

## Control of residual organic matter to reduce bacterial regrowth potential for wastewater reuse

Shijun Kim, Jeongsook Kim<sup>\*†</sup> and Yutaka Suzuki<sup>\*\*</sup>

Ulsan Regional Innovation Agency, #758-2 Yenamdong, Buggu, Ulsan 683-804, Korea

<sup>\*</sup>Busan Environmental Technology Center, #203 Daeyeondong, Namgu, Busan 608-737, Korea

<sup>\*\*</sup> Recycling Research Team, Material and Geotechnical Engineering Research Group, Public Works Research Institute, Minamihara 1-6, Tsukuba, Ibaraki 305-8516, Japan

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**Abstract**—Several problems have been reported about accumulated microorganisms in reclaimed water distribution systems. This paper presents the results of residual organic matter (OM) removal and apparent bacterial regrowth potential of treated wastewater obtained from laboratory-scale experiments using advanced biological treatments: two immobilization processes in series and a membrane bioreactor (MBR) process. Furthermore, a nanofiltration (NF) membrane process was applied to effluents of both advanced biological treatments. The immobilization process removed large molecular weight (MW) fractions >5,000 since immobilized microorganisms had sufficiently acclimated. The NF membrane was more effective in rejecting large MW fractions in the effluents of the immobilization and the MBR treatments. But it was difficult to reject small MW fractions <1,000 by NF. Neutral hydrophilic fraction of DOC was reduced by both advanced biological processes, and it can be thought that the microorganisms in the advanced processes could decompose and grow on some part of the neutral hydrophilic fraction. Quantity of attached microorganisms in the second immobilization reactor was significantly reduced compared to that in the first immobilization reactor. This suggests that apparent bacterial regrowth potential is controlled by the accumulation of effective microorganisms in the first reactor.

Key words: Wastewater Reuse, Bacterial Regrowth, Biological Advanced Treatment

### INTRODUCTION

Reuse of abundant municipal sewage effluent is gradually being regarded as a valuable method for obtaining new water resources in the near future. However, an obstacle to wastewater reuse is the existence of potentially residual organic matter (OM) [1-3]. Although residual OM has a low concentration, it can be a source substance for apparent bacterial regrowth by providing energy sources for biomass production [4]. As a result, a biofilm accumulation or blockage in distribution systems occurs as a result of the multiplication of viable bacteria. Lastly, the existence of residual OM can lead to deterioration of water quality, jeopardizing the feasibility of wastewater reuse [5].

Therefore, with the increasingly stringent requirements for reclaimed water quality, an effective treatment process for wastewater reuse is required. Reverse osmosis (RO) technology is gradually being applied in the field of water reuse, but because the cost remains high and it is strongly affected by energy consumption, an economical and efficient technology for eliminating residual OM is needed.

The objective of this study was to eliminate residual OM, thereby enhancing the value of sewage effluent as a water resource. This paper presents the results obtained from laboratory-scale experiments using an immobilization process and a membrane bioreactor process. Furthermore, NF membrane treatment was applied to effluents

of both advanced biological processes.

### MATERIALS AND METHODS

#### 1. Immobilization

A schematic of the experimental apparatus is shown in Fig. 1. Erlenmeyer flasks (2 liter) for immobilization carrier reactors (A and B) were prepared. The plastic media used is made of polypropylene, with a diameter of 4 mm and a length of 5 mm. Specific surface area is 1,410 m<sup>2</sup>/m<sup>3</sup> when the plastic media is densely packed. The packing volume ratio of the plastic media to the immobilization reactor was 30% to prevent sedimentation of the media on the bottom of the reactor. The plastic media was cleaned with milli-Q by ultrasonic apparatus in the beginning of the experiment. The plastic media, under slow agitation with a stirrer, was aerated by using an air diffuser, and samples were taken after certain aeration time. Hydraulic retention time (HRT) was set on 2 days. The experiment of fill and draw was carried out once a day.

Two immobilization reactors (A and B) were used to investigate apparent bacterial regrowth potential. An experiment using reactor A was performed to remove the residual OM, and another experiment, using reactor B, was performed to confirm apparent bacterial regrowth potential as quantity of attached microorganisms on the plastic media. Immobilized microorganisms were measured as follows: ninety pieces of plastic media added to 50 ml Milli-Q water were treated ultrasonically for 30 minutes to detach microorganisms. The suspended solution, including detached microorganisms, was measured with TN and TP, respectively.

