SIMULTANEOUS ORGANIC AND NITROGEN REMOVAL USING ANOXIC-AEROBIC MEMBRANE BIOREACTOR

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ABSTRACT

In this paper, anoxic - aerobic membrane bioreactor was constructed and used to investigate the possibility of simultaneous removal of organics and nitrogen. The membrane module was submerged in the aerobic zone of the bioreactor and was operated under an intermittent suction mode. The module was aerated from beneath the microfiltration module to provide the cross flow effect and the oxygen requirement for the biological process. The aeration around the membrane module and the intermittent mode served as the cleaning mechanism for the membrane; this resulted in an operation of the reactor at the flux of 2.7 – 5.4 $L.m^{-2}.h^{-1}$ for 180 days. Hydraulic retention time was ranged between 14.9 to 22.3 hours. MLSS of over 12000 mg/l was reached. The mixed liquor suspended solids (MLSS) internal recycling ratio (between the aerated compartment and anoxic compartment) was maintained at 300% influent flow rate. The average efficiency of the chemical oxygen demand (COD), removal was 99.3% for an average COD of 418 mg/l. 99.3% of influent ammonia was converted to oxidized nitrogen. Permeate inorganic nitrogen reduction was 65.3%.

Keywords: *membrane bioreactor, intermittent suction, organic removal, nitrogen removal, membrane flux.*

INTRODUCTION

Biological treatment is an important aspect of industrial and municipal wastewater treatment. Conventional activated sludge process (CASP) is commonly used worldwide [1]. However, CASP is characterized by various shortcomings such as the formation of the malodorous aerosols that frequently occur in open aerators. For that reason, wastewater treatment plants have been built far away from urban settlement. This causes high costs for maintenance of the sewerage system [2].

Also, large basin volumes are necessary in traditional wastewater treatment plants, since the degradation of the organic wastewater components is only guaranteed by long residence time. The size of basins, however, requires large space, and this gives negative drawbacks from economic as well as ecological point of view [2].

Increasing sludge concentration in the bioreactor to improve the efficiency of CASP is limited since a relatively low sludge concentration (2–4 g/l) is required to achieve good settling effect in the secondary clarifier. This limitation has been an inherent disadvantage of CASP [1].

Moreover, current legislation for wastewater effluent discharge has necessitated enhanced treatment process which capable of removing higher percentage of chemical oxygen demand (COD), nitrogen, phosphorus, suspended solids as well as pathogenic bacteria and viruses. One of the most promising newer technologies is membrane bioreactors (MBRs) [3].

MBRs can be defined as integrating biological degradation system of waste products with membrane filtration [4]. They have been proven quite effective in removing organic and inorganic contaminants as well as biological entities from wastewater. Advantages of the MBR include better control of biological activities, absolute control of the solids and hydraulic retention time, very high quality of the effluent, smaller plant size, and low sludge loading rate [3] and [5].

One of the biggest barriers in the MBR application is the membrane fouling. A series of methods have been put forward to overcome this problem. The most common one is to use cross-flow filtration and not the dead-end filtration [6]. Another type of MBR is submerged membrane system, which was first used by Yamamoto et al., [7]. In that system the membrane was scoured by the intense turbulence of water and the air from aeration in the tank. In addition, intermittent and lower suction pressure would be favourable to counteract the compaction of the cake layer on the surface of the membrane and to reduce the fouling of the membrane [3].

The aim of this study is to investigate the efficiency of the anoxic-aerobic membrane bioreactor on simultaneous organic and nitrogen removal as well as to study the membrane fouling under intermittent and low suction pressure.

MATERIALS AND METHODS

Equipments

A lab-scale immersed MBR was constructed and installed at the Public Health Engineering Laboratory, Department of Civil Engineering, Universiti Putra Malaysia. The schematic flow diagram of the system is shown in Figure 1. The bioreactor consists of two compartments; aerated and non-aerated with working volume of 20.4 litters (15.3 for aerated compartment and 5.1 for non-aerated compartment). A microfiltration membrane module (Table 1) was immersed in the aerated compartment for filtration. The MBR was aerated from beneath the microflitration module through a diffuser to provide the cross flow effect, the oxygen requirement for the biological process, and to mix the mixed liquor in the reactor.

