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The effects of operation conditions of carbon/nitrogen ratio and pH on nitrogen removal in intermittently aerated membrane bioreactor (IAMBR)

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ABSTRACT

In this study, the effect of chemical oxygen demand/nitrogen (COD/N) and pH on nitrification-dentrification was investigated by operating a sequencing batch reactor with the use of aeration duration control, coupled with a membrane bioreactor (MBR) system to improve the efficiency of nutrient removal. Among the COD/N ratios of 2, 4 and 6, the removal efficiencies of NH_4^+, NO_3^- and NO_2^- in permeate, were the highest at 92.31, 80.69 and 93.63% respectively, at the COD/N ratio of 6. This ratio is an important factor to be considered for successful nitrogen removal. For pH values of 5.5, 7.6 and 9 there was a great difference in the removal of NH_4^+, NO_3^- and NO_2^- with pH 7.6 were the highest at 92.31, 84.51 and 93.63%, respectively. This implies that a pH above 7.6 should be maintained for a nutrient removal efficiency of approximately 90%.

Keywords: Nitrification; Denitrification; COD/N ratio; Membrane bioreactor; Nitrogen; Wastewater

1. Introduction

The development and application of a membrane bioreactor (MBR) for wastewater treatment is the most important recent technological advances in terms of biological wastewater treatment. Advanced MBR wastewater treatment technology is being successfully applied at an ever-increasing number of locations around the world [1]. In recent years, MBR was proposed as an alternative to conventional activated sludge systems, where the traditional secondary clarifier is replaced by a membrane unit for the separation of the treated water from the mixed liquid without the risk of sludge bulking. Compared with the conventional activated sludge systems, MBR technology has many advantages [2]. Effective nitrogen removal by means of simultaneous nitrification and denitrification (SND) is one of the major specific assets of MBR [3–7]. Because of different environmental conditions of nitrifiers and denitrifiers, total nitrogen removal in wastewater treatment plants is most commonly achieved in a two-stage system or in sequencing batch reactor system where nitrification and denitrification were achieved by temporal separation. However, recent studies have revealed that these two important steps can occur concurrently in the same reactor [8–

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12], and this process is termed SND. The processes always consumed much time and required complex configuration, higher energy, and operation cost. Depending on these considerations, SND has gained significant attentions, which are due to the potential of SND that eliminate the need of separating tanks required by the conventional treatment plants, the simplified design, less space and time of the whole plants [13]. From previous studies [14–21], it was found that there are a lot of factors which influence the nitrification and the denitrification in membrane bioreactor, such as the concentration of sludge flocs, dissolved oxygen, Food/Microorganism ratio, C/N ratio, and pH. The chemical oxygen demand (COD/N) ratio of influent is one of the most critical parameters for wastewater nitrogen removal process, because it directly effects functional microorganism populations, including autotrophic ammonium (NH_4–N) oxidized bacteria, NO_2–N oxidized bacteria and heterotrophic denitrifies. Theoretically, the stoichiometric requirement for denitrification was 2.86 g COD/g N, considering the electron transmitting balance between organic substrate and NO_3–N. However, it has been reported that in a combined nitrification/denitrification process, COD/N requirements in practice was higher than 2.86 g/g [22,23]. Carrera et al. [24] quantified the influence of influent COD/N ratio on a biological nitrogen removal process. They observed that nitrification rate decreased when the influent COD/N ratio increased from 0.71 to 3.4, and the relationship between nitrification rate and COD/N ratio could be defined by an exponential function. Choi et al. [25] have evaluated the performance of an intermittently aerated membrane bioreactor (IAMBR) process across several COD/N ratios. Their results showed that the increase of the COD loading rate led to a higher denitrification rate and better assimilation of organic matter and nutrients. Furthermore, a COD/N ratio over 7 was required for nitrogen removal in the IAMBR system. Other important factors that are to be considered for nitrogen removal are alkalinity, temperature, and pH [26,27]. The purpose of this study is to show the effects of operation conditions of carbon/nitrogen ratio and pH on nitrogen removal in simultaneous nitrification-denitrification process sustained in a lab scale membrane bioreactor.

