Removal of organic matter and nitrogen from distillery wastewater by a combination of methane fermentation and denitrification/nitrification processes

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Abstract: The distillery wastewater of Guangdong Jiujiang Distillery, which is characteristic of containing high organic matters and rich total nitrogen, was treated by a combination of methane fermentation and denitrification/nitrification processes. 80% of COD in the raw wastewater was removed by methane fermentation at the COD volumetric loading rate of 20 kg COD/(m³·d) using the expanded granule sludge bed (EGSB) process. However, almost all the organic nitrogen in the raw wastewater was converted into ammonia by ammonification there. Ammonia and volatile fatty acids (VFA) remaining in the anaerobically treated wastewater were simultaneously removed utilizing VFA as an electron donor by denitrification occurring in the other EGSB reactor and nitrification using PEG-immobilized nitrifying bacteria with recirculation process. An aerobic biological contact oxidization reactor was designed between denitrification/nitrification reactor for further COD removal. With the above treatment system, 18000—28000 mg/L of COD in raw wastewater was reduced to less than 100 mg/L. Also, ammonia in the effluent of the system was not detected and the system had a high removal rate for 900—1200 mg/L of TN in the raw wastewater, only leaving 400 mg/L of nitrate nitrogen.

Keywords: ammonia; COD; VFA; methane fermentation; denitrification; nitrification; distillery wastewater; immobilized nitrifying bacteria

Introduction

Distillery wastewater mainly includes spent wash, which is one of the most complex, troublesome industrial organic effluents. This kind of wastewater has previously been treated aerobically after dilution with other wash waters. However, aerobic treatment has always been difficult because of the acidity of the waste, its high temperature and high oxygen demand (Wheatley, 1991).

Undoubtedly, anaerobic digestion is the most suitable way for the treatment of high strength effluents such as distillery wastewater (Rajeshwari et al., 2000). The BOD content of many high strength effluents from food, fermentation, beverage and pulp and paper industries can successfully be reduced by anaerobic digestion (Nigmet et al., 2003). Goodwin et al. (2001) studied the anaerobic biotreatment of malt whisky distillery wastewater using an upflow anaerobic sludge blanket(UASB) reactor. Stable operation was observed at an organic loading rate(OLR) of 5.46 kg COD/(m³·d) or less when the pot-ale feed was diluted, but failure occurred when undiluted pot ale was fed to the digester even though OLR was less than 5 kg COD/(m³·d). Garcia-Calderon et al. (1998) studied the application of the down-flow fluidization technology for the anaerobic digestion of red wine distillery wastewater. Once the down-flow anaerobic fluidized bed system reached the steady-state, organic load was increased step-wise by reducing hydraulic retention time (HRT), from 3.3 to 1.3 d, while maintaining constant the feed TOC concentration. The system achieved 85% TOC removal, at an organic loading rate of 4.5 kg TOC/(m³·d). The main advantages of the down-flow fluidization configuration are that a settler is not necessary; no clogging and the low energy requirement. Investigations were carried out by using rigid polyurethane foam as a packing material in the anaerobic contact filter (series) to treat distillery spentwash (Vijayaraghavan and Ramanujam, 2000). Experiment demonstrated that at 4 d HRT the overall COD removal ranged from 98% to 73% for an influent COD concentration ranging from 1500 to 19000 mg/L. Nigmet et al. (2003) draw a conclusion that the two stage UASB reactor configuration is an efficient system for malt whisky wastewater treatment until up to 33866 mg/L influent COD concentration. Following the UASB experiments, aerobic experiments in batch reactors were also conducted and further COD and BOD removal were up to 55% and 70%, respectively. The study of anaerobic digestion of distillery waste in the two-stage reactor enabled the identification of two main phases of the process. The acidogenic reactor performed satisfactorily in terms of conversion of initial COD to volatile fatty acids(VFAs). VFAs produced in the first stage were readily used as substrates in the acetogenic/methanogenic stage (Blonskaja et al., 2001). Goodwin et al. (1994) used two identical UASB reactors operated in parallel as duplicates for the treatment of malt whisky pot ale and achieved COD reductions of up to 90% for influent concentrations of 3526—52126 mg/L. This study was conducted for 327 d. When the OLRs of 15 kg/(m³·d) and above were used, the COD removal efficiency

dropped to less than 20%, in one of the duplicate reactors.

