An A_2O-MBR system for simultaneous biological nitrogen and phosphorus removal from brewery wastewater at various nitrate recirculation ratios

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Abstract. Anaerobic/Anoxic/Oxic – Membrane BioReactor (A2O-MBR) system including A2O unit at short solids retention time (SRT) for accumulation of PO43--P and MBR at long SRT for nitrification of NH4+-N was used to enhance simultaneous removal of nitrogen and phosphorus from brewery wastewater. The model of A2O-MBR system made from polyacrylic with the capacity of 49.5 liters was operated with organic loading rate of 0.75 kgCOD/m3.day. Nitrate recycling ratio was increased from 100 to 300% while sludge recirculation ratio was maintained at 100%. The results showed that for the nitrate recycling ratios of 100, 200, 300%, average NH4+-N and total nitrogen (TN) removal efficiencies of the model were 95.7 and 72.4, 99.2 and 86.7, 99.3 and 89.6%, respectively. The removal efficiencies of chemical oxygen demand (COD) and total phosphorus (TP) were over 90 and 75%, respectively, regardless of nitrate recirculation ratio. The output values of COD, NH4+-N, TN and TP were within the limits of Vietnam National Technical Regulation on Industrial Wastewater (QCVN 40:2011/BTNMT), column A, throughout the experiments. The model with recommended system configuration and optimum operational conditions could treat not only nitrogen but also phosphorus well due to appropriate nitrate recycling ratios.

1 Introduction

Agriculture is the hub of economic activity in Pakistan. It is the basic for economic development and growth of the economy. It directly contributes to gross domestic product and provide employment to the total labor force of the country. Major part of population depends earning of agriculture in many ways, therefore development of agriculture is development of country.

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By Vietnam Beer Alcohol Beverage Association, the Vietnamese beer market grew by a 5.0 percent year on year in 2017. Under a development strategy by 2020, Vietnam's beer industry would have output of 4.1 billion liters by 2020, 4.6 billion liters by 2025 and 5.5 billion liters by 2035. Along with this growth, serious problems with environmental pollution may be caused by a huge amount of brewery wastewater. This amount of wastewater must be treated before discharge into environment. To brewery wastewater, a combination anaerobic-aerobic treatment system has been used and traditional aerobic biological treatment processes such as activated sludge (suspended growth) or biological filter (attached growth) are often implemented [1, 2, 3, 4]. However, these processes have not yet treated thoroughly nitrogen and phosphorus from brewery wastewater to meet QCVN 40:2011/BTNMT, column A.

A_2O process commonly used in wastewater treatment is able to remove organic matter together with nitrogen and phosphorus with its own inherent advantages such as short hydraulic retention time (HRT), high pollutant removal efficiency and good shock loading capacity [5, 6]. The process consists of three anaerobic, anoxic, oxic compartments and one settling tank which are arranged in sequence with nitrate circulating flow from the oxic compartment to the anoxic compartment and sludge circulating flow from the settling tank to the anaerobic compartment. In this process, nitrification by nitrifiers occurs in the oxic compartment; denitrification by denitrifiers in the anoxic compartment; absorption of β-polyhydroxybutyrate (PHB) for phosphate release by Phosphorus Accumulating Organisms (PAOs) in the anaerobic compartment and then oxidation of PHB for phosphorus accumulation in the oxic compartment. Excess sludge discharge occurs in the settling tank [7]. It is apparent that the higher the nitrate recirculation ratio is, the more the denitrification rate reaches. Nitrogen removal efficiency can be further improved if a higher nitrate recycling ratio is adopted. However, high nitrate recirculation ratios ($\geq$ 400%) should be avoided from an economical point of view [8, 9].

In the past decade, it was reported that A_2O process performance in terms of organic degradation together with simultaneous nitrogen and phosphorus removal could be improved by incorporating a biological reactor into A_2O unit, so-called A_2O – Biological Reactor system [9, 10, 11, 12].

The A_2O unit containing microorganisms with short SRT is employed mainly for removal of organic matter and phosphorus together with denitrification. The biological reactor containing microorganisms with long SRT is employed mainly for nitrification of NH_4^+-N and recirculation of NO_3^--N. What type of biological reactor needs to be considered when choosing for the incorporation.

