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MBR에서 간헐포기에 의한 오염저감 효과

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Effects of Fouling Reduction by Intermittent Aeration in Membrane Bioreactors

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요 약: 30 LMH의 정유량 플럭스로 운전하는 MBR에서, 휴지 및 역세정에 따른 한외여과 분리막의 오염을 조사하였다. 또한, 연속적인 공기세정과 비교하여 분리막 여과저항을 최소화하기 위한 간헐적인 공기세정을 평가하였다. 여과 조건은 14.5분 여과와 0.5분의 휴지를 유지하였으며, 역세정 시간은 휴지 시간과 동일하게 운전하였다. 공기세정이 정지하는 동안에 분리막 표면의 겔층 위에 케잌이 빠르게 축척되었으며, 역세정으로 겔층과 케익층의 복합층은 쉽게 제거되었다. 역세정 후에 공기세정이 정지하는 동안 분리막 표면에 케잌이 형성되어 공경 내부의 오염현상을 억제하였다. Pearson 상관성을 조사한 결과, 간헐적인 공기세정에서 공기 세정이 정지하는 시간과 분리막의 오염은 매우 연관성이 높다는 것을 알았다. 즉, 간헐적인 세정에서 공기세정이 정지하는 시간이 갈수록 오염억제에 효과적이었다.

Abstract: The effects of relaxation and backwashing on fouling in ultrafiltration were investigated using full-scale membrane bioreactors (MBRs) which operated at a constant flux of 30 LMH. This paper also estimated the feasibility of using intermittent aeration strategies for minimizing the hydraulic resistance to filtration in comparison with the continuous aeration for running MBRs. Multiple cycles of filtration (14.5 min each) and relaxation (0.5 min each) were repeated. Similarly, a backwash was conducted by replacing a relaxation after each filtration cycle for the comparative performance test. The attached cake thickness on the membrane rapidly increased, caused by subsequent no aeration leading to easier combining with gel layer and the formation of heterogeneous layer on the membrane surface. During periodic backwashing, it is expected that gel and thin cake layer might sufficiently be removed by heterogeneous layer. After periodic backwashing, subsequent cake layer formation during time of no aeration was rapid than frequent no aeration, acting as a prefilter and preventing further irreversible fouling. Based on the Pearson correlation analysis, overall period fouling (dTMP/min) and average of all cycles (dTMP/min) were strongly correlated with the on-off period of aeration for operating MBRs.

Keywords: *Cake formation, Membrane bioreactor, Ultrafiltration, Intermittent aeration, Continuous aeration*

1. Introduction

The combination of activated sludge units and membrane filtration for biomass retention generally results in high effluent qualities and compact plant configurations. Complete solids removal, high rate and high efficiency organic removal and small footprint are common characteristics regardless the wastewater type to be treated or

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the commercial process used[1,2].

Beside various advantages, membrane fouling remains a major obstacle to their wider application in wastewater treatment. Membrane fouling is the most serious problem affecting system performance and maintenance cost. Fouling on the membrane surface and within the pores reduces long term flux stability necessitating membrane cleaning which then add the overall cost. Moreover, it reduces membrane permeability, thereby, increasing the membrane cleaning frequency, reducing the plant treatment capacity and shortening the membrane module lifespan[3].

Various techniques are used to reduce fouling. Basically fouling of membranes in membrane bioreactors (MBRs) systems can be minimized by increasing transmembrane pressure (TMP), promotion of cross flow to limit the thickness of the boundary layer and/or periodical application of cleaning measures to remove the cake layer and foulants.

A critical flux concept was proposed initially below which no fouling would occur[4]. But, recent studies indicate that the choice of filtration conditions close to the critical flux does not fully prevent the gradual fouling of the membrane, which is supposed to arise from deposition and subsequent interaction of solute and colloidal fractions with the membrane[5].

Most commonly crossflow aeration and backwashing techniques are employed to minimize membrane fouling. Crossflow aeration conditions within a MBRs promote scouring of the membrane surface to lower fouling layer formation. A critical air flow rate could exist beyond which any further increase in aeration has little effect on fouling suspension[6]. The rate and extent of fouling are also affected by the properties of mixed liquor suspension that the membrane modules are in contact with.

