### Study on biological filler-coupled biological process for phosphorus removal

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### ABSTRACT

Based on the self-made new iron–carbon micro-electrolytic packing, a composite packing was developed, coupled with a membrane bioreactor, and the simulation of low C/N urban sewage was used as the research object. Use anaerobic aerobic bioreactor, adopt intermittent cultivation, and then continuous cultivation, control hydraulic retention time (HRT) to 3, 4, 5, 6, and 7 h by adjusting influent, PH of influent is 5.5, 6.0, 6.5, 7.0, 7.5, and 8.0. The high-efficiency start-up effect and optimized phosphorus removal characteristics of the composite-filled bioreactor were investigated. The results show that the intermittent start-up and continuous cultivation method can reduce the start-up time to 30 d, the film hanging effect is good; when the water inlet HRT = 7, PH =  $7.5 \pm 0.1$ , the total phosphorus removal rate reached 90.1%; biological phase observation, and scanning electron microscopy chart showed that iron–carbon micro-electrolytic materials have no segregation and passivation; comparison test it is proved that the composite filler reactor removes total phosphorus removal, and the composite filler physical and chemical phosphorus removal and biological phosphorus removal.

Keywords: Composite filler; Phosphorus removal; Iron–carbon micro-electrolysis; Urban sewage; Bioreactor

### 1. Introduction

With the acceleration of urbanization, the discharge of domestic sewage is increasing, and the eutrophication of water bodies is exacerbated by the excessive phosphorus content in domestic sewage [1]. Phosphorus removal in urban domestic sewage is currently an urgent problem to be solved. In recent years, iron–carbon micro-electrolysis technology has a wide range of applications in the treatment of urban domestic sewage, but a large number of studies show that there are still many problems with this technology: traditional micro-electrolytic fillers have iron–carbon separation, are difficult to recycle, and have low processing efficiency; the surface of iron chips will form after a period of operation A layer of metal oxide and hydroxide film passivates iron filings, causing the micro-electrolysis process

to be interrupted [2,3]. At present, most studies use iron filings and activated carbon particles mixed to fill column reactors, such as SUN, etc. [4] adopting a sufficient mixture of iron filings and coke as a micro-electrolytic filler to treat industrial wastewater. During 50 d of treatment, the filler was severely crusted, and the subsequent treatment effect was significantly reduced. In order to efficiently remove phosphorus in urban domestic sewage, the traditional ironcarbon micro electrolyze the existing problems of phosphorus, and use a homemade iron-carbon micro-electrolytic filler, natural zeolite, and columnar activated carbon in a certain volume ratio to form a composite filler. Widely used membrane bioreactor [5] to achieve high-efficiency phosphorus removal. Some studies have shown that [6] based on the iron-carbon micro-electrolysis technology coupled with biological assimilation, which exerts the advantages

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of internal electrochemical physicochemical reactions and promotes biochemical effects. It is suitable for low C/N ratio wastewater treatment for phosphorus removal.

In order to solve the problem that the iron-carbon filler is easy to compact in the micro-electrolysis reaction [7,8], and achieve the purpose of efficient phosphorus removal, research, and use the iron-carbon micro-electrolytic filler [9] independently developed by this research group to develop a composite filler, coupled with sewage dephosphorization bioreactor, established a high-efficiency phosphorus removal system combined with biological methods, and physical and chemical effects The research object is low C/N ratio domestic sewage. The influence of different factors on the treatment effect of composite filler wastewater phosphorus removal reactor was investigated, and the best experiment was determined. Conditions; analysis of surface conditions after filler reaction through observation of biological phases and scanning electron microscopy (SEM) diagrams; discussion of the mechanism of phosphorus removal to provide theoretical and technical support for sewage phosphorus removal.

