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Effect of phosphorus removing agents on sludge activity and floc structure in saline wastewater treatment

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ABSTRACT

The cyclic activated sludge technology process was used to treat high-salinity mustard tuber wastewater and municipal wastewater. The effect of phosphorus removing agents on sludge activity and floc structure was investigated. Results indicated that polysilicate ferric, ferric chloride and aluminum sulfate improved the structure and density of sludge flocs. These three phosphorus removing agents had little effect on oxygen uptake rate (OUR) of heterotrophic bacteria and nitrifying bacteria and sludge dehydrogenase activity. The effect of aluminum sulfate on OUR of nitrite bacteria was significantly stronger than polysilicate ferric and ferric chloride. In the stable operation of the high-salt mustard wastewater and the urban sewage co-processing system of condition. The salinity was 5 g/L and the influent total phosphorus (TP) concentration was 7.3–8.7 mg/L, the effluent TP concentration was 2.1–3.6 mg/L in the biological phosphorus removal system. Results showed that the polysilicate ferric, ferric chloride and aluminum sulfate can enhance nitrogen removal from high-salinity mustard tuber wastewater. Biological and chemical phosphorus removal models were established, which used polysilicate ferric, ferric chloride and aluminum sulfate as the phosphorus removal agents, respectively. Phosphorus removal agents was conducive to further reduce the chemical oxygen demand and suspended solids values in the effluent from the treatment system, and the sequence of the enhanced effect was polysilicate ferric > ferric chloride > aluminum sulfate.

Keywords: Saline wastewater; Sludge activity; Phosphorus removing agent; Floc structure

1. Introduction

Fuling mustard tuber, one of the pillar industry in the Three Gorges reservoir area, generates a large amount of pickling and elutriation wastewater. Such effluent from mustard tuber pickling plant is characterized by high organic load, high level of nitrogen and high salinity. The direct discharge of such wastewater without prior treatment is known to be a great threat to the water environment in Three Gorges reservoir region [1]. It has been observed that high salinity can strongly inhibit the aerobic biological treatment of wastewater. Due to the high salt concentration (>10 g NaCl L^{-1}) cause plasmolysis and significant reduction of cell activity, because of its high concentration and complex composition, industrial saline wastewater is more difficult to deal with. Degradation of total or specific organic substances was the main purpose of most studies concerning nutrient removal from industrial saline wastewater [1].

The bio/chemical synergistic dephosphorization technology was established on the process proposed by biological phosphorus removal, it was difficult to achieve the first-level discharge requirement standard of total

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phosphorus content. However, the phosphorus removing agents would have a certain impact on the biological treatment system. The cyclic activated sludge system (CASS) can obtain a high pollutant removal efficiency in the low carbon source domestic wastewater in small scale wastewater treatment plants (WWTPs), which is a variant of the sequencing batch reactor system, combining a biological selector and a fed-batch reactor. Overall, the CASS has a simple, repeated, time-based sequence which incorporates fill-aeration, settlement and decanting in a basic cycle. Because of configuration flexibility, operational simplicity and a more economical footprint, the process has been widely used in urban sewage and various industrial WWTPs, especially for smaller facilities in China [2].

Phosphorus removing agents include three distinct classes, such as synthetic organic polymeric flocculants, inorganic coagulants and composite flocculants. Traditional flocculants, such as polyaluminum chloride or its polymer, are the most widely used inorganic coagulants in water treatment plants. Because it has some advantage characteristics, such as low toxicity and low price. Whereas, it has shortcomings, such as the formation of small flocs, high dosages, low removal efficiencies and easily affected by water quality [3]. In order to avoid the shortcoming, the aluminum sulfate, ferric chloride and polysilicate ferric were chosen in this study.