In this study, two identical EGSB (expanded granule sludge bed) reactors were respectively used for COD and nitrogen removal by methane fermentation and denitrification/nitrification circulation from Guangdong Jiujiang Distillery wastewater. Nitrification process adopted PEG-immobilized nitrifying bacteria pellet supplied by Japan Hitachi Plant Engineering & Construction Co., Ltd for obtaining high nitrification efficiency. Additionally, the activated sludge unit, that is, the aerobic contact oxidization reactor before the nitrification reactor is necessary to prevent the growth of BOD oxidizing bacteria in the nitrification reactor at the most. Fig.1 shows the overall treatment system process. The performance of the system was evaluated at a laboratory scale. This study mainly focused on effectiveness of an EGSB process for methane fermentation of distillery wastewater and nitrogen removal performance by a biological denitrification using an anerobic EGSB reactor and immobilized nitrification with circulation.
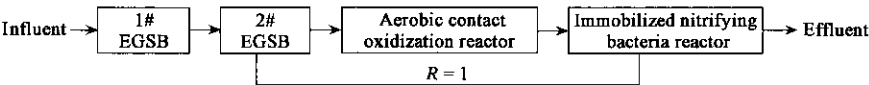
Influent → | 1# EGSB | → | 2# EGSB | → | Aerobic contact oxidization reactor | → | Immobilized nitrifying bacteria reactor | → Effluent
R = 1

Fig.1 Overall treatment system process with recirculation

1 Materials and methods

1.1 Characterization of distillery wastewater

The distillery wastewater from Guangdong Jiujiang Distillery contains high organic matters and rich total nitrogen. Its main components are shown in Table 1.

Table 1 Composition of distillery wastewater from Guangdong Jiujiang Distillery

Parameter	Concentration
COD, mg/L	18000—28000
pH	4.5—5.5
NH₄-N, mg/L	200—300
TN, mg/L	900—1200

1.2 Seeding sludge

Anaerobic granule sludge (MLSS=32976 mg/L, MLVSS=27398 mg/L, VSS/SS=83%), provided by Guilin Liquan Beer Company, was used as a seed sludge for methane fermentation and denitrification. Activated sludge in aeration tank of the old wastewater treatment system of the distillery was used as seed sludge for nitrification.

1.3 Methane fermentation experiments

As the pH of the distillery wastewater was about 5, it was adjusted to 7 by addition of NaOH. Additionally, 0.1 g/L KH₂PO₄ and 2 ml/L of a stock solution containing trace elements were also added. The stock solution consisted of (L⁻¹): FeCl₃·4H₂O, 2000 mg; H₃BO₃, 50 mg; ZnCl₂, 44.167 mg; CuCl₂· 2H₂O, 40 mg; AlCl₃, 30 mg; MnCl₂·4H₂O, 500 mg; (NH₄)₆Mo₇O₂₄·4H₂O, 175.578 mg; CoCl₂·6H₂O, 150 mg; NiCl₂·6H₂O, 100 mg. After that, the distillery wastewater can be used as the influent of the methane fermentation. The EGSB reactor used for methane fermentation had a working volume of 18 L, in which 8 L seed sludge was added. Temperature was kept at 35℃ by circulation of water through a water-jacket. The maximum COD volumetric loading rate was studied with stepwise increases in the feeding rate of the influent. During the experiment, HRT was maintained about 20 h and COD of the influent varied from 4000 to 20000 mg/L.

1.4 Denitrification experiments

The denitrification reactor, with a working volume of 18 L, was the same as the methane fermentation reactor. 8 L anaerobic granule sludge was put into the reactor. The anaerobically treated effluent was used as the influent of the denitrification with NO₃-N addition. The feeding rate of the NO₃-N was increased stepwise. During the experiment, the pH of the reactor was kept at 7.0 and influent COD varied from 1000—3000 mg/L; HRT was maintained 15 h.

