Experimental study on the simultaneous nitrification and denitrification of limited aeration biofilm reactor

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Abstract. To improve the nitrogen removal effects of domestic sewage and reactive mode, the two-stage limited aeration biofilm reactor was used to carry out an experiment. The pollutants removal effects and influencing factors were analyzed. The results indicate: Under the condition that the 1st reaction chamber DO of 1.5~2 mg/L, the 2nd reaction chamber DO of 1~1.5 mg/L, HRT=5 h, and influent ρ(COD) = 147~181 mg/L, ρ(NH$_4^+$-N) = 35.1~43.6 mg/L, ρ(TN) = 36.3~44.1 mg/L, the maximal concentration of COD, NH$_4^+$-N, and TN in the last effluent were 18.8 mg/L, 0.35 mg/L and 14.2 mg/L, with the mean removal rates of 91.1%, 99.5%, 66.6% and the SND ratio of 62.7% ~ 68.9%. Under the condition of 4.17h≤HRT≤5h, the effluent concentration of COD, NH$_4^+$-N met the Category Ⅲ of environmental quality standards for surface water (GB3838-2002), and the effluent concentration of TN met the Class A discharge standard of pollutants for municipal wastewater treatment plant (GB18918-2002) with the SND ratio of 65.7%. Under the condition that the 1st reaction chamber DO of 1.5~2 mg/L, the 2nd reaction chamber DO of 1~1.5 mg/L, HRT=4.17 h, the SND effects can be improved by the means of additional sodium acetate in the 2nd reaction chamber. The TN removal rate and SND ratio were respectively 75.4% and 75.8% at the optimal dosage of 40 mg/L.

1. Introduction
At present, the effluent water quality of domestic sewage treatment plants generally meets the requirements of Class A of GB 18918-2002 of the “Pollutant Discharge Standards of Urban Wastewater Treatment Plants”. With the increasing requirements for prevention and control of eutrophication of water bodies, some areas lacking receiving water bodies even require the effluent NH$_4^+$-N, COD and TP concentration of newly constructed sewage treatment plants to reach the limit value of Class III standard of “Environmental Quality Standard for Surface Water” GB3838-2002, and it is clearly required that ρ(TN)≤10mg/L. COD and TP can be further strengthened and removed through advanced treatment after biochemistry, while nitrogen removal is mainly achieved through biological treatment process [1-2]. Due to the limitation of influent water quality and process conditions, it is difficult for traditional processes to achieve low cost and deep nitrogen removal [3-4]. Aiming at the development of new biological nitrogen removal process for domestic sewage, domestic and foreign research mainly focuses on oxidation ditch, A/O, staged water intake method, SBR, biological filter, MBR and other processes, relatively little research on new biological membrane denitrification reactor [5-12]. SND (simultaneous nitrification and denitrification) can achieve the simultaneous removal of NH$_4^+$-N, TN and COD in the same reactor, which can save carbon source and simplify the process
flow, break the limitation of the space setting of the conventional biological treatment process, and strengthen the simultaneous denitrification Nitrogen effect [13]. The existing SND technology research is mainly aimed at the SBR reactor, and the process optimization is achieved through intermittent aeration, and there is a lack of research on the biofilm method [14]. This study takes oxygen-limited aeration biofilm reactor as the research object, discusses the control method of stably realizing SND and high-efficiency denitrification in a single reactor, and provides new ideas for the design of domestic sewage biological treatment process.

2. Materials and methods

2.1. Test device

As shown in Figure 1, the oxygen-limited aeration biofilm reactor is composed of C1 and C2 two-stage reaction chambers connected in series. The effective volumes of C1 and C2 are 1.5 m³ and 1 m³, respectively, and the total effective volume is 2.5 m³. Both C1 and C2 are equipped with a mesh-shaped cross-linked porous elastic carrier with a filling rate of 70%, an aeration tube and a sludge discharge tube. The carrier shape is a cube with a side length of 50 mm, and the porosity is 91.2%. The sewage is introduced into the upper part of C1 by the water pump, and the effluent of C1 flows into C2 through the lower water flow channel. The aeration pipelines of C1 and C2 are independent of each other. The air supplied by the two rotary fans is input into the bottom of the reaction chambers of C1 and C2 through the aeration pipes, respectively. The air volume is controlled by an online DO and PLC. For the external carbon source test, the sodium acetate solution used to supplement the carbon source is introduced into the lower part of C2 through a metering pump and mixed with sewage.

