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Treatment of reverse osmosis concentrate by electrolysis and MBR process

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ABSTRACT

In this study, the reverse osmosis (RO) concentrate treatment technology, which combines the electrolysis process with the MBR process, has been suggested to treat the RO concentrate generated in the water recycle process, which applied the RO membrane. The electrolysis process constructed in this study is to reduce the MBR process load by removing the non-degradable matters and the nitrogen compound, which are the weakness of biological treatment. When treating the RO concentrate through the electrolysis, the disinfection by-products—like residual chlorine, THMs and so forth—are generated. Since this reaction of the by-products affects the microbes, PAC was injected within the MBR reactor to desalinate the treated water. As the MBR process shows considerably high treatment efficiency for the organic matters and can increase the concentration of the activated sludge within reactor, it is effective to treat the nitrogen and phosphorus, which cause the eutrophication of water, but since there is a limit in treating the RO concentrate with this single process, such a hybrid process was suggested. The comparative experiment was performed depending on whether the electrolysis-applying pretreatment of RO concentrate has been applied or not, and whether the PAC has been injected within MBR or not. As a result, the DOC, T-N and T-P removal efficiency of the suggested process was each approximately 98.0, 40.0 and 45.6%, respectively. The experiment results showed that the suggested process has great potential in RO concentrate treatment.

Keywords: Membrane bioreactor; Electrolysis; Wastewater reclamation; Reverse osmosis concentrate; Powdered activated carbon

1. Introduction

By the influence of the global warming and the water pollution, the phenomenon of water shortage occurs seriously, and accordingly the interests in solving the water shortage and developing new water resources in on the increase. Therefore, the water reuse has gained lots of attention in the aspects of securing additional water sources and energy saving. Recently, applying the membrane technology such as ultrafiltration, reverse osmosis (RO), etc. to the water reuse process are getting in the limelight. In the wastewater reuse field, applying the membrane of RO is a form derived from the seawater desalination process, and the active researches about applying the membrane of RO are under progress all over the

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world. When the RO process is applied to the wastewater reuse system, it has the advantage to obtain highly pure water, but the RO concentrate treatment problem follows inevitably [1].

In case of RO concentrate, it has the characteristics of having low concentration of carbon organic matter and high concentration of salt and nitrogen. Being different from the general urban sewage or industrial wastewater, in which the nitrogen is composed mostly of the ammonia nitrogen, the nitrogen in the RO concentrate is composed of the ammonia nitrogen and the nitrate nitrogen in similar proportion, or in the slightly higher proportion of the nitrate nitrogen. As for this reason, when it is biologically treated, the microbes may extinct due to the lack of organic matters, and the treatment efficiency is reduced due to the lack of organic matters in the denitrification stage [2]. In addition, in the microbiological aspect, the wastewater containing high salt concentration causes the plasmolysis in the microbes leading to the dysfunction in the cell, and eventually to the destruction of cell [3].

The purpose of this study is to safely treat RO concentrate, which has been pointed out as a flaw when designing the RO membrane-applying wastewater reuse process, through the combination of electrolysis and MBR process. Electrolysis, as a pretreatment, is to oxidize non-degradable organic matters, to reduce of nitrate nitrogen and to gain the effect of disinfection. Electrolysis is considered to be effective in the treatment of RO concentrate because, in the case of RO concentrate, TDS does exist to a degree, therefore conductivity being also present, and thereby it is possible to treat it at a relatively low voltage. The activated sludge in the MBR removes the organic matter in the water by the reaction of microbes and PAC biologically removes the organic matter by physicochemical adsorption and by the biofilm generated on the membrane surface. Moreover, when the PAC cannot absorb organic matters anymore, its absorption ability is reclaimed by attached microbes, and thereby the removal of organic matters is sustainable. For such reason, the PAC-added MBR process is considered to contribute to the efficient and safe removal of organic matters, nitrogen and phosphorus.

Hence in this study, the artificial RO concentrate was made from the RO concentrate discharged in MBR-RO process, which is actually being used in wastewater reuse pilot plant. First, non-degradable organic matters and nitrogen in the artificial RO concentrate were treated by the pretreatment of electrolysis. Then, the raw water mixed from the artificial wastewater and the artificial RO concentrate in the proportion of 1:1 was inpoured into the PAC-added MBR process, and through this process, the removal characteristics of organic matters, nitrogen and phosphorus were evaluated. Furthermore, a comparison experiment was implemented by operating the MBR process without PAC addition under the same conditions.