2. Material and methods

2.1. Characteristics of the influent

The process of The Ain Benian municipal wastewater treatment plant (WWTP) which is located 20 km, Algiers west is constituted of a primary treatment and secondary treatment (conventional activated sludge). The influent is taken from the primary settling. The bioreactor is sown by activated sludge coming from the WWTP of Ain Benian (Algiers) at mixed liquor suspended solids concentrations equal to 3 g/L. Table 1 shows the composition of the influent.

Fig. 1 represents the experimental apparatus used in this study which consists mainly of an aerobic reactor and anoxic reactor intermittently. It is a glass cylindrical biological reactor, with useful volume of 40 L. It is equipped with an agitator in pales. The aeration is realized through an air pump, typifies fine bubbles, at the bottom of the reactor. The membrane bioreactor is consisted of an effective volume of 30 L. An air compressor, type Resun P 1500 was fed to the bioreactor. A tubular mineral membrane is constituted of alumina Al_2O_3 α as support and Al_2O_3 γ as membrane layer with an internal diameter of 15 mm and an external diameter of 53.5 mm.

The operating conditions are listed in Table 2.

2.2. Testing methods

The conventional analysis parameters including pH, dissolved oxygen, COD, NO_2^-, NO_3^-, and NH_4^+ were analyzed according to the standard methods. The concentration of the dissolved oxygen was measured by using the oximeter YSI 550A.

3. Results and discussions

3.1. Influence of the COD/N

The performance of the sequencing aerobic/anoxic/membrane bioreactor was investigated by studying the effect of COD/N ratio as represented in Fig. 2.

Table 1
Mean Characteristics of the influent

Parameter	Values
BOD_5 (mgO_2/L)	176.40
COD (mgO_2/L)	273.80
SS (mg/L)	212.00
NT (mg/L)	50.15
NH_4^+ (mg/L)	24.04
NO_3^- (mg/L)	17.05
NO_2^- (mg/L)	1.74
Conductivity (µs/cm)	1,523.83
Turbidity (NTU)	1.55
pH	7.74

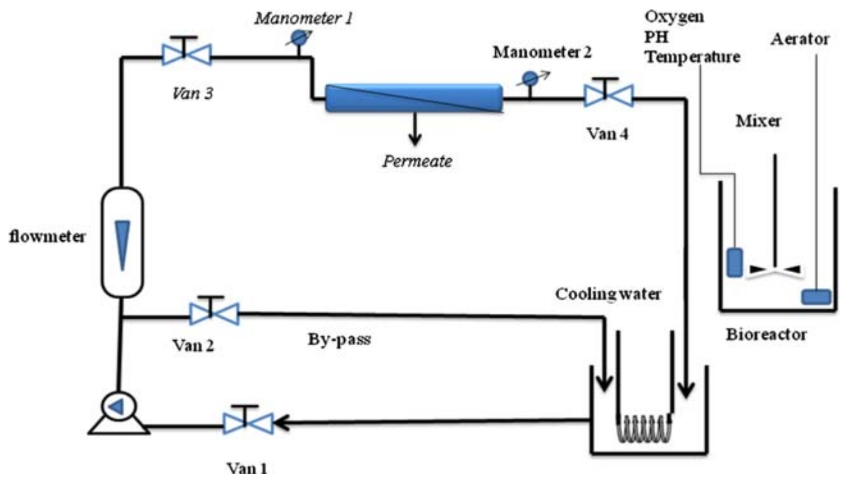

Fig. 1. Experimental set up.

Table 2
Conditions of global functioning

Operation conditions	Values
Bioreactor	
Temperature (℃)	25
pH	5.5, 7.6, 9
COD/TN	2, 4, 6
Dissolved oxygen (mgO_2/L)	2–4
Rotation speed (tr/min)	100
Aeration on/off (min/min)	2/2 h
Feed rate (L/j)	20
Membrane bioreactor	
Temperature (℃)	30
Transmembrane pressure (TMP, bar)	0.8–1
The rate of circulation (m/s)	4
Hydraulic permeability	45

The nitrification rate during the aerobic phase was determined by the disappearance of ammonium (NH_4^+) during this period, according to Eq. (1) [28].

$$r_n = -\frac{dc_{NH_4-N}}{dt} \quad (1)$$

The denitrification rate is calculated with the following Eq. (2).