1.5 Aerobic contact oxidation unit experiments

The aerobic contact oxidation reactor had a working volume of 15 L and 10 L activated sludge was put into the reactor. The denitrified effluent was fed. DO in the reactor maintained 3.5 mg/L by aeration. During the experiments, the COD volumetric loading rate was increased stepwise and COD removal efficiency was investigated.

1.6 Nitrification experiments

Nitrification process adopted the technology of embedded nitrifying bacteria pellet provided by Japan Hitachi Plant. The pellet was a small cubic with volume of 27 mm³. 2 L pellets was put into the nitrification reactor with a working volume of 18 L. The pellets in the reactor were fluidized under the 6.0 mg/L DO by aeration. Aerobically treated effluent was introduced the reactor and Na₂CO₃ was added so as to maintain the pH of the reactor at 7.5. During the experiment, HRT was kept at 15 h and the operating temperature was maintained at 30℃. Sometimes it was

necessary for the influent to be diluted because influent COD should less than 300 mg/L to prevent the growth of the BOD oxidation bacteria.

1.7 Overall sequential system with recirculation experiments

After the start-up of each reactor finished, the effluent from the nitrification reactor was recirculated to the denitrification reactor maintaining a recirculation feeding ratio of 100% together with the anaerobically treated effluent instead of the NO_3-N addition. Moreover, the pH of the denitrification unit also did not be adjusted. The denitrified effluent was fed into the aerobic contact oxidation reactor and was then routed to the nitrification reactor. The pH of the nitrification reactor still need to be maintained at 7.5 by the addition of Na_2CO_3 or HCl solution. The COD and ammonia removal efficiency of the whole system connected in series were investigated.

1.8 Analytical methods

All analyses were done in accordance with the standard methods for the examination of water and wastewater (APHA, 16th ed.).

2 Results and discussion

2.1 Anaerobic treatment using an expanded granule bed reactor

The distillery wastwater was first anaerobically treated with stepwise increases in the COD volumetric loading rate from 3 to 25 kg COD/(m³·d). As shown in Fig.2, the COD removal efficiency maintained more than 80% and VFA concentrations in the effluent was kept less than 2000 mg/L when the COD volumetric loading rate (CVLR) ranged from 3 to 20 kg COD/(m³·d). Effluent alkalinity tended to rise and fall with feed COD, but was over 2000 mg/L at all times.

On the day 20, effluent COD and VFA concentrations began to rise significantly and COD removal efficiency declined quickly because the CVLR was increased greatly. To prevent reactor

Fig.2 Effluent quality and COD removal efficiency during anaerobic EGSB treatment
TA. total alkalinity

failure, pH in the influent was adjusted to 9 from that day, then still kept at 7.0 on the day 28. VFA levels fell and COD removal efficiency improved, suggesting a return to stability.

After the CVLR was raised to more than 20 kg COD/(m³·d) on the day 45, COD removal efficiency fell sharply, effluent VFA increased and biogas production yield declined, indicating organic overload.

COD removal efficiency, gas production yield, effluent VFA and COD concentrations were determined at each COD volumetric loading rate as depicted in Fig.3. A COD removal efficiency of 80% was achieved even at the COD volumetric loading rate of 20 kg COD/(m³·d), but it decreased to 70% and VFA concentration in the effluent reached 3000 mg/L when the CVLR was raised 25 kg COD/(m³·d). The gas production yield increased with the CVLR increasing. 250 L/d of the gas production yield was achieved at the CVLR of 20 kg COD/ (m³·d), however, it began to fall when the CVLR was more than 20 kg COD/(m³·d). Only 120 L/d of the gas production yield was achieved at CVLR of 25 kg COD/ (m³·d). So, the maximum CVLR was determined to be 20 kg COD/(m³·d) corresponding to a HRT of 28 h.