It is a common practice in many submerged MBRs to backwash the membrane on a periodical basis. By using a reverse TMP for certain periods of time, permeate is forced through the membrane in reverse direction, which assists to dislodge the cake layer and enhance back diffusion of solute deposit[7-10].

For larger particles such as emulsion droplets and activated sludge, fouling was observed during crossflow filtration[11]. Large particles will be dislodged at a lower crossflow velocity than small particles, and small particles could selectively accumulate on the membrane surface during crossflow filtration. The selective accumulation of small particles was observed in crossflow filtration experiments[12].

At intermittent filtration operation, shortening the filtration time could reduce the membrane fouling. When filtration time was decreased from high to low, the permeability increased more than two times at ratio of filtration/ceasing of 3.75, compared with about one time at ratio of filtration/ceasing of 6[13]. It was obvious that the permeability depended on the filtration time.

Recent intermittent cyclic aeration mode results are still not clear to explain membrane fouling phenomenon[14,16]. Further, there have been little studies related to effect of no aeration time on intermittent aeration on membrane performance.

Finding the importance of optimizing no aeration time, this study aims to identify the effect of no aeration time including backwashing and relaxation conditions on the optimum no aeration time. Additionally, an insight into the effects on the hydraulic resistance is assessed, and Pearson correlation analysis was done to identify the major contributor to membrane fouling.

2. Materials and Methods

2.1. Submerged membrane module (YEF; Yonsei End Free)

Fig. 1 shows the submerged membrane module used in the study. A module has 18 fan shape elements which are assembled to the center connection pipe[17-19]. The duplicate center pipe transfers water and air. The ultrafiltration membrane fibers made up of polyacrylonitrile (PAN) have an effective surface area of 24 m^2. Table 1 shows the characteristics of the membrane used. The present study was carried out to obtain further insight into the characteristics of filtra tion of the submerged YEF module.

Table 1. Characteristics of the Membrane Module Used

Membrane material	I.D. (mm)	O.D. (mm)	Pore size (kDa)	Effective length (m)	Effective area (m^2)
PAN	0.8	1.4	100	1.8	24

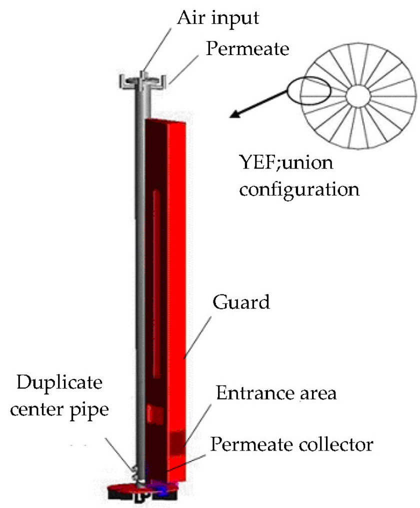

Fig. 1. A schematic diagram of one element setup of YEF module.

2.2. Superficial liquid velocity setup

The experimental setup used to measure the superficial liquid velocity is a cylinder reactor tank (Fig. 2) for pure water tested and the oxic tank for mixed liquor in the MBRs (Fig. 3). For this test, the equipment was composed of velocity meter (BFM002, Valeport), signal transmitter and personal computer for the data acquisition. Air holes existed at bottom of YEF module. The air is introduced through air holes with holes of 3-mm, connected to a duplicate center pipe. Bubbling flow along the membrane surface inner guard was created by aeration which allows an aeration intensity up to 0.0313 $m^3/m^2 \cdot hr$. Aeration intensity value was based on the ratio of air flow rate to membrane area. It units is m^3 air per m^2 per unit time, and thus nominally expressed as $m^3/m^2 \cdot hr$. It is easy to calculate but it is probably not the pertinent parameter to characterize the flow pattern in the membrane module.

