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# MBR반응기의 막오염방지를 위한 활성탄과 응집제를 이용한 전처리에 관한 연구

# Using Coagulant and Activated Carbon as Pretreatment for Membrane Fouling Control in MBR (Membrane Bioreactor)

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**Abstract**

The aim of this study was to investigate the characteristics of membrane fouling caused by soluble organic materials in a membrane bioreactor process. For the removal of filterable organic materials (FOC) smaller than 1$\mu$m, coagulants and activated carbon were added. A membrane bioreactor using a submerged 17$\mu$m metal sieve was operated in laboratory scale to examine the possibility of membrane fouling control.

As the dosage of GAC and coagulant increased, the residual FOC concentration decreased and the permeate flow rate increased markedly. The permeate flux increased with an increased PACl addition at the range from 0 to 50 mg/l. At coagulant dosage of 27mg/l, the removal of FOC was about 46% and the flux increased to 3.5 times compared to the case without PACl addition. The permeate flux increased gradually with an increase in GAC dosage. At GAC dosage of 50mg/L, the permeate flux was about 2 times higher compared that for raw water. The particle in the range of 0.1~1.0$\mu$m were removed effectively by the addition of GAC and coagulant. Higher osage of GAC and coagulant, led to higher removal of FOC.

A different set of experiments was also performed to investigate the effect of pretreatment on the permeation ability of MBR system using the metal sieve membrane. After 40 hours of operation, the permeate flux was about 1,000 (L/m$^2$-hr), which is 20 times higher compared to the results in literature. It is likely that combined pretreatment using coagulant and activated carbon was the most effective to resolve membrane fouling problems. Moreover, the continuous operations could be successful by applying this pretreatment method.

**Key words:** membrane, fouling, MBR, coagulant, activated carbon
**주제어**: 막, 막오염, MBR, 응집제, 활성탄

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## 1. INTRODUCTION

The membrane bioreactor (MBR) process eliminates the weakest link in the activated sludge process and makes effluent quality independent of settling characteristics of the biomass. Simplicity of operation, development of higher flux membranes with low fouling potentials and lower membrane costs have contributed to its world wide applications in small systems (Adham et al., 1996). In fact, membrane processes showed comparable or lower costs compared with conventional treatment for small systems of < 20,000$m^3$/d, or 5MGD (Wiesner et al., 1994).

However, MBR processes have some problems, in particular membrane fouling and concentration polarization. The origin of membrane clogging has often been based on notices of deposit or concentrated polarization layers on the surface of the membrane, notions of pore blockage due to steric overloading or progressive pore clogging. When this occurs, a thick gel layer (which can be both biological or abiotic in composition) is formed on and into the membrane causing the permeate flux to decline very fast. Especially in microfiltration and ultrafiltration, the flux decline is very severe with the process flux often being less than 5% that of the pure water flux.

Flux decline can be caused by several factors, such as concentration polarization, adsorption, gel layer formation and plugging of the pores. All these factors induce additional resistances on the feed side to the transport across the membrane. The extent of these phenomena is strongly dependent on the types of membrane process and feed solution employed.

Fouling occurs mainly in microfiltration/ ultrafiltration where porous membrane which are implicitly susceptible to fouling are used. Therefore, pressure driven processes will be emphasized but also here the type of separation problem and the type of membrane used in these processes determine the extent of fouling. Roughly three types of foulant can be distinguished; (1) organic precipitates: macromolecules, biological substances, etc. (Mukai et al., 1997; Bura et al., 1998), (2) inorganic precipitates (metal hydroxides, calcium salts, etc.), and (3) particulates (Mulder, 1996). Gan et al (1997). studied membrane fouling during the filtration of beer with inorganic membranes and concluded that the flux decreased mainly because of internal fouling caused by carbohydrates.

There have been many investigations about membrane fouling mechanisms, methods to restrict fouling, and methods to enhance the flux. To enhance organic removal and to reduce membrane fouling causing by potential hydrophobic substances, the use of PAC for raw water pretreatment or in combination with the UF system has been practiced. Some studies have indicated that the addition of PAC could increase membrane flux (Pribazari et al., 1992; Kim et al., 1996) and enhance organic removal (Adham et al., 1991).

The aim of this study is to investigate membrane fouling characteristics caused by the soluble organic materials in the membrane bioreactor system process. For the removal of the filterable organic materials (FOC) less than 1$\mu$m, coagulants and granular activated carbon were added as a pretreatment before MF filtration. As a result, MF permeate flux decline and the corresponding membrane fouling caused by the specific soluble organic matter in the absence/presence of pretreatment can be better understood.

