

평막을 이용한 침지형 막여과시스템에서 고농도 분말활성탄 주입에 의한 수처리성능 개선 효과

The Effects of PAC (Powdered Activated Carbon) on Water Treatment Performance of an Immersed Membrane System Using Flat-sheet Membrane Module

가이상취안 · 김한승*

Xiang-Juan Gai · Han-Seung Kim*

명지대학교 환경생명공학과

(2007년 1월4일 논문 접수; 2007년 3월 5일 최종 수정논문 채택)

Abstract

A submerged flat-sheet membrane separation system integrated with PAC (powdered activated carbon) was used in this research in order to investigate the effects of PAC on the efficiencies of operation and treatment and to evaluate the performance of the system. The experiments were carried out under operating conditions of a filtration rate of 0.38 m/d, water temperature of 20±28°C, and PAC dose of 0 g/L (Run-A) and 20 g/L (Run-B). The influent concentrations of TOC (total organic carbon), NH₄⁺-N (ammonia nitrogen) and UV₂₅₄ (UV absorbance at 254 nm) were 2.48 mg/L, 1.4 mg/L and 2.53 1/m, respectively. TOC removal of 43.2 and 73.6%, ammonia nitrogen removal of 4.9 and 15.9%, and UV₂₅₄ removal of 20.6 and 31.6% were obtained for Run-A and Run-B, respectively. During an experimental period of 33 days, no change was found in TMP (Run-B), but the TMP in Run-A increased by 5 kPa after 29 days. This research showed that the filtrate quality and the performance efficiency were enhanced when PAC was introduced into the filtration system.

Key words: Flat sheet membrane; Microfiltration; Organic removal, PAC; Reuse

주제어: 평막, 정밀여과, 유기물 제거, 분말활성탄, 재이용

1. Introduction

Treatment processes for wastewater reclamation are selective depending on the purpose of water reuse and the

required water quality. Commonly, the reclaimed water quality is established in terms of BOD or COD, turbidity, color, odor, coliform bacteria and viruses (Seo, et al., 1996). Membrane separation technology, which is increasingly adopted in the field of water and wastewater

*Corresponding author Tel: +82-31-330-6695, FAX: +82-31-336-6336, E-mail: kimhs210@mju.ac.kr (Kim, H.S.)

treatment, has shown a good performance in removing these kinds of contaminants. Membrane filtration systems would require either nanofiltration or reverse osmosis membrane for meeting the water quality in wastewater reuse. However, the need of severe pretreatments has hitherto rendered this approach uneconomical and impractical (Bjarne Nicolaisen, 2002). The MF (microfiltration) membrane with a nominal pore size of 0.2 μm could eliminate all fecal germs and ascarid eggs. COD and BOD reduction could be also achieved in MBR systems over sixty percent, but color was not eliminated sufficiently (Seo, et al., 1996). This says that although MF or UF membrane systems showed almost perfect removal for turbidity or colloidal particles larger than the membrane pore size, it has a limitation that it cannot remove contaminants which are smaller than the membrane pore size. In order to get a better removal performance, a hybrid process of membrane filtration and activated carbon adsorption was proposed. Activated carbon has effective roles to control fouling on the membrane as well as to remove the dissolved organic contaminants from natural water. And the membrane plays as a barrier to reject the particulate matter including activated carbon.

The capability of the hybrid system combining PAC (powdered activated carbon) and hollow fiber MF membrane was investigated for further removal of organics and pathogenic microorganisms by feeding synthetic secondary sewage effluent (Kim, et al., 2005). The hollow fiber membranes are reported to have some difficulty in handling and high cost in membrane replacement, compared to that of flat sheet membrane. Moreover, its small tube diameters make the fibers somewhat susceptible to plug the cartridge inlet (Munir Cheryan, 1998). Hal et al. (2005) compared the different fouling modes between the submerged hollow-fiber and flat-sheet membranes induced by high strength wastewater with concurrent biofouling, and found rather different performances of the membrane.