$$r_d = \frac{dc_{NH_4-N}}{dt} - \frac{dc_{NO_2-N}}{dt} - \frac{dc_{NO_3-N}}{dt} \quad (2)$$

The equation proposed by Katie et al. [29] was used to calculate the efficiency of the SND process.

$$E_{SND}\ (\%) = \left(1 - \frac{NO^-_{X_{remained}}}{NH^-_{4_{oxidized}}}\right)100 \quad (3)$$

During the nitrogen variation in the cycle, the result showed better rate of nitrification (0.12 mg/L h) is obtained between 60 and 90 min in aerobic phase, moreover, the best denitrification rate is reached between 220 and 240 min in anoxic phase with COD/N ratio equal to 2. During the experiment, the best result is obtained at $t = 120$ min for the COD/N ratio of 6 with a nitrification rate of 0.13 mg/L h in aerobic phase and the denitrification rate is 0.096 mg/L h at anoxic phase. The results clearly show that the availability of organic carbon is essential to the activity of heterotrophic denitrifying bacteria to carry out denitrification [29]. The required time for denitrification was longer than the nitrification. The results showed that the denitrification for the COD/N ratios of 2 and 4 attains the best result if the aeration time is too long. (Table 3)

For the COD/N ratios of 2, 4, and 6, the efficiency of the SND is respectively 30.55, 62.44, and 86.34%. When the carbon source is in excess, good efficiency of SND is obtained for the COD/N ratio of 6.

3.2. Removal of the COD, turbidity, and conductivity

Average concentrations of COD in the influent and the effluent and the conductivity with different ratio are summarized in table 4. COD removal efficiency was greater than 93%. It indicates that the elimination of COD is independent of the COD/N ratio. Most of the organic matter (COD) has been eliminated by