More recently, MBR is an attractive process that has been increasingly used for advanced biological wastewater treatment. With membrane filtration replacing secondary clarification, MBR possesses a number of merits such as biomass enrichment, perfect nitrification, small footprint, ensured sludge-effluent separation, easy manipulation of HRT and SRT, and excellent effluent quality with little organic and solid contents [13, 14, 15, 16]. Thus, MBR was selected as Biological Reactor in the combined system because of the capacity to achieve enhanced nitrification rate and produce high quality effluent [17, 18].

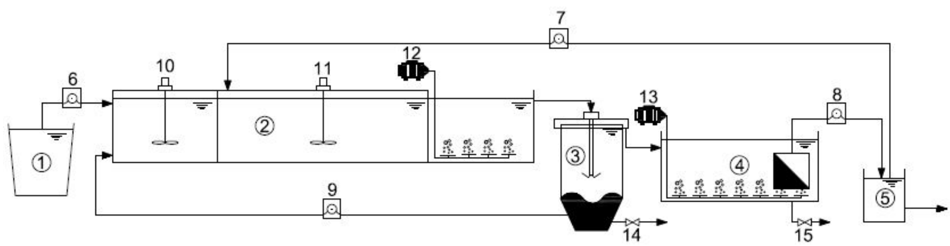
In this study, an A_2O-MBR system was used to evaluate the effects of nitrate recycling ratio on the combined system's simultaneous nitrogen and phosphorus removal performance via continuous flow by treating real brewery wastewater. The role of MBR in the combined system and its contribution to COD, nitrogen and phosphorus removal were also investigated.

2 Materials And Methods

2.1 Experimental system set-up

The polyacrylic model of A_2O-MBR system included Anaerobic/Anoxic/Oxic unit having an approximate dimension of 480 mm L x 150 mm W x 600 mm H with the corresponding working volume of 36.0 liters which was divided by baffles to create three compartments (anaerobic, anoxic and oxic) in ratio of 2:4:2 [12] and MBR having an approximate dimension of 180 mm L x 150 mm W x 600 mm H with the corresponding working volume of 13.5 liters. Total working volume of the model was 49.5 liters. Settling tank had an approximate dimension of 150 mm D x 300 mm H with the working volume of 7.2 liters. In the MBR, a polyethylene hollow-fiber membrane module (0.4 μm pore size, 0.32 m^2 effective area, Mitsubishi Rayon Co., Ltd, Japan) was immersed.

Aeration was provided through fine air diffusers from the bottoms in the oxic compartment and MBR while sludge in the anaerobic and anoxic compartments were suspended by paddle mixers at 50 rpm. Effluent was withdrawn through the membrane module by a suction pump that was designed for intermittent operation with a duty cycle of 8 minutes ON / 2 minutes OFF. To mitigate membrane fouling, backflushing was carried out every 24 hours for 15 min. Dissolved oxygen (DO) concentrations of the oxic compartment and MBR were determined by DO meter and controlled from 2 to 4 mg/L [19]. Schematic representation of the experimental system was represented in Figure 1.



1/Wastewater tank: 200 liters (PE, Vietnam); 2/Anaerobic/Anoxic/Oxic unit with three compartments: 36.0 liters (Polyacrylic, Vietnam); 3/Settling tank: 7.2 liters (Polyacrylic, Vietnam); 4/MBR with a polyethylene hollow-fiber membrane module: 13.5 liters (Polyacrylic, Vietnam); 5/Middle tank: 50 liters (PE, Vietnam); 6/Feed pump: 11 liters/hour (Sandur, India); 7/Effluent pump: 16 liters/hour (Sandur, India); 8/Suction pump: 11 liters/hour (Blue & White, United State); 9/Sludge pump: 11 liters/hour (Sandur, India); 10/Paddle mixer 1: (IWAKI, Japan); 11/Paddle mixer 2: (IWAKI, Japan); 12/Blower 1: 38 liters/min (RESUN, Ap 001, China); 13/Blower 2: 38 liters/min (RESUN, Ap 001, China); 14/Sludge valve 1: ∅13 (Copper, Vietnam); 15/Sludge valve 2: ∅13 (Copper, Vietnam).

Fig. 1. Schematic representation of the experimental system.

2.2 System operating conditions

The wastewater treatment experiment was conducted in four phases in the laboratory at room temperature (~ 25^0C). In the short initial phase, so-called phase 0, seed sludge was given to 50% volume of the model with Mixed Liquor Suspended Solids (MLSS) concentration about 5000 mg/L. Influent wastewater with average COD concentration of 500 mg/L diluted with tap water was pumped into the model. Nitrate recirculation ratio was 100% and sludge recycling ratio was 100% [9]. The phase 0 ended when COD removal efficiency remained stable at 80%. There was no sludge discharged except sampling to keep large amounts of biomass.

