The Performance of Ammonia Nitrogen Removal by using Membrane-Aerated Biofilm Reactor as Domestic Wastewater Treatment

Kanty Driantami and Setyo S. Moersidik
Department of Civil Engineering and Environmental Engineering, Universitas of Indonesia, 16424 Depok, West Java, Indonesia

Abstract: High concentration of NH$_3$-N in wastewater discharges from sewage treatment plant can cause eutrophication of the surface water that has the negative impacts for aquatic ecosystems. Membrane-Aerated Biofilm Reactor (MABR) has been proposed as a wastewater technology to reduce NH$_3$-N concentration in domestic wastewater. This study observed the performance of NH$_3$-N removal in domestic wastewater using MABR. Domestic wastewater contains concentration of NH$_3$-N from 73-104.8 mg/L (0.12-0.24 kg NH$_3$-N/m$^3$.d) and COD from 352-408 mg/L (0.56-1.05 kg COD/m$^3$.d). MABR was supplied by oxygen at the pressure of 20 kPa and study performed for 3 Hydraulic loading rate (HRT) variations which were 8, 10 and 12 h. After 33 days of running, the result showed COD/N ratio were about 3.9-5.72 with maximum efficiency of COD and NH$_3$-N removal occurred when HRT 12 h, reached 88 and 89.58%, respectively. This indicated that NH$_3$-N could remove by MABR Rat low COD/N ratio. Furthermore, autotrophs bacteria that responsible for oxidized NH$_3$-N to NO$_3$ and NO$_2$ have slower growth rates compared with heterotrophs bacteria. Thus, longer HRT provided benefit for nitrification process and high NH$_3$-N removal efficiency has been achieved.

Key words: Membrane aerated biofilm reactor, ammonia nitrogen removal, domestic wastewater, autotrophs bacteria, hydraulic retention time, Indonesia

INTRODUCTION

Untreated domestic wastewater in Indonesia contains the high concentration of organic matter and nitrogen with the average concentration of 615.01 and 84.76 mg/L, respectively (Idman, 2008). The excessive effluent concentration of NH$_3$-N from sewage treatment plant that released to water body can cause the environmental problem such as eutrophication and toxicity for aquatic life. Because the increases in NH$_3$-N concentration, pH increase and it will give impact on the high level of toxicity in the water body. The rule of domestic wastewater in Indonesia regulated on the regulation of minister of environment and forestry number 68 the years 2016. The regulation has the strict standard quality of NH$_3$-N with the permitted effluent concentration of 10 mg/L. Hence, the treatment which can remove the high concentration of NH$_3$-N from domestic wastewater is required. One of the parameters that affected the nitrification process of oxidized NH$_3$-N was Chemical Oxygen Demand (COD). Because, COD directly affects the growing competition between autotrophs and heterotrophs bacteria population (Marhossamni et al., 2010).

Membrane-aerated biofilm reactor is the treatment that proposed to treating domestic wastewater, specifically for simultaneous organic carbon and nitrogen removal (Liu et al., 2010, Truttim and Sohsalam, 2016, Roya et al., 2015). The membrane has different function in MABR. If other technology used the membrane as filtrate water, MABR used it as a media to supplied oxygen to immobilization microorganisms which degrade the organic matter and oxidized NH$_3$-N (Lin et al., 2015). The transferred oxygen to the membrane directly will provide the small air bubble which prevents the air stripping of volatile organic compounds and produces high Oxygen Transfer Efficiency (OTE) so it can reduce the cost of aeration process (Cerqueira et al., 2013; Wang et al., 2016). High OTE also provides high concentration of high dissolved oxygen which gives advantage for heterotroph and autotrophs bacteria. Therefore, MABR is a suitable technology for treating domestic wastewater that contains the high concentration of NH$_3$-N (Syron et al., 2015).

The autotroph bacteria which have a role in nitrification have a slower growth rate compared heterotroph bacteria. Because both of them, need oxygen for their growth thus COD/N ratio needs to be considered in simultaneously COD and NH$_3$-N removal process. Low COD/N ratio gives a benefit of nitrification process. While the high of ratio COD/N will be disturbed the nitrification
process that causes the competition of oxygen between autotroph and heterotroph bacteria (Lin et al., 2016). A pilot scale research before, resulted in removal efficiency for TSS, COD, NH$_4$-$N$ and TN were 91, 83, 95 and 66%, respectively at the aeration efficiency of 6 kgO$_2$/kWh (Cote et al., 2015). Another research that focused on the effect of COD/N ratio on the performance of nitrogen removal showed that COD/N ratio optimal in removing nitrogen has ranged from 3-10 with the removal efficiency up to 93.1% (Lin et al., 2016).

