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Utilization of micro-electrolysis, up-flow anaerobic sludge bed, anoxic/oxic-activated sludge process, and biological aerated filter in penicillin G wastewater treatment

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ABSTRACT

System which included micro-electrolysis (ME), up-flow anaerobic sludge bed (UASB), anoxic/oxic (A/O) activated sludge process and biological aerated filter (BAF) were investigated in penicillin G-processing wastewater treatment. The main quality of the wastewater was shown as follows: chemical oxygen demand (CODcr) of 1,4507–1,5280 mg L^{-1}, ammonium nitrogen (NH_3–N) of 598–826 mg L^{-1}, suspended solid of 1,850–2,190 mg L^{-1}, and pH of 3.5–5.2. Moreover, penicillin G residue in the wastewater was 130–150 mg L^{-1}. ME was utilized as the pretreatment method, UASB and A/O process were designed for the secondary treatment, and BAF was used as the advanced treatment. The results revealed that biodegradability of the wastewater could be effectively improved by ME, and most of CODcr and NH_3–N in the wastewater could be removed by UASB and A/O process, and the residual COD and NH_3–N could be removed by BAF. The average COD and NH_3–N in final effluent was about 275.3 and 19.8 mg L^{-1}, respectively, and the treated wastewater quality reached the requirement of the national discharge standards (wastewater quality standards for discharging into municipal sewer pipelines, China, COD ≤ 300 mg L^{-1}, NH_3–N ≤ 25 mg L^{-1} in C standard). Especially, about 80% of penicillin G residue in the wastewater could be removed or converted by ME reactor, and the wastewater could be effectively deposed in this system.

Keywords: Penicillin G wastewater; Biodegradability; Micro-electrolysis; UASB; BAF; System methods

1. Introduction

As the pharmaceutical industry developed rapidly, antibiotics have become important pharmaceutical drugs due to their high consumption rates in both veterinary and human medicine [1]. Meanwhile, a large amount of wastewater was generated from the antibiotics production, and the wastewater treatment has attracted more and more attention due to the high difficulty in its treatment [2]. As a kind of typical

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antibiotics, penicillin G has been widely used in curing bacterial infection and restraining pathogenic micro-organism infection [3,4]. Much attention has been paid to the penicillin G-processing wastewater treatment by enterprises and experts.

It is well known that penicillin G is an important beta-lactam antibiotic which is with great difficultly oxidized and degraded, and the processing wastewater has complex components, high concentrated organics, strong chromaticity, and pungent smell [5–7]. Therefore, the penicillin G-processing wastewater has become one kind of industrial wastewater which was difficultly disposed of. In recent years, methods for the penicillin G-processing wastewater treatment have mainly focused on physic-chemical methods and biochemical processes [8,9]. The physic-chemical methods are mainly advanced oxidation processes, such as fenton oxidation, photocatalysis oxidation, electrochemical oxidation, and ozonation, etc. [10–12]. Although these physic-chemical methods could obtain ideal results in the penicillin G-processing wastewater treatment, due to the high investment and operating cost, these methods were impossible to be applied in practical wastewater treatment project. The biochemical process utilized in the penicillin G-processing wastewater treatment was mainly focused on aerobic biological treatment, such as sequencing batch reactor-activated sludge process and membrane bioreactor [13,14]. However, the aerobic biological treatment could not get ideal treatment effect. On one hand, low organic loading rate (OLR) of the aerobic biological treatment process highly decreased the removal effect of the organics, especially in the high-concentration, non-biodegradable wastewater treatment [15–17]; on the other hand, the aerobic biological process could be strongly restrained due to high concentration inorganic salt and drug residue. For all the above reasons, it was significant to develop economical and effective methods for the penicillin G-processing wastewater treatment.

