Effect of Micro-electrolysis and Micro-nano Bubbles Coupled with Peroxymonosulfate Treatment of Rural Domestic Sewage

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Abstract With the continuous deepening of rural revitalization strategy and the increasingly strict sewage discharge standards, rural domestic sewage treatment technology is facing higher challenges and requirements. The combined process of micro-electrolysis + micro-nano bubbles coupled with peroxymonosulfate was constructed in this study, and the treatment effect and application value of this technology were explored with the actual rural domestic sewage as the treatment object. The experimental results showed that under the conditions of HRT of 120 min, PMS dosage of 0.15 mmol/L, pH = 7, MBs air intake of 15 ml/min, current intensity of 15 A, and Fe/C mass ratio of 1 : 1, the removal rates of COD, ammonia nitrogen and total phosphorus can reach 88.55%, 77.18% and 74.67%, respectively. Under the condition that the pH value of sewage was not adjusted, the non-biochemical simultaneous decarbonization, denitrification and phosphorus removal of rural domestic sewage can be achieved by micro-electrolysis and micro-nano bubbles coupled with peroxymonosulfate. The concentrations of effluent COD, ammonia nitrogen and total phosphorus met the requirements of the first level standard of the *Discharge Standard of Water Pollutants for Rural Domestic Sewage Treatment Facilities* (DB45T2413 – 2021). And the comprehensive operating cost was about 1.15 yuan/m^3.
Key words Integrated equipment; Rural domestic sewage; Micro-nano bubbles; Peroxymonosulfate; Micro-electrolysis
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The *Five-year Action Plan for Improving Rural Living Environment* (2021 – 2025) points out the need to accelerate the treatment of rural domestic sewage. The *Proposal of the Central Committee of the Communist Party of China on Formulating the* 14^{th} *Five-year Plan for National Economic and Social Development and the Long-range Objectives for* 2035 proposes to promote rural sewage treatment and improve rural living environment according to local conditions[1]. Therefore, rural domestic sewage treatment has become a key investment area of China and a new growth point for the environmental protection industry.

For a long time, the treatment of rural domestic sewage in China has mainly adopted treatment technologies from Japan and European and American countries. This does not match the characteristics of rural domestic sewage in China, such as small and scattered discharge scale, low pollutant concentration, large daily variation coefficient, and intermittent discharge. There are generally problems such as large land occupation, high operation and maintenance costs, and management difficulties. Therefore, it is of great practical significance to research and develop treatment technologies that are stable and easy to manage, have simple processes and high treatment efficiency, occupy less land, and save operating costs for rural domestic sewage in China.

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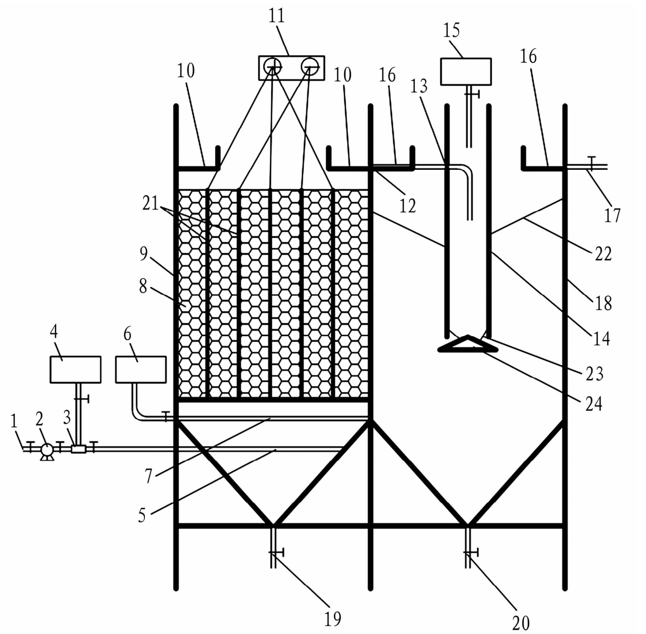
This study constructed a non-biochemical synchronous decarbonization, denitrification, and phosphorus removal sewage treatment system, and formed a new rural sewage treatment technology that is land saving, has a high reaction rate, a simple process, high efficiency, less sludge volume, and low operation and maintenance costs and difficulties, and does not require adjusting the pH value of sewage.

