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The effect of aeration and non-aeration time on simultaneous organic, nitrogen and phosphorus removal using an intermittent aeration membrane bioreactor

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Abstract A laboratory-scale membrane bioreactor (MBR) was fed with synthetic wastewater to investigate the possibility of simultaneous removal of organic, nitrogen and phosphorus by intermittent aeration. The MBR consists of two compartments using a microfiltration membrane with 0.2 µm pore size and a surface area of 0.35 m^2. Hydraulic retention time was set at 24 hours and solid retention time 25 days. MLSS concentration in the reactor was in the range of 2,500–3,800 mg/L. The MLSS internal recycling ratio was maintained at 100% influent flow rate. Intermittent aeration was applied in this study to provide an aerobic–anaerobic cycle. Three stages of operations were conducted to investigate the effect of aeration and non-aeration on simultaneous organic and nutrient removal. In Stage 1, time cycles of aeration and non-aeration were set at 90/150 min and 150/90 min in the first and second compartment, the removal efficiency was 97%, 94% and 70% for COD, nitrogen and phosphorus respectively. In Stage 2, time cycles of aeration and non-aeration were set at 60/120 min and 120/60 min in the first and second compartment, the removal efficiency was 97%, 96% and 71% for COD, nitrogen and phosphorus respectively. In Stage 3, time cycles of aeration and non-aeration were set at 120/120 min and 120/120 min in compartment 1 and 2, the removal efficiency was 98%, 96% and 78% for COD, nitrogen and phosphorus respectively. Results show that longer non-aeration time in the second compartment provided better performances of biological phosphorus removal.

Keywords Flat sheet type immersed membrane; intermittent aeration; membrane bioreactor (MBR); nitrogen removal; organic removal and phosphorus removal

Introduction

In domestic wastewater treatment, the combination of the activated sludge process with membrane filtration for organic carbon removal was first introduced in the 1960s (Smith *et al.*, 1969; Bemberis *et al.*, 1971; Hardt *et al.*, 1970). A high degree of organic oxidation was achieved while producing solid free effluent. Later studies succeeded to obtain nitrogen removal by simultaneous nitrification and denitrification (Suwa *et al.*, 1992; Chiemchaisri *et al.*, 1992; Nagaoka, 1999; Isaacs, 2000). This process could be operated with a very long solid retention time (SRT), resulting in complete retention of slow growing nitrifying bacteria in the system. Intermittent aeration can achieve nitrogen and phosphorus removal by simultaneous nitrification and denitrification, P-uptake and P-release in the same reactor in accordance with time cycle of aeration and non-aeration (Chiemchaisri *et al.*, 1993; Seo and Lee, 1995). However even though intermittent aeration was successful in removing nitrogen, phosphorus removal was difficult to achieve at a higher level. In addition it showed unstable nitrogen removal in its application to treat domestic sewage of rural settlements because of incomplete denitrification (Ueda *et al.*, 1996). The double tank type intermittent aeration activated process has been studied for simultaneous removal of organic, nitrogen and phosphorus, and stable organic (96.2% as COD), nitrogen (91.6% as TN) and phosphorus (66% as TP) removal have been obtained (Seo *et al.*, 2000).

This study aims to remove the organic, nitrogen and phosphorus simultaneously using two-stage intermittent aeration. The specific objective is to observe the effect of aeration and non-aeration time on simultaneous organic, nitrogen and phosphorus removal using intermittent aeration MBR.

Materials and methods

Equipment

A lab-scale immersed MBR was installed at the Environmental Engineering Laboratory, UTM. The schematic flow diagram of the system is shown in Figure 1. This system fed with a capacity of 25 litres of synthetic wastewater per day. The bioreactor consists of two compartments with working volume of 25 litres (8 litres for first compartment and 17 litres for second compartment). A flat sheet microfiltration membrane was immersed in the second compartment for suction type filtration. In this study, seven pairs of membrane sheets were set in parallel providing the total membrane area of 0.35 m^2. The membrane was made of polyolefins with pore size 0.2 μm and supplied by Yuasa Coporation (Japan). The MBR system used four timers to control the stirrer, air pump, influent pump and suction pump.

