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Evaluation of simultaneous organic matter and nitrogen removal in a novel anaerobic/anoxic/oxic membrane bioreactor system for treatment of synthetic paper-recycling wastewater

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ABSTRACT

Membrane bioreactor (MBR) offers significant advantages for effluent quality, footprint, sludge production, and operation. In this study, the performance of an MBR was evaluated in a hybrid airlift membrane bioreactor (HAMBR) operating under various hydraulic retention times (HRTs). Results demonstrated that HAMBR could effectively remove organic and nutrient pollutants. The removal percentages of chemical oxygen demand, ammonium and total nitrogen for permeate and supernatant were in the range of 87%–99%, 44%–96%, and 45%–95%, respectively. The results indicated that the HAMBR can be used effectively for simultaneous nitrification-denitrification in the treatment of wastewater, even at low HRTs. Regarding membrane fouling rate, HRT of 36 h could significantly mitigate membrane fouling compared with the shorter HRTs and it was an optimal HRT value for removal of organic matter. Therefore, HAMBR was introduced as a superior MBR regarding performance efficiency and membrane fouling in the treatment of wastewater.

Keywords: HAMBR; Membrane fouling; Hydraulic retention time

1. Introduction

The growth of the human population has caused an increase in the demand for industrial establishments to fulfill human requirements. Therefore, this occurrence has created some problems as the over-exploitation of resources, which leads to pollution of the environment [1]. Integrated pulp and paper plants are considered a water-consuming industry and they are a major origin of water pollution (After metals and chemical industries, this industry is the third water consuming industry) [2–4].

More than 250 chemicals appeared at various stages of pulp and paper process have been detected in their wastewater and discharging pollutants into the aquatic resources can make important environmental problems [1,5]. This type of wastewater includes large amounts of toxic compounds originated from raw materials or produced during the manufacturing and is one of the hardly remediated wastewater [6–8]. Therefore, related to the process of pulp and paper mills, various treatment technologies have been evaluated to minimize the harmful effects of effluent on the environment and the design and proficiency of these technologies will differ from mill to mill [9–11]. However, in general, the primary clarification, secondary treatment, and tertiary processes are the main methods applied in pulp and paper plants [12]. Generally, the conventional treatment approaches

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are not able to reach the requirements of water quality for the papermaking process and the final effluent consists of high value of organics (more than 40%) with low biodegradability and therefore, it needs advanced treatment. Because most investment is for environmental protection and also the supplementary treatment becomes more important in the future, the integrated membrane bioreactor was applied to treat the wastewater of paper mill [13–15]. An MBR integrates an activated sludge (AS) process with a physical separation via membrane. This technique has many benefits compared with conventional treatment, including high efficiency, small footprint, high disinfection ability and also less sludge production [16]. One of the main advantages of an MBR system is the retention of biomass via a membrane, which enables the handle of sludge retention time (SRT) [17].

An HRT of 1.1 ± 0.1 d was an optimal value for chemical oxygen demand (COD) reduction, and also, the cake formation was specified as the dominant mechanism of membrane fouling [18]. The wastewater treatment of pulp and paper plants by MBR technology was evaluated to acquire high-quality water and sustainable reclamation. It was reported that the COD and BOD reduction were 86% and 98%, respectively, and also ammonia and TKN were reduced to 90% [19]. An MBR was evaluated for the treatment of kraft pulp mill foul condensates with an HRT of 19 h. The results showed that the treatment at high temperatures was technically feasible and has acceptable potential for industrial applications [20]. It was shown that the treated effluent by the integrated membrane process could cover the whole standards of process water in a paper mill and can be reused in the pulp and paper manufacturing process perfectly [13]. The application of a thermophilic membrane bioreactor for kraft evaporator condensate treatment was studied and the results indicated that this treatment was practicable in terms of COD removal [21].