2. Materials and methods

In this study, the experiment was conducted by making artificial wastewater and artificial RO concentrate for the constant condition of the raw water. The artificial wastewater was made based on the property of the raw water flowed into the wastewater treatment plant of the local government of Gyeonggi-do, and the artificial RO concentrate was made based on the property of the RO concentrate discharged into the MBR-RO pilot plant installed at different wastewater treatment plants of the local government. The major characteristics of the influent water are shown in Table 1.

The MBR system used in the experiment is composed of 1st anoxic, 2nd anoxic, anaerobic and oxic reactors, and the membrane was installed in the oxic reactor. The membrane used in the experiment is a submerged-type plate membrane of polyvinylidene fluoride (PVDF) material and two frames are used within the membrane reactor. In the membrane reactor, when air is flowed into the posterior-end reactor for air washing of membrane, DO concentration increases. Since the denitrification efficiency was reduced in the anoxic reactor, and as the anaerobic condition is not made, the Luxury uptake was disturbed. To minimize such phenomenon, the Anoxic reactor was dualized. In addition, in order to prevent the reduction of the treatment efficiency in MBR process by the disinfection by-products of the water treated by electrolysis, 3.25 g/L of PAC—which is same proportion with MLSS concentration—was added. To examine the effects of PAC, the two MBR processes were operated under the same condition and the experiment was conducted with MBR 1 with PAC and MBR 2 without PAC. Thereby, the initial MLSS concentration of MBR1 and MBR2 was approximately 6,500 and 3,740 mg/L, respectively. PAC used in experiment was the KB-B model and has internal surface area of 500–1,000 m^2/g. PAC was used after placing it in the distilled water, then cleaning it by stirring and then leaving it for 24 h. Detailed operation conditions are shown in Table 2 (Fig. 1).

The electrolysis system used in experiment consists of electrolysis reactor, power supply and electrodes. In addition, since sufficient stirring effects within the reactor cannot be expected only with the bubbles generated during electrolysis, the agitator was installed at the centre of reactor (Table 3). The electrodes were

Table 1
Characteristics of the synthetic water and the synthetic RO concentrate

Item	Average value	
	Synthetic wastewater	Synthetic RO concentrate
T-N, mg/L	37.5	47
T-P, mg/L	5.0	1.5
Cl^-, mg/L	0	300
TDS, mg/L	350	745
DOC, mg/L	227.5	17.5
UV_{254}, abs	0.022	1.25
SUVA	0.01	7.0
pH	7.79	6.79

Table 2
MBR system operating characteristics

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
PAC concentration in MBR 1, g/L	5
Influent flowrate, L/d	17.3
Flow direction: Outside → In	
Return sludge (% influent)	
Small anoxic reactor, %	100
Medium anoxic reactor, %	100
SRT, d	50
Permeate flux, $m^3/m^2/d$	0.3
Time: On/OFF, min	9:1

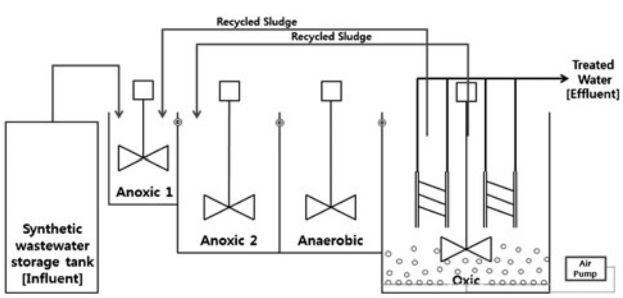

Fig. 1. Diagram of MBR system.

insoluble using Ti/IrO_2 for anode and Ti for cathode, and total eight electrodes were installed; four electrodes at each side of the agitator. The distance between the electrodes was set to 1 cm. The reaction time of electrolysis is based on one hour since the batch test showed that the oxidation of organic matters and oxidation and reduction of nitrogen compounds are finished within one hour. Detailed operational condition is shown in Table 4 (Fig. 2).

The experiment has been conducted for 55 d and the raw water, mixed of the artificial wastewater and the artificial RO concentrate with the same ratio, was used. For the RO concentrate, the changes in the water quality of RO concentrate and the impact on the MBR process were evaluated depending on the presence/absence of the pretreatment process by electrolysis. So as to secure the organic matter required for the growth of microbes, RO concentrate was mixed with the wastewater.