### 2. Experimental methods and materials

### 2.1. Experimental equipment and methods

### 2.1.1. Experimental setup

As shown in Fig. 1, the reactor system is composed of an anaerobic biological filter and an aerobic biological filter in series. The reactor is based on a composite filler (iron–carbon micro-electrolytic filler, natural zeolite, and biological activated carbon) as the core. Anaerobic technology integration makes the sewage always in an aerobic and anaerobic alternating environment. Both the anaerobic biological filter and the aerobic biological filter use the bottom water inlet method, and the raw water enters the anaerobic water from the bottom inlet through the peristaltic pump. In the aerobic biological filter, it flows out from the anaerobic biological filter water outlet under the action of gravity, flows from the bottom inlet into the aerobic biological filter, and the aerobic biological filter effluent returns to the anaerobic biological filter. The bottom of the tank is equipped with an aerator, which is used lo aerate and increase oxygen. The two filters are made of plexiglass and the effective volumes of the anaerobic and aerobic biological filers are 4.7 and 2.8 L, respectively. The accumulation volume of the composite filter material filled in the reactor system is about 70%–80% of the effective volume of the reactor. The effective volumes of the anaerobic and aerobic biological filters after being filled with composite filters are 2.0 and 1.0 L, respectively.

### 2.1.2. Composite filler

In the experiment, the homemade iron–carbon microelectrolytic filler natural zeolite and columnar activated carbon were mixed in a certain volume ratio to form a composite filler. The iron–carbon micro-electrolytic filler, natural zeolite, and activated carbon in the composite filler of the anaerobic and aerobic biological filter were 2:1:1, 1:1:1.

The self-made iron–carbon micro-electrolytic filler [10] is prepared by mixing certain iron powder and powdered activated carbon, and adding a small amount of binder and catalyst. As shown in Fig. 2, the iron–carbon micro-electrolytic filler is spherical and has a particle size of 10–20 mm. Iron filings account for 85%–90%, powdered activated carbon accounts for 10%–15%, and the volume ratio of iron to carbon is 1:2. Before use the filler is immersed in a 5% sulfuric acid solution for 1 h, and then 5% diluted hydrogen is used. Soak the sodium oxide solution for 1 h.

### 2.1.3. Experimental water

The experimental water is configured with simulated sewage, and the simulated sewage water quality indicators COD Cr,  $NH_4$ –N,  $NO_1$ –N, and TP are prepared with glucose, ammonium chloride, potassium nitrate, and potassium dihydrogen phosphate, respectively. In order to ensure the needs of microorganisms for the growth and reproduction of microelements, 1 L of experimental water is added with 1 mL of nutrient solution [11]. The substances and



Fig. 1. Composite packed reactor (1) peristaltic pump, (2) liquid flow meter, (3) support layer, (4) composite filter material, (5) water outlet valve, (6) gas flow meter, and (7) oxygen pump.



Fig. 2. Iron-carbon micro-electrolytic filler.

concentrations are shown in Table 1.The test drugs are of analytical grade.

### 2.2. Detection method

For the measurement methods of the main water quality indicators involved in the test, refer to "water and wastewater monitoring and analysis methods" (Fourth Edition) [12]. The specific water quality indicators and analysis methods are shown in Table 2.

### 2.3. Start-up of composite packing reactor

This experiment uses a batch culture followed by a continuous culture method. During continuous culture, the flow rate is gradually increased to the design flow rate 0.4 L/h. Due to the low content of organic matter in the water, in order to increase the film hanging speed, additional carbon sources are added to the culture medium used to rapidly acclimate the denitrifying organisms. Membrane,

Table 1 Nutrient solution formula

Reagent name	Concentration (g/L)
FeCl <sub>3</sub> ·6H <sub>2</sub> O	1.5
H <sub>3</sub> BO <sub>3</sub>	0.15
CuSO <sub>4</sub> ·5H <sub>2</sub> O	0.03
KI	0.18
MnCl <sub>2</sub> ·4H <sub>2</sub> O	0.12
$ZnSO_4 \cdot 7H_2O$	0.12
Na <sub>2</sub> MoO <sub>4</sub> ·2H <sub>2</sub> O	0.06
CoCl <sub>2</sub> ·6H <sub>2</sub> O	1.61
MgSO <sub>4</sub> ·7H <sub>2</sub> O	1.9
EDTA	10

