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

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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 332-468 mg/L (0.56-1.05 kg COD/m^3d). 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 MABRat low COD/N ratio. Furthermore, autotrophs bacteria that responsible for oxidized NH_3-N to NO_2^- and NO_3^- 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 (Idaman, 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.Becauseasthe 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 waste water 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-Nfrom 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 (Mirhossaini *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

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process that causes the competition of oxygen between autotroph and heterotroph bacteria (Lin *et al*., 2016). A pilot scale researched 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; Narakhetudomsak and Tondee, 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_3-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 (polyacrylonitrile) 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

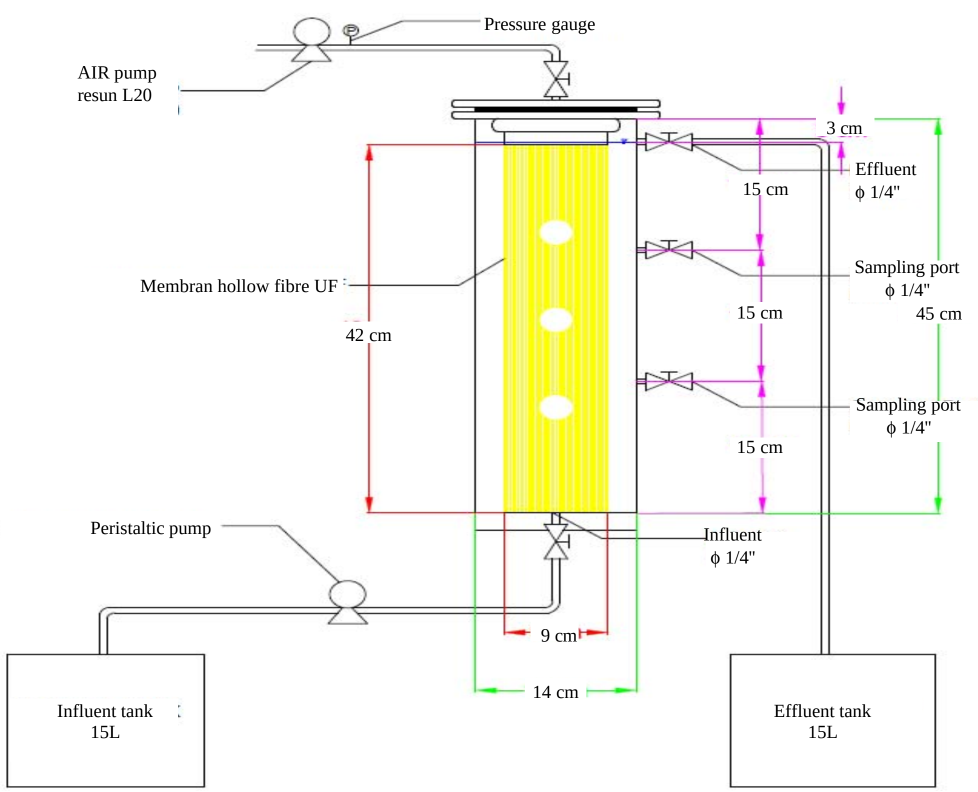

Fig. 1: Scheme of membrane aerated biofilm reactor

available for biofilm attachment was 51 m^2. The reactor was constructed with plexiglass and had working volume 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 test. 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 Resun 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 Jababeka, Bekasi. The seeding process took place with the continuous nsystem because it generated the effective nitrogen removal compared to the batch b system (Sun *et al*., 2009). After the biofilm formed on 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 NH_3-N concentrations from the effluent conducted to evaluate the performance of MABR. COD and NH_3-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, Oxi200) 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 floc 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

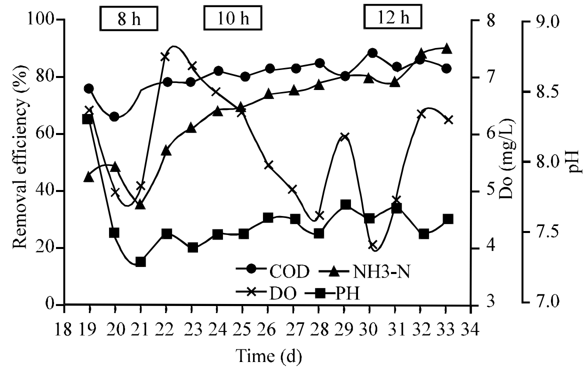

Fig. 2: Consetration of dissolved oxygen and pH condition during the treatment of MABR

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 upto 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 NH_3-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 NH_3-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 NH_3-N to Nitrite and Nitrate (Hong *et al*., 2012). As the high of activity of nitrifying bacteria, the removal efficiency of NH_3-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

