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Efficiency of a hybrid granular bed-contact oxidation biofilm baffled reactor for treating molasses wastewater

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ABSTRACT

A hybrid granular bed-contact oxidation biofilm baffled reactor (GBCOBBR) was studied with molasses wastewater for combined carbon and nitrogen removal. The GBCOBBR was operated at various organic loading rates ranged from 1.5 to 7 kg chemical oxygen demand (COD) $m^{-3}d^{-1}$ and ammonia nitrogen loading rates ranged from 0.05 to 0.204 kg NH_4^--N $m^{-3}d^{-1}$. Carbonaceous matter and nitrogen were removed simultaneously in the GBCOBBR at different recycle to influent ratios ranged from 1.5 to 2.5. The results demonstrated that when the loading rates of COD and ammonia were 3 kg COD $m^{-3}d^{-1}$ and 0.085 kg NH_4^--N $m^{-3}d^{-1}$, COD removal efficiencies of 90.2–91.5% were observed in the anaerobic unit, and over 92% by the two-stage treatment configuration (anaerobic and aerobic units) at all recycle to influent ratios. At all the recycle to influent ratios studied, almost all ammonia was converted to nitrate nitrogen with only small traces of nitrite nitrogen in the nitrification unit and total nitrogen removal efficiencies of 80.5–82% were observed.

Keywords: Anaerobic baffled reactor; Combination filler; Granular bed; Contact oxidation biofilm; Molasses wastewater; Nitrogen removal

1. Introduction

With the development of the economy, the molasses industry has developed quickly and produced much wastewater. Molasses wastewater is one of the main pollutants of natural water [1]. In western China, where there is lack of water resources, molasses wastewater is an important source of pollutants in water because it contains high concentration of organic materials. Molasses wastewater is very difficult to collect and is treated together because most of the molasses processing factories are located in towns and countryside dispersedly. Therefore, the design and manufacture of mobile equipment for the treatment of the molasses wastewater is extremely urgent in this part of China.

In recent decades, anaerobic biological treatment of high strength molasses wastewater has become an established pollution control technology. Integration of denitrification and anaerobic carbon removal in a

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single reactor was reported to be an alternative method to the conventional treatment process of anaerobic carbon removal followed by nitrification-denitrification processes for nitrogen removal [1]. The utilization of the configuration makes the carbon source available in the wastewater as an electron donor for the denitrification process. It will eliminate the operational problems regarding three independent treatment units (i.e. anaerobic system, denitrification and nitrification units) and an additional cost for external substrate for the completion of denitrification. The organic carbon will be removed after denitrification, with the combined system. Ammonia can be nitrified in the following nitrification unit and it not only converts ammonia to nitrate, but also completes carbon removal. Then, the effluent of the nitrification unit can be recycled to the anoxic chamber for denitrification.

Table 1
The characteristics of the molasses wastewater

Characteristics of molasses wastewater	The minimum value	The maximum value
COD (mg/L)	600	3,000
Ammonia (mg/L)	50	85
Total phosphate (mg/L)	6	23
pH	6.7	7.5
Temperature (℃)	35	

Most of the studies on combined denitrification and carbon removal have been conducted with completely mixed reactor configurations [2–4]. However, the studies encountered different problems, such as nitrate inhibition to anaerobic biomass activity, nitrate conversion to ammonium.

In order to reduce nitrate inhibition to anaerobic biomass activity, two-phase anoxic-anaerobic configurations have been studied [5–8]. The first phase was used for acidogenesis and denitrification process. In addition, the second phase was used for high removal efficiency of carbon only. However, the first phase might only obtain partial or incomplete denitrification under the nitrate loading conditions. Thus, nitrogen oxides could find their way into the second phase anaerobic chamber. The baffled system was reported to be the front compartments for nitrate removal [9]. The nitrate removal was complete and 50% of the nitrate was converted to ammonia with molasses wastewater.

A hybrid granular bed-contact oxidation biofilm baffled reactor (GBCOBBR) combines the properties of a granular bed system and contact oxidation biofilm system. The granular bed system was reported to be capable of creating distinct zones of denitrification and carbon removal within the system, especially at high organic loading rates [10,11]. The GBCOBBR may prove to be a more suitable system for the combined removal of carbonaceous and nitrogenous pollution in a single reactor than the traditional anaerobic bioreactors due to the plug flow, phase separation and granular bed characteristics. The objective of the study was to examine the feasibility of a GBCOBBR for both organic carbon and ammonia removal at various recycle to influent ratios with molasses wastewater.