Table 2

Main water quality indicators and detection methods

shorten the membrane hanging period. In the intermittent
aeration stage, a certain amount of glucose is directly sup-
plemented to the inlet water, and in the continuous aeration
stage, the C/N [13,14] of the inlet water is gradually reduced
by adjusting the amount of glucose added to adapt to low
C/N ratio water quality. The start-up process is broken
down into three stages, and the time node arrangement is
shown in Table 3.

In the intermittent water-feeding cultivation stage, the anaerobic biological filter adopts the circulating membrane method. After mixing 500 mL of inoculated activated sludge and the culture solution, the peristaltic pump is used to drive the activated sludge from the water inlet at the bottom of the anaerobic reactor. The composite filter was completely submerged. Adjust the inlet water flow rate of the peristaltic pump to 0.5 L/h, drive the culture solution into the anaerobic reactor, and the effluent enters the culture solution container. Prepare the same concentration of culture solution every day to circulate the membrane of the anaerobic reactor. Style cycle.

The aerobic biological filter adopts the inoculation membrane method. First, 500 mL of pre-cultured activated sludge is injected into the water inlet of the bottom of the aerobic biological filter with a peristaltic pump, and then the culture liquid is replenished from the water inlet of the bottom of the filter to the top. At the water outlet, turn on the aeration pump for suffocation, and adjust the aeration volume to make the dissolved oxygen in the aerobic biological filter between 4.0 and 5.0 mg/L. After 24 h of continuous aeration, the aeration was stopped, the supernatant was drained after the reactor was left to stand, and then the same volume of fresh culture medium was added from the water inlet at the bottom of the reactor to continue the sulking for 24 h. After repeating the operation for 5 d, the aerobic organism was removed. Filter media.

On the first day of the continuous water intake phase, adjust the water inlet flow rate to 0.2 L/h, and then increase the flow rate by 0.1 L/h every 3 d. After 6 d, the flow rate

Water quality index	Detection method
Total nitrogen (TN)	Potassium persulfate oxidation-ultraviolet spectrophotometry
Total phosphorus (TP)	Digestion of potassium persulfate-molybdenum antimony anti-spectrophotometry
Total iron (TFe)	Atomic absorption spectrophotometry (AAS)
PH	PH meter
Dissolved oxygen (DO)	Portable dissolved oxygen analyzer
Temperature ( <i>T</i> )	Portable dissolved oxygen analyzer

Table 3

Node arrangement of biological filter startup time

Stage	Training method	Duration (d)
Stage 1	Intermittent feed culture	5
Stage 2	Continuous water inflow without nitrate reflux	10
Stage 3	Continuous water inflow with nitrate reflux	15

reaches the design flow rate of 0.4 L/h, keeping the water inlet flow rate constant and stable for 4 d. Starting from the 16th day, the aerobic filter nitrate effluent was returned to the anaerobic biological filter, and the reflux ratio is 50%, and then the reflux ratio is increased by 50% every 4 d, and the reflux ratio is 150% after 8 d, keeping the reflux ratio unchanged., keeping the reflux ratio constant and operating stably for 7 d. Control the inlet pH =  $7.2 \pm 0.2$ , solved oxygen in aerobic filter DO =  $3.0 \pm 0.2$  mg/L, hydraulic retention time (HRT) of anaerobic filter, and aerobic filter is 3 h.

### 3. Results and discussion

## 3.1. Phosphorus removal characteristics of composite filler-coupled biological method during startup

The analysis of the change of total phosphorus in intermittent water incubation is not significant, so the total phosphorus concentration is monitored from the continuous water inflow phase. The change of the total phosphorus removal rule on the composite packed reactor system in the start-up phase is shown in Fig. 3.