<sup>†</sup>To whom correspondence should be addressed.

E-mail: jkim@km-c.ac.kr

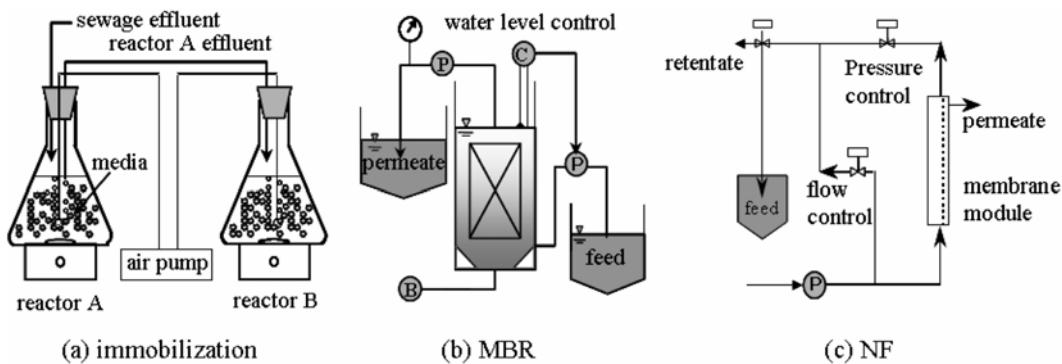


Fig. 1. Schematic diagram of experiment apparatus.

## 2. MBR

As shown schematically in Fig. 1, the MBR consisted of a water level control sensor with a dead-end microfiltration membrane submerged vertically in the reactor (4 liter). The flat sheets of polyolefin membrane (Kubota, Japan) had a nominal pore size of 0.4  $\mu\text{m}$  and an effective surface area of 0.11  $\text{m}^2$ . Filtration direction was from the outside to the inside of the two flat sheets. A diffuser in the bottom of the reactor provided air bubbles at approximately 5 l/min. The feed water was prepared in a stainless steel vessel and pumped into the reactor by using the water level control sensor. The filtrated effluent was pumped out of the MBR with a peristaltic pump so that the HRT became 3 hours. The fill and filtrate volume was 12.5% (0.5 l) of the full reactor liquid volume. Neither were suspended solids removed from the reactor nor was membrane cleaning carried out during the experiment periods.

An immobilization reactor C was set to receive the MBR effluent to confirm apparent bacterial regrowth potential of the water. At the beginning of experiment, global parameter (TOC, TN and TP) concentration of treated effluent was changeable. Effluent concentration of immobilization process was high more than municipal secondary treated sewage which was oligotrophic condition.

## 3. NF

The NF apparatus (Nitto Denko, Japan) consisted of a flat sheet type membrane test cell and a magnetic type pressurization gear pump. Fig. 1 shows a schematic diagram of NF, which has a nor-

mal salts rejection of 10% and an effective membrane surface area of 60  $\text{cm}^2$ . Operating pressure was set to approximately 500 kPa.

## 4. Source of Treated Sewage

The source of the treated sewage was the secondary effluent of a pilot plant using the conventional activated sludge process at the Kasumigaura Municipal Wastewater Treatment Plant located in Ibaraki prefecture, Japan. The effluent, which had been used as feed water, was stored in a storage room that was constantly maintained at 5 °C. The suspended solid (SS) concentration of the secondary effluent used in the experiment was 1 mg/l or less.

## 5. Gel Permeation Chromatograph (GPC) Analysis

MW distribution of the residual OM associated with each effluent was determined by GPC (Shodex GPC SYSTEM-21, Japan) analysis. The GPC separation was performed with Shodex SB-802 HQ and SB-806 MHQ (300 mm × 8 mm) columns. Detection was achieved with a differential refractive index (RI) detector at 254 nm (Shodex RI-71, Japan). Milli-Q water (Millipore, US) was deflated with He gas for 10 minutes and used as a mobile phase at a rate of 0.5 ml/min. The column temperature was maintained at 40 °C and injection volume was 330  $\mu\text{l}$ . Prior to the GPC analysis, all samples from the original water were concentrated by 10-fold using a rotary evaporator apparatus (EYELA N-1000, Japan).