Nevertheless, the membrane fouling phenomenon is a major drawback for the widespread application of MBR systems and has been extensively evaluated as a function of operation condition [22,23]. In this phenomenon, the small and soluble particles penetrate inside the membrane and adsorb into the membrane pores along with other organic and inorganic matters. This causes the permeate flux to decrease below the capacity of the membrane filtration, and consequently a large increase in the consumed operational energy takes place [24–26]. Membrane fouling limits the broad application of membranes in wastewater treatment. Serious attempts have been made to obtain a clear insight into the dominant fouling mechanisms, but because of its complication, the handle of fouling has been restricted. It is described that fouling is influenced by different factors including membrane pore size, pollution loading rate, particle distribution, membrane material, and operation conditions. The membranes should be regularly cleaned with physico-chemical methods to ensure that they will have an effective and long operating life [26–29].

Approximately, complete reduction of organic matters and nitrification can be carried out under aerobic conditions in MBRs, while for the denitrification process some modifications are necessary, such as modification of the bioreactor configuration and the addition of an anoxic tank before the aeration process [30,31].

Recently, hybrid MBR systems, including sequential or alternating anoxic-oxic zones, have been developed successfully and were effective in the degradation of organic matter and nutrients [32–34]. In the integrated anoxic-oxic systems, the adjustment of dissolved oxygen (DO) is a very important factor, which determines the prosperity of nutrients and organic pollutants removal. The DO value is completely affected by the growth rate and concentration of biomass in the reactor and the reduction of DO effect on some factors such as the biodiversity and succession of the microbial community [35–37]. An integrated fixed bed membrane bioreactor with an HRT of 36 h was developed to remove pollutants from real paper-recycling wastewater. The removal efficiencies of COD and total nitrogen (TN) for permeate and supernatant were in the range of 92%–99% and 68%–92%, respectively. Also, the membrane fouling was evaluated by transmembrane pressure (TMP) monitoring [38]. In another study, a hybrid airlift membrane bioreactor (HAMBR) system composed of oxic, anoxic and anaerobic zones was developed for simultaneous removal of organic matter and nitrogen from real paper-recycling wastewater. The removal efficiencies of COD and TN for permeate and supernatant were in the range of 88%–99% and 61%–90%, respectively. The results showed that HAMBR can be applied effectively to the simultaneous removal of organics and nutrients from real wastewater [39].

HRT is one of the main parameters in biological wastewater treatment. In the case of MBR operation, it has an obvious effect on the amount of membrane fouling. Generally, a decrease in HRT resulted in the development of severe membrane fouling in the membrane bioreactor, though its effect seems to be mainly indirect than direct [40–43]. Besides, it has been clarified that the prolonged HRT improved the formation of a diverse biocoenosis and subsequently biodegradation efficiency of micro-pollutants was increased [44,45].

This contribution describes the performance evaluation of the HAMBR bioreactor in the removal of pollutants from synthetic paper-recycling wastewater. The specific objective of this study is to investigate the performance of HAMBR for organic matters and nutrients removal. Besides, membrane fouling was evaluated at different hydraulic retention times during the treatment of wastewater, to uncover the mechanisms controlling fouling and pollutants removal and to evaluate the important features of the novel bioreactor.

2. Materials and methods

2.1. HAMBR reactor set-up and experimental process

The configuration and operating conditions of HAMBR are all shown in Fig. 1 and Table 1. The MBR was operated for 100 d at ambient temperature and it had a recirculation pump, which was working with recirculation of around 400% of the inflow. A flat sheet microfiltration membrane (Kubota, Japan) made of chlorinated polyethylene with an area of 0.106 m^2 and a pore size of 0.4 μm was used in the riser. Air was supplied through a diffuser under the flat sheet membrane module. The aeration has three main effects in MBR systems: (i) it supplies oxygen for activated sludge, (ii) it provides the driving force for the circulation of the suspension inside the HAMBR and (iii) the membrane scouring.