It can be seen that the total phosphorus concentration in the inlet water during the start-up phase is basically about 2 mg/L. The anaerobic filter and aerobic filter effluent have a relatively stable treatment effect in the continuous inlet culture phase, and the overall phosphorus removal rate does not change much. The total phosphorus concentration in the effluent was stable at about 0.1-0.25 mg/L, and the total phosphorus removal rate was as high as 90%. The total phosphorus concentration in the effluent was 0.11 mg/L on the 1st day of the continuous incubation stage, that is, the 5th day of the start-up phase. The concentration is gradually increasing. Analysis of the reasons, in addition to the anaerobic release of aerobic phosphorus by phosphorus accumulating bacteria and biological assimilation to remove phosphorus, the porous surface of natural zeolite, activated carbon, and iron-carbon filler has a certain adsorption of total phosphorus; The adsorption effect is the best on the 1st day of the start-up phase. When the adsorption reaches a certain saturation state, the adsorption



Fig. 3. Effect of total phosphorus removal by the system during startup.

amount will decrease sharply, and the microorganisms with phosphorus removal function in the system have not fully proliferated. The effluent from the 2nd to 5th days does not show a low total phosphorus concentration.

According to the theory of denitrifying polyphosphate bacteria aerobic phosphorus absorption and anaerobic phosphorus release [15], biological phosphorus removal should take place in the aerobic phase, but this experimental data is exactly the opposite of this conclusion. There are two main reasons for the analysis: first, the filter process different from activated sludge method, TP can be removed by sludge removal, and only a small part of the shed biofilm is removed, so the total phosphorus removal rate in the aerobic section is very low, on the other hand, the COD in the anaerobic section is higher, and the under oxygen conditions, acidification is easy to produce free hydrogen ions, so that zero-valent Fe in the internal electrolytic filer becomes Fe<sup>2-</sup> and a small amount of Fe<sup>3+</sup>. These iron ions and free phosphate form inorganic precipitates and are fixed in the biofilm or the biofilm exits the reactor system, so the anaerobic filter maintains a high removal rate of total phosphorus. In the anaerobic stage, the surface of the iron-carbon filler, especially the iron surface, is prone to microbial growth. In addition, it can create a lower redox potential (ORP) environment, which provides a more suitable growth condition for the phosphorus-removing bacteria, which is beneficial to improve the phosphorus-removing bacteria biomass and biological activity, improve phosphorus removal efficiency [16].

# 3.2. Influencing factors of composite filler coupled biological phosphorus removal

### 3.2.1. Influence of hydraulic retention time on phosphorus removal

Control the operating temperature of  $25^{\circ}$ C– $30^{\circ}$ C using constant temperature water bath(HHS–11-2), the reflux ratio of the nitrating solution is 100%, the dissolved oxygen of the aerobic filter is  $3 \pm 0.1$  mg/L, the ratio of the hydraulic retention time of the anaerobic aerobic filter is 1:1, and the inlet water pH =  $7.5 \pm 0.1$  The total HRT was controlled by adjusting the inlet flow rate to be 3, 4, 5, 6, and 7 h, with a period of 5 d. The effect of phosphorus removal efficiency of the anaerobic and aerobic biological filter system under different HRT was studied. The experimental results are shown in Fig. 4.

It can be seen from Fig. 4 that the total phosphorus concentration in the inlet water is basically maintained at about 1 mg/L. With the increase of HRT, the average total phosphorus concentration in AF effluent decreases from 0.67 to 0.27 mg/L. The concentration decreased from 0.43 to 0.12 mg/L, and the total phosphorus removal rate gradually increased and then stabilized with the extension of HRT. When HRT = 7, the total phosphorus removal efficiency reached 90% and remained stable.