This study investigated the effects of the addition of powdered activated carbon to the hybrid system using a

Table 1. Components of raw water

Organic/inorganic matter	Concentration
Polypeptone, mg/L	10
Yeast extract, mg/L	5
Glucose, mg/L	1.5
NaCl, mg/L	5
MgSO ₄ · 7H ₂ O, mg/L	0.2

flat-sheet membrane on improving the treated water quality for water reclamation or reuse.

2. Materials and method

2.1. Raw water

The compounds and concentration of the raw water used in this study are listed in **Table 1** (Kim, et al., 2001). Five stock solutions (concentrated 1000 times) were made for five compounds, respectively, and were stored in a refrigerator before use. Each stock solution of adequate quantity (20 mL) was introduced into a raw water tank (20 L) for dilution and mixing in order to obtain the final concentration listed in **Table 1**.

2.2. Powdered activated carbon (PAC)

The PAC used as adsorbent in the experiments was obtained from Osung Envitech (Korea). Darco KB-B was used in this experiment. The particle size range of the PAC is between 100 and 325 mesh, and the surface area is approximately 1500 m²/g.

2.3. Membrane

Flat sheet membrane modules were obtained from Kored Inc. (Korea). The membrane has nominal pore size of 0.2 μm and an effective area of 0.04 m². The material of the membrane was polyethersulfone (PES). The dimension (mm) of the membrane module is 150Wx300Hx6T, and the operating method is suction type with operating pressure of -0.6~0 kgf/cm²,

2.4. System description

The PAC-MF system consisted of a combination of PAC and MF as shown in **Fig. 1**. The raw water was

prepared by diluting and mixing the stock solutions in a raw water tank. This was then fed into the reactor by a peristaltic pump. And the water was filtered periodically by a suction pump. The water level of the filtration tank was maintained using a level sensor which controlled the influent pump and suction pump to keep the water volume constant in the reactor. Moreover, a pressure gauge was used to measure TMP (transmembrane pressure). In order to fluidize PAC and to prevent the accumulation of PAC onto the membrane, aeration was applied at the rate of 5 L/min by an air blower through a diffuser attached to the bottom of the reactor. The experiment was carried out under ambient conditions at a temperature range 20-28°C. The PAC was placed once at the start of the experiment into the filtration tank and no additional PAC was done. The volume of the reactor was 4 L and the amount of PAC input was 80 g for Run-B, corresponding to the concentration of 20 g/L. In this experiment, Run B was with 20 g/L powdered activated carbon in the reactor and Run A was without any PAC in the reactor.

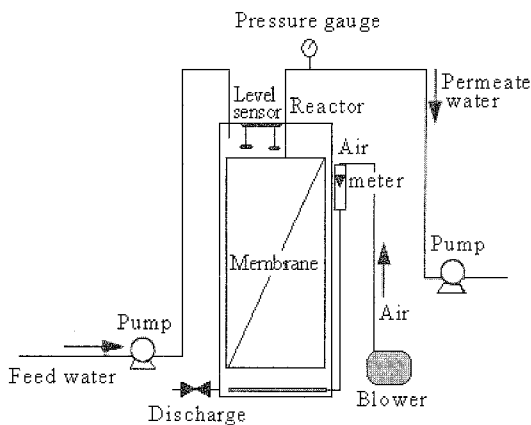


Fig.1. Schematic of the bench-scale submerged membrane system.

2.5. Analytical Methods

Samples from the influent, reactors and effluents were taken once a day during the operation period. Since the samples taken from the filtration tank contained suspended solids and/or PAC, a 0.45 μm filter was used in order to filter the samples. These were then stored in a 100 mL glass bottle, capped and put into a refrigerator prior to measuring.

The total organic carbon (TOC) was measured by using Shimadzu TOC 5000A, the UV absorbance at 254 nm (UV₂₅₄) with HACH DR/4000U spectrophotometer, the turbidity with HACH 2100N TURBIDIMETER, and the ammonia nitrogen (NH₄⁺-N) with pH ISE meter. Both the nitrite (NO₂⁻) and the nitrate (NO₃⁻) were determined using a standard method (Lenore, et al., 1998) and the HACH DR/4000U spectrophotometer. TMP and temperature were checked for each run at a scheduled time interval.