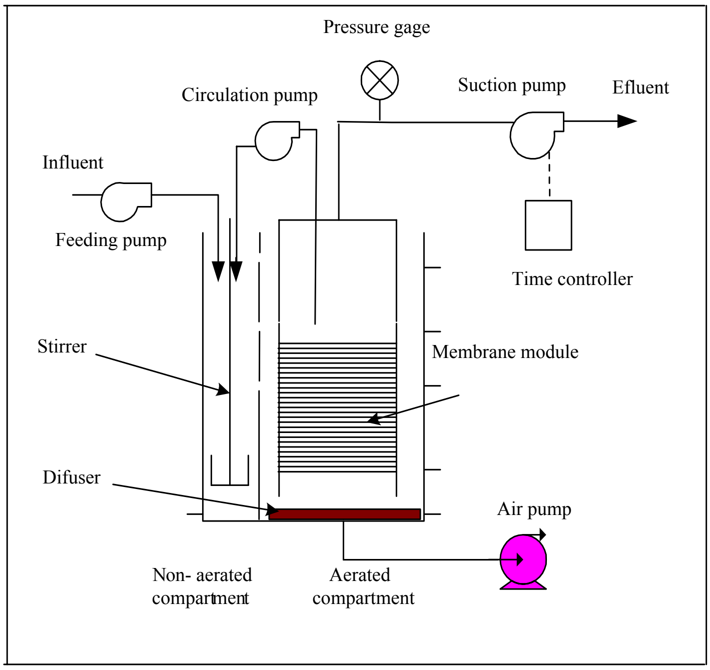

Figure 1: Schematic diagram of the MBR

Table 1: Specifications of membrane

Membrane material	Polyethylene
Outer diameter	540 µ m
Inner diameter	350 µ m
Pore size	0.4 µ m
Surface area	0.2 m2
Manufacture	Mitsubishi Rayon (Japan)

Operating conditions

The operation period of the reactor was about 180 days. Permeate was extracted by a suction pump under intermittent operation in a 10-min cycle; 8-min on and 2-min off. MLSS was measured in interval of 2-4 days samples and the excess sludge was removed from the reactor to maintain the MLSS concentration at around 9000 to 12000 mg.l^{-1}. Hydraulic retention time ranged between 14.9 to 22.3 hours.

Synthetic wastewater

Synthetic wastewater was used in this study instead of actual wastewater to control the inconsistency nature of nutrient concentration in raw wastewater. The composition of wastewater is shown in Table 2. A concentrated solution was prepared and kept in refrigerator at 4°C. The stock solution was diluted with distilled water to a desired COD concentration and a portion of clay suspension was added before being fed to the reactor. In this synthetic wastewater, glucose and glutamic acid were used as carbon source, CH_3COONH_4 and NH_4CL were used as nitrogen source, KH_2PO_4 and K_2HPO_4 were used to provide orthophosphate phosphorus (PO_4^{3-}-P) in the synthetic wastewater. Sodium bicarbonate ($NaHCO_3$) was used for alkalinity to keep pH at around natural [8]. Laguna clay suspension (165 ml/l) was added to increase the suspended solids in the synthetic wastewater.