The riser moves upward from the bottom entrance of module to exit of liquid fluid and air flow on top of the guard (Fig. 2). The superficial liquid velocity

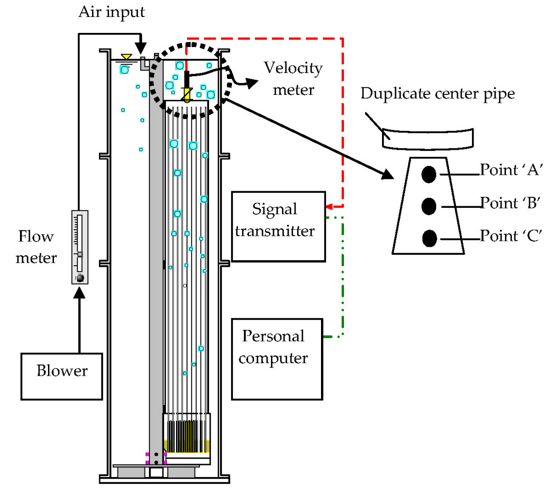

Fig. 2. Experimental setup for measuring superficial liquid velocity and measuring point of velocity meter.

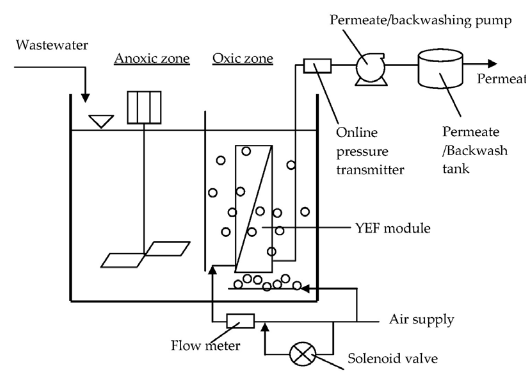

Fig. 3. A schematic of the MBRs process.

measured by velocity meter which instituted on freely membrane bundles inside guard of YEF module (Fig. 2). This study was run without filtration because the main objective was to characterize the flow induced by the aeration. A further study will be carried out with the filtration in order to determine the impact of the filtration on the flow hydrodynamics.

2.3. Membrane bioreactor setup

In this study, the MBRs for the treatment of real wastewater at an existing wastewater plant (WWTP) was operated by directly submerging the pilot scale YEF module into oxic basin in order to investigate the performance and fouling of membrane module. The hydraulic retention time (HRT) as well as biomass

Table 2. Operating Conditions Characterization in MBRs

Conditions	Mean value
HRT (hr)	12
MLSS (g/L)	5.6 ± 1.1

Table 3. Average Characteristics of Influent Water (n = number of analysis)

Items	Mean concentration*	n
COD (mg/L)	81.2 ± 59	62
T-N (mg/L)	33.5 ± 12.2	48
NH_3-N (mg/L)	25.4 ± 8.7	75
T-P (mg/L)	4.1 ± 2.5	52

*Values are given ± standard deviation.

Table 4. Operating Conditions Applied in the Different Runs

Run[1)]	Operating parameters for continuous or intermittent aeration					
	filtration (min)	relaxation (sec)	backwash (sec)	backwash flux (L/m^2 hr)	aeration off/on (sec)	MLSS (mg/L)
C-C[2)]	-	-	-	-	-	6,400
C-R[3)]	14.5	30	-	-	-	6,400
C-B[4)]	14.5	-	30	45	-	7,100
I[5)]-B1	14.5	-	30	45	10/10	3,450
I-B2	14.5	-	30	45	10/20	5,700
I-B3	14.5	-	30	45	15/20	4,600
I-B4	14.5	-	30	45	20/20	4,500
I-R	14.5	30	-	-	20/20	4,250

1) Aeration-filtration
2) C : Continuous filtration or aeration
3) R : Relaxation
4) B : Backwashing
5) I : Intermittent aeration

(MLSS) of MBRs are shown in Table 2. The characteristics of raw wastewater are listed in Table 3. The real submerged MBRs as shown in Fig. 3, which was located in Yonsei University Wonju campus, consisted of anoxic and oxic basin. The filtration system consisted of a reactor column a YEF element, permeate pump (gear flex type, DAESUNG Co., Ltd), blower (roots type, JUNGSUN Co., Ltd), pressure gauge (PMC731, Endress Hauser), electronic flowmeter (Promag 30, Endress Hauser) and computer. The pump operated in forward and backward directions for the suction of permeate and the backwashing.