To reduce the load on the microbes caused by the disinfection by-products, which are generated by the electrolysis, PAC was added in the MBR process and the behaviour characteristics of the microbes were observed depending on the PAC addition.

The raw water and the treated water were analysed, and the measurement items were T-N, T-P, DOC and UV_{254}. MLSS was measured using GF/C filter after drying more than 2 h at 105˚C. The analysis methods for each item are shown in Table 5.

3. Results and discussion

3.1. MBR operation characteristics

In this study, the operation time of MBR process was 55 d. To stabilize the microbes within MBR process, MBR was operated only with the artificial wastewater (1–7 d) and with the raw water mixed with RO concentrate and the wastewater by 1:1 (8–27 d) without pretreatment. Later on, to recover the activity of the microbes, MBR was operated using artificial wastewater (28–34 d), and after recovering the activity of the microbes, MBR was operated with the raw water mixed of the RO concentrate pretreated with electrolysis with the wastewater by 1:1 (35–55 d). The changes in MLSS concentration during the operation are shown in Fig. 3. The mean MLSS concentration in the MBR1 (with PAC) and MBR2 (without PAC) were approximately 9,975 and 5,427 mg/L, respectively, and when inpouring the artificial wastewater only, MLSS concentration was maintained relatively constant. When the raw water mixed of the RO concentrate with the wastewater by 1:1 was inpoured (8–27 d), the mean concentration of MBR1

Table 3
Characteristics of membrane

Membrane: TORAY (Japan)	
Shape	Flat type
Pore size	0.08 μm
Material	PVDF
Filtration flux	0.2–1.5 m^3/m^2 d
pH	5–10
Temperature	5–4℃
Membrane area	0.03 m^2
Dimensions	130 W × 130H × 7T

Table 4
Electrolysis system operating characteristics

Parameter	Set value
Total volume, L	3
Electrode type	
Anode	Ti/IrO_2
Cathode	Ti
Amount of electrodes, ea	8
Total area of electrodes, cm^2	2,520
Current density, A/cm^2	0.05
Total HRT, h	1

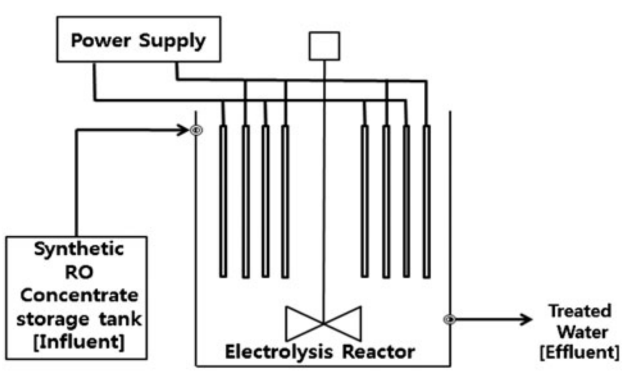

Fig. 2. Diagram of electrolysis system.

and MBR2 were reduced by approximately 39.5 and 41.5%, respectively. When the raw water mixed of the RO concentrate pretreated with electrolysis with the wastewater by 1:1 inpoured, the MLSS concentrations in the MBR1 and MBR2 were reduced by approximately 27.6 and 29.6%, respectively. When the RO concentrate is pretreated with electrolysis, the decreasing rate of the MLSS concentration was by approximately 11.9% lower than if not pretreated with electrolysis.

The MLSS concentration is one of the indexes representing the activity of the microbe, and in case of not applying with the electrolysis, MLSS has more reduced than the MLSS applying with the electrolysis. It is deemed that the influence of the RO concentrate on the activity of the microbe is greater than the influence of the disinfection by-products and the residual chlorine. In addition, in case of not applying the electrolysis, the MLSS concentration showed a continual decrease tendency, even though a decrease range difference was present. On the other side, it represented the relatively great reduction in the early stage of operation, but the longer the MBR was operated, the more it was maintained gradually stable. This means that microbes adapted relatively easily to the influent raw water with pretreatment, and thus the number of survived microbes was increased. In addition, it was observed that the MLSS decrease amount in the MBR1 with PAC was 2% lower compared to MBR2 without PAC, regardless of electrolysis.