Fig. 1 Schematic diagram of limited aeration biofilm reactor

2.2. Test method

The test water was domestic sewage from a community in Hangzhou. The raw water quality during the test period: NH₄⁺-N 32.2~43.6mg/L, TN 32.9~45.1 mg/L, TP 2.18~2.73 mg/L, COD 132~189mg/L, pH 7.35~7.56.

The test process is divided into three stages: reactor start-up, stable operation and parameter optimization. In the reactor start-up phase, after inoculation with 2L of compound microbial inoculum at one time, the aeration has begun. After the biofilm cultured in the carrier, the inlet water flow was gradually increased to 0.5m³/h according to the actual treatment effect of the reactor. During this period, the concentration of C1 and C2 DO was increased, controlled to 2±0.2 mg/L and 1.5±0.5 mg/L, respectively. In the stable operation stage, the influent flow rate is 0.5m³/h (HRT=5h), and the concentration of C1 and C2 DO is controlled to 1.5~2 mg/L and 1~1.5 mg/L, respectively, and the removal effect of the reactor on the main pollutants is analyzed. In the parameter optimization stage,
the concentrations of C1 and C2 DO were controlled to 1.5–2 mg/L and 1–1.5 mg/L, respectively, and the nitrogen removal effect was further optimized by adjusting the dosage of HRT and sodium acetate.

Analysis and test methods: NH$_4^+$-N, TN, COD were determined by Nessler reagent colorimetry, TNT persulfate digestion method, and rapid digestion spectrophotometry.

3. Results and discussion

3.1. Analysis of pollutant removal effect

Under the conditions of HRT=5h, $\rho$(DO)$_{C1}$=1.5–2 mg/L, $\rho$(DO)$_{C2}$=1–1.5 mg/L, the removal effect of COD, NH$_4^+$-N and TN is shown in Fig.2~Fig.4. In the case of influent $\rho$(COD)=147–181mg/L, C1 effluent $\rho$(COD)=36.5–47.3mg/L, C2 effluent $\rho$(COD)=11.6–18.8mg/L, after second level of biochemical reaction, the COD removal rate of the reactor is 89.4%–92.8%, the average removal rate is 91.1%, and the COD concentration of the effluent reaches the GB3838-2002 class III standard limit ($\rho$(COD)≤20mg/L).

Under the condition of HRT=5h, the removal effect of NH$_4^+$-N by oxygen-limited aeration biofilm reactor is shown in Fig. 3. In the case of influent $\rho$(NH$_4^+$-N)=35.1–43.6mg/L, C1 effluent $\rho$(NH$_4^+$-N)=0.02–0.35 mg/L. After second level of nitrification, the removal rate of NH$_4^+$-N by the reactor is 99.2%–99.9%, the average removal rate is 99.5%, and the concentration of NH$_4^+$-N in the effluent reaches the GB3838-2002 class III standard limit ($\rho$(NH$_4^+$-N)≤1mg/L) requirements.