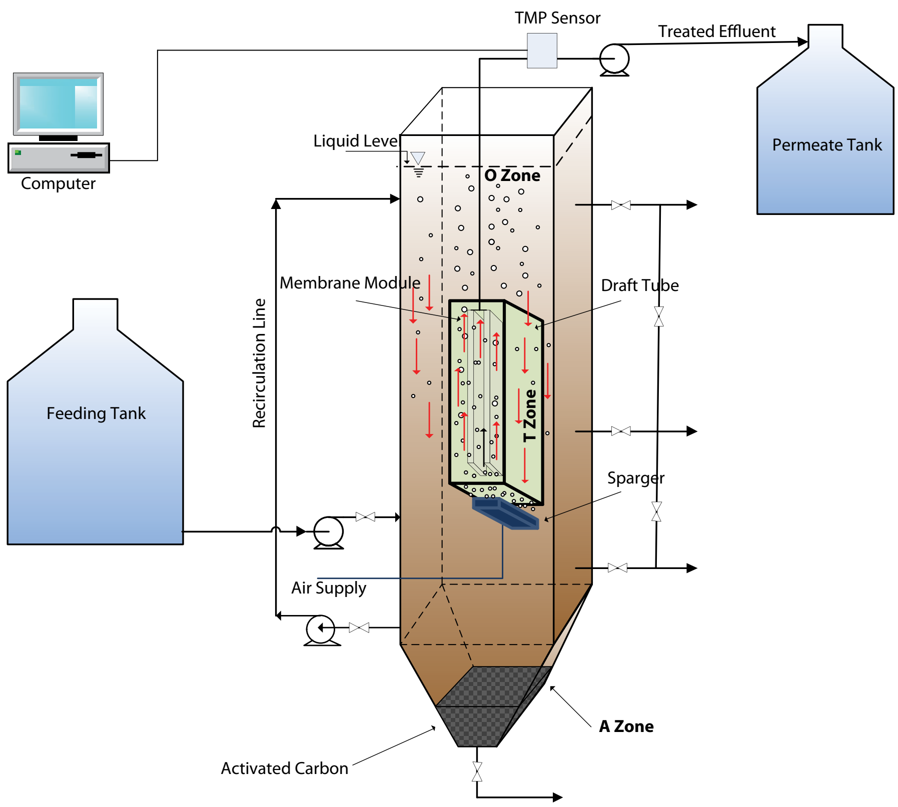

Fig. 1. Schematic diagram of the demonstrative HAMBR pilot plant.

Table 1
Operating conditions of the HAMBR

Phases	Stage I	Stage II	Stage III
HRT (h)	36	24	18
pH	7–7.5	7–7.5	7–7.5
Organic Loading Rate	average	average	average
OLR (kg COD/m³/d)	0.91	1.36	1.82
Temperature (°C)	20–25	20–25	20–25

Table 2
Quality of artificial wastewater

Items	Value
COD (mg/L)	1,365
NH_4–N (mg/L)	93
TN (mg/L)	125
TP (mg/L)	8

The pressure gauge was installed between the membrane module and the effluent discharge pump in order to monitor the variation trend of the TMP [46]. The bottom of HAMBR was filled by granular activated carbon media in order to make biofilm and increasing the removal efficiency of pollutants [47].

The HAMBR system was analyzed through several conventional indexes including mixed liquor suspended solids COD, NH_4^+, NO_2^-, NO_3^- and TN. Artificial wastewater, used as an imitation of paper-recycling wastewater, was composed of glucose, NH_4NO_3 and $(NH_4)_2HPO_4$ tap water (Table 2).

2.2. Analysis

The concentrations of MLSS, COD, NH_4^+, NO_2^-, NO_3^-, TN and total phosphorus were analyzed according to Standard Methods [48]. DO value was determined with a portable digital DO meter (MI 605, Martini). pH value was measured with a pH meter (691 pH Meter, Metrohm, Switzerland). The TMP trend was monitored by a pressure gauge which was located between the membrane module and the peristaltic suction pump.

3. Results and discussion

3.1. DO concentration in the HAMBR

The HAMBR was operated for 100 d and the DO values were obtained from the oxic (O), transition (T) and anaerobic (A) zones. Besides, the MLSS value was determined by sampling from the (O) zone. The results of the MLSS and the DO values are shown in Fig. 2. During HAMBR operation, the biomass concentration increased continuously with the rejection of MLSS by the membrane module. The experiment was started with 2 g/L MLSS and biomass concentration of the HAMBR was increasing because no sludge withdrawn took place.

The MLSS in the HAMBR increased slowly from the 1st to the 20th day, which demonstrated that the microorganisms were in the adaptation stage. From the 21st to the 65th day, the growth rate of MLSS gradually enhanced, which indicated that the microorganisms had acclimated to the condition in the bioreactor. After the 66th day, the growth rate of MLSS gradually decreased and MLSS concentration reached 6.1 g/L after 100 d of HAMBR operation. The trend of MLSS was in line with the findings of Tang et al. [49].