Irrelevant from whether PAC was added or not, the first reason why MLSS decreases is because ammonia nitrogen and usable organic matters in the RO concentrate, which are needed in the nitrification, is substantially low. The second being, when the nitrification occurs, heavy metals, toxic chemicals, biological toxicity which is a growth-restricting factor, are much contained in RO concentrate. It is considered that to solve this, the mix rate of wastewater and RO concentrate should be changed or solved through the injection of an external carbon source. The activated carbon is the best available treatment recommended by US EPA [4] to control the disinfection by-products such as THMs, HAAs, etc. When comparing it to other treatment technologies such as cohesion, oxidation, ion exchange, etc. the activated carbon absorption is relatively effective in treating the raw water contained the disinfection by-products. It is reported that the method of adding PAC directly into the MBR process, the removal of organic matter occurs by the absorption of PAC, and then in the biological stage, the non-degradable organic matters are removed in a high rate [5,6]. Furthermore, the increase of microbial activity and the absorption effect of soluble microbial products, and the increase of biolysis through this method are also reported [7–9]. It is deemed that the PAC is useful for the activity of the microbe besides removing disinfection by-products and residual chlorine (Fig. 4).

Table 5
Methods of analysis for each parameter

Parameter	Method	Equipment
T-N	Chromotropic acid method	HACH, DR 2800
T-P	Molybdovanadate method	HACH, DR 2800
DOC	Infrared spectrophotometer	AURO'RA®, TOC Analyzer
UV_{254}	Ultra violation spectrophotometer	SHIMAZU, UV 1800
NH_3-N	Nessler method	HACH, DR 2800
NO_2^--N	Diazotization method	HACH, DR 2800
NO_3^--N	Chromotropic acid method	HACH, DR 2800
pH	pH meter	Thermo, Orion 3star

3.2. Removal characteristics of electrolysis on the RO concentrate

The RO concentrate was pretreated under reaction time of 1 h and the current density of 0.05 A/cm^2, and its results are shown in Table 6. When electrolyzing the RO concentrate, the reduction rates of the DOC and UV_{254} were approximately 16.6 and 31.8%, respectively, and Specific Ultraviolet Absorbance (SUVA) was reduced by approximately 18.0%. This can be considered that the decomposition of the non-degradable organic matters occurred changing them into the biodegradable matters. UV_{254} is the index representing the characteristics of the carbon compound. Since, when the non-degradable matters are much contained, it represents the high absorbance, it is used as the index of evaluating the dissolved organic matters [10]. Applying DOC and UV_{254}, SUVA represented the characteristics relation of dissolved organic matters, and its definition and equation are as follows.

$$\text{SUVA} = \text{ABS/DOC} \times 100\,(\text{cm}^{-1}\text{of absorbance per mg/L of DOC}) \quad (1)$$

If the SUVA is 4 or higher, it means that the unsaturated carbon bonds per dissolved organic carbon are many and it represents that the DOC has complex carbon bonds and higher aromaticity, and that the proportion of the hydrophobic materials including humic acid is relatively high. In contrast, if SUVA is 3 or less, it represents that the DOC is composed of saturated aliphatic series of carbon, lots of biodegradable organic carbon compounds are contained and the proportion of hydrophilic materials among the dissolved organic matters is high [11–14] (Fig. 5).

When pretreating with the electrolysis, it showed about 43.2% of T-N removal rate. T-N was removed by the oxidation reaction of the ammonia nitrogen for 30 min in the initial stage, but after 30 min, T-N was removed by the reduction reaction by the nitrate nitrogen. However, the removal rate of nitrate nitrogen was approximately 16.9%, representing relatively low removal rate. Nitrate nitrogen is removed by reduction reaction on the cathodes.

In case of T-P, it represented about 42.6% of removal rate. It is deemed that T-P was removed as the metal ion in the RO concentrate reacted with the phosphorus having characteristics to form the metal oxide and precipitate easily. Generally, the phosphorus removal through the electrochemical method is known that chemical removal occurs by combining the metal ion eluted from the electrode with the