3. Results and Discussion

3.1. Raw water condition

The average concentrations of the raw water were as follows: TOC 2.48 mg/L; Turbidity 0.6 NTU; NH₄⁺-N 1.4 mg/L; UV₂₅₄ 2.53 1/m. As reported by Tao et al. (2005) in Singapore, typical MBR product quality is as follows: TOC 4.8-5.0 mg/L; Turbidity < 0.2 NTU; NH₄⁺-N 0.3-1.0 mg/L. Dockko et al. (2001) treated the wastewater using a submerged nanofiltration membrane bio-reactor (NF MBR), and obtained the following: TOC 2.0-3.5 mg/L. The objective of this study is for water reclamation and reuse. And the raw water was supposed to be made as the water quality of secondary effluent. The effluent of the MBR is one kind of secondary effluent. In this case, this system can be used to treat the secondary

Table 2. Comparison of water quality in terms of TOC, NH₄⁺-N, UV₂₅₄ and turbidity

	TOC (mg/L)	NH ₄ ⁺ -N (mg/L)	UV ₂₅₄ (1/m)	Turbidity (NTU)
Raw water in this study	1.6-5.7 (2.48)	1.4	2.53	0.6
MBR product[8]	4.8-5.0	0.3-1.0	-	< 0.2
NF MBR effluent[9]	2.0-3.5	-	-	-

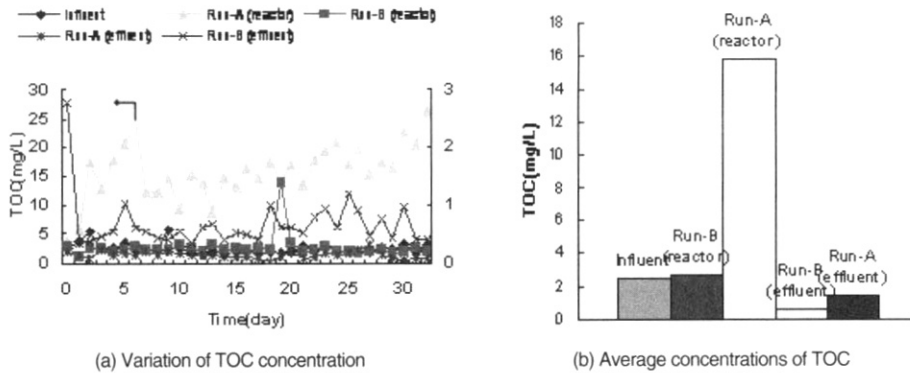


Fig. 2. Behavior of TOC concentration.

effluent from the typical MBR plants. Table 2 shows the comparison of water quality between the raw water and the effluents of MBR plants.

3.2. Transmembrane pressure (TMP)

One of the important parameters in membrane systems is TMP which increases with the operating time. TMP control is strongly related to the efficiency of the systems. Management cost can be lowered through efficient operation such as the elongation of the filtration time reaching a certain TMP (Kim, et al., 2005).

The duration of the experiment was 33 days and almost 500 L of raw water was treated. In the filtration rate of 0.38 m/d, the value of TMP did not yield any obvious increase during the whole experiment. In other words, this system can run more than one month without any problem under this condition.

3.3. The organic removal

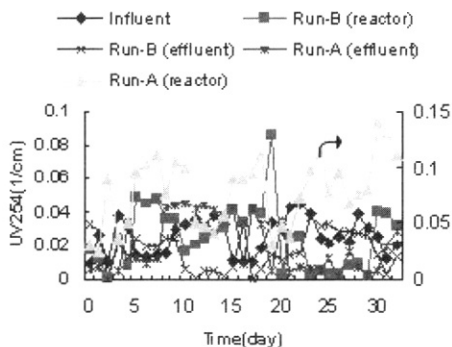
The influent concentration of TOC ranged from as low as 1.6 mg/L to as high as 5.7 mg/L and the average influent concentration of TOC was 2.48 mg/L.