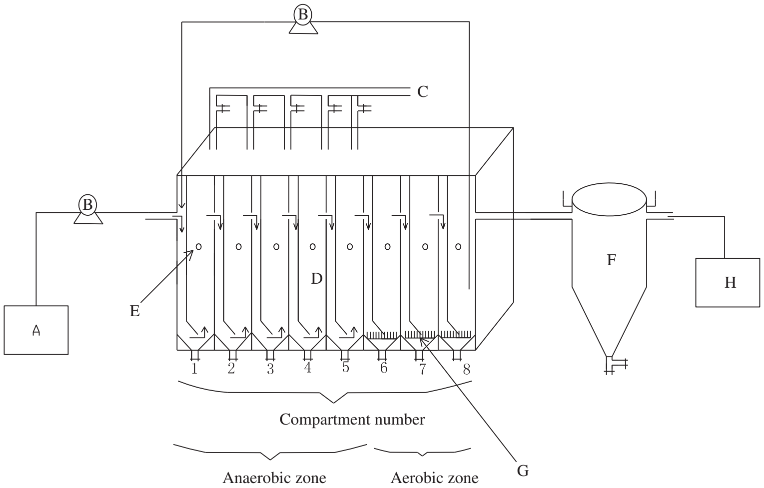

Fig. 1. The experimental layout for GBCOBBR. Note: (A) feed tank (50 L), (B) peristaltic pump, (C) gas outlet, (D) baffled reactor (21.6 L), (E) sampling port, (F) separation tank (5 L), (G) Aeration device, and (H) the water tank.

2. Materials and methods

2.1. Synthetic wastewater

According to the surveys and literatures about the molasses wastewater in China, the characteristics of the wastewater are shown in Table 1. The molasses wastewater was simulated in the laboratory. The organic substance of molasses wastewater is glucose and it is the main source of chemical oxygen demand (COD). The ammonia is NH_4HCO_3 and the phosphorous is KH_2PO_4. $NaHCO_3$ is the buffer of the influent and the pH of the influent is about 7–8.

2.2. The baffled reactor configuration

As shown in Fig. 1, the baffled reactor with a working volume of 21.6 L was consisted of eight discrete compartments of equal volume. The anoxic compartments of the reactor were initially seeded with 7 L of anaerobic sludge containing 32 g/L total suspended solids (TSS) (23 g/L volatile suspended solids (VSS)). The other two chambers were seeded with 4 L of screened aerobic sludge containing 26 g/L TSS (18 g/L VSS). Each compartment was further divided into two parts by slanted edge (45˚) baffles to encourage mixing within each compartment, and down-comer and up-comer regions were created within each compartment. The liquid flow is alternatively upwards and downwards between compartment partitions. It provided effective mixing and contact between the wastewater and biomass at the bottom of each up-comer region [12].

The liquid sampling ports were located in the middle of each compartment. The sludge sampling ports were located at the bottom of each compartment. The influent feed was pumped using peristaltic pump. The outlet of the baffled reactor was connected to a sedimentation basin to trap the solids. The combination filler was installed in the aerobic zone for extending SRT and the nitrifying bacteria growth. The specific surface area of combination filler was about 2,400 m^2/m^3 and its porosity was more than 90%. In addition, its density was about 0.8 g/cm^3.

2.3. Experimental procedure

The experimental program is outlined in Table 2. The experiment contained four events. The No I event was studied for the influence of C/N ratio on the performance of the baffled reactor through changing influent COD and keeping other experimental variables constant during 80 days. The No II event was used to assess the influence of the gas/water ratio (GWR) on nitrogen and COD removal efficiencies when all other experimental variables were constant during 80 days. In the following 60 days, when the concentrations of COD and ammonia were constant and GWR was 20:1, the internal recycling ratio was changed from 150 to 250%. In the last 210 days, the optimal hydraulic retention time (HRT) was obtained through changing HRT and keeping other elements constant.