Under the condition of HRT=5h, the removal effect of oxygen-limited aeration biofilm reactor on TN is shown in Fig.4. In the case of influent $\rho$(TN)=36.3–44.1mg/L, C1 effluent $\rho$(TN)=14.3–18.7mg/L, C2 effluent $\rho$(TN)=12.3–14.2mg/L, after second level of denitrification, the removal rate of TN by the reactor is 63.4%–69.5%, the average removal rate is 66.6%, the SND rate is 62.7%–68.9%, and the TN concentration of the effluent meets GB 18918-2002 level A ($\rho$(TN)≤15mg/L) requirements.
In the case of large fluctuations in the concentration of influent pollutants, the reactor's removal of major pollutants is stable. Under oxygen-limited aeration conditions, both C1 and C2 reaction chambers can achieve the simultaneous removal of NH₄⁺-N, COD and TN, and have a strong anti-impact load capacity, which achieves a deep treatment effect on NH₄⁺-N and COD. The operation mode of this reactor is different from the conventional aerobic biofilm reactor. Under oxygen-limited aeration conditions, the nitrifying bacteria and heterotrophic bacteria on the surface of the carrier maintain a high consumption rate of DO, and the bubbles are difficult to penetrate directly. The surface biofilm enters all the pores inside the carrier, thus forming a more stable local anaerobic microenvironment, prompting the reactor to carry out the SND reaction under the condition of sufficient carbon source. In addition, the reactor does not achieve denitrification through anaerobic section or sequential batch aeration. The micro-anaerobic zone does not affect the efficient removal of NH₄⁺-N and COD in the aerobic zone.

3.2. Effect of HRT on the processing result

Under the conditions of ρ(DO) C1 = 1.5~2 mg/L and ρ(DO) C2 = 1~1.5 mg/L, the effect of HRT on the treatment effect of the reactor is analyzed through the adjustment of the influent flow rate, removal effects of HRT on COD, NH₄⁺-N and TN are shown in Fig.5–Fig.7. In the case of 1.79h ≤ HRT ≤ 3.57h, prolonging HRT can significantly improve the COD removal rate. When HRT ≥ 3.57h, the effect of continuing to extend HRT on COD removal rate is not obvious, η(COD) = 89.5%~91.1%. When HRT ≥ 2.08h and effluent ρ(COD) ≤ 45.6 mg/L, it can meet the requirements of Class A of GB 18918-2002. When HRT ≥ 3.57h, effluent ρ(COD) ≤ 17.6 mg/L, it can meet the requirements of GB3838-2002 Class III standard.

In the case of 1.79h ≤ HRT ≤ 4.17h, prolonging HRT can significantly improve the removal rate of NH₄⁺-N. When HRT ≥ 4.17h, the effect of continuing to extend HRT on NH₄⁺-N removal rate is not obvious, η(NH₄⁺-N) = 98.9%~99.5%. When HRT ≥ 2.5h, effluent ρ(NH₄⁺-N) ≤ 4.51 mg/L, which can meet the requirements of Class A of GB 18918-2002. When HRT ≥ 4.17h, the NH₄⁺-N concentration of the effluent can meet the requirements of GB3838-2002 Class III standard.

In the case of 1.79h ≤ HRT ≤ 3.13h, the TN removal rate increases with the extension of HRT, this trend is particularly significant in 1.79h ≤ HRT ≤ 2.5h. When HRT ≥ 3.13h, effluent ρ(TN) ≤ 14.2 mg/L, which can meet the requirements of Class A of GB 18918-2002, η(TN) = 64.6%~66.2%.
According to the overall impact of HRT changes on the removal effect of COD, NH$_4^+$-N and TN, prolonging HRT can improve the effluent quality to a certain extent, but too long HRT will affect the treatment efficiency of the reactor. In the range of 4.17h≤HRT≤5h, the change of HRT basically does not affect the treatment effect. The COD and NH$_4^+$-N concentrations in the effluent of the reactor all meet the requirements of GB3838-2002 Class III standard. The effluent ρ(TN)≤13.3mg/L, SND The rate reached 65.7%. Considering the requirements of reactor treatment efficiency and effluent water quality, it is reasonable to control the reactor HRT at 4.17h.