2.4. Aeration mode experiments

Two different aeration (i.e. continuous and intermittent) and three filtration modes were applied and evaluated in terms of hydraulic performances and its resistances (Table 4). The filtration flux was 30 LMH ($L/m^2 \cdot hr$). With all aeration modes, aeration intensity was applied 0.0313 $m^3/m^2 \cdot hr$, that is, it was represented by 120 L/min of air flow rate. Filtration mode consisted of the continuous, the relaxation and the backwashing mode. The relaxation featured a non-filtration time of 30-s every 15-min. The backwashing has the backwashing flux of 30 and 45 LMH, duration

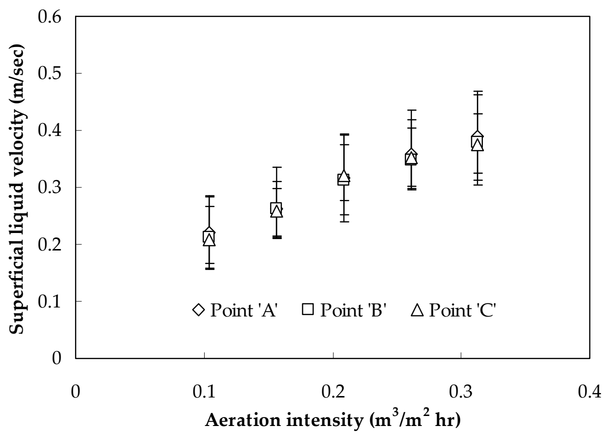

Fig. 4. Superficial liquid velocity at different measuring points.

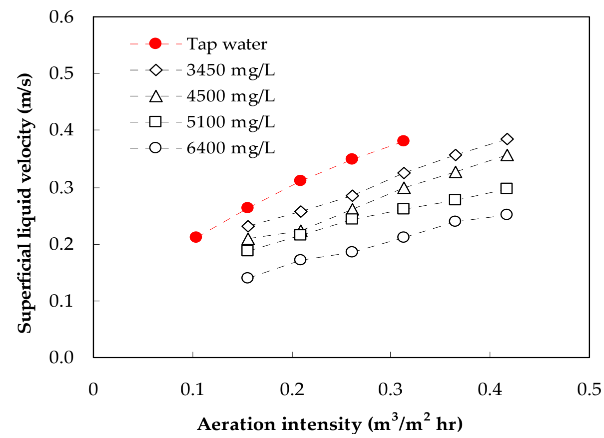

Fig. 5. Aeration intensity vs. superficial liquid velocity on different MLSS concentration.

of 30-s every 15-min.

2.5. Hydraulic analysis of the membrane fouling

To evaluate the fouling distribution of the different aeration modes, fouling propensity was monitored by continuous TMP measurement and a detailed analysis of the hydraulic resistances based on Darcy's law (Eq. (1)) and the resistance in series model (Eq. (2))[20].

$$J = \frac{TMP}{\mu \times R_{total}} \quad (1)$$

$$R_{total} = R_m + R_c + R_f \quad (2)$$

Where J is the permeate flux (L/m^2·h), R_{total} is the total membrane resistance (m^{-1}) and μ is the dynamic viscosity of the permeate water (Pa·s). The total membrane resistance (R_{total}) is composed of the intrinsic membrane (R_m), the cake resistance (R_c), and the fouling resistance (R_f) (Eq. (2)).

The intrinsic resistance of the membrane (R_m) was calculated by clean water tests. Total resistance of the fouled membrane (R_{total}) was calculated at the end of the filtration experiment (Eq. (2)). A clean water test was conducted to obtain the cake resistance after rinsing and flushing by clean water (R_c). Subsequently, the membrane module was soaked into NaOCl solution (3,000 mg/L) to remove the remaining material still attached to the membrane. The membrane was soaked for 12-h under homogeneous chlorine solution to remove the internal foulant into the membrane (R_f). Final, residual resistance represented irreversible fouling (R_{ir}).

3. Results and Discussions

3.1. Superficial liquid velocity of YEF (Yonsei End Free) module

The use of aeration to control fouling is highly important in the MBRs. Moreover, aeration is a key problem for the process as it constitutes one of the main operation costs. In the different concentration of mixed liquid suspended solid (MLSS), superficial liquid velocity of YEF module was investigated to determine dependence on aeration intensity. The one of the main objective of this research is that YEF module presents specific flow pattern as to superficial liquid velocity.