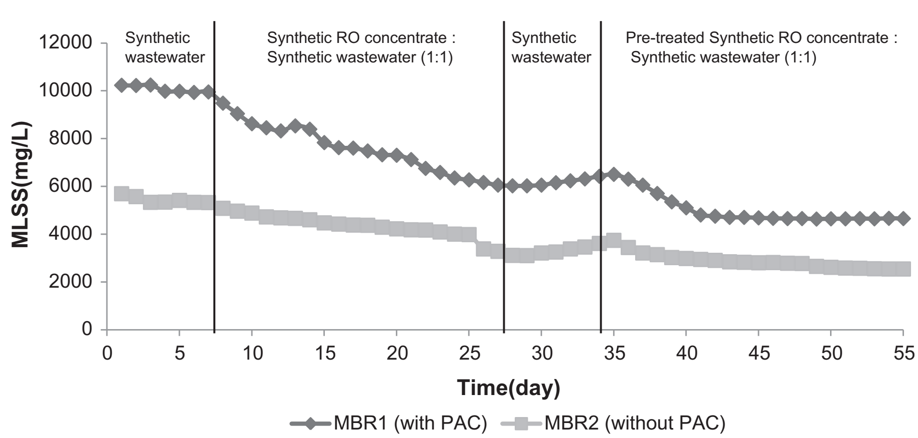

Fig. 3. Changes in MLSS concentration according to MBR operation time MBR.

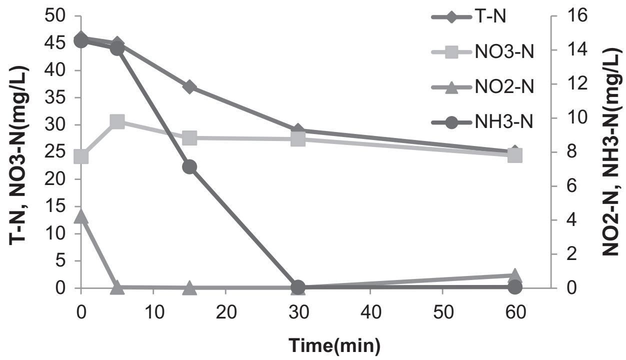

Fig. 4. Nitrogen compound removal characteristics by electrolysis.

Table 6
Changes in the ingredients of RO concentrate using electrolysis

Parameter	Influent	Effluent	Removal ratio (%)
DOC, mg/L	17.5	14.6	16.6
UV_{254}	1.235	0.842	31.8
SUVA	7.036	5.767	18.0
pH	7.11	7.36	–
Temperature, ℃	26.4	34.5	–
T-N, mg/L	47	26.7	43.2
NH_3-N, mg/L	14.55	0.07	99.5
NO_2-N, mg/L	4.238	0.753	82.2
NO_3-N, mg/L	24.2	20.1	16.9
T-P, mg/L	1.55	0.89	42.6

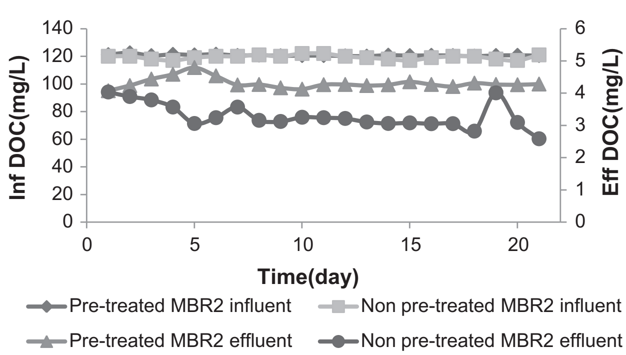

Fig. 5. Changes in DOC concentration of MBR depending on the application of electrolysis.

dissolved phosphate ion during the electrolytic reaction when soluble electrodes such as Al, Fe, etc. are used [15]. However, such reaction is deemed to occur because diverse heavy metal ions were already contained in the RO concentrate, although the insoluble electrodes were used in this study.

3.3. MBR removal characteristics according to application of electrolysis

In this study, in order to find out what influence the electrolysis has in the MBR operation, the removal characteristics of the MBR process excluding the activated carbon were examined by applying the electrolysis as pretreatment of MBR process. In case of organic matter, the mean removal rate in MBR2, in which the electrolysis was not performed, and in the MBR2, in which the electrolysis was performed was approximately 97.2 and 96.4%, respectively. Although the DOC removal rate of the process without pretreatment was 0.8% lower than the process with pretreatment, the SUVA of the treated water was analysed higher. It is deemed that lots of non-degradable matters were contained in the effluent from MBR without pretreatment, and in case of the effluent from MBR with pretreatment, the non-degradable matters were converted into biodegradable matters (Fig. 6).