Fe^{2+} competed with phosphate bacteria for phosphate inhibition and inhibited biological phosphorus removal [4,5]. Complexing chemical reagents were used to remove P from wastewater, such as synthesize nanoparticles, incinerated sewage sludge ash and ethylenediaminetetraacetic acid, which can enhance P release significantly from FePs, AlPs and biosolids during the mesophilic anaerobic fermentation. The high salinity could cause inhibiting the aerobic biological treatments of wastewater. Also, the high salt concentration is more than 10.0 g/L NaCl leads to plasmolysis and organism metabolism of organism reduction during the wastewater treatment process. It was proved that the application of activated sludge and the use of salt-tolerant organism in biological treatment are possible during the wastewater treatment process. Previous studies showed that the Fe^{3+} and Al^{3+} can increase the respiration rate of activated sludge by using bio/chemical synergistic dephosphorization technology [5–7]. It was found that the effect of Al^{3+} was stronger than that of Fe^{3+} and Al^{3+}. The respiratory rate of both bacteria and heterotrophic bacteria was inhibited, while the inhibition of nitrifying bacteria was not obvious. Result indicated that the aluminum salt was added into the aerated biological filter, it can enhance phosphorus removal and has little effect on the type and quantity of microorganisms [8–12]. Previous studies have shown that the application of polysilicate ferric as a coagulant can make the sludge floc become denser and improve the sedimentation performance [13–19]. Based on the effect of phosphorus removing agents on activated sludge in biological treatment system, the author used bio-chemical enhanced phosphorus removal method to study the effect of dephosphorization agent on sludge activity and floc structure.

2. Materials and methods

2.1. Test device

The test device is shown in Fig. 1. The effective volume of the CASS reactor was 0.5 m^3, and the effective volume ratio of the bioselective zone to the main reaction zone of the reactor was 1:5. The biological selection zone used an underwater agitator to thoroughly mix the influent and the reflux nitrifying solution in the main reaction zone. The main reaction zone used an aeration pump to supply oxygen through the microporous membrane aeration disk, and the nitrifying liquid was returned to the biological selection zone, the bottom of the separator connecting the biological selection zone and the main reaction zone was opened.

2.2. Test methods

2.2.1. Operating conditions

Four CASS reactors with an effective volume of 0.5 m^3 were used for the test. The inoculated sludge was used for the stable operation of the high-salt mustard wastewater and the urban sewage co-processing system. By adjusting the mixing ratio of the high-salt mustard wastewater to control the salinity of the co-processing system to 5 g/L. The CASS reactor has a cycle running time of 8 h, a drainage ratio of 40%, a reflux ratio of 150%, a DO of 2–3 mg/L in the main reaction zone, a DO of less than 0.5 mg/L in the biological selection zone, and the mixed liquid suspended solids (MLSS) was maintained at around 3,000 mg/L.

2.2.2. Phosphorus removing agents and dosage method

Aluminum sulfate, ferric chloride and polysilicate ferric were used as chemical reagents enhanced phosphorus

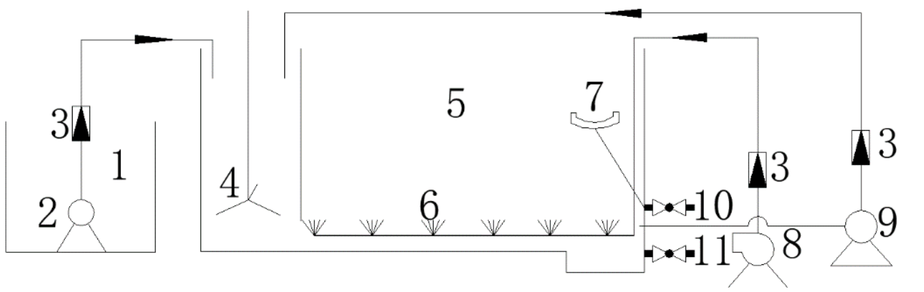

Fig. 1. Flowchart of CASS process. 1, Regulation pool; 2, inlet pump; 3, flow meter; 4, blender; 5, CASS reactor; 6, aeration head; 7, decanter; 8, aeration pump; 9, return pump; 10, drain pipe; 11, mud tubes.

removal. 0.5 h in front of the aeration stop time, aluminum sulfate, ferric chloride and polysilicate ferric were added to the main reaction zones of the three reactors, respectively. In another group of reactors, no phosphorus removing agents was added as a control group. The total influent phosphorus of the system was 7.3–8.7 mg/L. The system effluent TP was stabilized to the first-class B emission standard by adding three kinds of phosphorus removing agents, such as aluminum sulfate, ferric chloride and polysilicate ferric [10,11,20,21]. The effect of three phosphorus removing agents on the floc structure, oxygen consumption rate and dehydrogenase activity of activated sludge was studied [19,22].