Synthetic wastewater

Synthetic wastewater was used for the experiment instead of actual wastewater due to the inconsistent nature of nutrient concentration in raw wastewater, particularly phosphorus content. The composition of the synthetic wastewater is shown in Table 2. Concentrated feed solution was stocked in a refrigerator at 4°C. It was later diluted with tap water to the desired COD concentration before being fed to the reactor. In this synthetic wastewater, acetic acid was used as a carbon source because it is easily biodegradable and suitable as a representative of organic matter in domestic wastewater. Acetic acid can be found in the actual raw domestic wastewater because acetic acid is easily produced during anaerobic conditions in the storage tanks of wastewater or in sewer pipes. KH_2PO_4 was used to provide the PO_4 in the synthetic wastewater. It was used by bacteria during cell synthesis, maintenance and energy transport and remove by bacteria during the process of luxury uptake of phosphorus. Ammonium chloride was used as a nitrogen source because organic

Table 1 Membrane classifications

Membrane material	Polyolefins
Membrane configuration	Plate and flame
Pore size	0.2 μm
Surface area	0.35 m^2
Manufacturer	Yuasa Corporation (Japan)

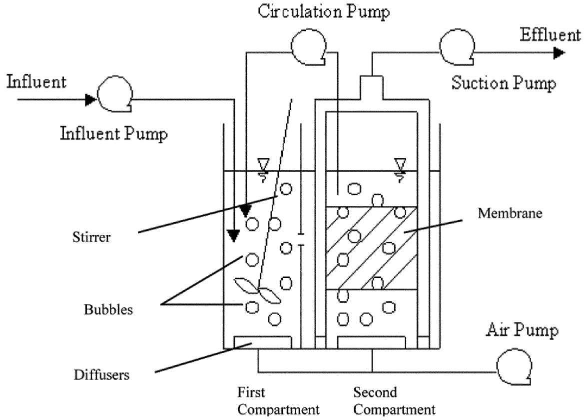

Figure 1 Schematic diagram of the MBR

Table 2 Composition of synthetic wastewater (mg/L)

Composition	Concentration (mg/L)
CH_3COOH	500–650
NH_4Cl	100
KH_2PO_4	16
$FeCl_3.6H_2O$	4
$CaCl_2$	11
$MgSO_4$	17
KCl	8
NaCl	8
$NaHCO_3$	350

nitrogen in wastewater is rapidly converted to ammonium nitrogen in the aeration tank and later stages of nitrogen conversion are rate-limiting. Sodium bicarbonate was used for alkalinity to keep pH at around neutral (Nagaoka, 1999).

Operational schedule

Tables 3, 4 and 5 display the operation cycle for Stages 1, 2 and 3. The objective of all stages was to investigate the effect of aeration and non-aeration on the simultaneous organic, nitrogen and phosphorus removal. In these operations, the wastewater was circulated from the second compartment to the first compartment with a similar flow rate as the influent flow rate. MLSS concentration in the reactor was in the range of 2,500–3,800 mg/L during the operation of Stages 1, 2 and 3. Nagaoka *et al.* (1998) found the critical value of the flux membrane for maintaining good performance to be around 0.1 m^3/m^2.day with MLSS between 5,000 to 10,000 mg/L. In this study, the operating membrane flux was higher than the 0.1 m^3/m^2.day (0.14 to 0.18 m^3/m^2.day), therefore the system was conducted with the low MLSS (2,500 to 3,800 mg/L) to reduce the biofouling. The mean cell retention time (SRT) for all stages was 25 days. The excess sludge was removed from the first compartment and second compartment according to volume ratio of the compartment. The removed excess sludge volume in this stage was 1 litre per day (0.3 litre from first compartment and 0.7 litre from second compartment. All stages were in operation for one month. Table 6 shows the summary of the operation cycle time for Stages 1, 2 and 3.

Table 3 The operation time in one process cycle for Stage1

Period	First compartment	Second compartment
00:00–00:30	Influent pump on Aeration off	Aeration on
00:30–02:30	Influent pump on Aeration off	Aeration on Suction pump on
02:30–04:00	Influent pump off Aeration on	Suction pump off Aeration off

Table 4 The operation time in one process cycle for Stage 2

Period	First compartment	Second compartment
00:00–00:30	Influent pump on Aeration off	Aeration on
00:30–02:00	Influent pump on Aeration off	Aeration on Suction pump on
02:00–03:00	Influent pump off Aeration on	Suction pump off Aeration off

Table 5 The operation time in one process cycle for Stage 3

Period	First compartment	Second compartment
00:00–00:30	Influent pump on Aeration off	Aeration on
00:30–02:00	Influent pump on Aeration off	Aeration on Suction pump on
02:00–04:00	Influent pump off Aeration on	Suction pump off Aeration off

Analytical methods

The influent and effluent from the MBR were analysed for COD, NH_3-N, NO_2-N, NO_3-N, PO_4 and MLSS, which were determined according to the procedures of *Standard Methods for the Examination of Water and Wastewater* (1995). In this study, phosphate was used as an indicator for phosphorus removal due to its function in enhanced biological phosphorus removal (EBPR).