In the next three phases according to overall treatment performance in relation to the different nitrate recirculation ratios, denoted as 1, 2 and 3, respetively, nitrate recycling ratio was increased from 100 to 300% while sludge recirculation ratio was maintained at 100%. An average daily wastewater flow to the model was approximately 74.25 liters/day, corresponding to organic, nitrogen and phosphorus loading rates of 0.75 kgCOD/m^3.day, 0.12 kgTN/m^3.day, and 0.021 kgTP/m^3.day, respectively as in Table 1. Excess sludge was discharged from the A_2O unit and MBR to provide SRTs from 5 to 7 days and from 45 to 60 days, respectively.

Trans-membrane pressure (TMP) was used as an indicator of membrane fouling and monitored continuously by a data logging manometer. When TMP reached 40 kPa, membrane washing was performed physically and chemically following the guidelines of the manufacturer. In the phases 0, 1, 2 and 3, the membrane module was physically washed on a daily basis for 15 min. During the entire period of experiment, the TMP was maintained below 40 kPa. Therefore, the membrane module was not cleaned chemically.

Table 1. The experimental condition in different phases for the model of A_2O-MBR system.

Phase	Duration (day)	COD (mg/L)	NH_4^+-N (mg/L)	TN (mg/L)	TP (mg/L)	Nitrate recirculation ratio (%)	HRT (h)
1	1 - 45	513 ± 47	69 ± 9	78 ± 10	13 ± 3	100	18
2	46 – 90	505 ± 43	70 ± 10	82 ± 12	15 ± 3	200	18
3	91 - 135	512 ± 46	68 ± 10	77 ± 11	14 ± 3	300	18

2.3 Wastewater source

Brewery wastewater came from the outlet of the Upflow Anaerobic Sludge Blanket (UASB) reactor of Wastewater Treatment Plant at Nguyen Chi Thanh – Saigon Beer Manufacturing Factory, Ho Chi Minh City, Vietnam. The main characteristics of influent wastewater were presented in Table 1. Seed sludge for the model of A_2O-MBR system was taken from one of the two Sequencing Batch Reactors (SBRs) of this wastewater treatment plant. Seed sludge was light brown, well-settled with Slude Volume Index (SVI) of 98 and Mixed Liquor Volatile Suspended Solids/Mixed Liquor Suspended Solids (MLVSS/MLSS) ratio of 0.74.

2.4 Analytical methods

The samples were collected at the input and output positions of the experimental system. They were also collected in three compartments of the A_2O unit. For each nitrate recycling ratio, the model was operated for 45 days to achieve a steady-state condition and the samples were collected over a 3-day period during these days. For determination of the overall treatment performance in terms of organic and nutrient removals, the parameters of wastewater such as COD, Suspended Solid (SS), Total Kjeldahl Nitrogen (TKN), NH_4^+-N, NO_2^--N, NO_3^--N and TP were analyzed according to Vietnam National Standards (QCVN) together with Standard Methods for the Examination of Water and Wastewater (APHA) [20] at Research Institute for Aquaculture No.2 in Ho Chi Minh City. The value for TN was based on the sum of TKN, NO_2^--N and NO_3^--N. pH and DO were measured by pH (Mettler Toledo MP220, Switzerland) and DO (YSI 5000, United States) meters, respectively. The results below were based on average value and standard deviation by using Microsoft Office Excel software.

3 Results and Discussions

3.1 Organic removal efficiency

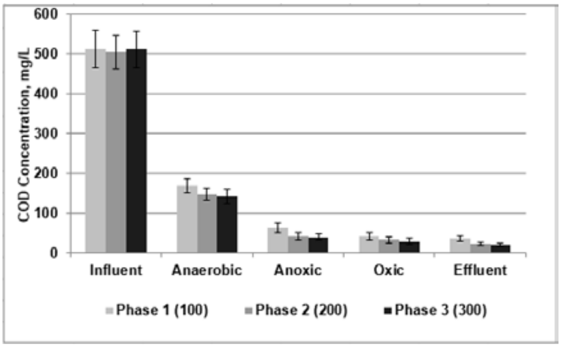


Fig. 2. Change of COD concentration at various nitrate recirculation ratios.