## 6. UF Fractionation

UF was applied to characterize the OM according to MW. The UF apparatus (Advantec UHP-76 K, Japan) consisted of a 300 ml

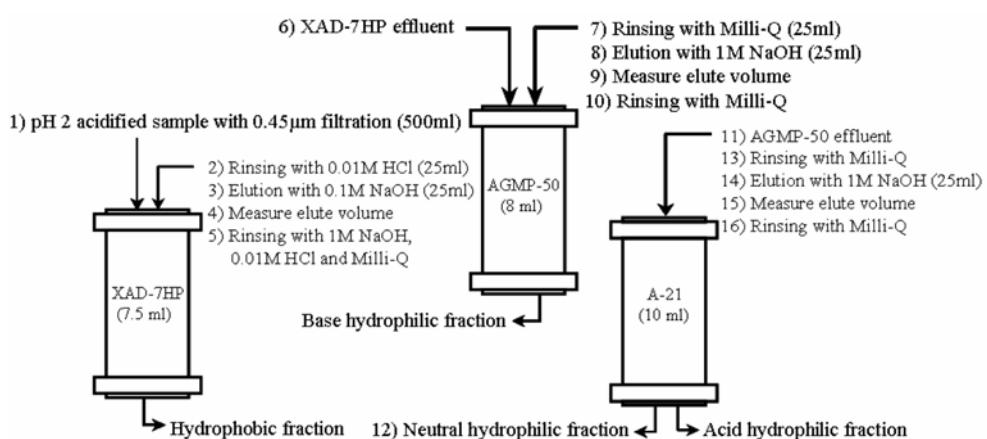


Fig. 2. Schematic diagram of procedure for dissolved OM fractionation.

stirred cell with tangential flow. Applied pressure of 98 kPa was used throughout the duration of each ultrafiltration process. Flat disc UF membranes (Amicon series, 76 mm diameter of regenerated cellulose) with nominal MW cutoffs of 1,000, 5,000, and 10,000 atomic mass units were used in the UF test cell. UF permeates were characterized, permitting calculation of the following discrete MW fraction ranges: <1,000, 1,000-5,000, 5,000-10,000 and >10,000. A 100 ml portion of fresh effluent was passed through each UF membrane, up to a total volume of 25 ml of ultrafiltrate. The organic content of each portion was determined as TOC.

## 7. DOC Fractionation

DOC fractionation procedure [6] shown in Fig. 2 was applied to water samples, and OM was classified into neutral hydrophilic fraction (NH<sub>1</sub>F), hydrophobic fraction (HoF), base hydrophilic fraction (BH<sub>1</sub>F) and acid hydrophilic fraction (AH<sub>1</sub>F).

## 8. Measurement of TOC, TN and TP

All effluents collected for the experiment were filtered through pre-washed 0.45  $\mu$ m membrane filters (Advantec, Japan), except for the MBR effluent. All of the experiments were conducted at 20 °C. DOC was measured after pre-treatment with 1 N HCl of 100  $\mu$ l and elimination of inorganic carbon with pure air bubbling by using a TOC analyzer (Shimadzu TOC-5000 A, Japan). TN and TP of the immo-

bilized microorganisms attached to the plastic media were measured by ultraviolet absorption spectrophotometry with a TRAACS 2000 analyzer (BRAN+LUEBBE, Germany).

## RESULTS AND DISCUSSION

### 1. MW Fractions Removal of the Residual OM

The MW distribution of secondary effluent was investigated by using the GPC in Fig. 3. MW was separated into four main peaks: <200, 200-700, 700-1,000 and >6,000, respectively. The peak MW of the residual OM was generally less than the MW 1,000 fraction. UF fractionation also revealed that small MW fractions <1,000 occupied relatively large amounts of the TOC portion of approximately 50%. Lastly, the results indicated that the secondary effluent consisted of mostly small MW organics.