1 Experimental apparatus and method

1.1 Experimental apparatus The experimental apparatus includes a peroxymonosulfate dosing box, micro-nano bubble generator, reaction tower, micro-electrolysis structure, PAM dosing box, and sludge – water separation tower. The lower end of the reaction tower is equipped with a sewage distribution pipe and a micro-nano bubble release pipe, and the micro-nano bubble release pipe is located above the sewage distribution pipe. The peroxymonosulfate dosing box is connected to the sewage distribution pipe, and the micro-nano bubble generator is connected to the micro-nano bubble release pipe. The micro-electrolysis structure is set in the reaction tower and located above the micro-nano bubble release pipe, and the micro-electrolysis structure is connected to the low-voltage DC charger. The outlet at the upper end of the reaction tower is connected to the inlet of the sludge – water separation tower, and the PAM dosing box is connected to the sludge – water separation tower. The structure of the experimental device is shown in Fig. 1.



Note: 1. Inlet of sewage lift pump; 2. Sewage lift pump; 3. Pipeline mixer; 4. Peroxymonosulfate dosing box; 5. Sewage distribution pipe; 6. Micro-nano bubble generator; 7. Micro-nano bubble release pipe; 8. Micro-electrolysis structure; 9. Reaction tower; 10. Overflow weir for reaction tower effluent; 11. Low voltage DC charger; 12. Reaction tower outlet; 13. Water inlet pipe of sludge – water separation tower; 14. Central vertical pipe; 15. PAM dosing box; 16. Overflow weir for effluent from sludge – water separation tower; 17. sludge – water separation tower outlet; 18. sludge – water separation tower; 19. Slag discharge outlet; 20. Sludge discharge outlet; 21. Electrode plate; 22. Central vertical pipe support; 23. Reflective plate support; 24. Reflective board.

Fig. 1 Structure of the test device

1.2 Test method

1.2.1 Operation mode. Peroxymonosulfate was added to the pipeline mixer on the inlet pipeline of the device, and the mixed solution continuously entered the reaction system. After treated by the micro-electrolysis layer, the sewage entered the sludge – water separation system, and the clean water was discharged from the upper part. This experiment was dynamic, with samples taken every 2 h, and the supernatant was analyzed and tested.

1.2.2 Experimental water quality. The experimental sewage was rural domestic sewage treated by septic tanks, with the following inlet water quality: COD≤300 mg/L, ammonia nitrogen≤35 mg/L, total phosphorus≤4 mg/L, pH of 6.0 – 9.0. The effluent quality standard adopted the first level standard of the *Discharge Standard of Water Pollutants for Rural Domestic Sewage Treatment Facilities* (DB45T2413 – 2021)[2], and the effluent quality was as follows: COD≤60 mg/L, ammonia nitrogen≤8 mg/L, total phosphorus≤1.5 mg/L, pH of 6.0 – 9.0.

1.2.3 Water quality testing methods. The concentration of ammonia nitrogen was determined by Nessler's reagent spectrophotometry (HJ 535 – 2009), COD_{Cr} was determined by dichromate method (HJ 828 – 2017), total phosphorus concentration was determined by ammonium molybdate spectrophotometry (GB 11893 – 89)[3], and the pH was measured using a glass electrode method.

1.2.4 Evaluation method. The calculation formula for pollutant removal rate was as follows:

$$\eta = \frac{c_{in} - c_{out}}{c_{in}} \times 100\%$$

where η is removal rate of pollutants, %; C_{in} is mass concentration of pollutants in the inflow, mg/L; C_{out} is mass concentration of pollutants in effluent, mg/L.

2 Results and analysis

2.1 Influence of hydraulic retention time on COD removal efficiency When the pH of the wastewater was 7, the PMS dosage was 0.15 mmol/L, MBs intake volume was 15 ml/min, the current intensity was 15 A, and iron – carbon mass ratio was 1 : 1, the variation of COD removal rate with hydraulic retention time (HRT) was shown in Fig. 2. As shown in Fig. 2, the COD removal rate rose with the increase of HRT. The COD removal rate can reach 85.98% at 120 min of HRT, but the increase in COD removal rate was not significant with the increase of HRT thereafter. This indicated that the optimal value for HRT was 120 min.