The initial values of DO in the anaerobic/anoxic/oxic zones were about 5 mg/L and changed during the operation of HAMBR, accordingly. During the stage, I, the reduction rate of DO in the anaerobic/anoxic/oxic zones were rather similar by the 24th day and after this point, the values of DO at the (A), (T) and (O) zones gradually exhibited significant differences. These variations were due to biomass growth and other side reactions. After the 25th day, the DO value in the anaerobic zone decreased rapidly and its value reached zero by the 40th day and remained unchanged over operation time. For both (O) and (T) zones, the DO value reduced modestly between 40 and 100 d and consequently, the values of DO in (O) and (T) zones were remained at about 3.7 and 1.1 mg/L, respectively.

3.2. Organic carbon removal

Fig. 3 represents the variations of effluent COD concentrations with operating time. The COD concentration in the permeate flow (COD_p) remained lower than 55 mg/L over the experimental period and fluctuated during the experiments and an average of 97% reduction value was observed.

The COD_p concentration value was mostly lower than COD in the supernatant ((CODs), which is in line with previous observation [50], that proves the helpful effect of dynamic membrane for increasing the COD removal. Nevertheless, the COD_s were low when operating at significantly high HRT. As a result, it proves the benefit of a decrease of organic loading in handling the bioreactor and the COD_s could remain over 185 mg/L when the HRT was 18 h. It is obvious that in the first stage, the performance of HAMBR in terms of COD removal is more effective and by reducing HRT, the efficiency of organic matter removal is reduced. The high COD reduction indicates that the biological population consumes organic matter continuously. This is supported by the growth in MLSS from 2 to 6 g/L within continuous operation [51].

3.3. Nitrogen removal

Fig. 4 shows the ammonium, nitrite, nitrate and TN concentrations over the experimental period for HAMBR. Generally, converting nitrogenous substances to nitrogen gas is obtained through aerobic nitrification-anoxic denitrification (through autotrophic nitrifying and heterotrophic denitrifying bacteria), respectively [52,53]. Nitrite is produced from the oxidation of ammonium and can be converted to nitrogen gas or nitrate by nitrification, which depends on the culture conditions and bacterial species [54].

Fig. 4 indicates that the concentration of ammonia decreases obviously and the maximal NH_4 removal percentage was 96%, which suggesting that the nitrifying bacteria gradually accumulate within the HAMBR. Moreover, the nitrite concentration was maintained at a low value during the whole period which shows most nitrites have been converted and complete nitrification has taken place. According to Figs. 4a and c, reduction of nitrate and TN concentration illustrates that a denitrification condition is formed in the bioreactor.

On the other hand, generally, nitrogenous substances in permeate were lower than supernatant which indicate the effectivity of membrane on nutrients removal. These results suggest that the bioreactor was stable and had excellent nutrients removal efficiencies.

3.4. Membrane performance

The online monitoring of TMP was performed during more than 100 d operations for evaluation of the fouling rate during the operation of HAMBR (Fig. 5). In MBRs, The rate

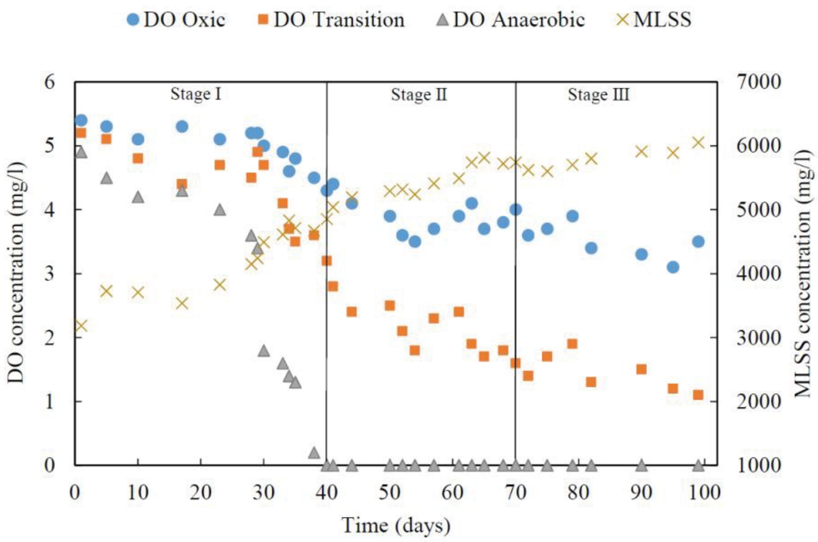

Fig. 2. DO and MLSS concentration vs. time in HAMBR.