The removal effect of aerobic biological phosphorus removal on total phosphorus is limited, and the increase of the biological phosphorus removal effect with the extension of HRT is much smaller than the physicochemical-biochemical phosphorus removal of the anaerobic



Fig. 4. Influence of HRT on TP removal effect.

filter. The reason for the analysis is mainly that for anaerobic biological filters, increasing HRT is beneficial to the anaerobic acid-producing reaction of organic matter. The hydrogen ions produced promote the zero-valent iron hydrogen production in the iron-carbon filler, which is also the process of alkali production. The iron ions released by the iron–carbon filler can react with free phosphate to form a precipitate in a neutral environment; meanwhile, the deoxidation of the iron-carbon filler creates a good environment for the anaerobic phosphorus release of polyphosphate bacteria and promotes the biological phosphorus removal; The aerobic biological filter mainly removes total phosphorus through the assimilation of microorganisms and aerobic phosphorus absorption by polyphosphate bacteria. Prolonging HRT is also beneficial to the removal of total phosphorus, but the aerobic biological filter has a limited removal effect on total phosphorus. With prolongation, the improvement of the biochemical phosphorus removal effect is much smaller than the physicochemical-biochemical coupling phosphorus removal of the anaerobic filter.

#### 3.2.2. Effect of pH of incoming water on phosphorus removal

Control operating temperature  $25^{\circ}C-30^{\circ}C$  using constant temperature water bath(HHS–11–2), the reflux ratio of the nitrating solution is 100%, aerobic filter dissolved oxygen 3 ± 0.1 mg/L, anaerobic aerobic filter hydraulic retention time ratio 1:1, adjust inlet water pH to 5.5, 6.0, 6.5, 7.0, 7.5, and 8.0, the TP removal characteristics of the composite packed reactor system under different pH conditions were studied, and the experimental results are shown in Fig. 5.

It can be seen from Fig. 5 that the effluent pH of the anaerobic filter and aerobic filter increases with the increase of the pH of the inlet water. When the pH of the inlet water is 5.5, 6.0, 6.5, 7.0, 7.5, and 8.0, the anaerobic. The average pH of the effluent of the filter and aerobic filter is 6.3, 6.6, 6.8, 6.9, 7.1, 7.5, and 6.8, 7.2, 7.3, 7.2, 7.3, and 7.6. At 0.12, 0.15, 0.19, 0.25, 0.28, and 0.35 mg/L, and 0.08, 0.1, 0.14, 0.18, 0.22, and 0.26 mg 1 L, the average TP concentration in the effluent of the anaerobic and aerobic filters is continuously increasing, and the pH is the effect of the phosphorus removal effect of the composite packing reactor system is significant,



Fig. 5. Effect of pH of inlet water on TP and pH of outlet water.

and the increase in pH is not conducive to the physical and chemical removal of phosphorus. Therefore, when the inlet water pH = 7–8, the TP of AF effluent water rises sharply, but the appropriate increase in pH is beneficial to enhance biological aerated filter (BAF). The activity of microorganisms and the effect of assimilation and phosphorus removal also make it possible to maintain the TP concentration of effluent water below 0.25mg 1 L when the TP concentration of BAF inlet water is continuously increasing when pH = 7–8.

The reasons for the analysis are mainly as follows: the pH of the feed water directly affects the rate of galvanic reaction of the iron-carbon filler, which determines the reaction effect of the precipitated iron ions and phosphate; on the other hand, the biochemical reaction generally depends on the enzyme inside the cell Promote the reaction, if the pH is too high or too low, it will lead to the decomposition of the key groups of the enzyme protein and even the degeneration and inactivation of the enzyme protein; when the pH gradually increases, the micro-electrolysis of the ironcarbon filler weakens, and the release of iron ions reduces the precipitation with phosphate; when the pH of the feed water is increased to about 7-8, the average TP concentration of anaerobic effluent water rises rapidly high, but alkaline water can supplement the alkalinity consumed by nitrification in aerobic filters. At the same time, most microorganisms are suitable for growing in a slightly alkaline environment to enhance biological activity and biological nitrogen and phosphorus removal.