The YEF module of liquid velocity is very uniform at all the measuring points as shown in Fig. 4. It was indicated that configuration of YEF module was profitable rectilinear, therefore, the superficial liquid velocity increased with increasing the aeration intensity. With different concentration of MLSS, the superficial liquid velocity of YEF module increased with increasing the aeration intensity as shown in Fig. 5. The superficial liquid velocity depends on the MLSS concentration, decreased with increasing the MLSS concentration. The

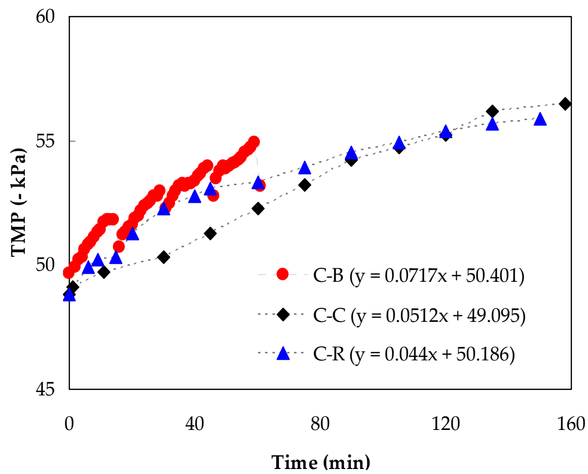

Fig. 6. TMP vs. time of the different filtration modes with continuous aeration mode.

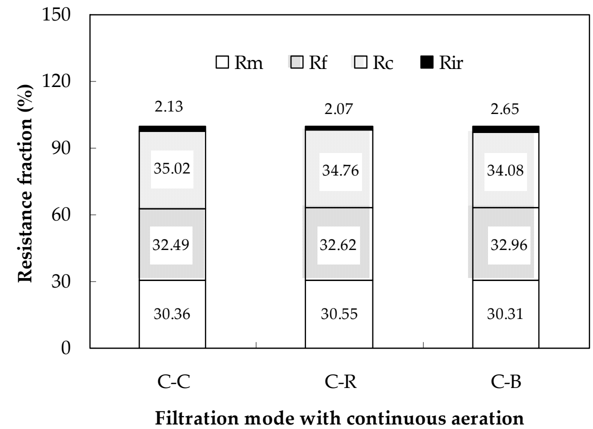

Fig. 7. Resistance fractions of the different filtration modes with continuous aeration mode.

reduction rate of the superficial liquid velocity was found to be 3 × 10^{-4} m · m^2 · hr/sec · m^3 per 1,000 mg MLSS/L. Operation and maintenance data reported that aeration intensity is operated with an air requirement from 0.28 to 0.75 m^3/m^2 · hr with fluxes between 10 and 20 LMH[1]. The effects of superficial liquid velocity have to be discussed in relation with the different aeration intensity and MLSS concentration. An increase in aeration intensity induced an improvement of superficial liquid velocity measured above 0.2 m/sec beyond 0.313 m^3/m^2 · hr of aeration intensity in all the applied MLSS concentration, as shown in Fig. 5. The effect of aeration intensity on superficial liquid velocity improvement is higher for lower MLSS concentration. Accordingly, superficial liquid velocity was dependent on MLSS concentration.

3.2. Continuous aeration mode

The TMP was monitored for the different filtration modes with continuous aeration mode. The TMP results in Fig. 6 were obtained during filtration time. It is common practice to operate MBRs under sub-critical flux conditions[21]. Due to the higher increase of fouling rates when flux higher than 30 LMH was applied (Fig. 6). It can be assumed that the critical flux of this system has been exceeded.

Filtration time was very short for the backwashing mode. However, the backwashing mode featured that the highest fouling rate at 0.0717 kPa/min and the relaxation mode was the lowest at 0.044 kPa/min. The relaxation mode featured a lower final TMP than the continuous mode. The TMP of the continuous mode was lower than the relaxation mode during 120-min of filtration, after increasing TMP rate.

In continuous mode, the formation of cake layer was reached. Some increase in the TMP after 120-min of filtration due to the accumulation of gel layer was caused by SMP or EPS on the membrane surface[22-24]. It indicated that irreversible fouling of continuous aeration mode increased than that of the relaxation mode (Fig. 7). Indeed, cake layer of the relaxation mode in the resistance fraction was almost the same with the continuous mode but assumed a loosely bound layer (Fig. 7). In the relaxation mode, the long time of relaxation exhibit a more efficient effect on TMP control[25,26]. In this study, the relaxation duration could be too short.