Table 2: Composition of synthetic wastewater (mg/l)

Composition	Concentration (mg/l)
Glucose	670 – 1080
Glutamic acid	285 – 460
CH_3COONH_4	220 – 350
$NaHCO_3$	750 – 3000
NH_4CL	33 – 53
KH_2PO_4	60
K_2HPO_4	80
$MgSO_4.7H_2O$	33
$FeCL_3.6H_2O$	2
$CaCL_2.2HO$	20
NaCL	25

Preparation of laguna clay suspension

Seventy-five grams of laguna clay was added to 5 liter of distilled water. The suspension was stirred slowly at 50 rpm for 1 hour in a 5-liter beaker for uniform dispersion of clay particles. The suspension was then allowed to stand for 24 hours to allow for complete hydration of the clay. The supernatant was decanted into a plastic container and kept in a room temperature. This suspension was used as a stock solution for the preparation of synthetic wastewater.

Analytical methods

The samples taken from influent, effluent and the bioreactor were analyzed for COD, oxidized nitrogen (NO_2 + NO_3), and MLSS were determined according to the procedure of Standard Methods for the Examination of Water and Wastewater [9]. Dissolved oxygen was measured by using the DO meter (Radiometer analytical, model IONcheck 20).

RESULTS AND DISCUSSIONS

Organic removal

The concentration of COD in the feeding tank varied between 360 mg/l and 480 mg/l with an average value of 418 mg/l as shown in Table 3 whereas the average COD of the effluent was 3 mg/l. Figure 2 shows the COD of influent, effluent and the percentage of COD removal. It is observed that the COD removal efficiency was 99.3 %. This indicates that organic matter can be highly degraded in the anoxic-aerobic MBR. From Figure 2 it can be seen that capacities of COD removal were high from the beginning of the experiment. This indicated that the heterotrophic bacteria which responsible of degrading the carbonaceous components, were enriched in the aerobic part of the reactor because the reactor was fed with a domestic sludge from IWK Kota Damansara sewage treatment plant (Malaysia) together with the synthetic wastewater for two weeks before starting the experiment. During this period the heterotrophic bacteria was acclimatized with the new waste. Ujang et al. [10] have conducted a study using an intermittent aeration in MBR with volumetric loading rate ranges between 0.62 to 0.7 kg COD $m^{-3}.d^{-1}$ and the removal efficiencies from 97 to 98%. Zhang and Verstraete [3] have carried out a study using staged anaerobic and aerobic MBR with the volumetric loading rate of 1.25 kg COD $m^{-3}.d^{-1}$ and the removal efficiency was 97%. In the present study, the volumetric loading rate was between 0.3 to 0.69

kg COD $m^{-3}.d^{-1}$. This shows that the COD removal efficiency in the present study is in agreement with the previous studies been published.

Table 3: Composition of synthetic wastewater (mg/l)

Items	Influent (mg/l)	Effluent (mg/l)	Removal (%)
COD	418	3	99.3
NH_3-N	10.076	0.067	99.3
(NO_2 -N+ NO_3 -N)	0.183	3.935	65.3

