane and Backwash Water Recovery System in the Immersed Membrane, Thesis of Konkuk University, 2007, pp. 13–14.
- [18] S.H. Joo, J.M. Park, Y.W. Lee, Evaluation on removal of organics and nutrients from reverse osmosis concentrate using activated carbon, *J. Korean Soc. Water Environ.* 28(3) (2012) 479–482.