2.2.3. Analytical methods

The properties of sewage and sludge, oxygen uptake rate (OUR), dehydrogenase activity, TP, chemical oxygen demand (COD), suspended solids (SS) were determined using the spectrophotometer according to standard method [23]. Dissolved oxygen (DO) were measured by a DO meter (HACH, HQ30D, USA), pH was measured by a pH meter (HACH, Sension2, USA).

3. Results and discussion

3.1. Effect of phosphorus removing agents on activated sludge floc structure

Fig. 2 shows the electron microscopic scanning of activated sludge flocs after the phosphorus removing agents were not added (control) and the polysilicate ferric, ferric chloride and aluminum sulfate were added, respectively.

It can be seen from Fig. 2 that the structure of the activated sludge flocs was loose when phosphorus removing agents was not added, and there was no obvious boundary between the floc particles, because the activated sludge particles had a large amount of extracellular polymeric substance (EPS), EPS combines with each other and forms larger biological aggregates. By adding the extracellular polymeric substance and ferric chloride, the affinity of Fe^{3+} and EPS hydrolysis was stronger than other cations, where Fe^{3+} replaces other cations from EPS by ion exchange [24]. Also, the Fe^{3+} have high valence, which could make Fe^{3+} combine with more negatively charged groups [1,25]. Therefore, the Fe^{3+} can improve the structure and density of the activated sludge floc, and the boundary between the

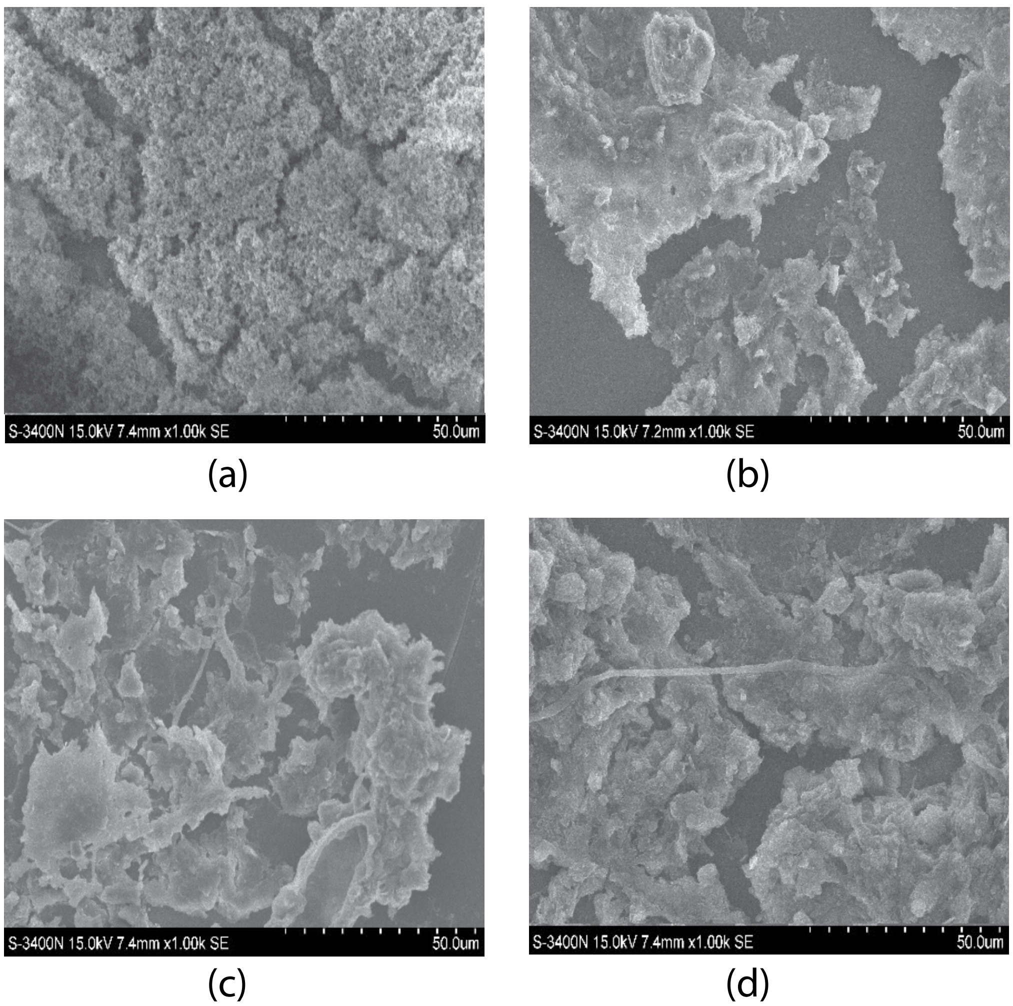

Fig. 2. Scanning electron microscope photos of sludge floc (×1,000). (a) Control, (b) added polysilicate ferric, (c) added aluminum sulfate and (d) added ferric chloride.