In case of T-N, the removal rate of MBR2 with electrolysis and of MBR2 without electrolysis was approximately 30.5 and 25.5%, respectively. The removal rate of the MBR2 with the electrolysis was 5.0% higher than the rate of MBR2 with the electrolysis, but it is regarded as a result of the MLSS concentration difference in the MBR.

Although the removal rate of MBR with pretreatment is lower than the removal rate of MBR without pretreatment, the mean T-N concentration of the effluent from MBR2 without pretreatment and of the effluent from MBR2 with pretreatment was approximately 31.2 and 24.1 mg/L, respectively. The concentration of the effluent from MBR with pretreatment is observed to be lower by mean 7.1 mg/L. This is considered to be due to the T-N concentration difference of the influent from MBR, which is caused by the

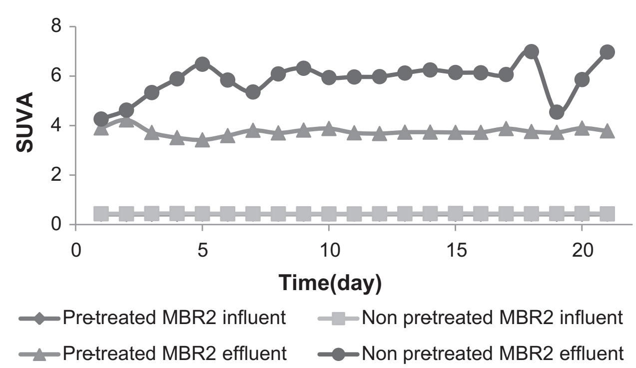

Fig. 6. Changes in SUVA of MBR depending on the application of electrolysis.

removal of ammonia nitrogen and nitrate nitrogen through electrolysis (Fig. 7).

It was observed that, in the early stage of operation, the removal rates of MBR with the electrolysis and that without the electrolysis were unstable. However, the T-N removal efficiency of the MBR with the electrolysis was relatively stable after the microbes were stabilized by adapting to the raw water. On the other side, it was observed that the T-N removal efficiency of the MBR without the electrolysis was still unstable. If the denitrification is to occur efficiently as part of the biological nitrogen removal, biodegradable organic matters are needed. However, it is deemed that the removal efficiency of T-N is unstable because the proportion of non-degradable organic matters was high in the effluent from the MBR without pretreatment.

In case of T-P, the removal rates of MBR2 without electrolysis and of MBR2 with electrolysis were approximately 26.5 and 20.9%, respectively. Generally, the treated water quality of MBR effluent was unstable. It is deemed that only applying the electrolysis pretreatment has no great impact on the removal efficiency of phosphorus in MBR process (Fig. 8).

3.4. Removal characteristics of MBR according to PAC addition

RO concentrate removal characteristics depending on the PAC addition in the MBR were examined when the RO concentrate pretreated with electrolysis was inpoured into the MBR. In case of DOC, the removal rates for MBR1 and MBR2 were approximately 98.0 and 96.4%, respectively. The MBR1 with PAC was treated more stably than MBR2 (Fig. 9). In addition, in case of SUVA, which represented the characteristics of non-degradable organic matters, MBR1 showed the stable removal efficiency as shown in Fig. 10. It is deemed that the biodegradable matters removed by absorption to PAC were converted from the non-degradable organic matters, and as the SUVA was less than 3 L/mg m, mainly the low molecular weight and hydrophilic organic matters were remained in the water [16].

In case of T-N, the removal efficiency of MBR1 and MBR2 was approximately 40.4 and 25.5%, respectively. The concentration of T-N in the effluent during early stage of operation was unstable as shown in Fig. 11,

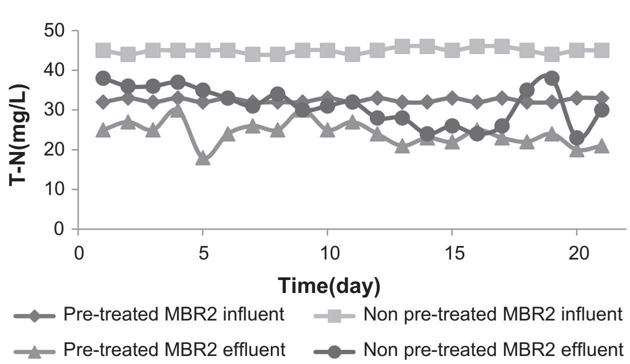

Fig. 7. Changes in T-N concentration of MBR depending on the application of electrolysis.