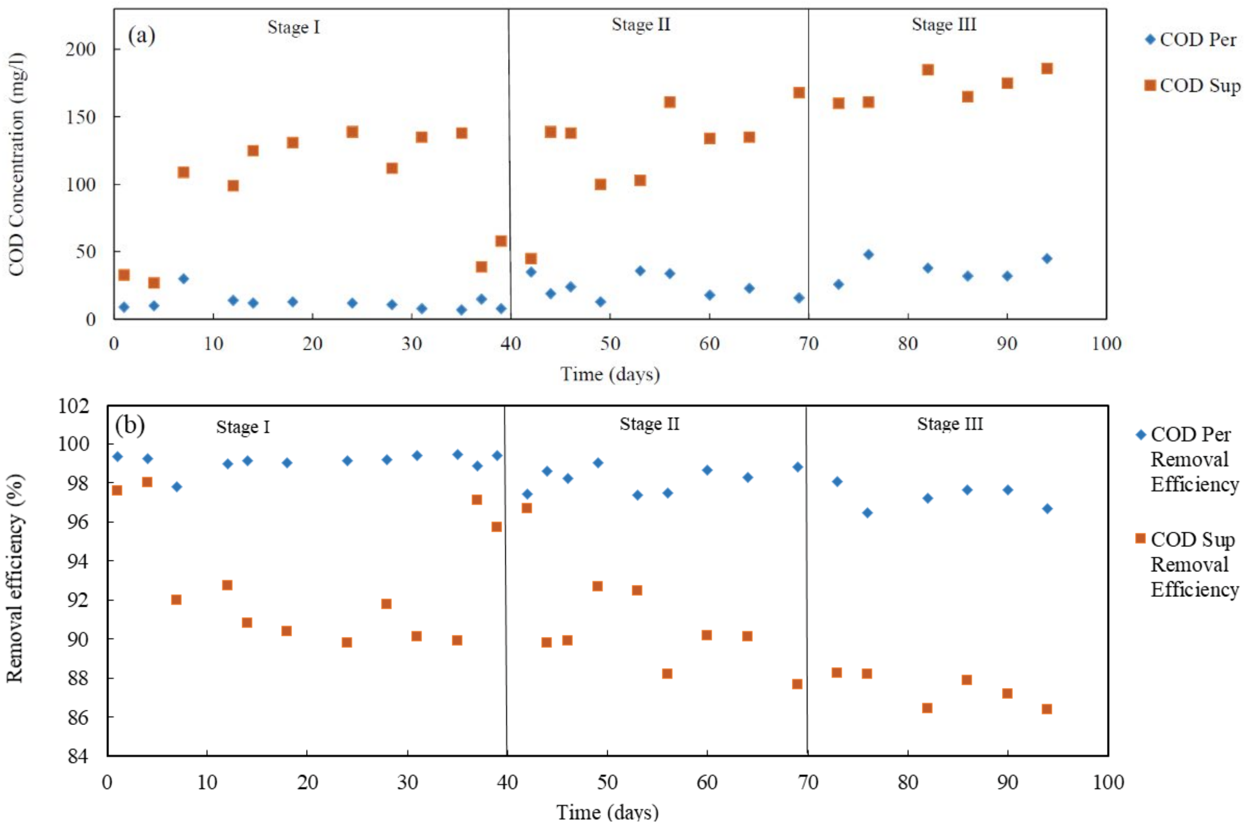

Fig. 3. (a) COD concentration in permeate and supernatant vs. time in the HAMBR, and (b) COD removal efficiency with and without membrane vs. time in the HAMBR.

of change in TMP is one of the important factors to evaluate the system performance [55], since TMP and membrane fouling rate are directly associated with a constant permeate flow rate [56].