activated sludge flocs also become obvious, the boundaries activated sludge flocs effect when adding aluminum sulfate was less than the former. In addition, the activated sludge floc particles were large when the polyferric silicate and ferric chloride were added in the wastewater process. It was considered that the polysilicate ferric had both the adsorption effect of the nano-sized silicon particles and the bridging effect of polymeric iron ions, which was stronger than the bridging effect of the separate polymeric iron ions in ferric chloride [7,8,26,27].

3.2. Effect of phosphorus removing agents on sludge activity

The effect of phosphorus removing agents on sludge activity was mainly caused by metal ions in the phosphorus removing agents. The presence of metal ions can cause toxic effects on microorganisms, which lead to weakening of metabolic function. Also, it seriously affected the stable operation of sewage biological treatment system. The effects of three phosphorus removing agents on activated sludge OUR and dehydrogenase activity were investigated [13,14].

3.2.1. Impact on oxygen uptake rate

In the experiment, the activity of nitrifying bacteria was inhibited by $NaClO_3$, and the effects of three phosphorus removing agents on OUR of nitrite bacteria, allyl group thiourea solution inhibits the activity of nitrite bacteria, and analyzed the effects of three phosphorus removing agents on OUR of nitrifying bacteria and heterotrophic bacteria, the allylthiourea and $NaClO_3$ inhibited the activity of nitrifying bacteria and nitrite bacteria, and analyzed the effects of three phosphorus removing agents on OUR of heterotrophic bacteria [25]. The OUR values of heterotrophic bacteria, nitrifying bacteria and nitrite bacteria when added three kinds of phosphorus removing agents and not adding phosphorus removing agents are shown in Table 1.

It can be seen from Table 1 that the addition of iron salt had little effect on the OUR of heterotrophic bacteria, nitrifying bacteria and nitrite bacteria. The addition of aluminum salt had a certain inhibitory effect on the activities of nitrifying bacteria and heterotrophic bacteria, but not obvious, however, the inhibitory effect on the activity of nitrite bacteria was stronger. Therefore, the addition of aluminum salt had a certain influence on the OUR of the activated sludge in the system, but would not damage the stable operation of the system.

3.2.2. Effect of phosphorus removing agents on dehydrogenase activity

The dehydrogenase activities of the activated sludge were 3.60, 3.54, 3.49 and 3.15 μg TF/(mg MLSS h), respectively, when the dephosphorization agent was not added and the polyferric silicate, ferric chloride and aluminum sulfate were, respectively, used as the dephosphorization agent. It indicated that ferric chloride as the dephosphorization agent had little effect on the sludge activity, while the use of aluminum sulfate as the dephosphorization agent, the dehydrogenase activity of the activated sludge was reduced, but not significantly.

3.3. Biological phosphorus removal

In the control group, when the salinity is 5.0 g/L salinity, the COD, TP and SS were detected of effluent from CASS process for three cycles a day. The results showed that when the influent COD, TP and SS were 352–397, 7.3–8.7 and 163–195 mg/L and the corresponding mean values were 371, 8.1 and 177 mg/L, respectively, the effluent COD, TP and SS were 42–53, 2.1–3.6 and 14–19 mg/L, with the mean values of 48, 3.2 and 17 mg/L, respectively. The rates were 84.6%–88.7% (average 86.5%), 52.6%–73.7% (average 62.5%) and 88.2%–91.3% (average 89.7%), respectively. It was known that the biological phosphorus removal efficiency was limited, and the effluent TP did not meet the discharge standard under the condition of 5 g/L salinity.