2.4. Analytical methods

All the chemicals used were of analytical reagent grade and laboratory distilled water was used during the experiments. COD, NH_4^+-N, NO_2^--N, NO_3^--N, TSS, and VSS were measured according to the standard methods, as set out by the American Public Health

Table 2
Timetable for COD and nitrogen removal experiments

Event no.	Event	Days
I	HRT = 24 h, GWR = 20:1, internal recycling ratio = 200% (a) 600 mg/L COD, 85 mg/L ammonia, 23 mg/L; (b) 850 mg/L COD, 85 mg/L ammonia, 23 mg/L; (c) 1100 mg/L COD, 85 mg/L ammonia, 23 mg/L; (d) 1350 mg/L COD, 85 mg/L ammonia, 23 mg/L	80
Ⅱ	600 mg/L COD, 50 mg/L ammonia, 6 mg/L TP HRT = 24 h, internal recycling ratio = 200% GWR = 17:1, 18:1, 19:1, 20:1	80
Ⅲ	600 mg/L COD, 50 mg/L ammonia, 6 mg/L TP HRT = 24 h, GWR = 20:1 Internal recycling ratio = 150%, 200%, 250%	60
Ⅳ	3,000 mg/L COD, 85 mg/L ammonia, 23 mg/L TP GWR = 20:1, internal recycling ratio = 200% HRT = 48 h, 36 h, 24 h,20 h,14 h,12 h,10 h	210

Association (APHA/AWWA/WEF, 2005) [9]. The effluent samples were filtered before analysis. The concentrations of NH_4^+-N and NO_2^--N were measured using a different colorimetric method and the concentration of NO_3^--N was analyzed by using the UV spectrophotometric method. DO and pH were measured with a DO meter (YSI, Model 55, USA) and an acid meter (PHS-25; Shanghai REX Instrument factory, China). Total nitrogen (TN) was measured offline with Multi N/C 3,100 (Jena, Germany). The influent flow and recycling ratio were controlled by the peristaltic pumps.

3. Results and discussions

3.1. Effect of the GWR

Experiment I was used to assess the influence of the GWR on nitrogen and COD removal efficiencies when all other experimental variables were constant. As shown in Fig. 2, experiment I was conducted at various GWRs of 17, 18, 19, and 20 and TN removal efficiencies were 70, 75, 80 and 85%, respectively.

The effect of gas/water ratio on ammonia removal was studied through maintaining a constant aeration flow rate of 1 L/min when the oxygen partial pressure was altered. As shown in Fig. 2(b), a positive correlation of ammonia removal and gas/water ratio demonstrated that ammonia removal efficiency increased with increasing gas/water ratio. It was proposed that the linear Eq. (1) between ammonia removal and the partial pressure of oxygen P_0 (atm) [10] is:

$$NH_{3\,\mathrm{removed(mg/L)}} = 108.76 \times P_0 + 75 \qquad (1)$$

Equilibrium between the gas and aqueous phases was assumed due to oxygen transfer experiments with the same filler, giving DO concentrations of 7.4, 14.8, and 29.6 mg O_2/L at oxygen concentrations of 21, 42, and 84%, respectively [13].

As shown in Fig. 2, the COD removal efficiency was ranged from 75 to 80% at the COD concentration of 600 mg/L. Then, the remaining COD was further removed when the effluent of anoxic zone flowed into the aerobic zone and COD removal efficiencies were ranged from 80 to 88% at various GWR. In addition, when GWR increased from 17 to 20, the COD removal efficiency was increased by 8%. It implied that the residual organic carbon was non-biodegradable. Transformation of organic carbon to inorganic carbon took place in the anoxic zone due to denitrification and fermentation reactions of various heterotrophic organisms [14].