Removal of MW fractions by both advanced biological treatments and NF treatment of the advanced biological effluents is shown in Fig. 4. The secondary effluent and advanced biologically treated water were characterized by the UF fractionations, based on TOC. A significant amount (60%) of the TOC of MW fractions >5,000 was removed by using the immobilization carrier process. On the other hand, the TOC portion of MW fractions <1,000 increased to more

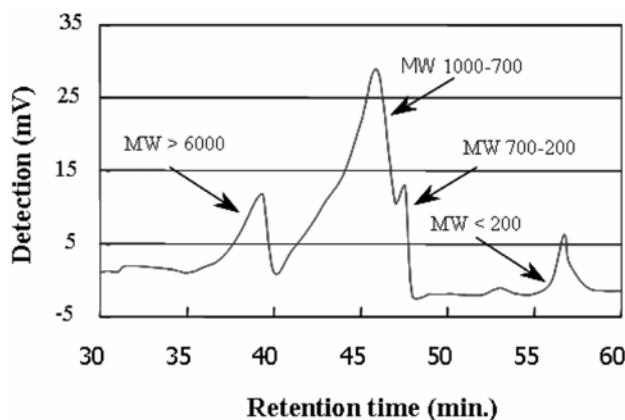


Fig. 3. MW distribution of the secondary effluent.

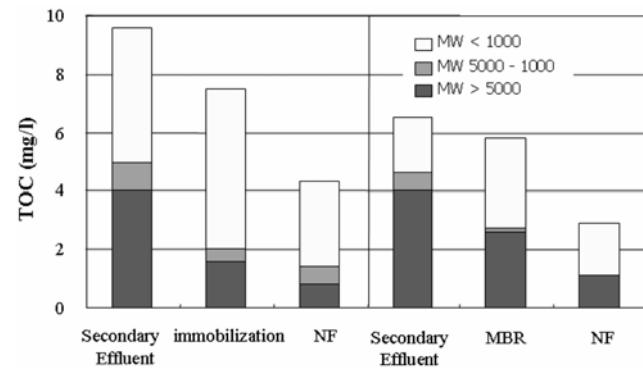


Fig. 4. Removal of MW fractions in immobilization process and MBR, and NF treatment of both the advanced biological treatment effluents.

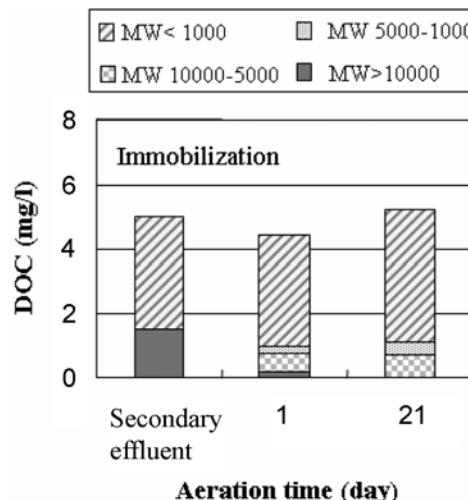


Fig. 5. Removal of MW fractions in the advanced biological treatments by long-term aeration.

than that of the original sewage effluent due to significant activity of immobilized microorganisms. The immobilization process removed large MW fractions  $>5,000$  since immobilized microorganisms had sufficiently acclimated during experiment periods of 60 days. The MBR exhibited a relatively poor removal efficiency of MW fractions  $>5,000$ . The MBR showed a slightly lower rate of residual OM removal than that of the immobilization process, presumably due to the shorter HRT. However, the MBR has the advantage of reducing the amount of microbial seed entering the reclaimed water distribution system.

Furthermore, the NF process rejected the MW fractions of both advanced biological treatment effluents. It was found that NF contributed particularly to the large MW rejection, but the low rate of rejection of small MW fractions  $<1,000$  resulted in low TOC removal efficiency. It is considered that small MW fractions pass through the membrane pores, because this NF membrane is a borderline tight UF membrane.

The MW fractions in the advanced biological treatment effluents transform according to long-term aeration phase, as shown in Fig. 5. In the immobilization process, the MW fraction composition following the long-term aeration phase (21 days) was certainly different from that after the short-term aeration phase (1 day). It was found that MW fraction  $>10,000$  is gradually decomposed by immobilized microorganisms and converted to small MW fractions  $<1,000$  during long-term aeration without source water feeding.