By now, few effective technologies have been found in the penicillin G-processing wastewater treatment. In this paper, a complete treatment system including micro-electrolysis (ME), up-flow anaerobic sludge bed (UASB), anoxic/oxic (A/O) process, and biological aerated filter (BAF) were investigated for the penicillin G-processing wastewater treatment. (1) ME, as one kind of physical–chemical methods, has been widely used in refractory wastewater treatment [18] such as pharmacy wastewater, petrochemical wastewater, dye wastewater, electroplating industry wastewater, and pesticide wastewater [19–24]. ME could destroy the structure of organic pollutants and enhance the ratio of BOD_5 (biochemical oxygen demand) to COD (BOD_5/COD) [18,25,26]. (2) UASB belongs to anaerobic biological treatment method, as a successful low investment technology [27] it could be utilized to decompose highly concentrated organic wastewater, including pharmaceutical wastewater, starch wastewater, beer wastewater, and papermaking wastewater [28–31]. (3) As one kind of stable aerobic biological processes, A/O method not only degraded organic pollutants, but also effectively removed nitrogen and phosphorus [32]. (4) BAF, which was developed from biological filters in Europe during the late 1980s, has been regarded as an effective advanced treatment technology. It was characterized by less sludge formation, stronger environmental shock resistance, and a smaller size of reactor [33,34] and it has been widely applied in secondary and tertiary wastewater treatments [35] including textile wastewater, petrochemical wastewater, slaughterhouse wastewater treatment, and other organic wastewater [36–39].

The reasons for choosing the above treatment system in treating the penicillin G-processing wastewater were as follows. Firstly, ME was utilized as the pretreatment method which was used to lower the bio-toxicity and enhance the biodegradability of the penicillin G-processing wastewater. Secondly, UASB, A/O, and BAF were used as biochemical processes to remove organic pollutants and nitrogen. Thirdly, the possibility of ME, UASB, A/O, and BAF as a complete system in treating the penicillin G-processing wastewater was investigated.

2. Materials and methods

2.1. Wastewater qualification

In this study, the practical penicillin G-processing wastewater was obtained from a penicillin G manufacturing enterprise in Hulun Buir (Inner Mongolia Autonomous Region, China). The wastewater was mainly generated from fermentation process and refining process and the components of raw wastewater are shown in Table 1. The penicillin G residue was 130–150 mg L^{-1}, and it has the characteristics of high biotoxicity and low B/C ratio (0.106–0.125). Therefore, it was necessary to apply effective pretreatment method to lower down the biotoxicity and enhance biodegradability.

2.2. Flow chart of the treatment system

The whole treatment system for the target wastewater was composed of three different portions. Coagulation and ME were utilized as pretreatment methods for the wastewater, the raw wastewater was

Table 1
The component of the raw penicillin G-processing wastewater

Indicator	Unit	Concentration range
CODcr	mg L^{-1}	14,507–15,280
NH_3–N	mg L^{-1}	598–826
SS	mg L^{-1}	1,850–2,190
SO_4^{2-}	mg L^{-1}	1,382–1,538
pH	–	3.5–5.2
Penicillin G residue	mg L^{-1}	130–150
BOD_5/CODcr	–	0.106–0.125

fed into a tank for coagulation, removing suspended solid (SS) in the wastewater to prevent the stoppage of ME. In order to reduce penicillin G residue and enhance the biodegradability the wastewater, the effluent of coagulation was introduced into the ME reactor. Secondly, secondary treatment units were composed of UASB and A/O, effluent of the ME reactor was collected into a regulating tank to adjust pH, and it was fed into the UASB tower to remove most of the organic pollutants. Then the wastewater flowed into the A/O tank for nitrogen, phosphorus and a portion of the organic pollutants' removal. Thirdly, BAF was used as the advanced treatment unit, effluent of the A/O tank was pumped into BAF reactor, and the main purpose of this procedure was to remove the organic pollutants and nitrogen.

2.3. Starting and running

2.3.1. Pretreatment

A pilot scale experiment was conducted in the wastewater treatment plant of the penicillin G manufacturing enterprise, and the designed scale for the wastewater treatment was 1.0 m^3 d^{-1}. Firstly, coagulation and ME were utilized as pretreatments, calcium chloride was added into the wastewater to remove the portion of SO_4^{2-}, reducing the influence of SO_4^{2-} in the later anaerobic biological treatment stage. Polyaluminum chloride and polyacrylamide were used as coagulant and coagulant aid in the coagulation process, respectively. The reactor for coagulation made from carbon steel had a length of 1.0 m, a width of 1.0 m, and a height of 1.2 m, and the effective volume was 1.0 m^3.