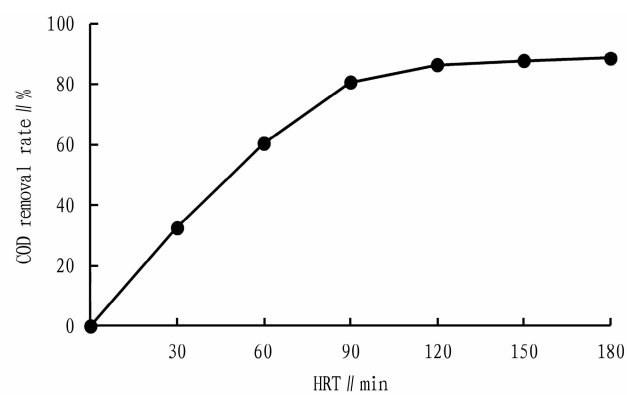


Fig. 2 Effect of HRT on COD removal

2.2 Influence of iron – carbon ratio on COD removal efficiency When the pH of the wastewater was 7, the PMS dosage was 0.15 mmol/L, MBs intake volume was 15 ml/min, the current intensity was 15 A, and HRT was 120 min, the variation of COD removal rate with iron – carbon ratio was shown in Table 1. From Table 1, it can be seen that the COD removal efficiency decreased with the increase of iron – carbon mass ratio, indicating that the optimal iron – carbon mass ratio was 1 : 1.

Table 1 Influence of iron – carbon mass ratio on COD removal effect

No.	Iron – carbon mass ratio	COD removal rate // %
1	1 : 1	85.79
2	2 : 1	81.23
3	3 : 1	75.19
4	4 : 1	66.52

2.3 Influence of PMS dosage on COD removal efficiency When the pH of the wastewater was 7, the MBs intake was 15 ml/min, the current intensity was 15 A, iron – carbon mass ratio was 1 : 1, and HRT was 120 min, the variation of COD removal rate with PMS dosage was shown in Fig. 3. As shown in Fig. 3, the COD removal rate rose with the increase of PMS dosage. When the PMS dosage was 0.15 mol/L, the COD removal rate can reach 85.98%. However, with the increase of PMS dosage, the COD removal rate did not increase significantly. This indicated that the optimal dosage of PMS was 0.15 mol/L.

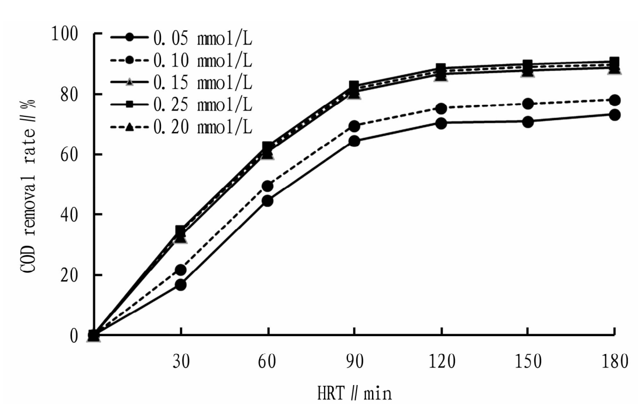


Fig. 3 Influence of PMS dosage on COD removal effect

2.4 Influence of MBs intake volume on COD removal efficiency When the pH of the wastewater was 7, the PMS dosage was 0.15 mmol/L, the current intensity was 15 A, the iron-carbon mass ratio was 1 : 1, and HRT was 120 min, the variation of COD removal rate with MBs intake was shown in Fig. 4. From Fig. 4, it can be seen that the COD removal rate showed a trend of first increasing and then decreasing with the increase of MBs intake[4]. When the intake volume of MBs increased from 5 to 15 ml/min, the COD removal rate gradually increased and reached the highest value at 15 ml/min of MBs intake, which was 86.23%. As the intake volume of MBs increased from 15 to 25 ml/min, the COD removal rate decreased to 83.32%. This indicated that the optimal intake volume for MBs was 15 ml/min.