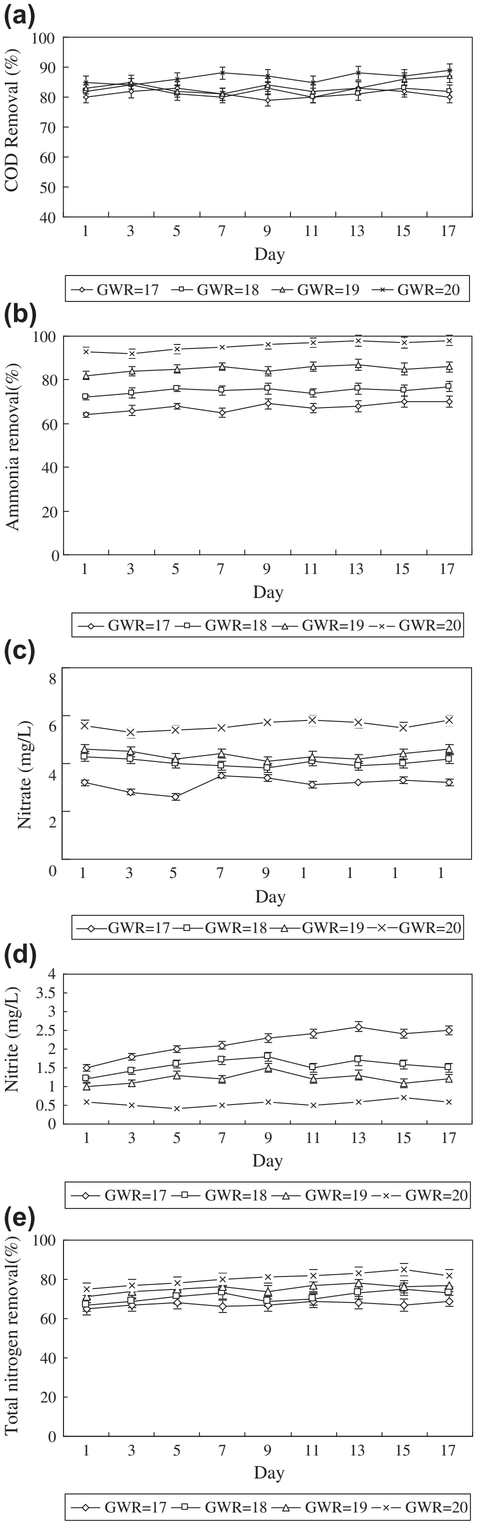

Fig. 2. Variation of removal efficiencies of (a) COD; (b) ammonia; (e) TN and variation of effluent concentrations of (c) nitrate and (d) nitrite at the gas/water ratio of 17, 18, 19 and 20.

The main role of the aerobic zone was to convert the ammonium ion into nitrite and/or nitrate. The average NH_4^+-N concentration of the influent was about 50 mg/L. When the GWR was 17:1, the NH_4^+-N concentration of effluent was about 15 mg/L. As the GWR increased, the NH_4^+-N concentration of effluent gradually decreased. When the GWR was 20:1, the NH_4^+-N concentration of effluent was kept less than 1 mg/L and the NO_2^--N concentration was ranged from 0.5 to 2.5 mg/L. In addition, the NO_3^--N concentration was less than 3.0 mg/L.

Denitrification was promoted by recycling the nitrite/nitrate formed via nitrification back to the anoxic zone. The average TN concentration remained below 10 mg/L and the average TN removal efficiency was 85% at the GWR = 20:1 (note that regulations stipulate the TN concentration less than 30 mg/L for discharging into surface water in China). In addition, the TN concentration of effluent was less than 10 mg/L, and it indicated that the GBCOBBR was functionally stable throughout the study.

3.2. *Effect of the internal recycling ratio*

Experiment II was carried out to assess the influence of the internal recycling ratio on nitrogen and COD removal efficiencies as all other experimental variables were constant. The effects of three various internal recycling ratios (R = 150%, R = 200%, R = 250%) on COD and nitrogen removal of the GBCOBBR were shown in Fig. 5.

The TN of the influent was only composed of NH_4^+-N and NH_4^+-N concentration was about 50 mg/L in the experiment II. The NH_4^+-N concentration decreased significantly in the anoxic zone due to the dilution of nitrate recycling stream as the internal recycling ratio increased from 150 to 250%. The NH_4^+-N concentration had a significant reduction in the aerobic zone and the average NH_4^+-N concentration of effluent was only 1.5 mg/L at all three recycling ratios (met the class A discharge standard). In addition, as shown in Table 3, the NO_3^--N concentration of effluent decreased significantly from 6.2 to 5.6 then to 5.3 mg/L. Thus, NO_3^--N was the main component of TN in the effluent. When GWR of the aerobic zone was 20, NO_3^--N was the only pollutant in the effluent.