## 2. EXPERIMENTS

For preliminary examination about the removal of FOC of feed water by PAC and coagulant dosages, some pretreatments were applied before membrane filtration tests. The additions of granular activated carbon (GAC), coagulant (PACl), and the combination of GAC + PACl were investigated under some experimental conditions. These conditions were summarized in Table 1. A stock solution for feed water was prepared by using mixed liquor of activated sludge, and it was stored at 4°C for subsequent uses. To estimate the FOC removal efficiencies by different PAC and coagulant dosages, jar

**Table 1.** Pretreatment conditions for the FOC removal

| | Experimental conditions |
|---|---|
| Run 1 | Coagulant (0 ~ 50mg/l as alum) |
| Run 2 | Granular activated carbon (GAC 0 ~100g) |
| Run 3 | GAC (50g) + coagulant (27mg/l as alum) |

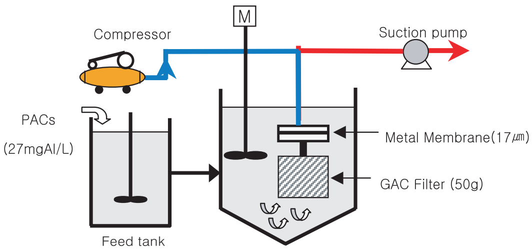


**Fig. 1.** Schematic diagram of MBR.

tests were carried out at a stirring speed of 150rpm for 15~30min using 1L beakers and the water pH was controlled about 7 using 0.1N NaOH or 0.1N HCl solutions.

Some dead-end filtration test were conducted to investigate the effects of FOC on the permeate flux. The Adventec dead-end filtration cell with 47mm in diameter and an effective filtration area of 17.3cm$^2$ was used to measure the feed water permeability, which is a fundamental and necessary property in characterizing a membrane. The water was continuously delivered to the filtration cell through a 8 liter reservoir which was pressurized by air compressor.

The water permeability data were obtained by periodically measuring the amount of time required for 200~500ml of feed water to flow through a 0.45 micrometer filter at a pressure of 206kPa. As shown in Figure 1, the MBR was consisted of a cylindrical bioreactor(effective volume 30L), a GAC filter, and a submerged membrane module(metal sieve, pore size of 17$\mu$m, surface area of 0.08m$^2$, $\phi$ 100mm). The membrane module was submerged in the reactor and vacuum was applied inside of it for filtration. To minimize membrane fouling, intermittent aeration was applied inside of the filter with 3 kgf/cm$^2$.

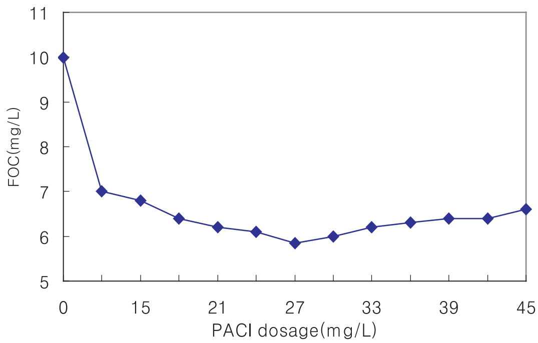


**Fig. 2.** Effects of PACl dosage on FOC removal.

## 3. RESULTS AND DISCUSSION

### 3.1. Pretreatment effects by PACl and GAC on permeate flux decline

Some dead-end filtration tests were conducted to investigate the effects of pretreatment by PACl and GAC on the permeate flux under these conditions: SS 30~500mg/l, FOC 2.5~17.3mg/l. The time required to filter a fixed volume (50ml) of water through a standard 1 and 5$\mu$m pore size membrane filter was measured. Gravity filtration and filtration under the pressure of 206kPa were conducted on the experiments.

Figure 2 shows the effects of PACl dosage on FOC removal. The permeate flux increased with increase in PACl dosage at the range from 0 to 50 mg/l (Figure 3). At coagulant dosage of 27mg/l, the removal of FOC was about 46% and the flux increased to 3.5 times that of no PACl addition. This result indicated that dissolved organic matter that passed a 1.0$\mu$m filter was more responsible for flux decline in MF filtration. These results confirmed the effect of filterable soluble organic material on the membrane fouling. Thus, it was believed that the sharp flux reduction mainly resulted from the build-up of FOC cake layer resistance and plugging of membrane pores by organic particles.