The effluent of coagulation process was collected in a regulating tank made of plastic, and pH of the wastewater was adjusted to about 3.0, which was beneficial for ME reactor [40,41]. Then the wastewater was pumped into the ME reactor with metering pump, the reactor made from stainless steel was composed of an upper cylinder and the lower cone, these two parts were isolated by a filter board. The upper cylinder reactor with a diameter of 1.0 m and a height of 1.5 m was filled with cobblestone and ceramics, cobblestone used as the under supporting layer had a height of 0.3 m, and ferric-carbon ceramics with a media height of 1.0 m was applied as the upper packing layer. Four tray aerators were fixed in the cone reactor to provide air for the ME reaction, and the height of this reactor was 0.86 m. The effective volume of the whole ME reactor was 0.74 m^3, the wastewater was pumped into the reactor from the under inlet in the cone reactor and the effluent was discharged from the upper outlet in the cylinder reactor, the whole reactor was backwashed every week.

2.3.2. Secondary treatment

After the pretreatment of the wastewater, UASB and A/O process were applied as the secondary treatments. The UASB reactor made from carbon steel was inoculated with some granular sludge, which was obtained from the wastewater plant of a pharmaceuticals company, the reactor had a diameter of 0.7 m, a height of 6.0 m, and an effective volume of 1.68 m^3, and the concentration of the sludge in the reactor was about 13.9 g VSS L^{-1}. The OLR in start-up stage was about 1.0 kg m^{-3} d^{-1}, and it was increased gradually until it reached the optimum OLR. Sodium bicarbonate was added to adjust pH of the wastewater and pH was controlled at about 7.0. Temperature of the wastewater was adjusted at about 35.0℃ by adding steam.

The wastewater was discharged into A/O tank after the anaerobic biological treatment, the reactor made from carbon steel was inoculated with some activated sludge, obtained from the same wastewater plant as the UASB reactor, and the concentration of the sludge was about 4.1 g MLSS L^{-1}. The reactor had a length of 4.0 m, a width of 1.0 m, and a height of 1.2 m, and the effective volume was 4.0 m^3, the volume of anoxic tank was one-third of that of the oxic tank's. Sodium bicarbonate was utilized to adjust pH of the wastewater, and pH of anoxic tank and oxic tank were kept at about 7.0 and 8.0, respectively. The operating temperature of the wastewater was maintained at about 30.0℃ by adding steam.

2.3.3. Advanced treatment

When discharged from A/O tank, the wastewater was discharged into a secondary sedimentation tank, and the effluent was then fed into BAF system for the

advanced treatment. The reactor used in BAF system was almost the same as the ME reactor except material quality, and it was made from carbon steel, the BAF system was inoculated with some activated sludge from the oxic tank, and the effluent of the secondary sedimentation tank was pumped into the system for biofilm culturing. The removal rate of CODcr in this system reached stable after about 10 d, meaning the end of the biofilm culturing, then the system entered into stable operation period which had an ideal removal rate of CODcr. Temperature and pH in this system were kept at about 30.0˚C and 8.0, respectively, and the reactor was backwashed every week.

2.4. Analysis methods

The measurement of CODcr, ammonium nitrogen (NH_3–N), SS, SO_4^{2-}, BOD_5, and pH was inducted according to national standard methods (State Environmental Protection Administration of China, 2002a, b), dissolved oxygen (DO) and temperature were monitored with DO meter (HQ 30d53LED™ HACH, made in USA) and thermometer (JR913, made in China), and volatile fatty acid in the UASB tower was measured by titrimetry [42].

Moreover, penicillin G residue in the wastewater was measured by high-performance liquid chromatography–mass spectrometry (LC-2010A, made in Japan). The equipment utilized for penicillin G quantification was manufactured by Shimadzu Corporation in Japan. The injection volume for all experiments was 10 μL and flow rate was set at 1.0 mL/min, the column temperature and determined wavelength were 20.0˚C and 254 nm, respectively. Moreover, the mobile phase in this study consisted of ultra-pure water, acetonitrile, 1 mol L^{-1} monopotassium phosphate solution and 1 mol L^{-1} acetic acid solution.