The changes in the daily samples and the average values of the five samples are shown in Fig. 2. The influent concentration is not stable due to the method of making raw water (dilution and mixing) as described in 2.1. Moreover, the addition of raw water every other day may have caused particulate accumulation in the raw water tank during the two days and lead to the fluctuation.

Based on Fig. 2, it can be seen that TOC removal in Run-B was better by 30% than Run-A. The average concentration of the effluent for Run-A was 1.42 mg/L, which corresponded to a removal rate of 43.2%. On the other hand, values obtained for Run-B were 0.66 mg/L and 73.6%. This result implied flat sheet membrane system combined with PAC has similar performance in TOC reduction compared to the hollow fiber membrane system with PAC (Kim, et al., 2005).

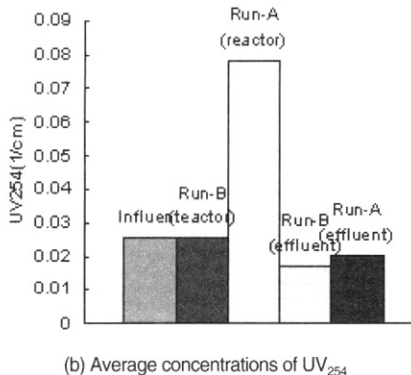
In Run-A, it was evident that TOC accumulated in the reactor during filtration. A possible reason is that the TOC was rejected into bulk rather than attach to the membrane surface. On the other hand, most of the TOC was adsorbed onto the surface of the PAC in Run-B. The capability to withstand a high loading of organic matter can be promoted by integrating PAC into the system, and that more stable and safe treatment performances can be obtained.

Fig. 3 shows the daily change of UV_{254} in the samples and the average concentrations for 33 days. The result is not obvious in Fig. 3 (a), but as shown in Fig. 3 (b), the removal in Run-B is better than in Run-A, showing a similar trend with TOC removal. The average value of the influent was 2.53 1/m. The average value of the effluent for Run-A was 2.01 1/m and the removal rate was 20.6%. On the other hand, Run-B resulted to 1.73 1/m and 31.6%. The function of PAC had also been detected for UV_{254} .

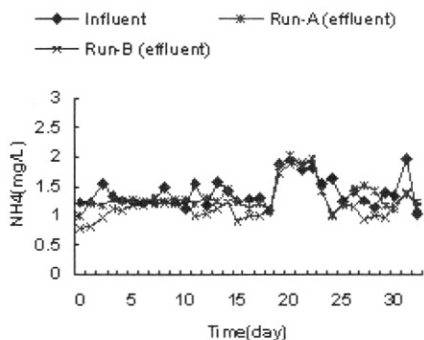


(a) Variation of UV254 concentration

Fig. 3. Behavior of UV254.

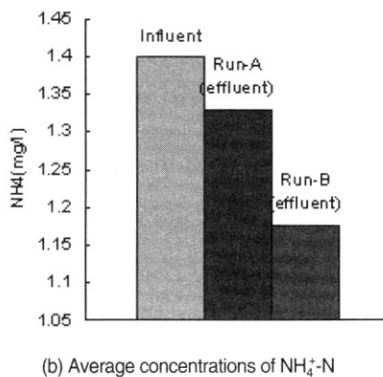


(b) Average concentrations of UV₂₅₄



(a) Variation of NH₄-N concentration

Fig. 4. Behavior of ammonia nitrogen.



3.4. Ammonia nitrogen removal

Nitrogen concentrations of ammonia, nitrite and nitrate were measured for the samples from the filtration tank as well as from the influent and the effluent. It was found that the concentration of the nitrogen compounds in the filtration tank was equal to that in the effluent for all samples throughout operation which implies that all the nitrogen compounds could not be removed by membrane rejection (Kim, et al., 2005).

Fig. 4 shows the daily change of the ammonia-nitrogen in the samples during the filtration period of 33 days. The average concentration of NH₄-N in the influent was 1.401 mg/L. The effluent concentration for Run-A was 1.331 mg/L and decrease rate was 5.0%; the values for the effluent for Run-B were 1.177 mg/L and 16.0%.