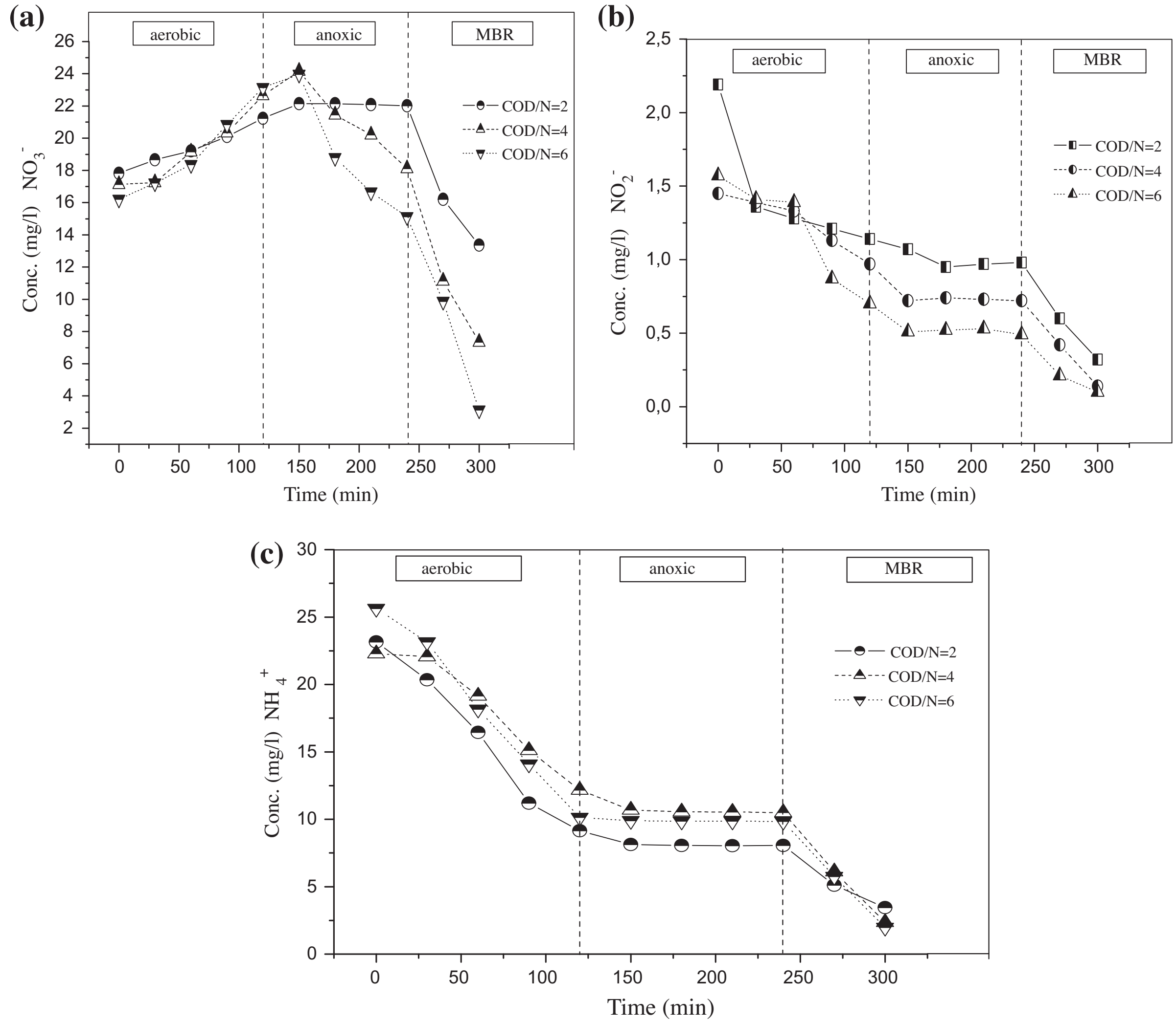

Fig. 2. Evolution of the nitrogen concentration for different values of COD/N: (a) NO_3^-, (b) NO_2^-, and (c) NH_4^+.

Table 3
Rates of nitrification and denitrification with different ratio COD/N

COD/N	Nitrification rate (mg NH_4^+/L h)	Denitrification rate (mg NOx/L h)
2	0.120	0.0029
4	0.098	0.0660
6	0.130	0.0960

biological degradation in the anoxic phase. The results are in agreement with the study of Fu et al. [30]. The turbidity is constant for all COD/N, removal efficiency was 99%. A significant reduction of the conductivity is achieved during the operation. For the COD/N ratio of 6, the removal efficiency was 68%.

3.3. Influence of pH on removal efficiency

Fig. 3 shows the evolution of the nitrogen concentration for different values of pH for COD/N ratio. The results show that the highest rate of nitrification occurred at pH 7.6 with a removal efficiency of NH_4^+, NO_3^-, and NO_2^- being 92.31, 84.51, and 93.63%, respectively. The nitrification process is inhibited when the pH is less than or greater than 7. Anthonisen et al. [31] have studied the effects of pH on the nitrification. Indeed, it favors the formation of free ammonia (NH_3 ammonia undissociated) or nitrous acid (HNO_2), which are inhibitors of the nitrification. The hypothesis that the form of NH_3 is the principal substrate of nitrifying populations would explain this phenomenon [30,32]. Moreover, the inhibition of bacteria appears to be accentuated when the pH increases. In fact, the more the pH increases, the more there is a

Table 4
Water quality of influent and effluent

COD/N	COD (mg L^{-1})		COD removal (%)	Conductivity (μs/cm)		Conductivity removal (%)
	Influent	Effluent		Influent	Effluent	
2	283.70	19.21	93	1,632	469	71
4	196.8	14.33	93	1,423	424	70
6	323.80	9.54	97	1,332	432	68