For the backwashing mode, it can be assumed that frequent backwashing extended the internal fouling of the membrane as in irreversible fouling (R_{ir}) (Fig. 7). It is expected that membrane pore can be almost opened by periodic backwashing. At the beginning of the filtration, the fine colloids and macromolecules infiltrating in the direction of permeate flow can also generate fouling inside of the membrane and contribute to the

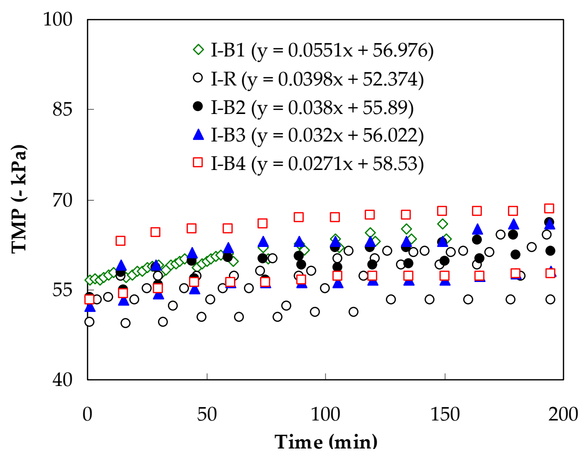

Fig. 8. TMP vs. time of the different intermittent aeration modes.

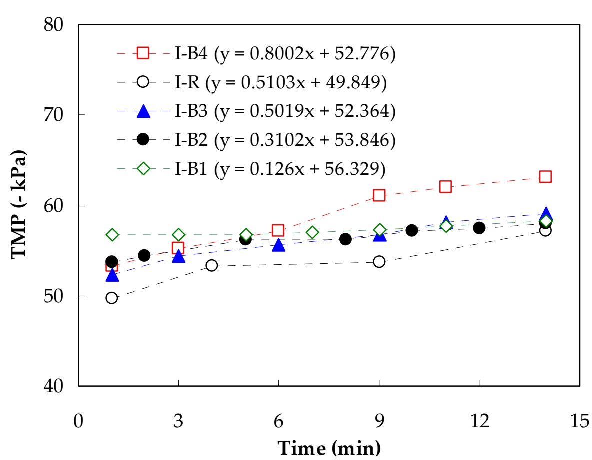

Fig. 9. TMP vs. time of the 1st cycle in the different intermittent aeration modes.

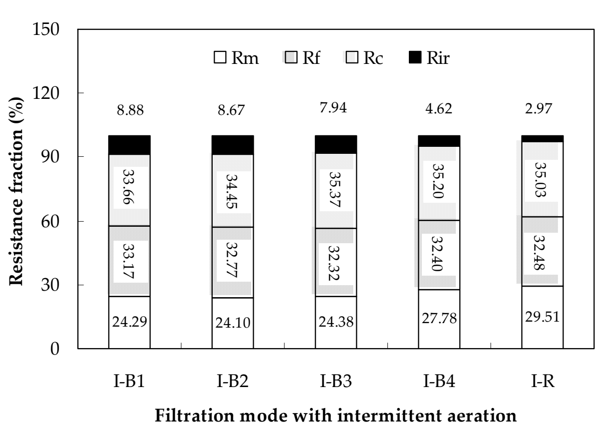

Fig. 10. Resistance fractions of the different filtration modes with intermittent aeration mode.

irreversible resistance (Fig. 7). Another effect possibly contributing to the membrane fouling is the continuous aeration mode of an internal face of the membrane. During the continuous aeration mode, a high fraction of small colloids and macromolecules can destroy biomass floc[27-29]. These studies indicate that their interaction play a major role in the membrane fouling. However, the limited filtration time of this experiment has to be considered. Because of the highest fouling rate observed in the backwashing mode for 60-min, it is expected that this may vary for longer filtration times.

3.3. Intermittent aeration mode

Differences in the TMP profiles were observed for five intermittent aeration modes with different filtration conditions during backwashing and relaxation modes (Fig. 8).