As shown in Fig. 3, most of the TN was removed in the anoxic zones, and the TN concentrations decreased to 20, 15, and 13 mg/L at the outlet of the anoxic zone at various internal recycling ratios. It was noticed that TN concentration had a slight decrease in the aerobic zone. As shown in Table 3, when nitrate recycling ratios were 150, 200, and 250%, the TN concentrations of the effluent were 15 mg/L (met the class B discharge standards), 10 mg/L (met the class A discharge standards) and 8 mg/L (met the class A discharge standards), respectively.

The removal efficiency of TN was increased by 14% as the internal recycling ratio increased from 150 to 250%. The TN removal efficiency was increased by 10%, when the internal recycling ratio increased from 150 to 200%. The results also showed that the optimum denitrification could be obtained at the internal recycling ratio of 200%.

In addition, although the higher internal recycling ratio was beneficial for TN removal, it could be economically non-profitable in case of an influent with low ammonia load. The economical cost of internal recycling ratio was directly related to its flow rate. Based on our study, both TN removal efficiency and economic potential should be taken into consideration, when a relatively higher internal recycling ratio was applied in a real situation. It was suggested that the application of various internal recycling ratios should depend on the actual conditions in the wastewater treatment plants [15]. Nitrate recycling improved the homogeneous distribution of microbial communities in the reactors, and increased the removal efficiency of TN [16]. As recommended, nitrate recycling ratio should be around 200%, and it could provide a proper compromise between removal efficiency and costs.

Table 3
Pollutant removal efficiencies by A/O system at various ratios

	$R = 150\%$			$R = 200\%$			$R = 250\%$		
Parameter	Inf. (mg/L)	Eff. (mg/L)	Rem. (%)	Inf. (mg/L)	Eff. (mg/L)	Rem. (%)	Inf. (mg/L)	Eff. (mg/L)	Rem. (%)
COD	600	78	87	600	81	86.5	600	69	88.5
NH_4^+-N	50	1.5	97	50	1.25	97.5	50	1	97
NO_2^--N	0	0.2	–	0	0.18	–	0	0.16	–
NO_3^--N	0	6.2	–	0	5.6	–	0	5.3	–
TN	50	15	70	50	10	80	50	8	84

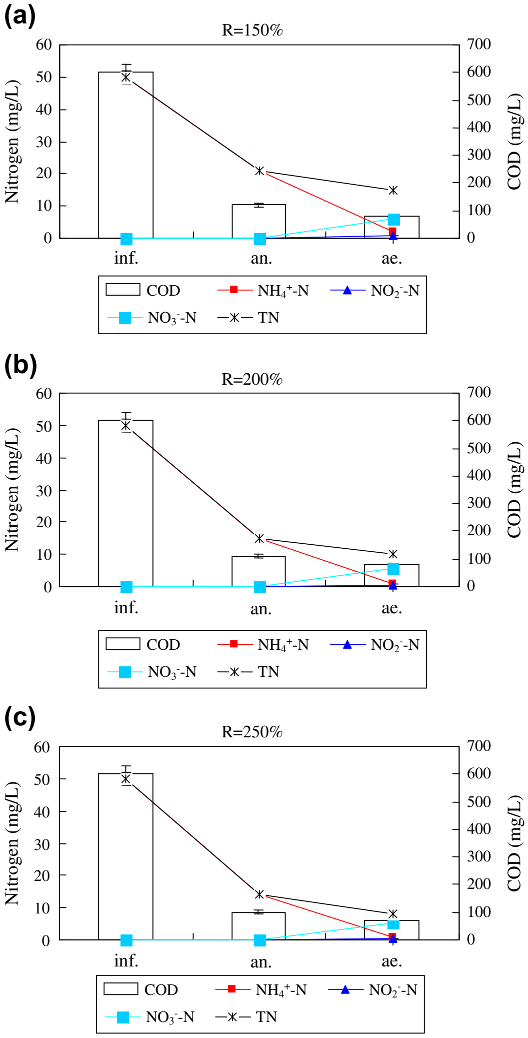

Fig. 3. Evolution of COD and nitrogen in reactor at various internal recycling ratios.