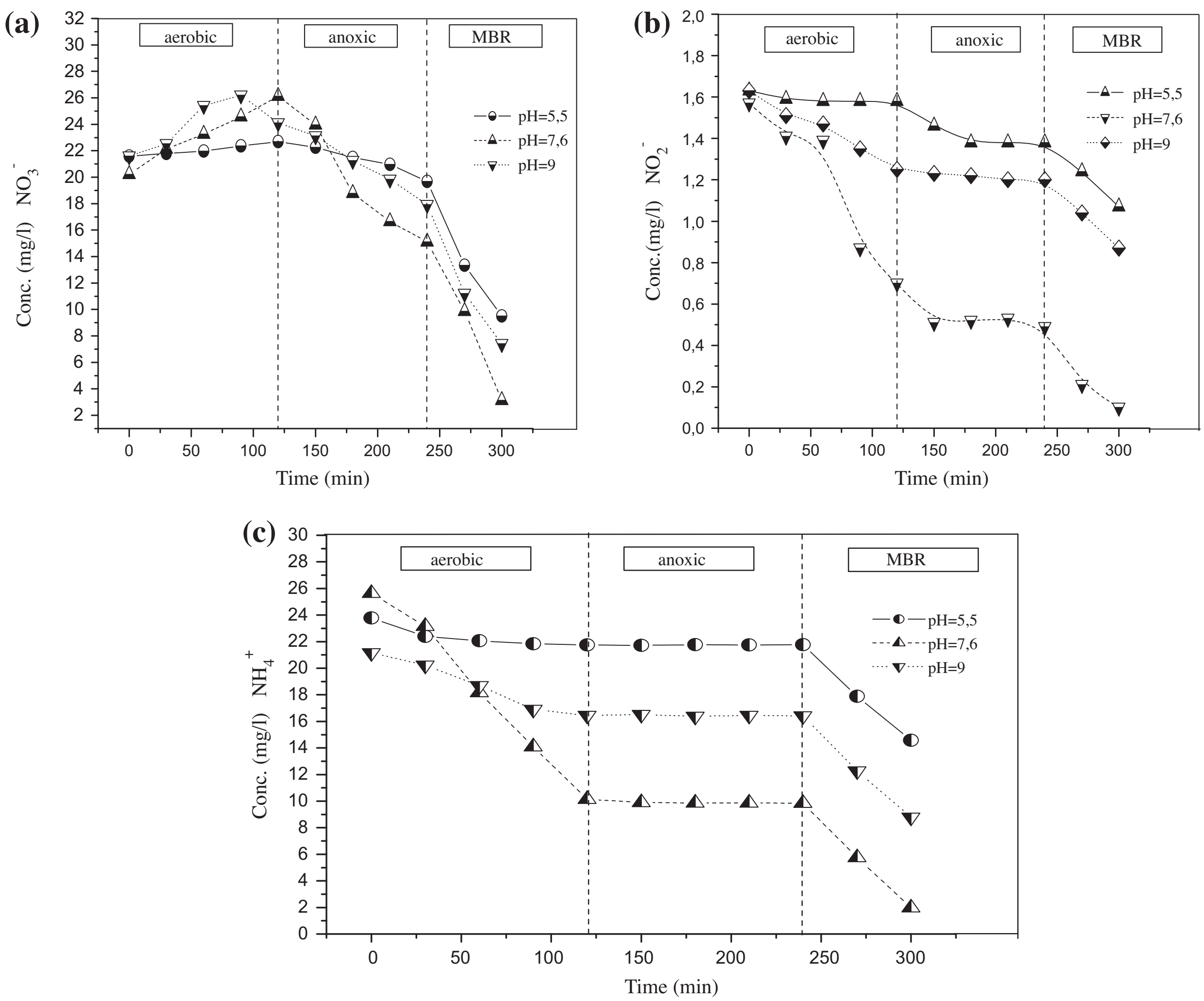

Fig. 3. Evolution of the nitrogen concentration for different values of pH: (a) NO_3^-, (b) NO_2^-, and (c) NH_4^+.

Table 5
COD and conductivity during the experiment

pH	COD (mg L^{-1})		COD removal (%)	Conductivity (μs/cm)		Conductivity removal (%)
	Influent	Effluent		Influent	Effluent	
5.5	265.2	8.00	97	1612.00	501.00	69
7.6	313.8	13.00	96	1754.00	487.00	72
9	265.2	17.00	94	1390.00	345.00	75

rapid change in slope in the profile of concentration of nitrites and nitrates. This means that heterotrophic bacteria had a great buffer power for varying pH. On the basis of the experimental results, we can conclude that the pH is a very important factor affecting the process of nitrification-denitrification in an MBR for efficient removal of nitrogen.

According to the results given in table 5, the elimination of COD was important with the approach of neutral pH.

4. Conclusion

This study enabled the study of the performance of the sequencing aerobic/anoxic/membrane bioreactor process. The intermittent aeration of the reactor allows to reduce the concentrations of NH_4^+, NO_3^-, and NO_2^-, with a cycle of 5 h. Nitrogen removal efficiency has been studied for the different COD/N ratios and different pH values, the best performance was obtained with a COD/N ratio equal to 6 and a pH equal to 7.6.

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