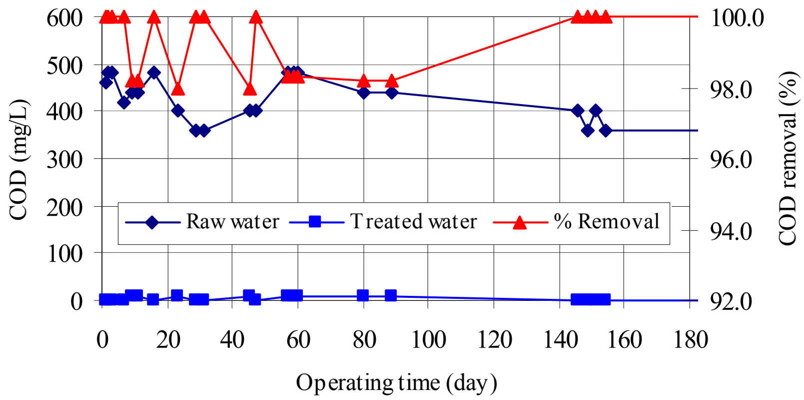

Figure 2: The variation of COD before and after treatment in MBR

Nitrogen removal

The ammonia concentration in this study varied between 6.0 mg/l to 14.3 mg/l and the average value was 10.1 mg/l. An average ammonia concentration in the effluent was 0.067 mg/l as it shown in table 3. Figure 3 displays the removal percentage of ammonia during the operation period. It is noted that the percentage removal was ranged from 98.2 % to 99.9 % with an average value of 99.3 %, which implies that almost all ammonia nitrogen of the influent has been converted to oxidized nitrogen. In the previous study that has been done by Ujang et al. [10], ammonia removal was recorded 99.8 %. This shows that the ammonia removal in the present study is almost similar to that previous study. The high removal of ammonia at the beginning of the experiment indicated that the nitrifying bacteria growth in the reactor was occurred during the two weeks acclimatization period.

The concentration of dissolved oxygen (DO) has a significant effect on the rate of the nitrifier growth and nitrification in biological waste treatment systems. Most often the operating DO for combined carbon oxidation-nitrification systems is 2 mg/l [11]. The DO concentration during the experimental run ranged between 3.9 mg/l to 5.1-mg/l. Thus the DO level for this study is more than the oxygen required for nitrification occurs. However, the excessive aeration is necessary for the scouring process.

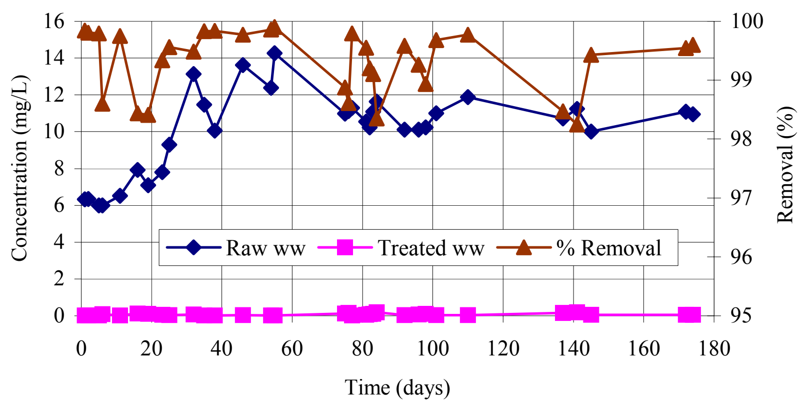

Figure 3: Ammonia variation in MBR

The mixed liquor was recycled from the aeration zone in the MBR to the anoxic zone with a recycling rate of 300 % of the inflow rate so as to achieve the denitrification process. This was done by a peristaltic pump. The biological process of denitrification involves the reduction of nitrogen, NO_3^-, to a gaseous nitrogen species. The gaseous product is primarily nitrogen gas, N_2, but may also be nitrous oxide, N_2O, or nitric oxide, NO. Thus denitrification converts nitrogen to harmless form, which has no significant effect on the environment [11].

Inorganic nitrogen is the sum of NH_3-N, NO_2-N and NO_3-N. In this work the ammonia nitrogen concentration of the effluent was very low (Table 3), therefore the inorganic nitrogen of the effluent was mainly oxidized nitrogen. Figure 4 shows the percentage removal of the inorganic nitrogen. It is observed that the percentage of removal during the first 25 days was in the range of 30 % to 40 % then it increased to 60 % - 73 %. The reason of this might be due to the operation condition, which was the MLSS circulation. The circulation to the anoxic compartment was started at the beginning of the experiment not during the acclimatization period therefore denitrifying bacteria growth was delayed. Thalasso et al. [12] studied the denitrification using methane as the sole carbon source in the presence of oxygen and under strict anoxic conditions. They demonstrated that under the strict anoxic conditions no significant denitrification occurred. However, in the presence of oxygen a successful denitrification was obtained in batch reactors. Houborn et al. [13] demonstrated that a maximum denitrification rate was reached in the batch culture when DO decreased below 1 mg l^{-1}. Ide et al., [14] reported that the activity of nitrifying organisms seems to be enhanced after exposure to oxygen. In this study, the DO in the anoxic zone was varied between 0-to 0.4 mg/l that might play a beneficial effect in the denitrification process.