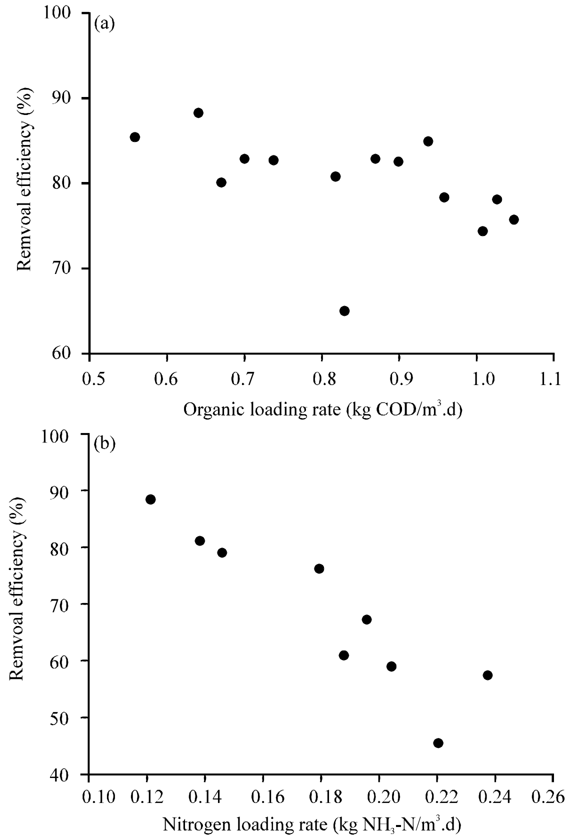

Fig. 3: a) organic loading rate on removal efficiency of COD and b) nitrogen loading rate on removal efficiency of NH_3-N

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/m^3.day (Fig. 3a). For an organic loading rate of 0.63 kgCOD/m^3.day, the removal of COD reached the maximum efficiency and while the organic loading rate of 0.54 kg/m^3.day, the minimum removal efficiency of COD occurred. At the removal of NH_3-N, the maximum efficiency was 89.58% for the lowest nitrogen loading rate of 0.122 kgNH$_3$-N/m^3.day. Whereas, when the nitrogen loading rate increased to 0.24 kgNH$_3$-N, it resulted in the minimum efficiency of 35.66% (Fig 3b). From the result, it can conclude the MABR performance of NH_3-N removal will reach the optimal efficiency when the nitrogen loading rate of domestic wastewater is lower than 0.122 kgNH$_3$-N/m^3.day.

After the treatment of domestic wastewater by using MABR conducted at three different hydraulic retention time it resulted nthat 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

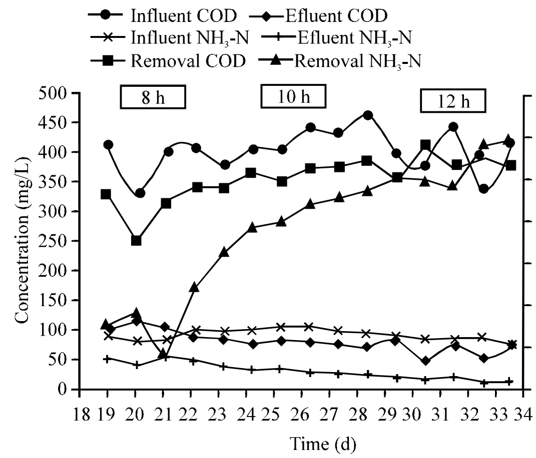

Fig. 4: Influent-effluent concentration and performance of removal efficiency COD and NH3-N with at different hydaraulic retention time

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 NH_3-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 NH_3-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 NH_3-N due to the decreased of influent NH_3-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(Q_2 = 82.77; $\bar{x}$ = 83.78) provided larger value compared to hydraulic time of 10 h (Q_2 = 82.59; $\bar{x}$ = 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

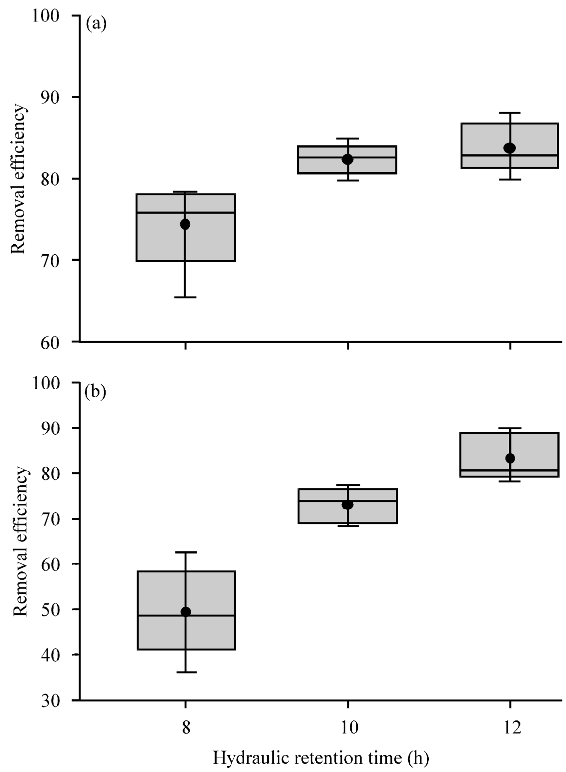

Fig 5: Box plot of MABR performance on a) COD removal efficiency and b) NH_3-N removal efficiency

to COD, the performance removal efficiency of NH_3-N showed (Fig. 5b) that mean and median in hydraulic retention time 12 h ($Q_2 = 80.23$; $\bar{x} = 83.1$) were the biggest value compared to the performance in 8 h and 10 h. According to the resulted, 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 o words remembered at the $p< 0.05$ label for three condition [$F (2, 12) = 9.383$, $p = 0.04$ 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 sugest the longer retention time do have an effect of the removal effiency of COD and NH_3-N. Specifically when the hydraulic reteion time conducted longer, the removal will achieve the higher efficiency. From the resulted 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 concentraion of dissolved oxygen always above 5 mg/L. This concentration was sufficient for the full nitrfication 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 autotrophs bacteria that responsible for oxidized NH_3-N to NO_2^- and NO_3^- have slower growth rates compared to heterotrophs bacteria. Thus, high NH_3-N removal efficiency achieved at the longer hydraulic retention time.

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