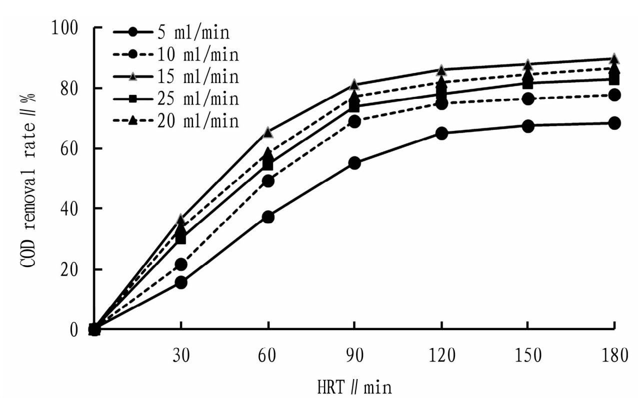


Fig. 4 Influence of MBs intake volume on COD removal effect

2.5 Influence of current intensity on COD removal efficiency When the pH of the wastewater was 7, the PMS dosage was 0.15 mmol/L, MBs intake volume was 15 ml/min, the iron-carbon mass ratio was 1 : 1, and HRT was 120 min, the variation of COD removal rate with current intensity was shown in Fig. 5. As shown in Fig. 5, the COD removal rate rose continuously as the current intensity increased from 5 to 15 A, while the COD removal rate decreased when the current intensity increased from 15 to 25 A[5]. This indicated that the optimal value for current intensity was 15 A.

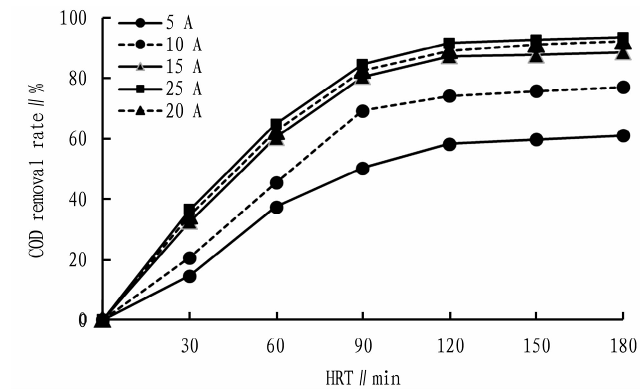


Fig. 5 Influence of current intensity on COD removal effect

2.6 Influence of initial pH on COD removal efficiency When the current intensity was 15 A, the PMS dosage was 0.15 mmol/L, the MBs intake volume was 15 ml/min, the iron-carbon mass ratio was 1 : 1, and HRT was 120 min, the variation of COD removal rate with the initial pH of wastewater was shown in Fig. 6. From Fig. 6, it can be seen that as the initial pH increased from 3 to 7, the COD removal rate continuously increased, while as the initial pH increased from 7 to 11, the COD removal rate actually decreased. This indicated that the optimal initial pH value was 7.

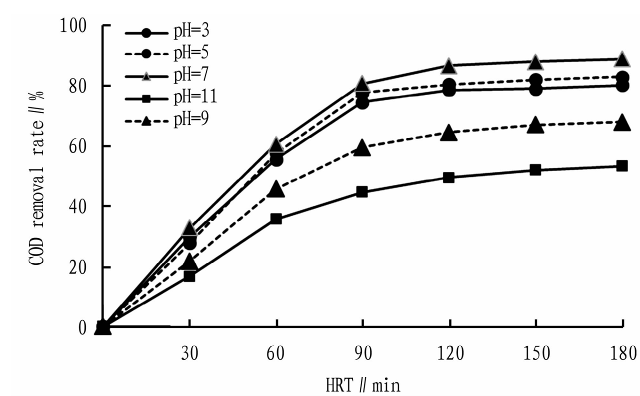


Fig. 6 Influence of initial pH on COD removal effect

2.7 Removal rate of ammonia nitrogen under the optimal condition When the pH of the wastewater was 7, the PMS dosage was 0.15 mmol/L, the MBs intake volume was 15 ml/min, the current intensity was 15 A, and the iron-carbon mass ratio was 1 : 1, the variation of ammonia nitrogen removal rate was shown in Fig. 7. From Fig. 7, it can be seen that when HRT was 30 min, the removal rate of ammonia nitrogen in sewage was 26.68%; with the increase of HRT, the removal rate of ammonia nitrogen in sewage rapidly increased. When HRT was 120 min, the removal rate of ammonia nitrogen reached 76.57%. This demonstrated the importance of HRT in improving the efficiency of ammonia nitrogen removal.