3.4. Biological/chemical enhanced phosphorus removal model

In order to control the phosphorus removal agent dosage, phosphorus removal agent was added into the reactor, which is dependent on the ratio of metal ions to total phosphorus, the different dosage ratio beta = n (M^{3+})/n (P) (M^{3+} represents trivalent metal ions, n represents the amount of substances) on total phosphorus. Results indicated in Fig. 3.

As can be seen in Fig. 3, it can be seen that the phosphorus removal effect of the three dephosphorization agents was in order: polysilicate ferric > ferric chloride > aluminum sulfate. Polysilicate ferric has the best phosphorus removal effect, and the TP concentration of effluent decreases with the increase of the dosage ratio. When ferric chloride, aluminum sulfate and polysilicate ferric were added, respectively, the total phosphorus concentration (y) of effluent satisfies the following relationship with the dosage ratio (x): $y = 5.466e^{-2.38x}$ ($R^2 = 0.998$), $y = 5.539e^{-2.00x}$ ($R^2 = 0.990$), $y = 5.060e^{-3.06x}$ ($R^2 = 0.977$).

When polysilicate ferric, ferric chloride and aluminum sulfate were used as phosphorus removal agents, respectively, the biological–chemical phosphorus removal model showed that if the effluent TP < 1.5 mg/L, the dosage ratio was 0.4, 0.6 and 0.8, respectively, the corresponding concentration of metal ions were 5.85, 8.78 and 5.64 mg/L, respectively; if the effluent TP < 1.0 mg/L, the dosage ratio was 0.6, 0.8 and 1.0, respectively, the corresponding concentration of metal ions were 8.78, 11.70 and 7.05 mg/L, respectively.

The biological phosphorus removal from the control reactor showed that the average concentration of total phosphorus in the influent of the treatment system was 8.1 mg/L, and the average concentration of total phosphorus in the effluent was 3.2 mg/L. If the effluent is TP < 1.5 mg/L, the investment ratio can be converted to the ratio of metal ions to total removal phosphorus, the corresponding ratios of 0.4, 0.6 and 0.8 should be 1.9, 2.8 and 3.8, respectively. Therefore, the actual dosage of phosphorus removal agent is much larger than the theoretical calculation. The main reasons are as follows: the pH value is 7.8–8.6 in the terminal reactor. In addition to the formation of phosphate precipitation, side reactions occur to form metal hydroxide precipitation. Hence, the dosage ratio of phosphorus removal agent is much higher than that of theoretical calculation. The mixing conditions of chemical reactions are not good, and the

Table 1
Effect of phosphorus removing agents on OUR of bacteria (mg O_2/L min)

Bacteria	Control	Ferric chloride	Polysilicate ferric	Aluminum sulfate
Heterotrophic bacteria	0.690	0.668	0.653	0.616
Nitrifying bacteria	0.260	0.254	0.247	0.208
Nitrite bacteria	0.144	0.138	0.135	0.94

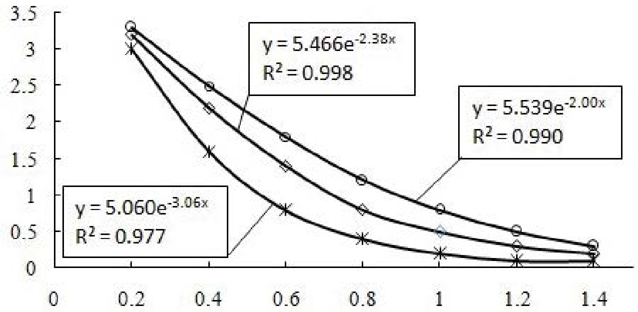

Fig. 3. Removal efficiencies of TP by different phosphorus removal agents.

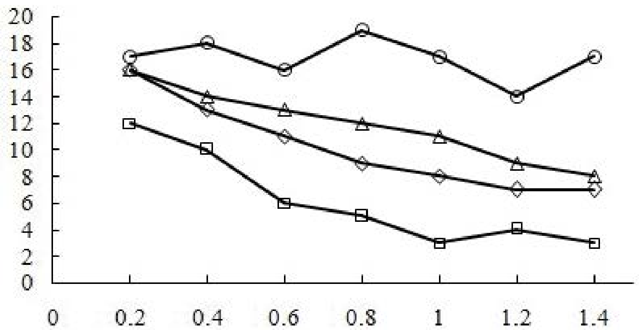

Fig. 4. Influence of phosphorus removal agents on effluent SS.

dosage increases because of the insufficient mixing degree of the reagent added directly into the reactor.