### 4. Discussion on total phosphorus removal mechanism

#### 4.1. Observation of biological phase after film hanging

After the film hanging, the biofilm flocs attached to the surface of the filter material in the anaerobic biological filter and the aerobic biological filter were taken, respectively, and observed with a microscope. It was observed that the surface morphology of the three filter materials was basically the same, among which the iron–carbon micro-electrolytic filler. The micrograph is shown in Fig. 6.

The color of the anaerobic biofilm is black, which may be caused by the adsorption of green ferrous ions on the biofilm, which proves to some extent the analysis of the TP removal mechanism of the anaerobic section. The biofilm floc is odorous, the sludge floc is dense, and the microorganisms mostly exist in the form of bacterial micelles. The biofilm mainly contains microorganisms such as phyllophyte, paramecium, and a few filamentous bacteria.

The biofilm on the surface of the filter material in the aerobic biological filter is reddish-brown, the biological community is densely distributed and the layers are complex. At the same time, there are black granular materials inside the bacteria, and the ordinary activated sludge has a loose tan appearance. Therefore, the aerobic biofilm may absorb the iron element on the surface of the filler, so it has a better flocculent shape. Structure and sludge settling. The surface of microbial cells is mainly composed of proteins, lipids, and the like. Trivalent iron ions can combine with functional groups such as hydroxyl and amino groups in these components to reduce the electron repulsion between cells and have a good flocculation effect on the organism. A large number of metazoans are found in aerobic biofilms,



Fig. 6. Microfilm of the surface of the electrolytic filler in the anaerobic filter (a) micrograph (100 times) and (b) micrograph (400 times).



Fig. 7. Microfilm of the surface of the electrolytic filler in the aerobic filter (a) micrograph (400 times) and (b) micrograph (1,000 times).

especially fixed bell-worm, as shown in Figs. 7a and b, indicating that the biofilms are growing well.

# 4.2. Observation of microscopic morphology of filler surface after film hanging

The S-520 (Hitachi, Japan) type SEM was used to test this new type of composite phosphorus removal filler. The SEM results of the electrolytic filler in the reactor after hanging the film are shown in Figs. 8 and 9.

It can be seen from Fig. 8a that there are pores of different sizes on the surface and inside of the iron–carbon micro-electrolytic filler, the surface is rough, the specific surface area is large, and no obvious passivation phenomenon is found. From Fig. 8c, iron can be found, there are many corynebacterium and filamentous bacteria on the surface of the carbon micro-electrolytic filler. The filamentous bacteria adhere to the filler and can grow in large numbers to form a network structure. It provides a suitable place for the attachment and growth of anaerobíc microorganisms and hydrophilícity that facilitates the growth of microorganisms. Layer, while avoiding sludge swelling caused by filamentous bacteria. From Fig. 9a, it can be seen that the biofilm on the surface of the iron–carbon micro-electrolytic

filler in the aerobic filter fills the gap, and aerobic microorganisms are more likely to fill the gap. Surface attachment.

The surface and the inside of the iron–carbon microelectrolytic material are filled with many pores, providing a stable growth environment for microorganisms. The surface of the filler is rough, the specific surface area is large, and the ability to capture microorganisms is strong. The large specific surface area enables the filler to carry high concentrations of organisms. the amount.

## 4.3. Validation of coupled biochemical phosphorus removal mechanism of composite filler

Take out about 200 mL of composite filter material from the stable operation stage, add anaerobic effluent to the 500 m mark, and let it stand for 24 h. Filter 100 mL filtrate (counted as 1#) for later use. After sonicating the remaining mixed solution for 1 h, then filter to obtain 100 mL filtrate (counted as 2#). Add about 0.5 mL of 10% dilute hydrochloric acid to the remaining mixed solution and react. After 30 min, 100 mL of filtrate was counted (counted as 3#). The TP and total iron content in 1#, 2#, and 3# filtrates were measured respectively. The measurement results are shown in Fig. 10.