3. Results and discussion

3.1. Treatment effect of ME

Biodegradability of the penicillin G-processing wastewater was crucial for biological wastewater treatment, therefore, ME reactor was utilized to remove the penicillin G residue and enhance BOD_5/CODcr of the wastewater. Moreover, a portion of CODcr was reduced in this reactor and hydraulic retention time (HRT) was about 6.0 h.

Treated by the ME system, most of the penicillin G residue was removed and BOD_5/CODcr of the wastewater was enhanced (from about 0.1 to about 0.3), meaning that the biodegradability of the wastewater might be greatly improved. About 80% of penicillin G residue in the wastewater was removed or converted, the concentration of penicillin G in the effluent of ME reactor was about 30 mg L^{-1}, the result revealed that the biotoxicity of wastewater was greatly reduced and the negative effect of penicillin G residue on anaerobic treatment was very small. Meanwhile, the influence of ME on NH_3–N of the wastewater is shown in Fig. 1. It could be seen that the concentration of NH_3–N in influent was about 600 and increased to about 1,200 mg L^{-1} in effluent, which indicated that the nitrogen groups of penicillin G might be destroyed by ME [43]. Also, this result revealed that most penicillin G in the wastewater was removed or converted. Moreover, BOD_5/CODcr of the effluent reached about 0.310, which was much higher than that of the influent's (0.106–0.125), the results showed that the biodegradability of the wastewater might be greatly improved by the ME system, it also demonstrated directly that the biotoxicity of the wastewater might be greatly reduced by this ME system, and the pretreatment of the penicillin G-processing wastewater was effective for its later biological treatment.

CODcr-removal effect of the ME system is shown in Fig. 2. It could be seen that a portion of CODcr could be removed by the ME system. In the first 30 d, more than 50% CODcr in the wastewater could be reduced by this system, and the high removal rate might be obtained from the combination effect of ME reaction, absorption of the carbon in the ceramic fillers, and flocculation from ferrous and ferric iron [40]. After 30 d, the removal rate of CODcr was lower than 50%, and it finally came down to around 10%. On the one hand, absorption of the carbon in the ceramic fillers might reach saturation, causing decrease of the removal rate of CODcr from carbon absorption. Whereas on the other hand, flocculation from ferrous and ferric iron was weakened as electrode material in

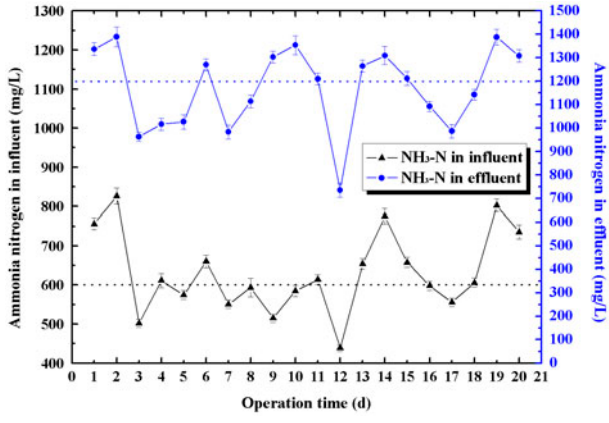

Fig. 1. The influence of ME on NH3–N.

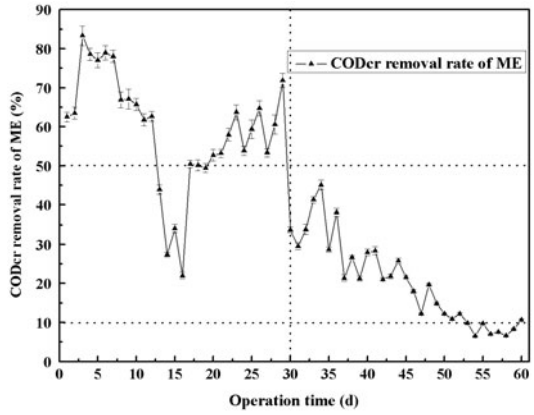

Fig. 2. CODcr removal effect of ME.