Results and discussion

System performance

Organic removal. The COD concentration in the effluent was observed to be an average of 16 ± 10mg/L as shown in Table 7. Figure 2 shows that the COD removal efficiency was

Table 6 The operation time cycle for Stages 1, 2 and 3

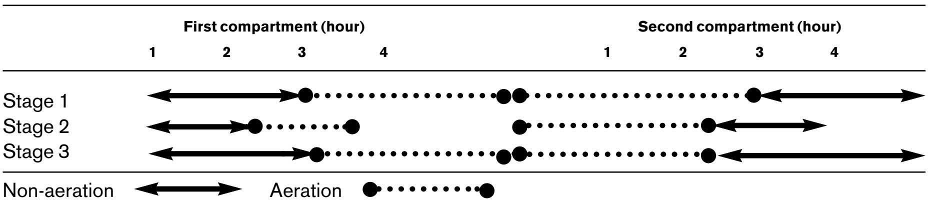

Table 7 Summary of the results for Stages 1, 2 and 3

Stage	Items	Influent (mg/L)	Effluent (mg/L)	Removal (%)
1	COD	625 ± 25	15 ± 10	97.6 ± 2.0
	Nitrogen	29.5 ± 4.0	1.08 ± 0.3	96.0 ± 1.0
	NH_3-N	29.5 ± 4.0	0.05 ± 0.03	99.8 ± 0.2
	NO_2-N	0	0.04 ± 0.01	–
	NO_3-N	0	1.1 ± 0.2	96.1 ± 1.0
	PO_4	12 ± 0.5	3.0 ± 0.4	71.8 ± 3.0
2	COD	678 ± 5	17 ± 10	97.5 ± 2.0
	Nitrogen	22.3 ± 0.3	0.9 ± 0.3	95.7 ± 1.0
	NH_3-N	22.3 ± 0.3	0.03 ± 0.02	99.8 ± 0.2
	NO_2-N	0	0.03 ± 0.02	–
	NO_3-N	0	0.9 ± 0.3	95.9 ± 1.0
	PO_4	10.9 ± 0.2	3.0 ± 0.4	71.2 ± 3.0
3	COD	674 ± 10	16 ± 10	97.8 ± 2.0
	Nitrogen	24.3 ± 0.3	0.9 ± 0.15	96.3 ± 1.0
	NH_3-N	24.3 ± 0.3	0.04 ± 0.03	99.8 ± 0.2
	NO_2-N	0	0.03 ± 0.01	–
	NO_3-N	0	0.8 ± 0.2	96.5 ± 1.0
	PO4	11.2 ± 0.5	2.56 ± 0.4	78.3 ± 3.0

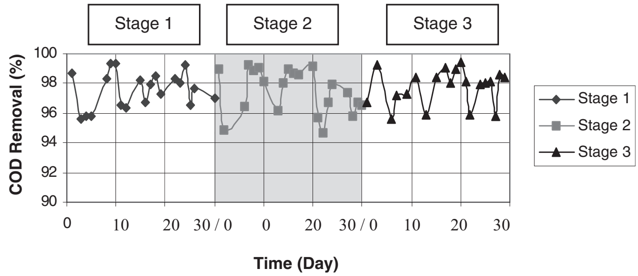

Figure 2 COD removal in intermittent aeration MBR

97.6 ± 2.0%, 97.5 ± 2.0% and 97.8 ± 2.0% for Stages 1, 2 and 3 respectively. The results shown that there was no significant difference on COD removal efficiency although operated with varying aeration and non-aeration time. This indicates that organic matter can be degraded under both aerobic and anaerobic conditions. The performance of the MBR on organic removal appears to be relatively insensitive to the cycle time, whereas it resulted in high COD removal efficiency.

Nitrogen removal. In this study, nitrogen in the influent is assumed to be equal to ammonia-nitrogen concentration, and the nitrogen in the permeate is assumed to be equal to inorganic nitrogen (the sum of NH_3-N, NO_2-N and NO_3-N). Figure 3 displays the removal percentage of nitrogen in Stages 1, 2 and 3. The nitrogen removal for Stages 1, 2 and 3 was 96.0 ± 1.0%, 95.7 ± 1.0% and 96.3 ± 1.0% respectively as shown in Table 7. The removal of nitrogen was similar in this study although operated with difference aeration and non-aeration time. The time cycles used by these three stages resulted in a high degree of nitrogen removal efficiency.