For intermittent aeration mode, Run I-B4 (20-s of no aeration time every 40-s) obtained the lowest fouling rate, and Run I-B1 (10-s of no aeration time every 20-s) resulted the highest fouling rate. Moreover, the fouling rate of I-B1 was higher than that of I-R (20-s of no aeration every 40-s with relaxation mode). The fouling rate decreased with increasing time of no aeration at the same intermittent aeration time, among the runs of I-B2, I-B3 and I-B4. Indeed, the fraction of irreversible resistance increased with decreasing with time of no aeration at all backwashing filtration modes (Fig. 10). With higher time of no aeration, the fouling rate of first cycle of all intermittent aeration modes increased (Fig. 10). The phenomenon can be explained by the fact that at time of no aeration by intermittent aeration mode possibly formed the thicker cake layer than that of continuous aeration mode. And then, at periodic backwashing, it is expected that gel and thin cake layer maybe sufficiently removed by heterogeneous layer. Especially, after periodic backwashing, subsequent cake layer caused by time of no aeration is rapidly formed as to the long time of no aeration, acting as a prefilter, and preventing further irreversible fouling (Fig. 10). Nevertheless, it can be indicated that

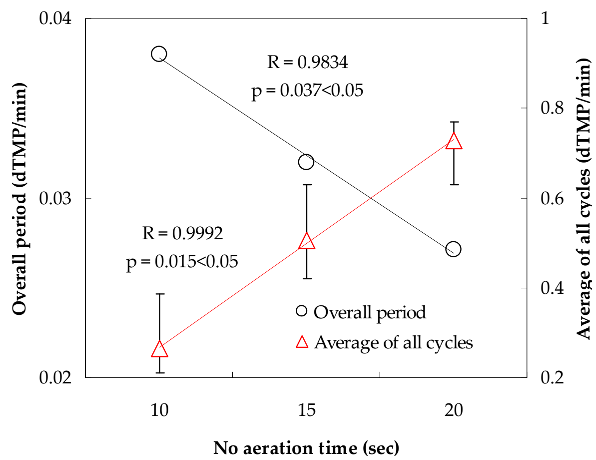

Fig. 11. Relationship between overall period (dTMP/min) and average of cycle (dTMP/min) for different off-time of intermittent aeration by Pearson correlation analysis.

irreversible fouling cannot be completely controlled by intermittent aeration.

At the aeration intensity for intermittent aeration mode of 10-s/10-s, cake layer during aeration-off period was not be effectively formed by subsequent aeration (Fig. 10). In the relaxation filtration mode with intermittent aeration mode of 20-s/20-s, it can be assumed that the cake layer might partially be removed by periodic relaxation time. Consequently, residual layer was prevented the fine foulants resulting the lowest irreversible fouling (Fig. 10).

3.4. Factors affecting fouling

Recently, factors affecting membrane fouling was estimated by Pearson correlation analysis[26,30]. Since Pearson correlation shows the clear relationship than linear correlation to predict the significant effect of operation parameters. Pearson correlation analysis was performed to identify the correlation between overall TMP rate and average of all cycles TMP rate for each experiment under different off-time of intermittent aeration. While analyzing the Pearson correlation in this study, only 20-s with on-time of intermittent aeration was applied, indeed in defined backwashing mode. R was an indicator of linear estimations and p was an indicator of correlation significance. Correlations were considered statistically significant at 95% confidence interval ($p < 0.05$). Correlation analysis was firstly carried out between overall TMP rate and average of all cycles TMP rate (Fig. 11). Linear relationship between the two parameters was found for both $R > 0.98$. Due to the definition of 20 sec with on-time of intermittent aeration, significant correlation could be proved for both parameters with $p < 0.05$. TMP rate has significant correlations with on-time of intermittent aeration. From linear and Pearson correlation analysis, two parameters were found to strongly affect off-time of intermittent aeration. Since these two values are interdependent, the cake layer formation with intermittent aeration did exhibit more positive effect on fouling control. Further, optimization of intermittent aeration should be considered at wide MLSS concentration and aeration intensity conditions.