The MBR process completely decomposed MW fractions  $>10,000$  during the long-term aeration phase. Many microorganisms had accumulated in the MBR; thus there is a possibility that it caused the autolysis. MW fractions  $<1,000$  increased more than that of the immobilization process.

The results of the GPC analysis, as shown in Fig. 6, clearly present the peak difference before and after both advanced biological treatments. Lastly, the results revealed that both processes transformed large MW fractions to small MW fractions during the long-term aeration phase. MW fractions  $<1,000$  presumably remain as refractory substances or metabolic organic matter.

## 2. Removal of DOC Fractions

Removal of DOC fractions by both advanced biological treatments

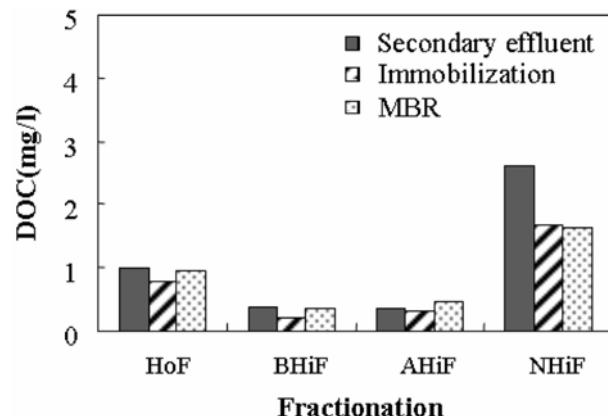


Fig. 7. Removal of DOC fractions in immobilization process and MBR.

HoF. Hydrophobic fraction

AHiF. Acid hydrophilic fraction

BHiF. Base hydrophilic fraction

NHiF. Neutral hydrophilic fraction

is shown in Fig. 7. DOC fractionation of the residual organic matter was occupied in the order of neutral hydrophilic fraction (NHiF), hydrophobic fraction (HoF), base hydrophilic fraction (BHiF) and acid hydrophilic fraction (AHiF). Not much difference was observed in the concentrations of HoF, BHiF and AHiF between the secondary effluent and the advanced process effluents, but NHiF concentration was reduced by both advanced processes. It can be thought that the microorganisms in the advanced processes could decompose and grow on some part of NHiF because of the longer solids retention time achieved in the advanced processes.

## 3. Control of Apparent Bacterial Regrowth

The apparent bacterial regrowth was estimated by using immobilization reactors B and C, which received effluents from reactor A and MBR, respectively. Little difference was found in the TOC concentration between the effluent of reactor A and that of reactor B (Fig. 8). However, the bacterial regrowth potential expressed as accumulated biomass on the plastic media was significantly reduced

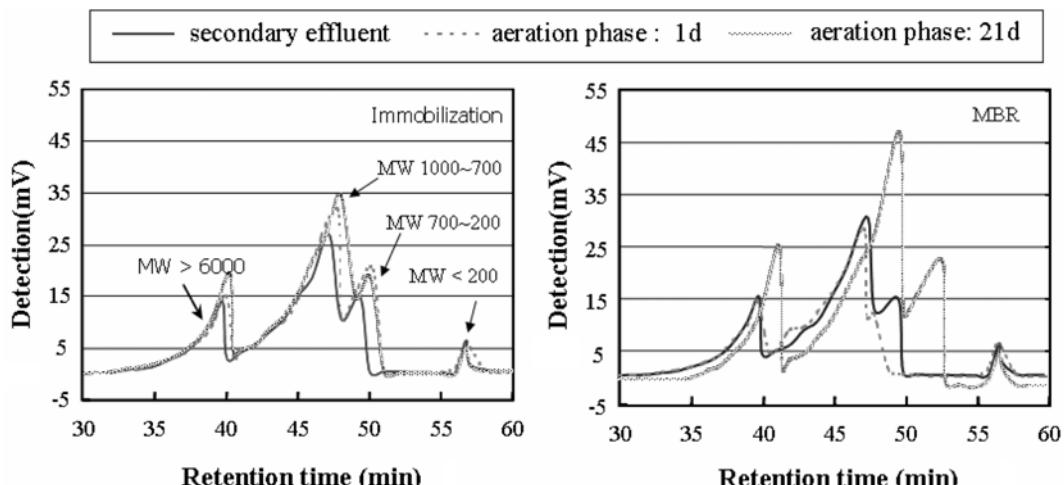


Fig. 6. MW distribution of advanced biological treatment by long-term aeration.