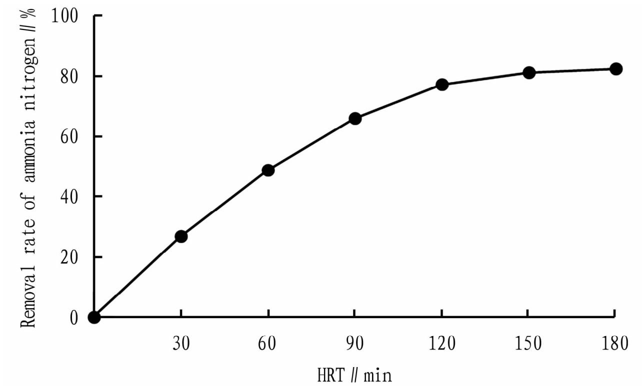


Fig. 7 Removal rate of ammonia nitrogen under the optimal condition

2.8 Removal rate of total phosphorus under the optimal condition When the pH of the wastewater was 7, the PMS dosage was 0.15 mmol/L, the MBs intake volume was 15 ml/min, the current intensity was 15 A, and the iron – carbon mass ratio was 1 : 1, the changes in total phosphorus removal rate were shown in Fig. 8. From Fig. 8, it can be seen that when the HRT was 30 min, the total phosphorus removal rate in the wastewater was 25.68%. However, with the increase of HRT, the removal rate of total phosphorus showed a significant upward trend. When HRT was extended to 120 min, the removal rate of total phosphorus increased significantly to 73.58%, indicating the importance of HRT in improving the efficiency of total phosphorus removal.

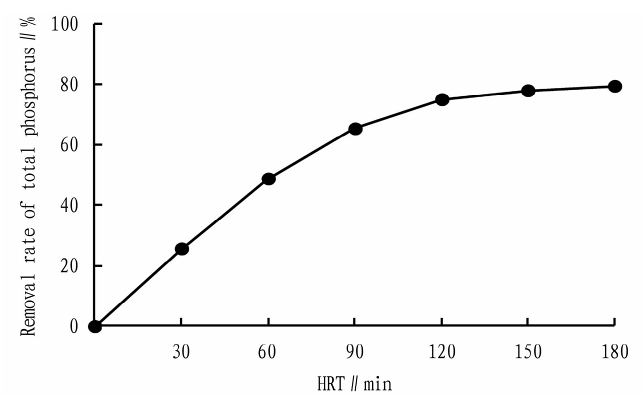


Fig. 8 Removal rate of total phosphorus under the optimal condition

2.9 Economic analysis According to the experiment, the treatment of rural domestic sewage by the micro-electrolysis + micro-nano bubbles coupled with peroxymonosulfate can achieve water quality standards. According to the experimental estimation, the main operating costs included the power consumption of the sewage lift pump, micro-nano bubble generator, and micro-electrolysis DC charger, which was approximately 0.6 kW · H/m^3. Calculated by 0.5 yuan/(kW · h) of electricity price, the electricity fee was 0.3 yuan/m^3. The dosage of reagent PMS was 0.1 kg/m^3. Calculated at 8 yuan/kg, the chemical cost was 0.8 yuan/m^3. Loss rate of micro-electrolysis packing was 25 g/m^3. Calculated at 2 yuan/kg, the consumables cost was 0.05 yuan/m^3. Therefore, the comprehensive operating cost was 1.15 yuan/m^3.

3 Conclusions

(1) When HRT was 120 min, the PMS dosage was 0.15 mmol/L, pH = 7, MBs intake volume was 15 ml/min, the current intensity was 15 A, and the iron – carbon mass ratio was 1 : 1, the removal rates of COD, ammonia nitrogen, and total phosphorus can reach 85.79%, 76.57%, and 73.58%, respectively.

(2) Without adjusting the pH value of the sewage, the micro-electrolysis + micro-nano bubbles coupled with peroxymonosulfate can achieve non-biochemical synchronous decarbonization, denitrification, and phosphorus removal of rural domestic sewage. The concentrations of COD, ammonia nitrogen, and total phosphorus in the effluent met the requirements of the first-level standard of the *Discharge Standard of Water Pollutants for Rural Domestic Sewage Treatment Facilities* (DB45T2413 – 2021), and the comprehensive operating cost was about 1.15 yuan/m^3.

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(From page 53)

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