Few researched about MABR used hollow fiber membrane as a media for biofilm to attached (Lin et al., 2015; Syron et al., 2015; Tian et al., 2016). The hollow fiber membrane has a wide specific surface area so that it can form a larger biofilm layer on the media. Cross-diffusion will occur inside the membrane where oxygen will diffuse through the membrane into the biofilm that attached to the outer surface of the membrane while the waste water pollutants will contact with the biofilm from inside the membrane (Panrare et al., 2008; Narikhetuadomkas and Tondlee, 2016; Syron and Casey, 2008). Due to the ability of MABR in simultaneous organic carbon and nitrogen removal, this research was focusing on the performance of MABR in removal high NH$_4$-$N$ from domestic wastewater. Media that used for MABR is hollow fiber membrane. Three variants of hydraulic retention time applied for this study which was 8, 10 and 12 h to observe the optimum time for removal of COD and NH$_3$-$N$.

**MATERIALS AND METHODS**

**Wastewater characteristics:** Domestic wastewater was obtained from one of the head offices of industrial in Jakarta, Indonesia. Domestic wastewater that used as influent of MABR contains the high concentration of NH$_3$-$N$ which ranged from 73-104.8 mg/L. COD and BOD concentration in domestic wastewater were typically 332-780 and 269-289 mg/L with pH was around 7.3-8.3. The biodegradability index (BOD$_5$/COD) of domestic wastewater ranged from 0.61-0.64.

**MABR operation:** This research performed on the laboratory scale with the scheme of MABR which showed in Fig. 1 Ultrafiltration Hollow Fiber Membrane (polyacrylonitrille) was used as a media to supply the oxygen to biofilm. The module membrane height and diameter was 420 and 90 mm, respectively with the membrane that has an inner diameter of 0.9 mm and an outer diameter of 1.6 mm. The membrane surface area that.
available for biofilm attachment was 51 m². The reactor was constructed with plexiglass and had a working volume of 6 L with 45 cm height and 14 cm diameter. There were 2 sampling ports on the reactor that used to take the effluent sample for laboratory tests. The wastewater flows through the reactor by up flow with a peristaltic pump at the flow velocity of 10.42, 8.33 and 6.34 L/min. This research conducted three different variants of hydraulic retention time which are 8, 10 and 12 h. The aerator Rean LP-20 was supplied the oxygen to biofilm through the membrane from above the reactor intermittently (3 h on, 1 h off) with the capacity of 25 L/min and pressure at 20 kPa. The temperature in the reactor adjusted at 25±1°C which was the average temperature in Indonesia.

In seeding process to cultivate biofilm, MABR was seeding with 2L activated sludge (MLSS 6000 mg/L) which collected from oxidation ditch unit in WWTP Jatinegara, Bekasi. The seeding process took place with the continuous system because it generated the effective nitrogen removal compared to the batch system (Sun et al., 2009). After the biofilm formed during days 6, sludge was discharged and replaced with domestic wastewater for acclimatization process. Acclimatization process has done for 12 days until the steady state has achieved and biofilm could adapt to domestic wastewater. The pH and DO were measured during the seeding and acclimatization process to observe the bacteria activity in the reactor.

Analytical methods: The laboratory tests for COD and NH₄-N concentrations from the effluent conducted to evaluate the performance of MABR. COD and NH₄-N were determined according to the National Standard of Indonesia (SNI 6989.72.2009) and standard methods (8038.2005). DO were measured by DO Meter (SensiDirect, Oxi2000) and pH was measured by pH meter (pH-009). ANOVA test was used to determine the optimum hydraulic retention time based on the significance of analytical statistics.