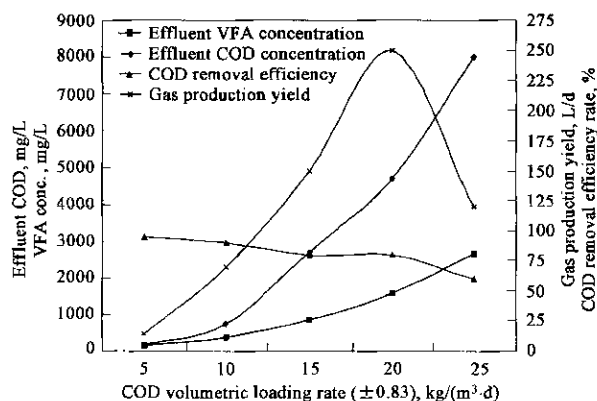Fig.3 Effect on COD volumetric loading rate on treatment efficiency during anaerobic EGSB treatment

Fig.4 shows the changes of NH_4-N and TN with time in the influent and effluent. NH_4-N concentration ranged between 50 and 400 mg/L and TN concentration varied between 400 and 1200 mg/L in the influent. After anaerobic treatment at a HRT of 20 h, a great deal of ammonia was accumulated because almost all the organic nitrogen in the raw wastewater was converted into ammonia by ammonification and NH_4-N concentration was close to the TN concentration in the effluent.

2.2 Removal of VFA remaining in the anaerobically treated effluent by NO_3 addition in a denitrification reactor using an expended granule bed
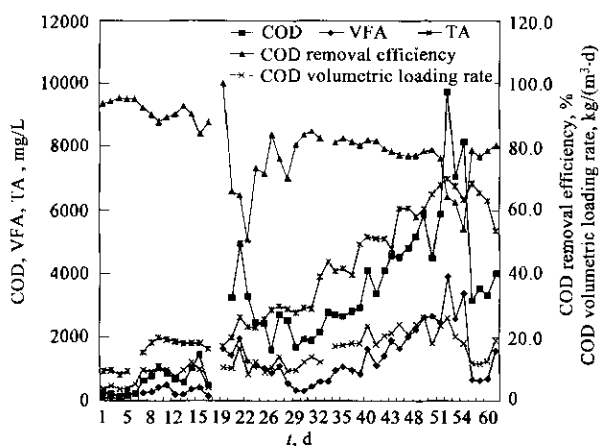
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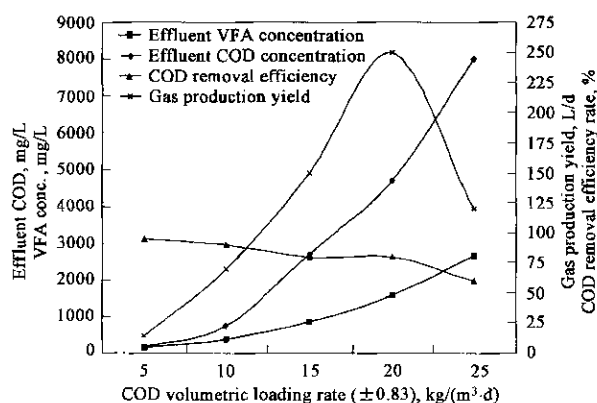Fig.3 Effect on COD volumetric loading rate on treatment efficiency during anaerobic EGSB treatment

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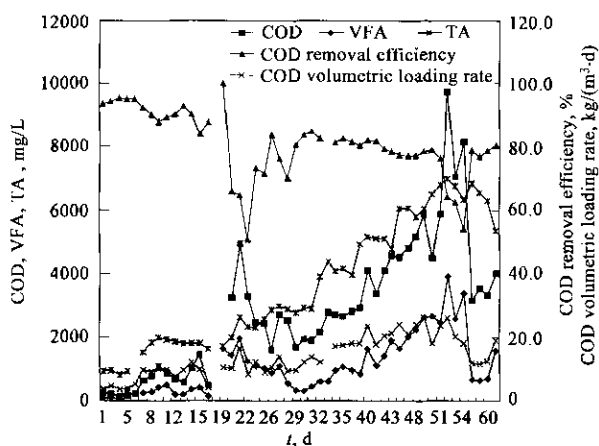
The anaerobically treated wastewater with NO_3-N
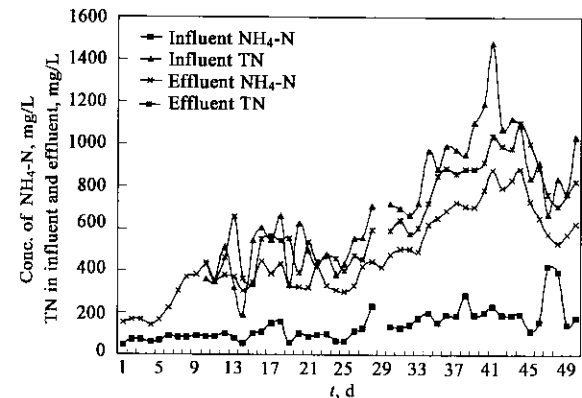
Fig.4 Changes of NH₄-N and TN concentrations in the influent and effluent with time during anaerobic EGSB treatment