From Fig. 5, a small amount of NO₂-N was observed.

The quantity of NO₃-N didn't show an obvious change, compared with influent.

From Table 4, the average concentration of NO₂-N in influent was 0.0034 mg/L. The effluent concentration for Run-A was 0.0076 mg/L and the increase rate was 135%; the effluent values for Run-B were 0.0104 mg/L and 194%. These values show an increase, compared to the influent. The average concentration of NO₃-N in the influent was 0.356 mg/L. The effluent concentration for Run-A was 0.355 mg/L and the decrease rate was 3.3%; the effluent values for Run-B were 0.363 mg/L and 1.1%.

The ammonia oxidation was found in this experiment. Both the decreasing of NH₄-N and the increasing of NO₂-N in Run-B are higher than Run-A. In the system using a hollow fiber with high dose of PAC (40g/L) (Kim, et al., 2005), the ammonia oxidation was evident, but no differences in ammonia concentration were observed

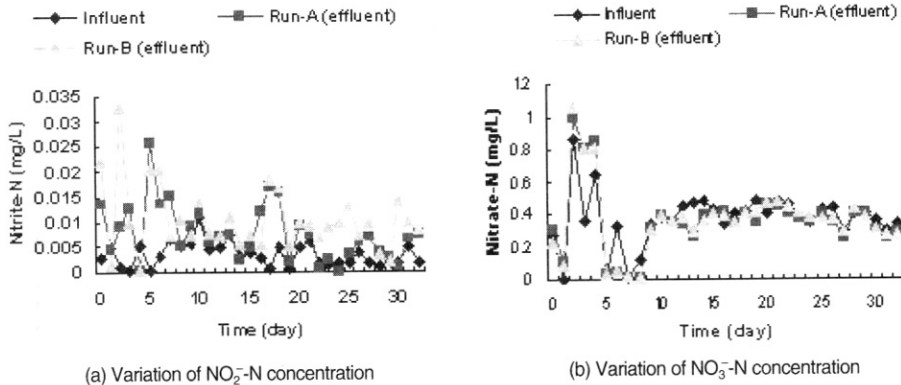


Fig. 5. Behavior of nitrite and nitrate nitrogen.

Table 3. Average concentrations and removal rates of nitrogen

	Influent	Reactor (Run-A)	Reactor (Run-B)	Effluent (Run-A)	Effluent (Run-B)	Removal (Run-A)	Removal (Run-B)
NH ₄ ⁺ -N(mg/L)	1.401	1.472	1.192	1.331	1.177	5.0%	16.0%
NO ₂ ⁻ -N(mg/L)	0.0034	0.0090	0.0097	0.0076	0.0104	(135%)*	(194%)*
NO ₃ ⁻ -N(mg/L)	0.367	0.373	0.402	0.355	0.363	3.3%	1.1%

* Increase

between the influent and effluents for both without PAC and 10 g/L of PAC. This means that the high dose of powdered activated carbon conduces to reacting of the ammonia oxidation.

3.5. Turbidity

Water clarity is important in products destined for human consumption and in many manufacturing operations (Lenore, et al., 1998). The removal of turbidity

is significant in water treatment.

Fig. 6 shows the daily change of the turbidity in the samples, yielding a better performance for Run-B than for Run-A. The average concentration of turbidity in the influent was 0.600 NTU. The effluent concentration for Run-A was 0.182 NTU and removal rate was 69.7%; the effluent for Run-B was 0.091 NTU and 84.8%.

The removals of turbidity are apparent in both Run-A and Run-B observed in the figures. The superiority of the

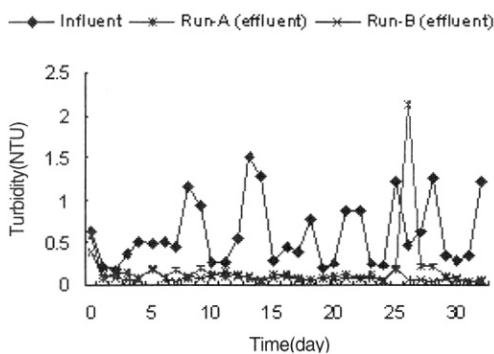


Fig.6. (a) Variation of turbidity.