To investigate the performance of GAC dosage on FOC removal, batch runs were done at different GAC dosages. Figure 4 shows the results of GAC dosages on permeate flux decline after 30 min contact. The permeate

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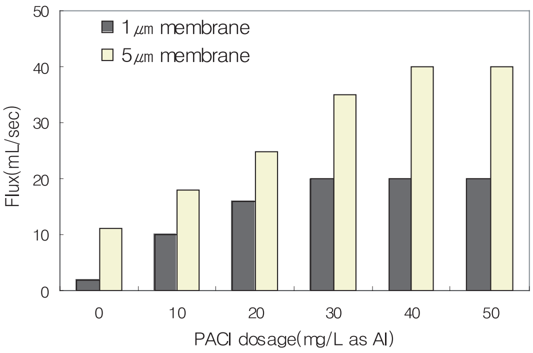


**Fig. 3.** Flux decline as a function of PACl dosage.

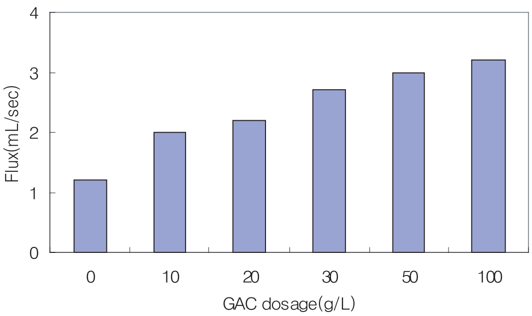


**Fig. 4.** Flux decline as a function of GAC dosage.

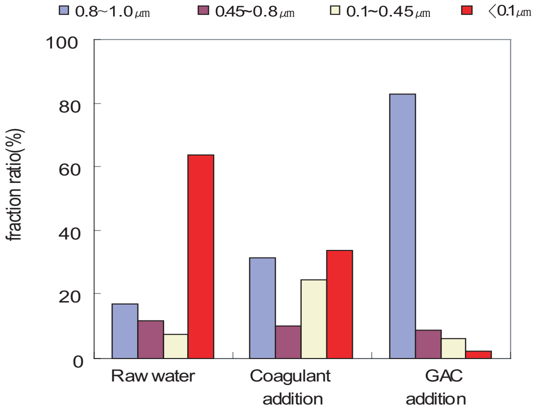


**Fig. 5.** Effect of pretreatment on particle size distribution.

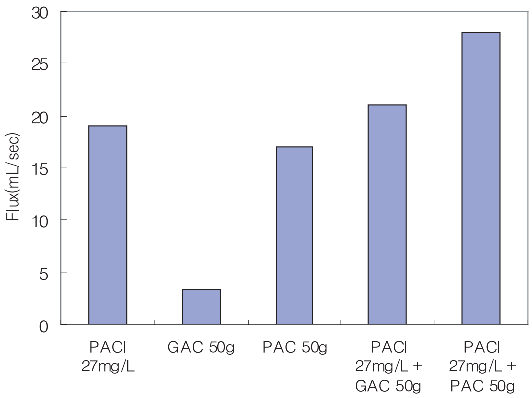


**Fig. 6.** Effects of coagulant and GAC dosage on permeate flux.

flux increased gradually with increase in GAC dosage. At GAC dosage of 50g/L, the permeate flux is about 2 times higher compared to raw water.

As showed in Figure 2~4, the addition of GAC or coagulant could decreased the residual FOC concentration and markedly increased the permeate flux. Thus, the combination of MF membrane processes with the addition of PAC + coagulant combination is being attractive because pretreatment helps remove dissolved organic matter in addition to the possibility in membrane fouling reduction.

The effect of pretreatment on particle size distribution is shown in Figure 5. In the case of raw sewage water, more than 60% of FOC was less than 0.1$\mu$m, and the portion of large particle (larger than 0.45$\mu$m) was 29%. The results from Figure 5 show that GAC and coagulant is effective in removing small particles less than 0.1$\mu$m. The particle substances in the range of 0.1~1.0$\mu$m were removed effectively by the addition of GAC or coagulant.