In the first stage (HRT of 36 h), a moderate increase in TMP was observed and afterward, at 42 d, the TMP reached 225 mbar. Thus, the membrane was taken from HAMBR. In the next step, it was washed with water to remove the cake layer. This caused a sharp decrease in TMP, but the initial TMP values were not achieved. Then during Stage II, the rate of TMP increasing was slow over operation period, but at 65 d, a sharp increase in TMP was observed and water cleaning was not effective, therefore, chemical cleaning was applied according to manufacturer's instruction after 72 d. During stage III, HRT of 18 h, the results showed an increase in the rate of membrane fouling and the air scouring was not able to clean the membrane module. At 83 d sharply, and after performing water cleaning, the TMP value 96 mbar. This conclusion was similar to previous MBR studies [42,57].

In this study, when HRT was decreased, a sharp rise in the TMP trend was achieved. This trend indicates that the decrease in HRT affects the rate. It was reported that a stepwise decrease in HRT resulted in a decrease in the DO value, which increased the growth of filamentous organisms in the AS. This variation led to an increase in extracellular polymeric substances (EPS) concentration and mixed liquor viscosity, which contributes to the large increase in membrane fouling [42]. These different soluble products and their interaction with the membrane material has a major role in the fouling phenomena [25]. It was reported when HRT value decreased the value of EPS, MLSS concentrations and the average particle size increased, which resulted in the reduction of sludge settle ability and increase in membrane fouling [57,58]. In general, significant membrane fouling takes place when the aeration rate is low and/or the permeate flux is high. At modest flux in the near-critical flux, fouling phenomena due to an increase in permeate flux can be handled by increasing the rate of aeration [59].

4. Conclusions

The proposed HAMBR showed very good performance in the pilot test using synthetic wastewater. Through an HAMBR experiment under the specified operational conditions, it was found that multi-zone conditions form gradually in the single bioreactor and will create a beneficial environment in order to simultaneous nitrification and denitrification for the removal of nutrients and also organics removal. An operational condition such as HRT is an effective parameter on membrane fouling and with decreasing HRT, membrane fouling occurred rapidly and HRT of 36 h was found as an optimal value for COD removal and prevention of fouling phenomena.