3.3. Effect of the concentration ratio of COD to TN (COD/TN)

Experiment III was conducted at the influent ammonia concentration of 85 mg/L and the HRT of 24 h when the COD/TN ratio was stepwise increased. Fig. 4 shows COD, ammonia, nitrite, nitrate, and TN concentrations of effluent at various COD/TN ratios.

As shown in Fig. 4, the removal efficiencies of COD and TN were not greatly different at four COD/N ratios. It indicated that the COD/N ratio did not make a strong influence on the COD and nitrogen removal.

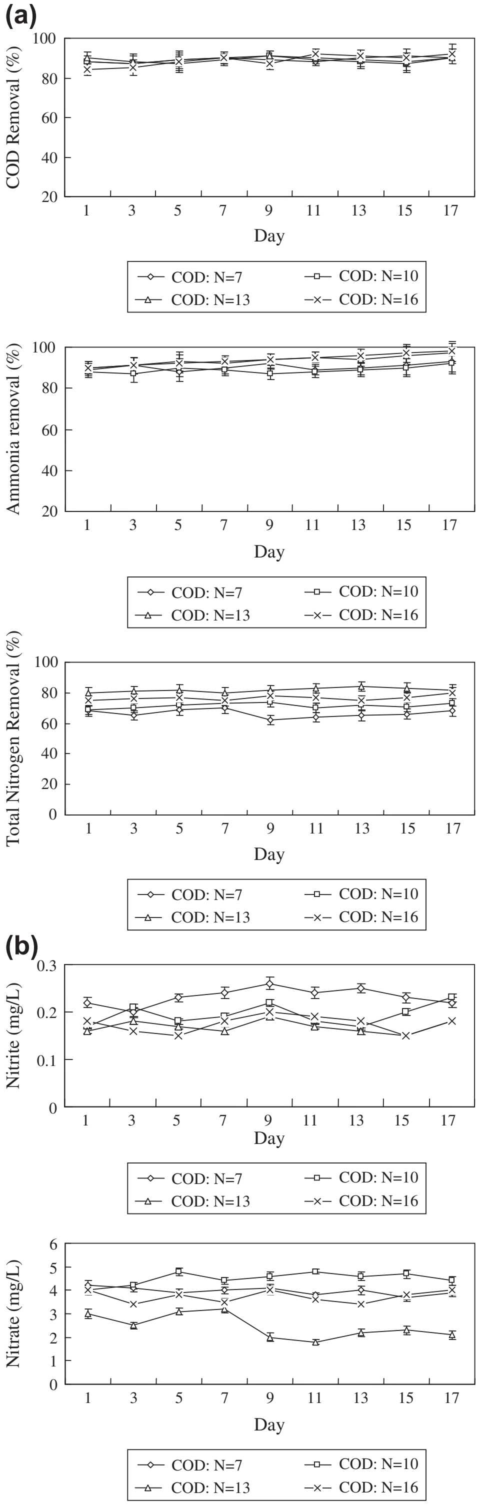

Fig. 4. Variation of removal efficiencies of COD; ammonia; TN (a) and variation of effluent concentrations of nitrate and nitrite (b) at the COD: N ratios of 7, 10, 13 and 16.

As shown in Fig. 4, when the COD/N ratio was 7, NO$_3^-$-N and NO$_2^-$-N accumulations were observed, and the TN removal efficiency was 70%. It demonstrated that the nitrification was more efficient than the denitrification. Although the ammonia and COD removal efficiencies remained about 95 and 90%, TN removal efficiency was various at different COD/N ratios. The TN removal efficiency increased from 70 to 80% as the COD/N ratio increased from 7 to 13. It indicated the high performance of denitrification. High COD/N ratios could accelerate the growth of heterotrophic denitrifying bacteria in the biofilm. Thus, it promoted the total denitrification efficiency [17]. The dependence on the organic carbon source determined the high sensitivity to changes of organics during heterotrophic denitrification. Therefore, the denitrification efficiency increased as the COD/N ratio increased from 7 to 13. However, when the COD/N ratio increased to 13, TN removal efficiency decreased to 75%. It illustrated that the denitrification efficiency decreased at the COD/N ratio of 13.