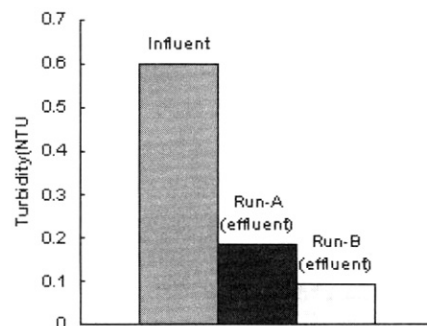


Fig.6. (b) Average turbidity.

high dosage PAC can also be seen, as PAC adsorbed some suspended and colloidal matters of small size, which can pass by the pore of membrane.

4. Conclusions

In this study, it was obviously proved that PAC acted as effective roles to enhance the operation performance and water treatment: to extend the continuous filtration time and to further remove organic and inorganic substances. The continuous filtration experiments were carried out with a PAC dosage of as high as 20 g/L for an experimental period of 33 days without any trouble. The results of this study are summarized as follows:

- The increase of TMP was reduced by introducing PAC, therefore less chemical cleaning can be provided compared to membrane system with no PAC.
- The dissolved matters as well as the particulate matters was effectively removed by integrating PAC with flat sheet membrane system;
- Ammonia oxidation was found in the reactor with PAC, and this means that PAC can provide biological activity by operation for long period;

This study showed a basic performance of the PAC combined membrane systems. The more study should be done on the mechanism of fouling mitigation by PAC and biological activity in filtration tank.) this system was found in this study.

Acknowledgement

This work was supported by 2005 Research Fund of Myongji University. And the membrane was provided by

Kored Inc. and PAC by Osung Envitech.

References

1. Gyu Tae Seo, Yutaka Suzuki, Shinichiro Ohgaki (Japan), (1996) Biological powdered activated carbon (BPAC) microfiltration for wastewater reclamation and reuse, *Desalination*, **106**, pp. 39-45
2. Bjarne Nicolaisen, (2002) Developments in membrane technology for water treatment, *Desalination*, **153**, pp. 355-360
3. Han-Seung Kim, Hiroyuki Katayama, Satoshi Takizawa, Shinichiro Ohgaki, (2005) Development of a microfilter separation system coupled with high dose of powdered activated carbon for advanced water treatment, *Desalination*, **186**, pp. 215-226
4. Munir cheryan, Ph.D. (1998) *Ultrafiltration and microfiltration handbook*, Technomic Publishing Company, Inc, USA
5. Faisal Ibney Hal, Kazuo Yamamoto, Kensuke Fukushi, (2005) Different fouling modes of submerged hollow-fiber and flat-sheet membranes induced by high strength wastewater with concurrent biofouling, *Desalination*, **180**, pp. 89-97
6. H.S. Kim, H. Katayama, S. Takizawa and S. Ohgaki, (2001) Removal of Coliphage Q β , and Organic Matter from Synthetic Secondary Effluent by Powdered-Microfiltration (PAC-MF) Process, Tel-Aviv: Membrane Technology for Wastewater Reclamation and Reuse, pp. 211-219
7. Lenore S. Clescerl, Arnold E. Greenberg and Andrew D. Eaton, (1998) Standard methods for the examination of water and wastewater (20th edition), USA
8. G. Tao, K. Kekre, Z. Wei, T.C. Lee, B. Viswanath, H. Seah, (2005) Membrane bioreactors for water reclamation, *Water Science & Technology*, **51**(6-7), pp. 431-440.
9. Seok Dockko and K. Yamamoto (Japan) (2001) Wastewater Treatment Using Directly Submerged Nanofiltration Membrane Bio-Reactor (NF MBR), Tel-Aviv Israel: Membrane Technology for Wastewater Reclamation and Reuse, pp. 21-31.