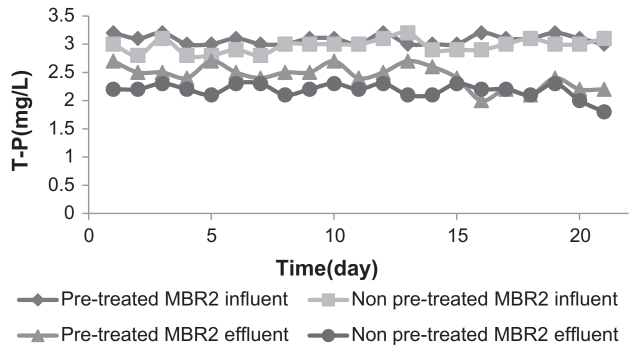

Fig. 8. Changes in T-P concentration of MBR depending on the application of electrolysis.

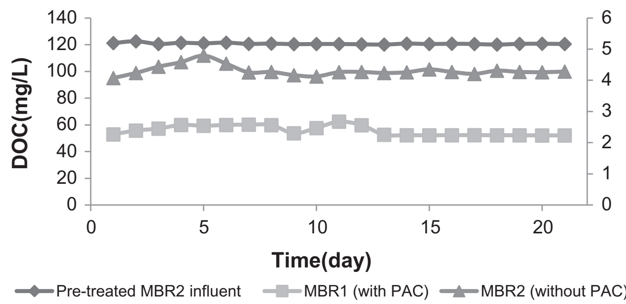

Fig. 9. Changes in DOC concentration of MBR depending on PAC injection.

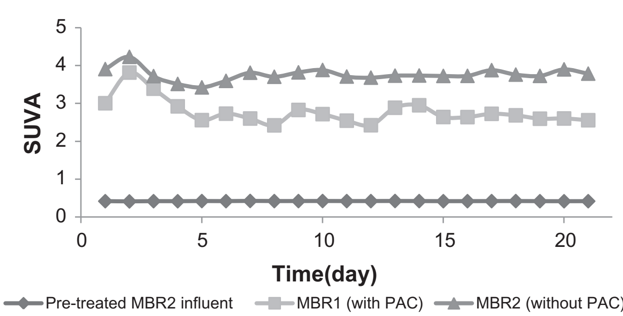

Fig. 10. Changes in SUVA of MBR depending on PAC injection.

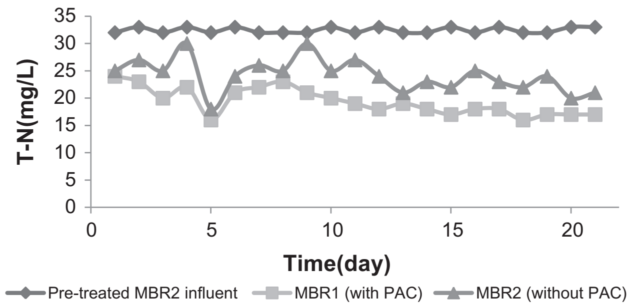

Fig. 11. Changes in T-N concentration of MBR depending on PAC injection.

which is deemed that the nitrification and denitrification were disturbed since the activity of the microbes dropped by the residual chlorine and disinfection by-products. However, it was observed that it was treated stably as the microbes adapted to the influent in the course of time. The removal efficiency of MBR1 with PAC was inclined to be stabilized more quickly than MBR2. It is deemed that the PAC influenced on the stabilization of the microbes by adhering and removing the disinfection by-product, residual chlorine, etc. The longer the MBR was operated, the more the removal rate of each reactor was restored, increasing to approximately 48.5 and 36.4%, respectively.

In case of T-P, the removal rate of MBR1 and MBR2 showed approximately 45.6 and 20.9%, respectively. As shown in Fig. 12, the treated water quality of MBR2 was constantly unstable, while the treated water quality of MBR1 was relatively stable. T-P removal efficiency of MBR1 was restored to the level in which only the artificial wastewater was inpoured. It reported that the presence of readily biodegradable COD (RBDCOD) had a crucial effect on the reaction for the biological phosphorus removal [17]. As a result of the role of PAC adsorbing non-biodegradable organic matter, the biodegradable portion increased in MBR1. And this can have a positive effect on the activity of PAOs, resulting in the better removal efficiency for T-P.