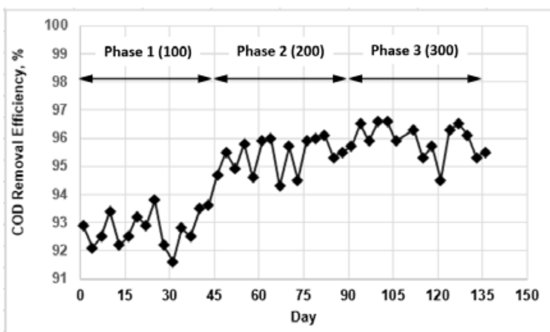


Fig. 3. COD removal efficiencies at various nitrate recirculation ratios.

Figure 2 showed COD concentrations at different positions of the model and Figure 3 indicated variation of COD removal efficiencies during the whole period of operation. It could be seen that COD concentration decreased significantly in the anaerobic and anoxic compartments. The decline could be attributed mainly by the dilution and uptake. The major part of influent COD was utilized in the anaerobic compartment by PAOs and in the anoxic compartment by denitrifiers [11, 21]. It changed slightly in the oxic compartment and the MBR. The additional organic removal was attributable to the step of membrane filtration which is beneficial to keep a higher COD removal efficiency [22, 23]. Accumulation of PO_4^{3-}-P by PAOs happened mostly in the oxic compartment. Nitrification of NH_4^+-N by nitrifiers happened mostly in the MBR. Before wastewater flowed into the MBR, large amount of COD in wastewater was removed. It was condidered to be advantageous for the nitrification because of non-inhibitory effects. Therefore, the growth of nitrifiers was favourable and the nitrification was enhanced as well. When the nitrate recycling ratios varied from 100, 200 to 300%, the effluent COD concentrations decreased from 37, 23 to 21 mg/L, which were much lower than the limit of QCVN 40:2011/BTNMT, column A and the corresponding removal efficiencies of COD were 92.8, 95.4 and 95.9%, respectively. A higher nitrate recirculation ratio will result in a higher NO_3^--N load in the anoxic compartment. Therefore, along with the increasing of nitrate recycling ratio, a slightly high percentage of COD removal in the anoxic compartment was due to denitrification COD uptake and aerobic oxidization as a result of DO recirculation [9, 24].

3.2 Nitrogen removal efficiency

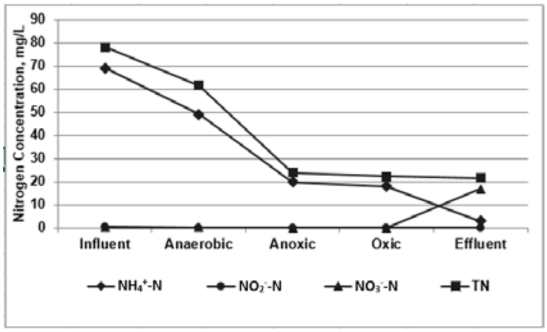


Fig. 4. Conversion of nitrogen concentration for a nitrate recirculation ratio of 100%.

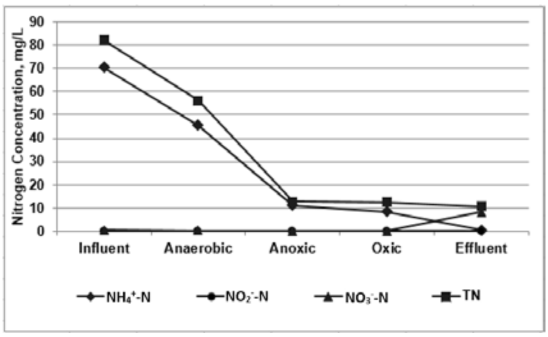


Fig. 5. Conversion of nitrogen concentration for a nitrate recirculation ratio of 200%.

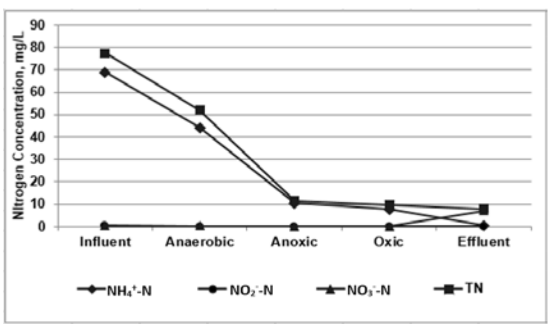


Fig. 6. Conversion of nitrogen concentration for a nitrate recirculation ratio of 300%.