The nitrification process in this study was observed at 99.8 ± 0.2% for Stages 1, 2 and 3. The DO level for Stages 1, 2 and 3 have been recorded between 3.0 to 5.5 mg/L during the aeration period. Tchobanoglous and Burton (1991) reported that DO concentrations above 1 mg/L are essential for nitrification to occur. Thus the DO level during the aeration period in Stage 1, 2 and 3 is suitable for the occurence of nitrification. The MBR system is shown to perform satisfactorily during the nitrification process and is not affected by the difference of aeration and non-aeration time that was used in this study. The aeration and non-aeration period used in Stages 1, 2 and 3 resulted in a high degree of nitrification in this MBR system. This indicates that the aeration and non-aeration time for Stages 1, 2 and 3 were sufficient for the occurence of the complete nitrification process.

The denitrification process for the Stages 1, 2 and 3 was observed at 96.1 ± 1.0%, 95.9 ± 1.0% and 96.5 ± 1.0% respectively. These indicate that all stages achieved a high degree of denitrification process. The results show that there was not much difference in the efficiency of the denitrification process although operated in varying aeration and non-aeration

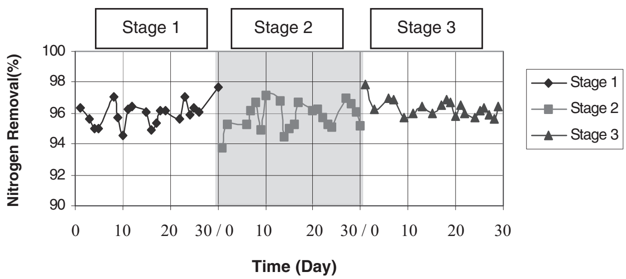

Figure 3 Nitrogen removal in intermittent MBR

periods. The results point out that the aeration and non-aeration time in Stages 1, 2 and 3 were adequate for the denitrification process.

Phosphorus removal. The removal percentages of phosphate for Stages 1, 2 and 3 are given in Figure 4 with removal efficiency 71.8 ± 3.0%, 71.2 ± 3.0% and 78.3 ± 3.0% respectively as shown in Table 7. Stage 3 showed the highest phosphate removal efficiency, followed by Stage 1 and then Stage 2. This was due to the non-aeration time in the compartment 2 for Stage 3 being the longest, i.e. 2 hours compared to others. The non-aeration phases in compartment 2 in one cycle time for Stages 1, 2 and 3 were 1.5, 1 and 2 hours respectively as shown in Table 6. With the long non-aeration period, the DO inside the compartment will drop to zero and a complete denitrification process will occur. Thus is provided a suitable condition for the uptake of acetate, which is stored as poly-hydroxybutyrate (PHB), into the polyphosphate accumulating organisms (PAO). The phosphate removal efficiency for Stages 1 and 2 was 71.8 ± 3.0% and 71.2 ± 3.0% respectively. In general, the phosphate removal efficiency was similar for both stages. Although Stage 1 which was operated for 90 minutes of non-aeration time in the second compartment had a longer non-aeration time compared with the Stage 2 which was operated for 60 minutes of non-aeration time in the second compartment, the differences of phosphate removal efficiency between these two stages were very insignificant.

In the present study, the membrane functioned as a physical barrier to retain the PAO in the reactor and filter out the biologically treated wastewater. Beun (2001) reported that the typical diameter for the PHB in the range of 0.2 to 0.5 μm. PHB is used by the PAOs to generate energy for growth and for polyphosphate synthesis during aerobic conditions. The PHB is stored inside the PAO during the anaerobic conditions (Smolders, 1995). These two earlier studies indicated that the size of PAOs is typically bigger than 0.5 μm. Therefore the microfiltration membrane (0.2 μm) that was used in MBR this study has retained the PAO in activated sludge. Thus is provided sufficient biomass for the enhanced biological phosphorus removal (EBPR) to take place during aerobic conditions and PHB accumulation during anaerobic conditions.

Membrane flux. The flux of membrane used in the Stages 1, 2 and 3 was maintained at 0.14, 0.14 and 0.18 m^3/m^2day respectively. The transmembrane pressure is measured using a pressure gauge. The measured transmembrane pressure for all stages ranges from 0 to 3.4 kPa within one operation cycle. The transmembrane pressure was indicated stable at 3.4 kPa during the operation period. The low transmembrane pressure in this MBR was due to the intermittent suction applied for the membrane. Chiemchaisri *et al*. (1992) reported that the intermittent suction could prevent the clogging of membranes to some extent (without any regular cleaning). Besides that, the membrane module was installed directly over the diffusers through which air was supplied. The sheering stresses generated by the

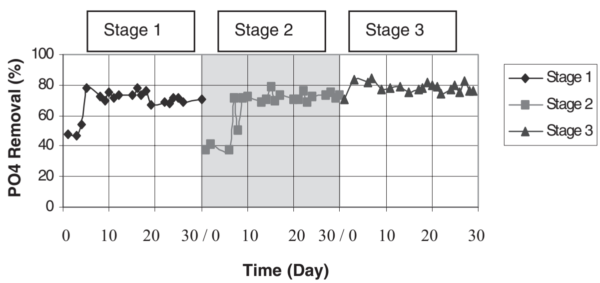

Figure 4 The removal percentage of phosphorus

uplifting flow of bubbling air prevent accumulation of solid on the membrane surface (Seo *et al.*, 2000).