addition was fed into the denitrification, in which the denitrifying bacteria would be acclimatized. Fig.5 shows VFA, COD and NO₃-N removal efficiency and changes of the NO₃-N volumetric loading during the experiment. The NO₃-N volumetric loading rate was increased stepwise from 40 to 800 gN/(m³·d), then was maintained about 400 gN/(m³·d) after the day 31.
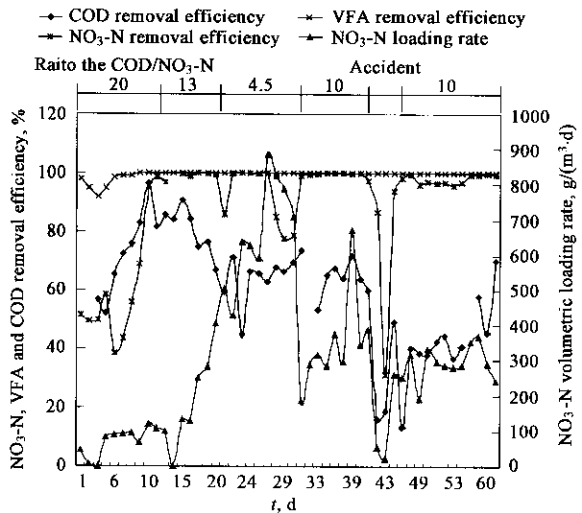

Fig.5 Changes of NO₃-N, VFA and COD removal efficiency during denitrification of anaerobic methane fermentation effluent using another anaerobic EGSB reactor with NO₃-N addition

The VFA removal efficiency was always almost 100% and the effluent NO₃-N concentration is below 1 mg/L up to the NO₃-N volumetric loading rate of 500 g/(m³·d) on the day 20, suggesting VFA remaining in the anaerobically treated effluent and NO₃-N can be removed simultaneously utilizing VFA as an electron donor by denitrification.

However, the loading ranging from 500 to 800 gN/(m³·d) and the average COD: NO₃-N ratio of 4.5 from the day 20 to 30 brought about the unstable denitrification efficiency because of lack of carbon source of electron donors. After the day 30, the loading of 400 gN/(m³·d) corresponding to an average COD : NO₃-N ratio of 10 was maintained, then the

removal rate of NO₃-N seemed to be increased again, suggesting without the shortage of COD.

From the day 1 to 10, the average COD: NO₃-N ratio of 20 was enough for denitrification and the NO₃-N removal efficiency increased gradually up to 100% with NO₃-N addition. This period can be regarded as the acclimatization of the denitrifying bacteria. Moreover, a high COD removal efficiency of 95% was also achieved during this period, which can be explained by simultaneous methanogenesis and denitrification. This is possible when the carbon source is easily assimilated and an adequate C:N ratio (Akunna et al., 1992). Some authors also used VFA produced by a previous anaerobic unit and studied the influence of the type of carbon source on the growth of denitrifiers and methanogens (Rustrian et al., 1997; Bernet et al., 1996). The denitrifying capacity of methanogenic sludge was studied in lab assays, using glucose, to establish the importance of the COD: N ratio. The end products, working at ratios higher than 53, are methane and ammonia. Values of COD:N ratio between 9 and 53 produce methanisation and complete denitrification, while with values lower than 9 the main process is denitrification (Akunna et al., 1992).