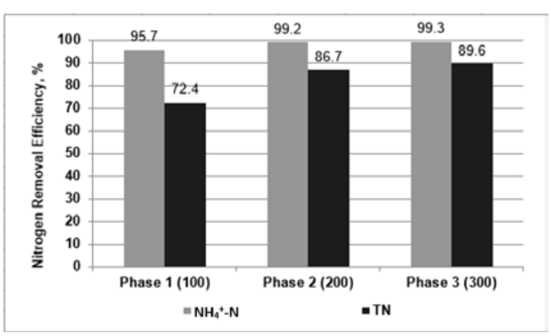


Fig. 7. Nitrogen removal efficiencies at various nitrate recirculation ratios.

The effects of three various nitrate recycling ratios (100, 200 and 300%) on nitrogen removal of the experimental system were revealed in Figures 4, 5, 6 and 7. NH_4^+-N and TN concentrations decreased significantly in the anaerobic and anoxic compartments. The decline could be attributed mainly by the dilution of the sludge circulating flow (ratio of 100%) in the anaerobic compartment and denitrification by denitrifiers in the anoxic compartment. It also showed that TN at the oxic compartment and MBR was mostly NH_4^+-N and NO_3^--N, respectively. Nitrification hardly occured in the MBR and a large amount of NH_4^+-N was completely transformed.

Due to membrane separation, a sufficiently long SRT necessary to prevent the washout of nitrifiers was applied in the MBR to improve the nitrification capability of activated sludge [13]. Very low NO_3^--N concentration in the anoxic compartment indicated that denitrification happened as much as possible [9].

The MBR and anoxic compartment played their roles very well to remove nitrogen. Moreover, a small amount of NH_4^+-N was metabolized for the growth of microorganisms in the model. For the nitrate recirculation ratios of 100, 200, 300%, average NH_4^+-N and TN removal efficiencies of the model were 95.7 and 72.4, 99.2 and 86.7, 99.3 and 89.6%, respectively, and the output values of NH_4^+-N and TN were within the limits of QCVN 40:2011/BTNMT, column A. It was fully reasonable with the change of COD stated above. Together with organic removal, nitrogen removal exhibited an incremental trend with the increase of nitrate recycling ratio. A proper denitrification could be obtained in the experimental system with a nitrate recirculation ratio of 200% based on the economic cost of nitrate recycling directly related to its flow rate.

3.3 Phosphorus removal efficiency

Figure 8 depicted phosphorus concentrations at different positions of the model and Figure 9 represented variation of phosphorus removal efficiencies for various nitrate recirculation ratios. TP concentration increased to the maximum level in the anaerobic compartment when PAOs released phosphate by utilizing COD in wastewater as mentioned above. TP concentration in the anaerobic compartment was much higher than that in the oxic compartment. This implied that the PAOs community was well developed in the A2O unit. Conditions that favor the growth of PAOs and anaerobic phosphorus release could be provided. TP concentration decreased in the anoxic compartment by the dilution of the nitrate circulating flow (ratio ranged from 100 to 300%).

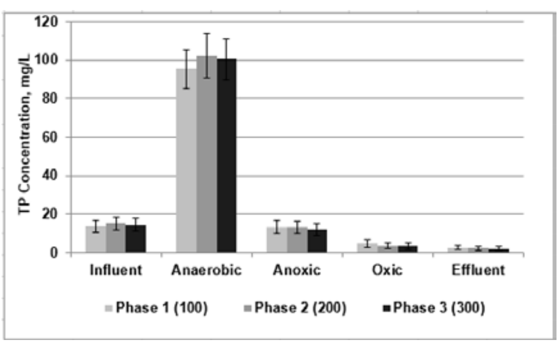


Fig. 8. Conversion of TP concentration at various nitrate recirculation ratios.

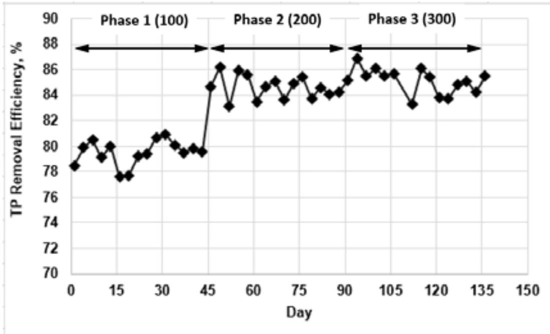


Fig. 9. TP removal efficiencies at various nitrate recirculation ratios.