Fig. 8. SEM image of iron–carbon micro-electrolytic filler after membrane attachment of anaerobic filter (a) SEM image (500 times), (b) SEM image (1,000 times), and (c) SEM image (2,000 times).



Fig. 9. SEM image of iron–carbon micro-electrolytic filler after aerobic filter hanging. (a) SEM image (5,000 times) and (b) SEM image (10,000 times).



Fig. 10. Comparison of TP and TFe content after different treatment methods.

It can be seen from 10 that the content of TP and TFe in the filtrate # 1, that is, the filtrate without any treatment, is 0.63 and 0.93 mg/L. The contents of TP and TFe in filtrate # 2, that is, the filtrate after ultrasonic treatment, were 1.17 and 1.15 mg/L, respectively, with a slight increase. The filtrate # 3, namely, the measured TP and TFe of the filtrate after acidification with hydrochloric acid. The contents were 9.02 and 14.36 mg/L, and both of them increased significantly. The ratio of TP and TFe content increase after acidification is  $\Delta$ (TP)/ $\Delta$ (TFe) = 0.594, which is very close to the molar mass ratio of P to Fe in FePO<sub>4</sub> 0.554. It can be inferred that the anaerobic filter can remove total phosphorus. Mainly by chemical precipitation, biological phosphorus removal has a low contribution to the phosphorus removal effect.

The analysis principle is that iron ions (Fe<sup>2+</sup>, Fe<sup>3+</sup>) produced by internal electrolysis can react with phosphates, and colloids produced by iron ion hydrolysis can adsorb and coagulate phosphate, and play a role in chemical phosphorus removal. In addition, iron ions It has the effect of stimulating the growth of microorganisms, and the effect of micro-current generated by micro-electrolysis can also stimulate the activity of denitrifying polyphosphate bacteria, which has promoted chemical and biological phosphorus removal to a certain extent. Synergistic biological phosphorus removal removes total phosphorus and strengthens the removal of total phosphorus.

### 5. Conclusion

- The phosphorus removal effect of the composite filler sewage phosphorus removal reactor is obvious at the start-up stage, and the reaction rate is stable in the later stage, which indicates that the iron–carbon microelectricity has not interrupted the phosphorus removal reaction, and no passivation and segregation has occurred.
- With the increase of HRT, the removal rate of total phosphorus gradually increased and then stabilized, and reached stability when HRT = 7. The removal effect of aerobic biological phosphorus removal on total phosphorus was limited, and the improvement of biochemical phosphorus removal effect with the extension of HRT was much smaller than that of physicochemical and biochemical coupling phosphorus removal in anaerobic filters.
- The pH obvious effect on the total phosphorus removal performance, the rise of pH does not favor the physical and chemical phosphorus removal for, so when the water pH = 7–8, AF TP water has risen sharply, but the moderate pH increase to strengthen BAF microbial activity and assimilation of phosphorus removal effect, also makes when pH = 7–8, in the case of BAF TP concentration increased the water, its water TP concentration can remain below 0.25 mg/L, composite filler in bioreactor optimum pH = 7.5 ± 0.1.
- The homemade iron carbon micro electrolysis filler used adhesive skeleton as filler and reaction after micro electrolysis ferroelectric material surface SEM Fig. show that iron–carbon micro electrolysis material surface and internal pore size, surface is rough, specific surface area

is big, does not appear the iron filings harden passivation problem. Among them, the anaerobic filter mainly by iron ion and phosphate radical reaction, producing chemical precipitation andiron ion hydrolysis colloidal of phosphoric acid root adsorption coagulation effect on total phosphorus removal, biological phosphorus removal contribution to phosphorus removal effect is very low.