RESULTS AND DISCUSSION

Start-up: During the seeding process, the pH was around 7.4-7.9. Biofilm can optimally grow with this condition because aerobic bacteria have optimal pH which ranged from 6.5-8.5 (Spellman, 2003). Bacteria that grow inside the membrane were from activated sludge has the aerobic bacteria such as Haliscomenobacter hydrossis and Sphaerotilus natans, Zoogloea ramigera from floe formed, Nitrosomonas dan Nitrobacter (Gerardi, 2006). At acclimatization process, pH increased from up to 8.1 on days 11. This pH condition gave benefit for nitrifying bacteria since Nitrosomonas and Nitrobacter have the pH optimal for growing which ranged from 7.2-8.8 and 7.2-9.0, respectively (Sajuni et al., 2010). DO concentration during the acclimatization process were always above 3 mg/L that provided sufficient oxygen for heterotrophs and autotroph bacteria. COD removal was observed to see the activities of bacteria in degradation organic matter. At day 17 of acclimatization process, COD removal efficiency has decreased from 94.49 up to 84.49% and increased again on the next day to 90%. This fluctuation of removal efficiency indicated the steady state of the biofilm in MABR.

The performance of cod and NH₄-N removal under different hydraulic retention time: Figure 2 showed pH condition and DO concentration in the reactor for three different hydraulic retention time. pH condition was fluctuating since the activity of nitrifying bacteria for oxidized the NH₄-N. The pH ranged from 6.9-8.1. The decreased of pH on day 21 occurred from 7.7-6.9 that cause of the high activity of nitrifying bacteria which consumed the alkalinity to oxidized NH₄-N to Nitrite and Nitrate (Hong et al., 2012). As the high of activity of nitrifying bacteria, the removal efficiency of NH₄-N increased from 33.66-53.95%. Whereas pH was increased up to 8.1 occurred because of the completed nitrification process. Fluctuation condition of pH did not affect the removal efficiency of COD. Although, there were decreasing of pH, removal efficiency of COD still has good performance above 80%. During the operation of MABR, dissolved oxygen was always above 5 mg/L. Therefore, competition between nitrifying and heterotroph did not occur because of an adequate oxygen in MABR and gave benefit for nitrifying bacteria to nitrification process. High dissolved oxygen during the operation proved that MABR has the high oxygen transfer.

An observation about the relationship between organic loading rate and nitrogen loading rate has
Fig. 3: a) organic loading rate on removal efficiency of COD and b) nitrogen loading rate on removal efficiency of NH₃-N

represented in Fig. 3. It showed that the performance of MABR of COD removal was about 65.36-88% when the organic loading rate ranged from 0.48-0.80 kgCOD/m³.day (Fig. 3a). For an organic loading rate of 0.63 kgCOD/m³.day, the removal of COD reached the maximum efficiency and while the organic loading rate of 0.54 kg/m³.day, the minimum removal efficiency of COD occurred. At the removal of NH₃-N, the maximum efficiency was 89.58% for the lowest nitrogen loading rate of 0.122 kgNH₃-N/m³.day. Whereas, when the nitrogen loading rate increased to 0.24 kgNH₃-N, it resulted in the minimum efficiency of 35.66% (Fig 3b). From the result, it can conclude the MABR performance of NH₃-N removal will reach the optimal efficiency when the nitrogen loading rate of domestic wastewater is lower than 0.122 kgNH₃-N/m³.day.

After the treatment of domestic wastewater by using MABR conducted at three different hydraulic retention time it resulted that the effluent concentration COD from 46-115 mg/L with the removal efficiency from 65.36-88% (Fig. 4). Almost all of the effluent had met COD standard quality of domestic wastewater in Indonesia of 100 mg/L, except for effluent on the first three days of the treatment. The good removal efficiency occurred due to a sufficient dissolved oxygen that used by heterotrophs bacteria to degrade the organic matter in the treatment of domestic wastewater in MABR (>3 mg/L). As increasing of influent of COD, the removal efficiency of COD tends to increased (Fig. 4). During the treatment of domestic wastewater high removal efficiency of >80% tends to occur at a high influent concentration of COD (>400 mg/L). It showed from days 26-27 at hydraulic retention time of 10 h, the stable removal efficiency of COD occurred at 82.58, 82.9 and 84.82%. Otherwise, at days 21 of hydraulic retention time 8 h when the effluent concentration was lower than 400 mg/L (332 mg/L), the removal efficiency reached the minimum efficiency of 65.36%. But the effluent of NH₃-N only met the standard quality of domestic wastewater in Indonesia of 10 mg/L at last days of the treatment with the concentration and the removal efficiency were 7.6 mg/L and 89.58%, respectively. Nevertheless, removal of NH₃-N at the hydraulic retention time of 12 h still in a good performance with the efficiency ranged from 78.07-89.58% (Fig. 4). Furthermore, this high of removal efficiency of NH₃-N due to the decreased of influent NH₃-N since days 25.