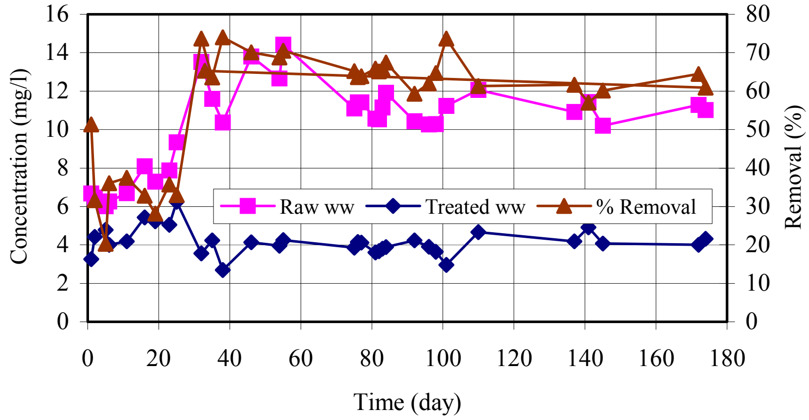

Figure 4: Inorganic nitrogen variation in MBR

Membrane flux

The membrane module was operated in the fixed flux procedure. The initial membrane flux was 5.4 l/m^2.h. The flux maintained at 5.4 l/m^2.h for 40 days and started to decrease gradually until it reached 3.6 l/m^2.h at day 77 (Figure 5) at which the suction pump has been stopped for 40 minutes due to some problem in the pump's tube. The tube has been changed and the suction speed was adjusted to the initial flux without cleaning the membrane. This mechanism of on the aeration and off the suction had partially cleaned the membrane. The flux kept decreasing gradually up to day 130 and after that it kept constant until day 180 at which the membrane was fouled. The aeration around the membrane module and the intermittent suction mode as well as the low suction pressure resulted in preventing the membrane clogging for 180 days. Chiemchaisri et al. [15] reported that the intermittent suction could prevent clogging of membrane to some extent (without any regular cleaning). Seo et al., [16] found that the sheering stress generated by the uplifting flow of bubbling air prevents accumulation of solids on the membrane surface. These studies are in agreement with the present study.

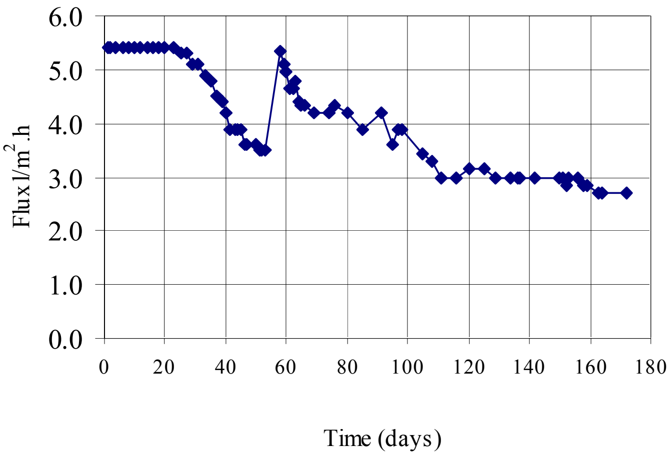

Figure 5: Variation of membrane flux with time

CONCLUSIONS

The anoxic-aerobic membrane bioreactor was successfully used to treat wastewater containing 418 mg/l COD and 10.076 mg/l NH_3-N. The average removal efficiency of COD was 99.3% whereas 99.3% of ammonia nitrogen was converted to oxidized nitrogen. From 30 to 73% of the oxidized nitrogen that converted from the oxidation of ammonia was denitrified. The MBR was operated for 180 days without cleaning of hollow fibres this is due to the intermittent suction mode, uplifting flow of bubbling air and low membrane suction pressure.

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