### References

- L. Zhang, W. Cao, Y. Ma, C. Han, Y. Qin, Y. Zhao, Z. Liu, C. Yang, Distribution and potential eutrophication of nitrogen and phosphorus in tidal reach and nearest Estuary of Daliao River, J. Environ. Sci., 37 (2016) 1677–1684.
- [2] H.Y. Zhao, Y.P. Ye, Y.J. Wen, Z.C. Zhang, Experimental study on treatment of high-concentration plasticizer wastewater by dynamic micro-electrolysis-Fenton reagent method, Ind.Water Treat., 28 (2008) 31–34.
- [3] W.C. Zou, S.X. Zhang, Z.B. Luan, Y. Mu, D.L. Zou, Preparation of micro-electrolysis structured packing for wastewater treatment, Environ. Prot. Chem. Ind., 28 (2008) 546–548.
- [4] S. Liang, W. Can, J. Ji, K. Xin, Treatment of mixed chemical wastewater and the agglomeration mechanism via an internal electrolysis filter, Chem. Eng. J., 215–216 (2013) 50–56.
- [5] R. Li, J. Cui, X. Li, X. Li, Phosphorus removal and recovery from wastewater using Fe-dosing bioreactor and cofermentation: investigation by X-ray absorption near-edge structure spectroscopy, Environ. Sci. Technol., 52 (2018) 14119–14128.
- [6] Z. Qi, D.S. Li, S.H. Deng, X. Yang, S.B. Zhu, Physicochemicalbiological coupling deep denitrification based on iron-carbon internal electrolysis, Water Treat. Technol., 42 (2016) 92–96+101.

- [7] W. Zou, S. Zhang, Z. Luan, Y. Mu, D. Zou, Preparation of microelectrolyte regularized packing for wastewater treatment, Environ. Prot. Chem. Ind., 28 (2008) 546–548.
- [8] D. Zou, M. Li, H. Zou, D. Fan, M. Sun, Experiments on the treatment of benzene-contaminated groundwater by a new type of iron-carbon micro-electrolytic filler, J. Jilin Univ., 40 (2010) 1441–1445.
- [9] Tongji University, Preparation of Iron-Carbon Micro-Electrolysis Filler, China, 2009 (accessed May 12, 2010).
- [10] L. Pan, J. Wang, H. Dong, A Powder Metallurgy Iron-Based Catalytic Internal Electrolytic Filler and Its Preparation Method, CN105712443A (accessed June 29, 2016).
- [11] W. Xing, W. Zhang, D. Li, J. Li, F. Jia, Y. Cui, F. Ren, An integrated O/A two-stage packed-bed reactor (INT-PBR) for total nitrogen removal from low organic carbon wastewater, Chem. Eng. J., 328 (2017) 894–903.
- [12] General Administration of Environmental Protection, Water and Wastewater Monitoring and Analysis Methods, 4th ed., China Environmental Science Press, Beijing, 2002.
- [13] J. Li, K. Cui, K. Wang, F. Tian, R. Meng, Study on the rapid cultivation of activated sludge in low C/N sewage treatment by A/O device, Water Treat. Technol., 43 (2017) 103–105.
- [14] W. Yin, Y. Chen, Y. Zhang, L. Xu, P. Wu, W. Liu, Start-up and stable operation of full autotrophic denitrification coupled with denitrification and phosphorus removal, Environ. Sci., 40 (2019) 5032–5039.
- [15] Y.Y. Wang, Y.Z. Peng, S.Y. Wang, Y.Z. Li, M.L. Pan, Denitrification and phosphorus removal theory, technology and influencing factors, China Water Wastewater, 19 (2003) 33–36.
- [16] X. Liang, L. Ma, Z. Zhang, Characteristics analysis of surface adhesion layer under the condition of catalytic iron and biological coupling process, China Environ. Sci., 35 (2015) 1343–1350.