3.5. Fouling mechanism on aeration mode

The initial of next TMP cycle less raised than the previous TMP cycle since the outer part of gel layer might be remove by relaxation and/or backwashing action. The removed gel layer might be controlled by physical method such as aeration but the remained layer might be require for chemical cleaning due to irreversible fouling such as internal pore blocking. On the other hand, the cake layer can easily removed by periodic backwashing. Further, it is expected that membrane pore maybe almost opened by periodic backwashing. At the beginning of the filtration, the fine colloids and macromolecules infiltrating in the direction of permeate flow can also generate fouling inside of the membrane and contribute to the irreversible resistance.

Based on the results presented in this study, fouling mechanisms for the continuous and intermittent aeration modes is proposed (Fig. 12).

3.5.1. Continuous aeration mode

At the beginning of filtration, a thin layer consisting of gel layer (solute and/or EPS/SMP) accumulates on the membrane surface (Fig. 12 (a)). The attached gel or thin cake thickness on the membrane is limited and

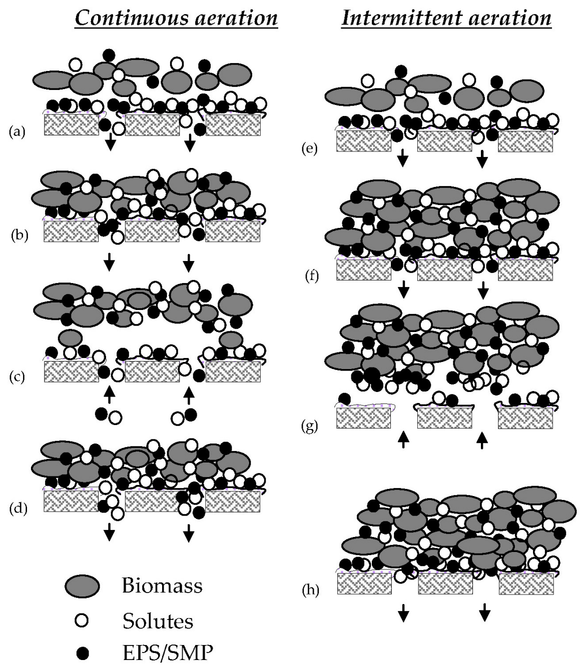

Fig. 12. Suggested fouling mechanisms for continuous and intermittent aeration modes with backwashing mode.

operates in a steady state due to application of continuous aeration (Fig. 12 (b)). During the periodic backwashing, gel and thin cake layer are removed from the membrane, however it will not be completely removed (Fig. 12 (c)). Moreover, it is expected that membrane pore can be almost opened by periodic backwashing. At the beginning of the filtration, the fine colloids and macromolecules infiltrating in the direction of permeate flow can also generate fouling inside of the membrane and contribute to the irreversible resistance (Fig. 12 (d)).

3.5.2. Intermittent aeration mode

At the beginning of filtration, a cake layer consisting of biomass accumulates on the membrane surface (Fig. 12 (e)). The attached cake thickness on the membrane rapidly increased, caused by subsequent no aeration leading to easier combining with gel layer and the formation of heterogeneous layer (biomass composites) on the membrane surface (Fig. 12 (f)). And then, during periodic backwashing, it is expected that gel and thin cake layer might sufficiently be removed by heterogeneous layer (Fig. 12 (g)). Especially, after periodic backwashing, subsequent cake layer formation during time of no aeration was rapid than frequent no aeration, acting as a prefilter (Fig. 12 (h)), and preventing further irreversible fouling (Fig. 10). Over time, decreasing irreversible fouling results in an enhanced process efficiency.

4. Conclusions

Backwashing with intermittent aeration did exhibit more positive effect on fouling control. The intermittent aeration provoked the formation of a cake layer on the gel layer which was previously formed during the aeration, resulting in increased TMP level. During backwashing, this composite layer is removed from the membrane surface. The cake layer can be removed effectively as the gel layer is reduced from membrane surface together. Consequently, at the beginning of the next filtration cycle, a cake and/or gel layer was covered instantaneously. These layers were then formed by intermittent aeration, acting as a prefilter and preventing further internal pore blocking. The thickness of the cake or gel layer was limited due to applied continuous aeration and hence cake layer was absent. This fouling layer is susceptible to further internal pore fouling resulting in further increase of TMP. During backwashing, the fouling layer remained on the membrane surface as the more firmly bound foulants, leading to a higher degree in pore fouling.

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