In addition, TP concentration also decreased significantly in the anoxic compartment due to its uptake by Denitrifying Phosphorus Accumulating Organisms (DPAOs), which could use nitrate and/or nitrite rather than oxygen as an electron accepter when exposed to an anoxic environment. In the oxic compartment, TP was further accumulated by PAOs to reach complete biological phosphorus removal.

[25] also showed that DPAOs played an important role in removing almost entirely phosphorus from wastewater when treating domestic wastewater by an A_2O-BAF system [9]. For the nitrate recirculation ratios of 100, 200, 300%,, average TP removal efficiencies of the model were 79.4, 84.6 and 85.1%, respectively. Phosphorus removal efficiency also reached the highest values at the nitrate recirculation ratio of 300%. For all three nitrate recycling ratios, output values of TP were within the limits of QCVN 40:2011/BTNMT, column A. In relation to the results obtained above, the more COD removal or cell growth is, the more phosphorus removal is.

3.4 Membrane fouling

Membrane fouling in MBR were inevitable. The TMP in the MBR of the model was monitored continuously to evaluate the membrane fouling during the entire running period. The TMP was in the range of 10 – 33 kPa and the flux was 9.6 L/m2.h (LMH). The membrane fouling rate in the MBR correlates well with the MLSS concentration [25]. Figures 10 and 11 show the variations of TMP and MLSS concentration during 135 days of operation. The MLSS concentrations ranged from 5600 to 6450 mg/L. In the phase 1 with the nitrate recirculation ratio of 100%, the TMP was in the range of 10 – 16 kPa for 45 days. During the phase 2 with the nitrate recycling ratio of 200%, the TMP increased gradually with time to 26 kPa on day 90. In the phase 3 with the nitrate recirculation ratio of 300%, the TMP increased almost linearly and reached about 33 kPa on day 135.

As mentioned above, the membrane fouling could be alleviated to a certain degree by the intermittent operation of the membrane (2 min rest in every 10 min operation), air bubbling and backflushing.

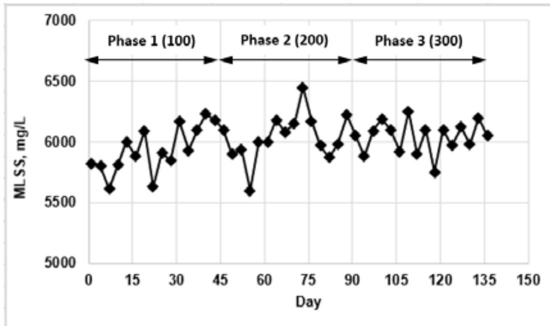


Fig. 10. Variation of MLSS concentration during the operational period.

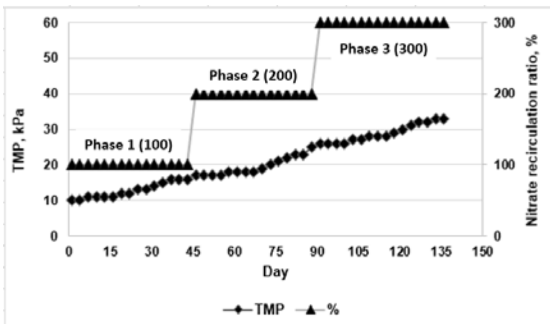


Fig. 11. Variation of TMP during the operational period.

4 Conclusion

In this study, the model of A_2O-MBR system was capable of achieving effluents with low nitrogen and phosphorus concentrations from brewery wastewater at all three nitrate recycling ratios. NH_4^+-N and TN removal efficiencies exhibited an incremental trend with the increase of nitrate recirculation ratio. COD and TP removal efficiencies had a slight increase when nitrate recycling ratio was increased. Treatment efficiencies of NH_4^+-N and TN were over 95% and 70%, respectively, during the whole experiment period. For nitrate recycling ratio of 300%, treatment efficiencies of COD, NH_4^+-N, TN and TP of the model were 95.9, 99.3, 89.6 and 85.1%, respectively. The model with recommended system configuration and optimum operational conditions could treat not only nitrogen but also phosphorus well due to appropriate nitrate recycling ratios.

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