In Figure 5, it was found that dissolved matter, particularly less than 0.1$\mu$m was most responsible for flux decline. Thus membrane flux improvement must be due to removal of these compounds. On the other hand, pretreatment with PAC + coagulant may be most effective tools for the preventing of membrane fouling problems. The effects of pretreatment on permeate flux are shown in Figure 6. A flux behavior for these results indicates that membrane fouling could be controlled by the addition of GAC and PACl as a pretreatment.

### 3.2. Effect of pretreatment on permeate flux rate at MBR system

Figure 7 shows the effects of pretreatment on permeate

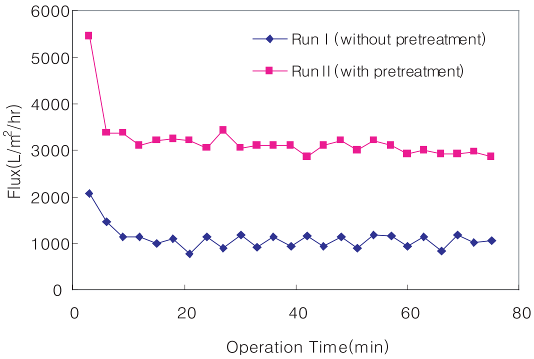


**Fig. 7.** Effects of pretreatment by coagulant and GAC on flux.

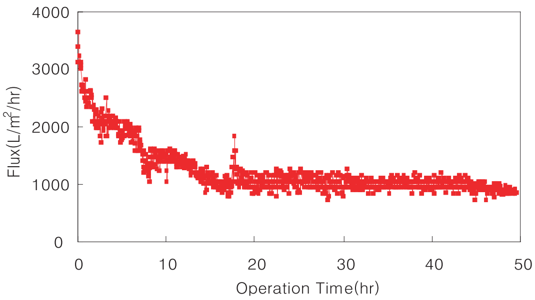


**Fig. 8.** Permeate flow rate at MBR system with the pretreatment.

flux at MBR system. With the pretreatment by addition of GAC of 50g and PACl of 27mg/L, the flux was about 3,500L/m$^2$/hr (LMH), which was about 3.5 times that of Run1 (without pretreatment). To investigate the long term permeation ability of 17$\mu$m metal sieve membrane, some tests were conducted under the experiment conditions (influent SS 210mg/L, intermittent filtration of 3 min. vacuum suction and 0.5 min. air backwashing).

The decline of permeate flux was shown in Figure 8. After 40 hours operation, the permeate flux was about 1,000 (LMH), which is 20 times high compared to the other's results(Brindle, 1996). This high flux could be obtained by using of metal sieve with 17$\mu$m pore size, which is large pore size compared to the conventional MF membrane, and by applying of periodic air backwashing, and by the pretreatment with GAC+PACl addition. In the case of long-term operation of MBR process with 1$\mu$m MF membrane, the permeate flux was about 400 (LMH) by the pretreatment with GAC + PACl addition.

Consequently, it was obvious that dissolved matter that pass a 1.0$\mu$m filter was the primary factor affecting flux decline, but it was effectively controlled with the integrated GAC + coagulant pretreatment. This study concluded that GAC addition after the coagulation reaction is the most effective process of the prevention of membrane fouling problems.

## CONCLUSIONS

The performance of a combined system composed of GAC + coagulant pretreatment and microfiltration processes was investigated in terms of filterable organic matter removal and membrane permeability during treatment of a MBR. As the dosage of GAC and coagulant increased, the residual FOC concentration decreased and the permeate flow rate increased markedly. The permeate flux increased with increased PACl addition at the range from 0 to 50mg/L. At coagulant dosage of 27mg/l, the removal of FOC was about 46% and the flux increased to 3.5 times that of no PACl addition. This results indicated that the FOC (= dissolved organic matter that passed a 1.0$\mu$m filter) was more responsible for flux decline in MF filtration.

The permeate flux increased gradually with increase in GAC dosage. At GAC dosage of 50mg/L, the permeate flux is about 2 times higher compared to raw water. The particle substances in the range of 0.1~1.0$\mu$m were removed effectively by the addition of GAC and coagulant. Higher dosage of GAC and coagulant led to the higher removal of FOC.

Some tests were conducted to investigate the pretreatment effect on permeation ability of SMBR system with 17$\mu$m metal sieve membrane. After 40 hours operation, the permeate flux was about 1,000 (LMH), which is 20 times high compared to the other's results. It is concluded that the pretreatment combined with coagulant and activated carbon is the one of the most effective process for the solution of membrane fouling problems, and the continuous operations could be successful to some extent.

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