3.5. Effect of phosphorus removal agent on effluent quality of treatment system

3.5.1. Effect of phosphorus removal agent on chemical oxygen demand of effluent

The COD value of effluent with phosphorus removal agent was lower than that of bioreactor, and the COD concentration of effluent decreased gradually with the increasing phosphorus removal agent. When the dosage was less than 1.5 mg/L, the COD value of the effluent treated with polysilicate ferric, ferric chloride and aluminum sulfate reactor decreased by 25, 18 and 16 mg/L, respectively. It is believed that the phosphorus removal agent can rapidly hydrolyze and form long-line polynuclear hydroxyl complex after adding the reactor. These complexes can absorb a large number of colloids and particles in water, thus further reducing the organic matter content in effluent. Therefore, the COD value of effluent was lower than that of effluent from simple bioreactor. When the dosage of phosphorus removal agent meets the TP requirement of effluent, the dosage of phosphorus removal agent will increase, and the COD value of effluent will rapidly decrease and a large number of carbon sources would be consumed, which was not conducive to denitrification in anaerobic stage. In addition, polymerized ferric silicate is more effective in reducing COD in effluent because polymerized ferric silicate fully exerts the adsorption of nano-silica particles and bridging effect of polymerized cations.

3.5.2. Effect of phosphorus removal agent on suspended solids of the effluent

The effluent SS values of different dephosphorization agents at different dosage ratios were compared with those of the effluent of the simple bioreactor as shown in Fig. 4. It can be seen that the SS value of effluent after adding dephosphorization agent was lower than that of biological reactor, and with the increase of dosage ratio, the SS value of effluent gradually decreases. The results showed that the surface chargeability of bacterial micelles and suspended particles or the distribution of colloidal particles and the potential distribution of colloidal particles were changed by exchanging hydrogen ions between metal ions and hydrolysates after adding phosphorus removal agent to the reactor. The double layer of the colloidal particles and the ζ potential of the colloidal particles are reduced, and the colloidal destabilization, flocculation and sedimentation are caused by the adsorption bridging effect, so that the SS content in the effluent was further reduced. It can also be seen from Fig. 4 that polysilicate ferric was more effective in reducing SS in effluent. When the dosage meets the requirement of total phosphorus in effluent, SS can maintain less than 5 mg/L. Because the polysilicate ferric gives full play to the adsorption of nano-silica particles and bridging effect of polymeric cations. The SS and colloidal substances in the sewage are adsorbed quickly and the effluent SS concentration decreases.

4. Conclusion

When the salinity of hypersalt mustard tuber wastewater and municipal wastewater synergistic treatment system was 5 g/L, the bio-chemical enhanced phosphorus removal efficiency was studied by using chemical phosphorus removal agent and the effect of phosphorus removal agent on effluent quality was analyzed. The following conclusions were drawn:

- When the influent total phosphorus was 7.3–8.7 mg/L, the effluent total phosphorus of biological phosphorus removal was 2.1–3.6 mg/L, and ferric chloride, aluminum sulfate and polysilicate ferric were used as phosphorus removal agents for bio-chemical enhanced phosphorus removal. The following phosphorus removal models were obtained: $y = 5.466e^{-2.38x}$ ($R^2 = 0.998$), $y = 5.539e^{-2.00x}$ ($R^2 = 0.990$), $y = 0.560e^{-3.06x}$ ($R^2 = 0.977$).

- The addition of phosphorus removal agent can further reduce the COD and SS concentration in the effluent of the system. The effect was in turn: polysilicate ferric > ferric chloride > aluminum sulfate.
- In the influent salinity of 5 g/L, total phosphorus was 7.3–8.7 mg/L, used bio-chemical enhanced phosphorus removal method to control the effluent TP concentration to reach the first-class B emission standard.
- The addition of polysilicate ferric, ferric chloride and aluminum sulfate improved the floc structure of the sludge and improved the compactness of the sludge flocs.
- Three agents had little effect on the OUR of heterotrophic bacteria and nitrifying bacteria and sludge dehydrogenase activities, but the effect of aluminum sulfate on nitrite bacteria OUR was significantly stronger than that of polysilicate ferric and ferric chloride.

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