Reactor analysis

Fluctuation of nitrogen, phosphate and DO concentrations in one cycle of operation is shown in Figure 5. Nitrification, denitrification, release and uptake of phosphate were observed during the one cycle operation in the first compartment as shown in Figure 5. The ammonia-nitrogen, nitrate-nitrogen and phosphate level in the second compartment was observed to be lower than the first compartment. This shows that some portion of ammonia-nitrogen, nitrate-nitrogen and phosphate have been removed in the first compartment.

Ammonia-nitrogen concentration was reduced to almost zero level while nitrate-nitrogen increased to 0.7mg/L during the aeration period in the second compartment. During the non-aeration period in the second compartment, the ammonia-nitrogen was observed to increase at the beginning of the non-aeration time, and then decreased at the end of the non-aeration period. Whereas nitrate-nitrogen was observed to decrease at the end of the non-aeration period. This is due to the DO concentration decreasing slowly during non-aeration in the second compartment. Although not very obvious, the release and uptake of phosphate was observed in one cycle of the non-aeration period and the aeration period in the second compartment.

General discussion

Trouve *et al.* (1994), Ghyoot *et al.* (1999), Fan *et al.* (1998) and Chaize and Huyard (1999) have conducted the studies with volumetric loading rate ranges between 0.45 to 1.5 kg COD $m^{-3}d^{-1}$ with corresponding removal efficiencies 86–96%. In the present study, the volumetric loading rate was between 0.62 to 0.70 kg COD $m^{-3}d^{-1}$ and the removal efficiencies 97% to 98%. This shows that the COD removal efficiencies in the present study were almost similar or even better than the previous studies that have been conducted. The nitrogen removal for Stages 1, 2 and 3 was 96.0 ± 1.0%, 95.7 ± 1.0% and 96.3 ± 1.0% respectively. The time cycles used by these three stages were given a high degree of nitrogen removal efficiency. This illustrated that the removal of nitrogen was similar in this study although operated with differences in aeration and non-aeration time. The phosphate removal efficiency for the Stages 1, 2 and 3 was 71.8 ± 3.0%, 71.2 ± 3.0% and 78.3 ± 3.0% respectively. Stage 3 showed the highest phosphate removal efficiency, followed by Stage 1 and then Stage 2. This was due to the non-aeration time in the second compartment for Stage 3 was the longest among these three stages. This finding is consistent with Seo *et al.* (2000) who studied the performance of MBR on simultaneous organic and nutrient removal. From his

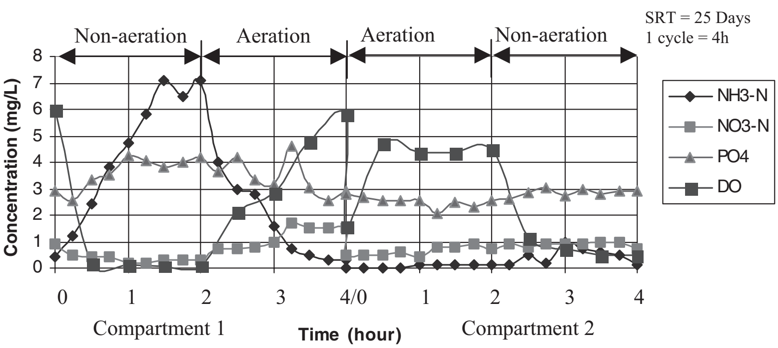

Figure 5 Variation of ammonia-nitrogen, nitrate-nitrogen and phosphate in one cycle operation period in MBR for Stage 3

study, it was reported that an expended period of non-aeration might be required for enough phosphorus release and subsequent uptake in aeration time.

Conclusion

The findings from this study served to emphasize that the operation cycle of the aeration and non-aeration period does not effect the organic and nitrogen removal in MBR. Whereas, longer non-aeration time in the second compartment is needed for a high degree of EBPR. To increase the performance of organic, nitrogen and phosphorus removal, the DO level has to be reduced to zero concentration during the non-aeration period in the second compartment.

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