However, the removal efficiency of COD deceased from 90% to 60% caused by the activation of the denitrification process with the NO₃-N volumetric loading increasing. The true reason was the lack of the carbon resource. Studies (Akunna et al., 1993; Hanaki and Polprasert, 1989) showed the denitrification took place in the bottom of the digester while methanogenesis occurred in the upper part, where nitrate was completely exhausted in anaerobic reactors. If lack of the carbon resource, nitrated was not exhausted completely and went into the upper part of the reactor where it would strongly inhibit the methanogenesis (Akunna et al., 1993).

Simultaneous methanogenesis and denitrification of the anaerobically pretreated wastewater for Guangdong Jiujiang Distillery wastewater treatment would be discussed in another paper.

On the day 41, an accident happened and the new sludge was filled into the denitrifying reactor. Still maintaining the COD: NO₃-N ratio of 10, COD and NO₃-N removal efficiency reached 70% and 99% after 20 days.

2.3 Further removal of COD in the denitrified effluent by aerobic contact oxidation process

The aerobic contact oxidation process was operated for 40 d. The denitrified effluent was treated and the changes of COD and ammonia were illustrated in Fig.6. During aerobic treatment, COD in the influent varied between 600 and 1000 mg/L and its effluent COD was always under 300 mg/L, satisfying the demand of the influent of the nitrification reactor.

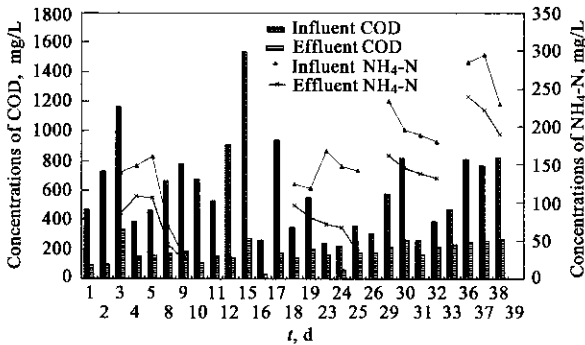Moreover, a NH$_4$-N removal rate of 25% could be achieved at the same time.

Fig.6 Changes of COD and NH$_4$-N concentrations in the influent and effluent during the aerobic contact oxidation process

2.4 Removal of ammonia by nitrification using embedded nitrifying bacteria pellets

Fig.7 shows the NH$_4$-N removal, NO$_3$-N and NO$_2$-N production change with time in immobilized cell bioreactor. The ranges of loading rate changes for

NH$_4$-N was 40—200 mgN/(L-pellet·h). When the loading was more than 130 mgN/(L-pellet·h), the effluent ammonia concentration was beyond 15 mg/L and cannot satisfy the demanded effluent quality. Then, the loading was maintained at 130 mg N/ (L-pellet·h) until the experiment ended.

Experimental results demonstrated the ammonia removal efficiency approximately 95% at loading 40—150 mgN/(L-pellet·h). Moreover, NO$_3$-N was dominant compared with NO$_2$-N in the effluent. However, NO$_3$-N concentration decreased and NO$_2$-N concentration had an increasing trend with the operation time suggesting NO$_2$-N was accumulated gradually by high free ammonia. Additionally, a total nitrogen removal efficiency of 10% —20% can be achieved in the later period of the experiment, which can be explained by stripping because of high free ammonia concentration during that period.

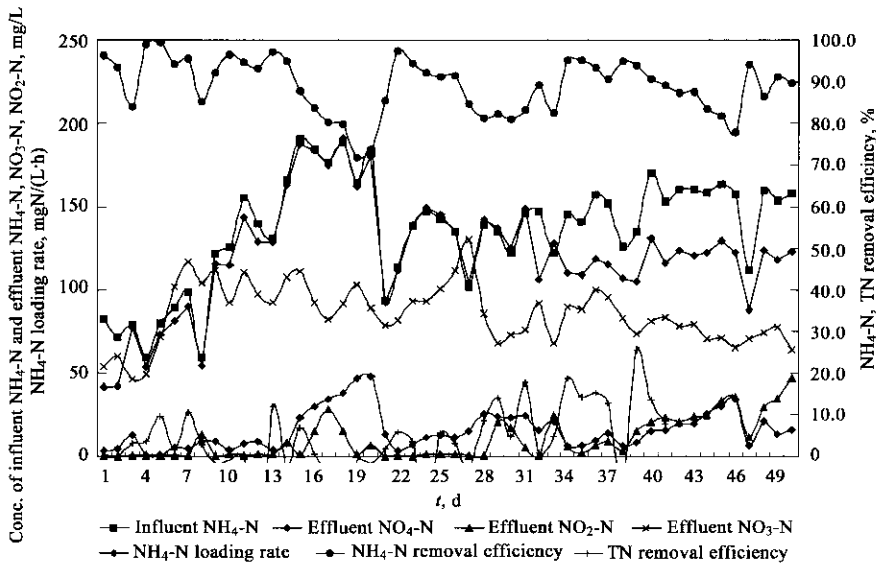The maximum nitrification rate in the immobilized nitrifying bacteria bioreactor remained at