From Fig. 5a, it showed that the performance of removal efficiency of COD in hydraulic retention time 10 and 12 h were not too different. If it observed by mean and media, hydraulic retention time of 12 h (Q₁ = 82.77, X = 83.78) provided larger value compared to hydraulic time of 10 h (Q₁ = 82.59, X = 83.22). Furthermore, the distribution data of 10 h tend to be centered and at 12 h the distribution data were comparatively wider. In contrast
to COD, the performance removal efficiency of NH$_3$-N showed (Fig. 5b) that mean and median in hydraulic retention time 12 h (Q2 = 80.23; X = 83.1) were the biggest value compared to the performance in 8 h and 10 h. According to the results, simultaneous COD and NH$_3$-N removal has the good performance. The MABR provided the removal efficiency of COD and NH$_3$-N >80% which could meet the standard quality of domestic wastewater in Indonesia.

The result was similar with ANOVA test that conducted to compare the effect of different hydraulic retention time of 8, 10 and 12 h on removal efficiency of COD and NH$_3$-N. There was a significant effect of hydraulic retention time of words remembered at the p<0.05 label for three condition [F (2, 12) = 9.393, p = 0.04 for COD; F (2, 12) = 32.779, p = 0.00 for NH$_3$-N]. Post hoc comparisons using the Turkey HDS test indicated that the mean score for the hydraulic retention time of 8 h (M = 0.74, SD = 0.05 for COD; M = 0.49, SD = 0.01 for NH$_3$-N) was significantly different than the hydraulic retention time of 10 h (M = 0.82, SD = 0.02 for COD; M = 0.72, SD = 0.04 for NH$_3$-N). However, the hydraulic retention time of 12 (M = 0.83, SD = 0.03 for COD; M = 0.83, SD = 0.05 for NH$_3$-N) did not significantly differ from the hydraulic retention time of 10 h. Taken together, these result suggest the longer retention time do have an effect of the removal efficiency of COD and NH$_3$-N. Specifically when the hydraulic retention time conducted longer, the removal will achieve the higher efficiency. From the result of research and statistical analysis, the optimum removal efficiency of COD and NH$_3$-N to achieve the effluent which meets the standard quality of domestic wastewater in Indonesia was 12 h.

**Effect COD/N ratio for performance of MABR:** During the treatment of MABR, the COD/N ratio ranged from 3.91-5.73. At days 26-28, increasing of COD/N ratio followed by increasing the removal efficiency of COD. It also influenced by another factor such as dissolved oxygen. When organic carbon was excessive, heterotrophs bacteria will use the carbon as a donor elector to obtained the energy faster. Otherwise, when COD/N ratio decreased the removal efficiency tend to increase. Decreasing of COD/N ratio followed by increasing of removal efficiency which occurred on days 21, 22, 25, 19 and 32. On days 32, removal of NH$_3$-N reached the high efficiency of 80% with the COD/N ratio was 3.9. This because of the small amount of organic matter in wastewater that will decrease the activity of bacteria heterotroph and nitrification process will running well.

**CONCLUSION**

This study conducted the observation about effect hydraulic retention time on removal efficiency of COD and NH$_3$-N simultaneously by using Membrane-aerated biofilm reactor. MABR reached the maximum removal efficiency of COD and NH$_3$-N in 88 at day 22 and 89.58% at day 23, respectively. During the treatment of domestic wastewater, the measured of concentration of dissolved oxygen always above 5 mg/L. This concentration was sufficient for the full nitrification process because it will prevent the competition between autotrophs and heterotrophs bacteria on oxygen use for their growth. The resulted show the low COD/N ratio which ranged from 3.9-5.73.

Optimum hydraulic retention time for domestic wastewater treatment that contains the high concentration of NH3-N was 12 h. At his hydraulic retention time, the MABR have the maximum efficiency removal and the effluent concentration of 88% and 46 mg/L for COD, 89.58% and 7.6 mg/L for NH$_3$-N. At that efficiency, the effluent meets the standard quality of domestic wastewater that permitted in Indonesia for COD and NH$_3$-N of 100 and 10 mg/L, respectively. The longer
hydraulic retention time gives the benefit for autotrophs bacteria. Because of autotrophic bacteria that responsible for oxidized NH₃-N to NO₂⁻ and NO₃⁻ have slower growth rates compared to heterotrophic bacteria. Thus, high NH₃-N removal efficiency achieved at the longer hydraulic retention time.

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REFERENCES