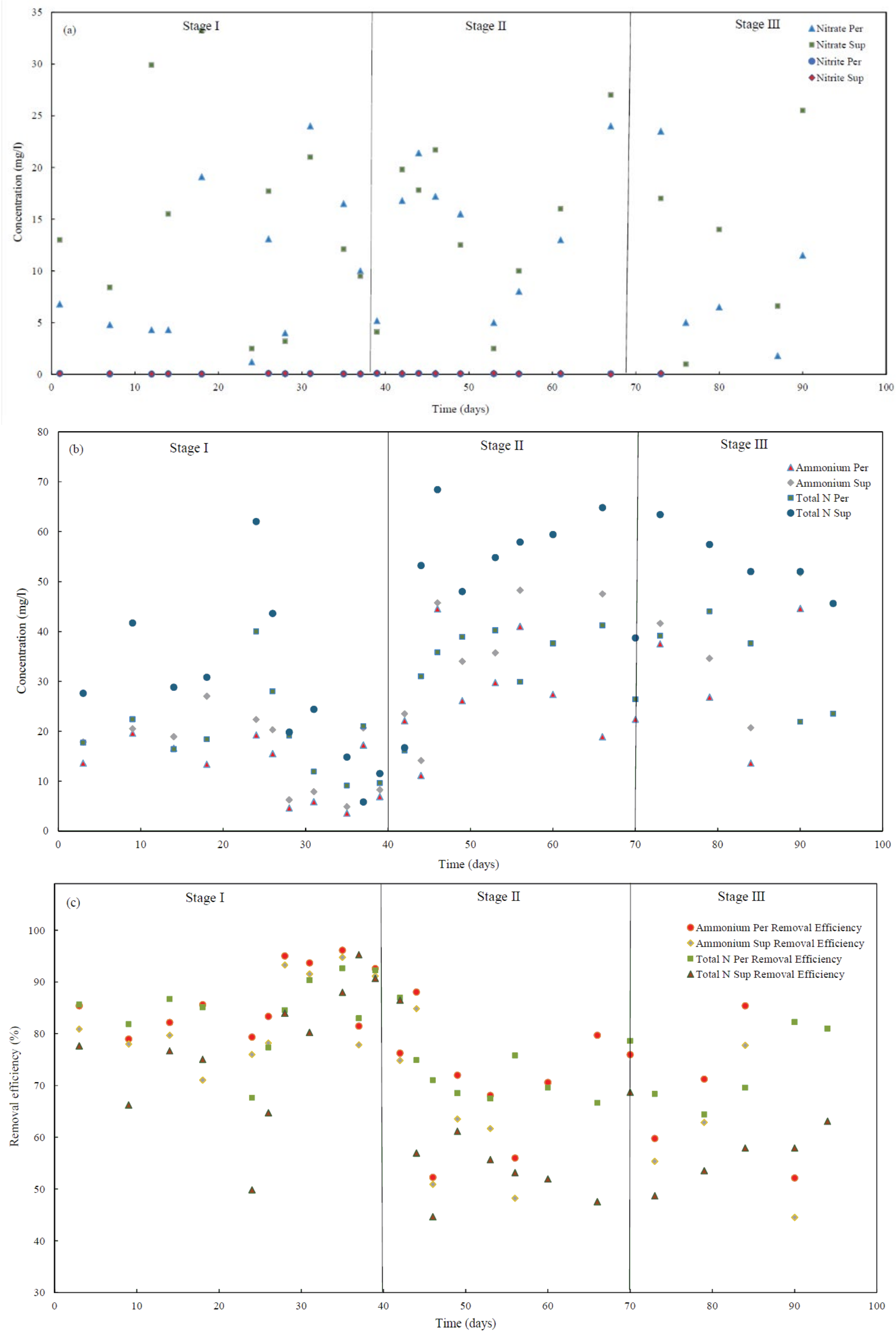

Fig. 4. Performance of the HAMBR: (a) Nitrite and Nitrate, (b) Ammonium and TN concentrations, and (c) Removal of NH_4 and TN.

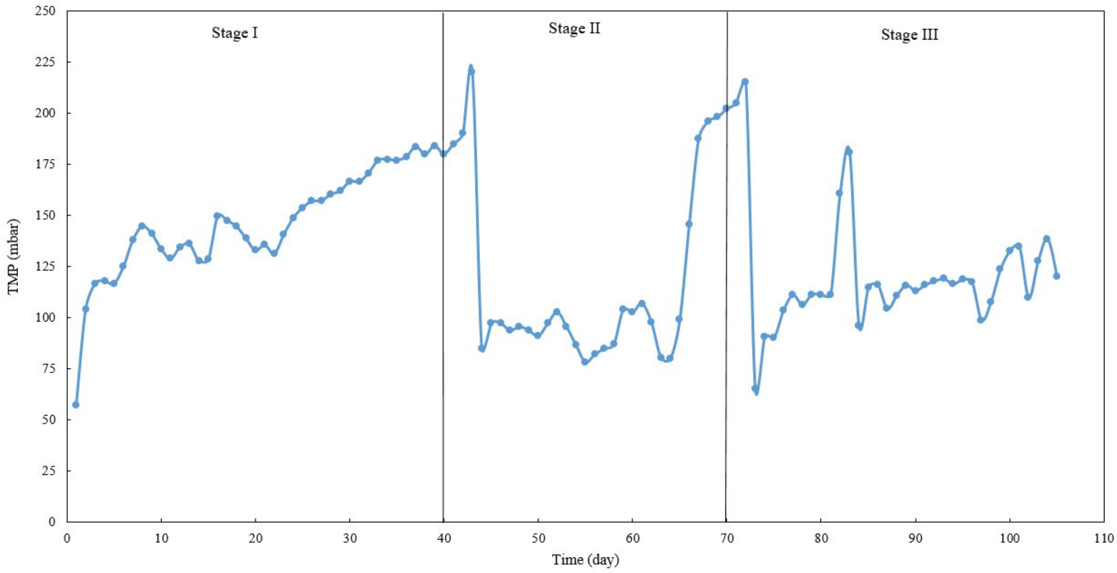

Fig. 5. TMP profile of different operating conditions.

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