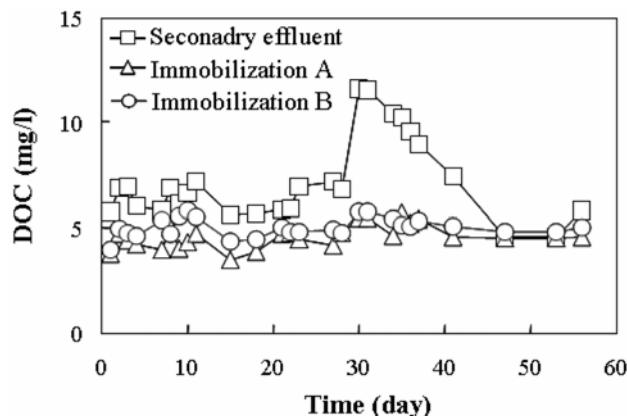


Fig. 8. Comparison of DOC concentration of secondary effluent and effluents from reactors A and B.

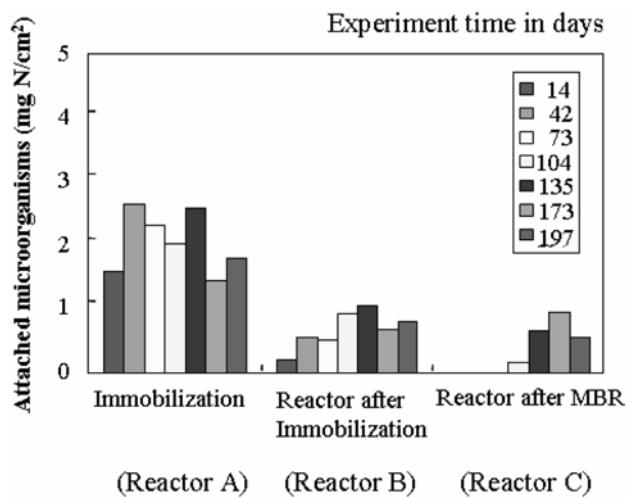


Fig. 9. Change of bacterial regrowth potential after immobilization process and MBR.

in the cases of reactors B and C compared to that of reactor A (Fig. 9). The quantity of attached microorganisms after 30 days of operation of immobilization reactor (reactor B) slightly increased 0.06 mg-P/cm<sup>2</sup> to 0.64 mg-P/cm<sup>2</sup>. But after an additional 30 days, the amount decreased slightly to 0.62 mg-P/cm<sup>2</sup>. This result indicates that the bacterial regrowth potential was effectively reduced from the secondary effluent by the both advanced treatment processes, which

could keep microorganisms capable of decomposing residual NHF. As a result, the apparent bacterial regrowth potential of the biodegradable residual OM is considered as being controlled.

## CONCLUSIONS

The MW distribution of the secondary sewage effluent of a conventional activated sludge process was separated into four peaks by using the GPC method. The peaks were mostly small fractions less than MW 1,000. Also, relatively large amounts of TOC of the residual OM were identified as small MW fractions, occupying 50% by UF fractionation.

The immobilization process removed large MW fractions >5,000, since immobilized microorganisms had sufficiently acclimated. The NF membrane was more effective in rejecting large MW fractions in the effluents of the immobilization and the MBR processes. It was difficult to reject small MW fractions <1,000 by NF; thus the total TOC portion had a low rate of removal efficiency.

The immobilization and the MBR processes exhibited a significant removal rate of MW fractions >10,000 during the batch long-term aeration phase. However, MW composition following the long-term aeration phase was significantly converted to small MW fractions <1,000 that might be hardly biodegradable.

Neutral hydrophilic fraction of residual organic matter was reduced by the advanced biological processes, though not much difference was observed in other fractions. It can be thought that the microorganisms in the advanced processes could decompose and grow on the some part of neutral hydrophilic fraction.

Bacterial regrowth potential was effectively reduced from the secondary effluent by both advanced treatment processes, which could keep microorganisms capable of decomposing residual neutral hydrophilic fraction.

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