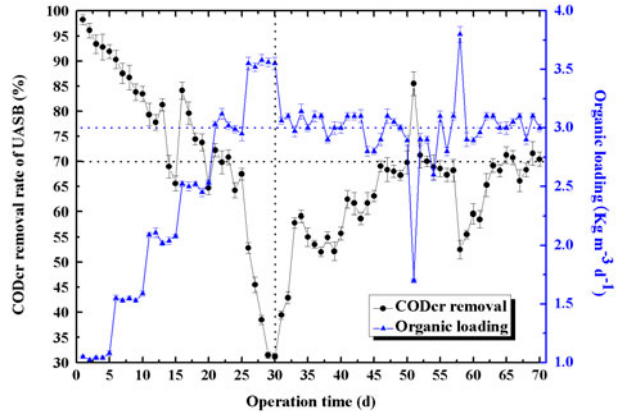

Fig. 3. The removal effect of UASB on CODcr.

the fillers was reduced during the wastewater treatment. Therefore, as the treatment process proceeded, the removal rate of CODcr was decreased gradually, and the final removal rate was about 10%.

To sum up, the biotoxicity of the penicillin G-processing wastewater could be greatly reduced and BOD_5/CODcr of the wastewater effectively enhanced by the ME System, the results revealed that biodegradability of the wastewater could be improved effectively by ME. Moreover, a portion of CODcr in the wastewater could be removed by the ME system. Therefore, ME was suitably used in pretreatment for the penicillin G-processing wastewater.

3.2. Treatment effect of UASB

Anaerobic treatment was necessary for high-concentration organic wastewater, the pretreated wastewater was fed into the UASB tower for treatment. The original OLR was about 1.0 kg m^{-3} d^{-1}, and the increasing step size of OLR was about 0.5 kg m^{-3} d^{-1} every 5 d. The final OLR was determined according to removal effect on CODcr.

Fig. 3 showed the removal effect of UASB on CODcr, it could be seen that about 70% of CODcr in the wastewater could be removed through the UASB system. It was obvious that the whole process could be divided into two stages, including bacterial culture stage (previous 30 d) and stable operation stage (later 40 d). At the bacterial culture stage, as OLR was increased from 0.5 kg m^{-3} d^{-1} to 3.5 kg m^{-3} d^{-1}, the removal rate of CODcr was decreased from about 95 to 30%, and the ratio of the volatile acids to bicarbonate alkalinity (VA/BA) valued between 0.4 to 0.8, indicating that the reactor was instable in this stage [44]. It could be deduced that the anaerobic bacteria was inhabited when OLR was increased. Moreover, as OLR increased, the concentration of SO_4^{2-} in the reactor was increased accordingly, and sulfate-reducing bacteria (SRB) bred rapidly, which would greatly restrict the production of methanogens [45], it also reduced the removal rate of CODcr. At this stage, although as the removal rate of CODcr was gradually decreased, the total removal quality of pollutants was also gradually increased. It revealed that anaerobic bacteria in the UASB tower gradually adapted to the wastewater. When OLR was stably kept at about 3.0 kg m^{-3} d^{-1} after 30 d, the removal rate of CODcr was gradually restored and stabilized at about 70%, and VA/BA ratio was lower than 0.1 which indicated stable operation of the reactor. The reason might be that the anaerobic bacteria for the penicillin G-processing wastewater had been cultivated completely and the methanogens were bred effectively, and the process entered into the stable operation stage.

Overall, utilization of UASB tower for organic pollutants' removal was satisfactory, and about 70% of the removal rate of CODcr could be reduced by the UASB system. The optimum OLR should be 3.0 kg m^{-3} d^{-1} according to effluent quality and investment costs.

3.3. Treatment effect of A/O system

There was still some organic pollutants residue in the effluent of UASB tower, and the wastewater was then introduced into the A/O tank to remove the organic pollutants residue. Moreover, because the concentration of NH_3–N in effluent of UASB tower was high, the reactor could be also used to remove NH_3–N of the wastewater. As OLR reached about 2.0 kg m^{-3} d^{-1}, the A/O system was started.