3.3. Effect of external carbon source on nitrogen removal effect

In the SND system of the oxygen-limited aeration biofilm reactor, the organic carbon source is both the energy source of the heterotrophic bacteria and the electron donor in the denitrification process, which has a significant impact on the activity of the heterotrophic bacteria and the effect of denitrification. Adding carbon source is the main means to improve the TN removal effect of domestic sewage. Under the conditions of HRT=4.17h, ρ(DO)$_{C1}$=1.5~2 mg/L, ρ(DO)$_{C2}$=1~1.5mg/L, sodium acetate is added to C2 to supplement the organic carbon source. The effect of sodium acetate addition on the treatment effect is shown in Fig. 8. When the dosage of sodium acetate is 20~100mg/L, increasing the dosage can further optimize the TN removal effect. The TN removal rate is increased from 69.1% to 86.4%, the SND rate is increased from 65.8% to 89.2%, and the effluent TN concentration can be reduced to 5.31mg/L. But on the other hand, the addition of sodium acetate will affect COD and NH$_4^+$-N removal to varying degrees. When the dosage of sodium acetate is greater than 40mg/L, continuing to increase the dosage will cause the COD concentration in the effluent of the reactor to increase. The dosage is ρ(COD)=23.2 mg/L under the condition of 80mg/L, exceeding GB3838-2002 Class III standard limits. The C2 HRT is only 1.67h, and the reaction time is relatively short. Some sodium acetate does not actually participate in the aerobic decomposition or denitrification in the local anaerobic zone, resulting in an increase in the effluent COD concentration. Relatively speaking, the addition of sodium acetate has little effect on nitrification. When the sodium acetate dosage ≤60mg/L, the effluent ρ(NH$_4^+$-N)≤0.81mg/L, which can meet the requirements of GB3838-2002 Class III standard. When the dosage of sodium acetate ≥80mg/L, the concentration of NH$_4^+$-N in the effluent showed a significant upward trend, which further affected the increase of TN removal rate. In summary, the addition of sodium acetate to C2 can improve the TN removal rate. Considering the cost of the agent and the overall removal effect of pollutants, the optimal dosage is 40mg/L. The corresponding effluent COD, NH$_4^+$-N and TN concentrations are respectively 16.5mg/L, 0.63mg/L and 9.62mg/L, and the TN removal rate and SND rate were 75.4% and 75.8%, respectively.
4. Conclusion

(1) Adopt second-level oxygen-limited aeration biofilm reactor to treat domestic sewage. When $\rho(\text{DO})_C1=1.5\sim2\text{mg/L}$, $\rho(\text{DO})_C2=1\sim1.5\text{mg/L}$, $\text{HRT}=5\text{h}$, $\rho(\text{COD})=147\sim181\text{mg/L}$, $\rho(\text{NH}_4^+-\text{N})=35.1\sim43.6\text{mg/L}$, $\rho(\text{TN})=36.3\sim44.1\text{mg/L}$, the maximum concentration of effluent COD, $\text{NH}_4^+-\text{N}$, $\text{TN}$ is $18.8\text{mg/L}$, $0.35\text{mg/L}$ and $14.2\text{mg/L}$, the average removal rate is $91.1\%$, $99.5\%$ and $66.6\%$, SND rate is $62.7\%$$\sim68.9\%$. The effluent COD and $\text{NH}_4^+-\text{N}$ meet the GB3838-2002 class III standard limit value requirements, and the TN concentration of the effluent meets the requirements of GB 18918-2002 Grade A ($\rho(\text{TN})\leq15\text{mg/L}$).

(2) In the range of $4.17\text{h} \leq \text{HRT} \leq 5\text{h}$, the change of HRT basically does not affect the treatment effect. The concentration of COD and $\text{NH}_4^+-\text{N}$ in the effluent of the reactor have reached the requirements of GB3838-2002 Class III standard, and the TN concentration of the effluent has reached GB 18918-2002 Level A requirements, SND rate reached $65.7\%$.

(3) Under the conditions of $\text{HRT}=4.17\text{h}$, $\rho(\text{DO})_C1=1.5\sim2\text{mg/L}$, $\rho(\text{DO})_C2=1\sim1.5\text{mg/L}$, adding sodium acetate to C2 can further optimize the SND effect. Considering the cost of reagents and the overall removal effect of pollutants, the optimal dosage is $40\text{mg/L}$, and the corresponding effluent COD, $\text{NH}_4^+-\text{N}$ and TN concentrations are $16.5\text{mg/L}$, $0.63\text{mg/L}$ and $9.62\text{mg/L}$, TN respectively. The removal rate and SND rate were $75.4\%$ and $75.8\%$, respectively.

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