Fig.7 Ammonia removal, nitrite and nitrate production changes with time during nitrification using embedded nitrifying bacteria pellets

155 mgN/(L-pellet·h) until the loading was increased up to 160 mg N/(L-pellet·h) after which the nitrification rate seemed to be decreasing as in Fig. 8.

2.5 Performance of the overall sequential system with recirculation from the nitrification to the denitrification reactor for COD and N removal

Table 2 shows the experimental conditions for COD and N removal by the overall sequential system with recirculation and quality of effluent from each step.

The COD and TN removal efficiency by anaerobic-aerobic process with recirculation were 90% and 66%. Moreover, NH$_4$-N accumulated in the anaerobically treated wastewater could also be removed approximately 100% by denitrification and nitrification utilizing VFA remaining in the anaerobically treated wastewater as electron donors.
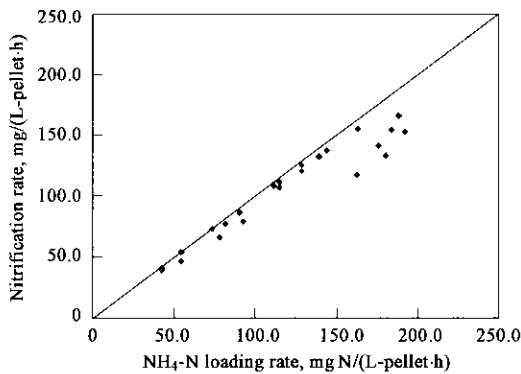

Fig.8 NH$_4$-N loading and nitrification rate during nitrification using embedded nitrifying bacteria pellets

The quality of effluent from the nitrification reactor was suitable to be discharged.

Table 2 Experimental conditions for COD and N removal by the overall sequential system with recirculation and quality of effluent from each step

	Distillery wastewater	Anaerobic treatment	Denitrification treatment	Aerobic contact oxidation treatment	Nitrification treatment
Conditions					
Working volume, L		18	18	15	18
Feed, L/d		15	30	30	30
HRT, h		28.8	14.4	12.0	14.4
Quality					
COD, mg/L	22000	4500	1050	150	90
NH$_4$-N, mg/L	350	1100	480	320	0
NO$_2$-N, mg/L	-	-	0	100	30
NO$_3$-N, mg/L	-	-	0	30	380
TN, mg/L	1200	1200	500	450	400
pH	7.0	7.5	8.6	8.0	7.5

3 Conclusions

In conclusion, the distillery wastewater of the Guangdong Jiujiang Distillery was effectively treated by the proposed system by a combination of methane fermentation, denitrification, aerobic treatment and nitrification with circulation processes. 80% of COD in the raw wastewater was effectively removed by methane fermentation using the EGSB process. The maximum COD volumetric loading rate of 20 kg COD/(m^3·d) was achieved corresponding to a HRT of 28 h. Moreover, NH$_4$-N accumulated in the anaerobically treated wastewater could also be removed approximately 100% by denitrification/nitrification utilizing VFA remaining in the anaerobically treated wastewater as electron donors without addition of electron donor. With the proposed overall treatment system, 18000—28000 mg/L of COD in the raw wastewater was reduced to less than 100 mg/L. Also, ammonia in the effluent of the system was not detected and the system had a high removal rate for 900—1200 mg/L of TN in the raw wastewater, only leaving 400 mg/L of nitrate nitrogen.

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