The theoretical requirement for complete denitrification, ignoring microbial growth, is 2.85 mg COD/mg nitrate-N reduced to N_2. At relatively low COD/N ratio, incomplete denitrification is possible. It could lead to the formation of nitric oxide (NO) and nitrous oxide (N_2O), and they were potent greenhouse gases [17,18]. NO and N_2O were not detected at any of the COD/N ratios tested. It confirmed that the complete denitrification occurred and it led to the production of nitrogen gas (N_2) in the nitrate reduction process.

3.4. Effect of HRT

The reactor was operated at different HRTs and the consistent stable carbon and ammonia removals were attained. At all HRTs, the reactor was operated until a steady-state was reached and the constant effluent COD and ammonia concentrations were obtained. The variations of COD and ammonia removal efficiencies were observed for a short period as HRT increased.

Fig. 5 shows the effect of HRT on the COD removal efficiency. The HRT was varied from 48 to 10 h during 210 days operation to investigate the effect of HRT on the efficiency of COD removal. As shown in Fig. 5, the microorganisms adapted to the increased HRTs in a short period and the efficiency of the reactor did not decrease greatly. The main reasons were the stability of the baffled reactor and the easily biodegradable components, such as glucose in the wastewater composition [18,19]. The COD concentration of the effluent gradually increased from 150 to 600 mg/L as the HRT decreased from 48 to 10 h. The total efficiency of COD removal decreased from 95 to 80%. The COD concentration of effluent was less than 400 mg/L, and the maximum efficiency of COD removal was more than 87%, when the HRT was more than 20 h. Therefore, the optimal HRT was between 20 and 48 h, and effluent water met the discharge standard.

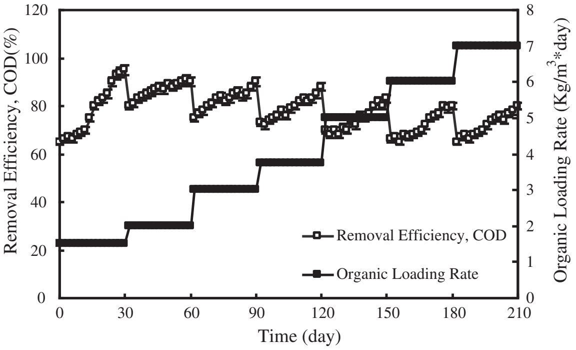

Fig. 5. COD removal efficiencies at different organic loading rates.

As shown in Fig. 5, the NH_4^+-N concentration of the inflow was 85 mg/L and that of the effluent was ranged from 5 to 15 mg/L, as the HRT was varied from 48 to 10 h. The TN removal efficiency decreased slightly as HRT increased. It was probably because the extended retention time of the aeration zone allowed the slow growing nitrifiers to oxidize ammonia. The HRT decreased, and it was beneficial for decreasing the heterotrophic inhibition of nitrification [20–22]. The NH_4^+-N concentration of the effluent was less than 15 mg/L and TN concentration was less than 30 mg/L. The results indicated that the bioreactor was highly efficient for the removal of NH_4^+-N from molasses wastewater.

4. Conclusions

The results of the study demonstrate that the GBCOBBR is an effective system for carbonaceous matter and nitrogen removal for wastewater containing organic matter and ammonia. Simultaneous organic removal and denitrification were observed in the GBCOBBR, and it showed the importance of a multi-phase granular bed system for combined carbon and nitrogen removal. In the GBCOBBR system, an active denitrification zone can be developed in the acidogenic dominant zone by recycling nitrified effluent to the feed of the reactor. Sufficient carbon was available for the completion of the denitrification process of the reactor.

In the study, the COD concentration of the effluent decreased to less than 400 mg/L and the maximum removal efficiency of COD increased to 91.8% at the HRT of 24 h, the recirculation of 200%, and the GWR of 20:1. The ammonia concentration of the effluent was maintained at 10 mg/L and TN concentration was maintained less than 30 mg/L, and it demonstrated that effluent of the reactor met discharge standard. The configuration is an effective solution to the treatment of wastewater for most small- and medium-sized molasses plants in China which possess low economic capacity to invest in environmental controls.

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