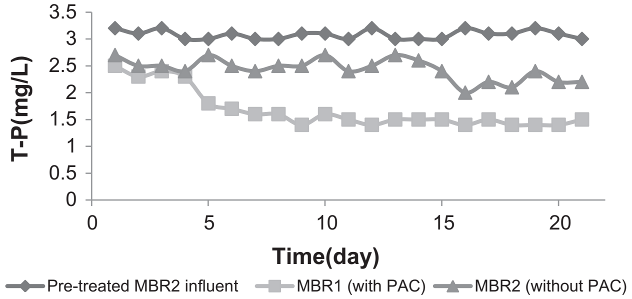

Fig. 12. Changes in T-P concentration of MBR depending on PAC injection.

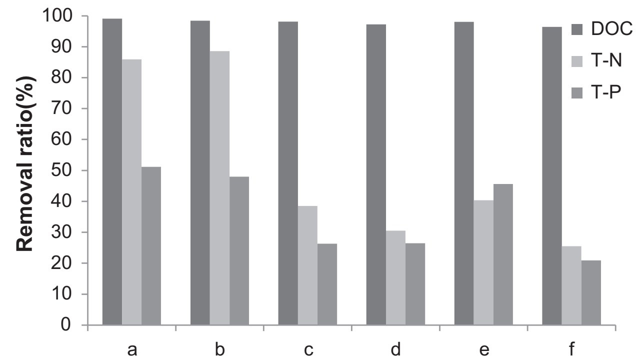

Fig. 13. Removal Efficiency of MBR Process depending on the raw water (a: wastewater—MBR1, b: wastewater—MBR2, c: wastewater + RO concentrate (1:1)—MBR1, d: wastewater + RO concentrate (1:1)—MBR2, e: wastewater + pretreated RO concentrate (1:1)–MBR1, f: wastewater + pretreated RO concentrate (1:1)—MBR2).

3.5. Removal characteristics of MBR depending on the raw water

The removal characteristics of MBR depending on the raw water are shown in Fig. 13. The main purpose of this study is to treat RO concentrate by the MBR process added with PAC and electrolysis (e). When comparing this process (e) and MBR process for the wastewater treatment (b), the difference in the removal rates of DOC, T-N and T-P was approximately −0.3, −44.2 and −2.4%, respectively. In case of DOC and T-P, a relatively similar removal rate was shown, but the removal rate of T-N was lower. However, in case of the MBR process to treat the RO concentrate, it represented that the T-N removal rate was higher by approximately 2–15%, and that the T-P removal rate was higher by 19–25%, compared to the other processes. In addition, since the disinfection by-product and non-degradable matters can be removed, it is deemed to be able to operate stably.

4. Conclusions

In this study, the nitrogen, phosphorus and organic matters removal characteristics of MBR were evaluated by inpouring the artificial RO concentrate made by simulating the RO concentrate generated from MBR-RO process for wastewater reuse. The treatment of RO concentrate was implemented by PAC-added MBR process with pretreatment of electrolysis. The conclusion is as follows.

(1) As a result of observing the changes of MLSS in the MBR process depending on the electrolysis and PAC addition, it is confirmed that the characteristics of RO concentrate has a greater negative influence on microbes activity than it has on the disinfection by-products and residual chlorine.
(2) When the artificial RO concentrate was pretreated with electrolysis, it was observed that the non-degradable organic matter was decomposed into biodegradable matters. In case of T-N, the ammonia nitrate was mostly removed, but the nitrate nitrogen removal rate was low. In case of T-P, it was removed as it reacted with metal ions in the RO concentrate.
(3) In the comparison experiment between MBR with PAC and MBR without PAC, the removal rate of the MBR with PAC was higher and was stable in the aspect of microbe activity. Furthermore, it was confirmed biodegradable matters, which were decomposed from the non-degradable organic matters by the electrolysis, were adhered and removed by PAC, and thus increased the removal rate of the hydrophobic materials of the high molecular weight.
(4) Observing the removal characteristics of MBR depending on the raw water, in the comparison of DOC and T-P removal rate of the suggested process and the removal rate of the MBR process for treating wastewater, there was no significant difference. However, in order to improve the removal of nitrogen compound, it is certain that additional research is needed.

Acknowledgements

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