It could be seen from Fig. 4 that the operation process in the A/O system was similar to that in the UASB system. The process could also be divided into two stages, including bacterial culture stage (previous 20 d) and stable operation stage (later 40 d). At the first stage, the removal rate of CODcr was decreased gradually from about 95 to 60%, the reason might be that bacteria in A/O system was cultivated at this stage, with an increased influent CODcr due to the increase of effluent in the UASB tower, the organic pollutants could not be degraded effectively. After 20 d, the removal rate of CODcr tended to reach stable, indicating that it had entered into the stable operation stage. Because the effluent of the UASB tower reached stable and the bacteria for A/O system were cultivated completely, the removal rate of CODcr was kept at about 80%. Therefore, NH_3–N of the wastewater could be mostly removed due to the strong ability of A/O system for NH_3–N degradation, and the removal rate of NH_3–N could reach about 90%.

From above, it could be concluded that organic pollutants residue and NH_3–N could be reduced effectively, and the removal rate of CODcr and NH_3–N reached about 80 and 90%, respectively. Therefore, UASB and A/O system were suitably utilized as the secondary treatment for the penicillin G-processing wastewater.

3.4. Treatment effect of BAF

After the pretreatment and secondary treatment, the wastewater still could not be discharged into municipal sewers, BAF system was then designed as an advanced treatment unit for the wastewater. HRT of the wastewater for the BAF system was set at 12 h. Fig. 5 showed the removal effect of BAF system on CODcr, the removal rate of CODcr could reach about 60%.

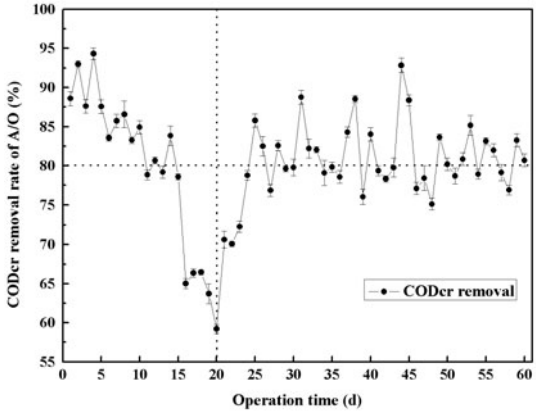

Fig. 4. CODcr removal effect of A/O.

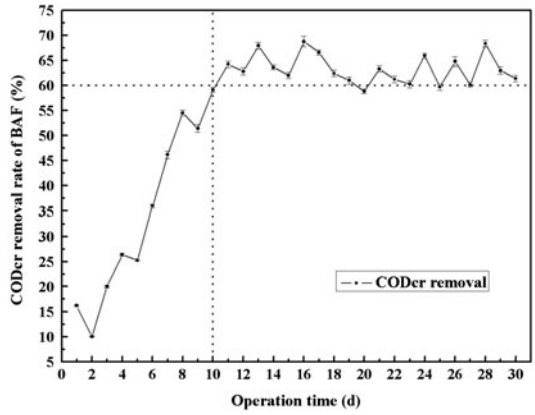

Fig. 5. The removal effect of BAF on CODcr.

In the first 10 d, the removal rate of CODcr was low and gradually increased. It could be deduced that the system was in the biofilm culturing stage, biofilm was not formed on the surface of the fillers fully, and the removal rate of CODcr increased gradually as biofilm was formed completely. When biofilm culturing was finished, the BAF entered into the stable operation stage and the removal rate of CODcr was kept at about 60%. Moreover, BAF system had strong ability of NH_3–N degradation [46], and about 85% NH_3–N in the influent could be further removed by this BAF system.

According to the measurement, the average concentration of CODcr and NH_3–N in final effluent was about 275.3 and 19.8 mg L^{-1}, the effluent could reach the requirement of the national discharge standards (wastewater quality standards for discharge to municipal sewers, China, CODcr ≤ 300 mg L^{-1}, NH_3–N ≤ 25 mg L^{-1} in C standard) (CJ 343-2010). Therefore, BAF system was feasible and a suitable method for the wastewater advanced treatment.

4. Conclusion

Combination utilization of ME, UASB, A/O, and BAF for the penicillin G-processing wastewater treatment was feasible, and the treatment effect was satisfactory; in this study, the conclusion could be summarized as follows:

(1) ME was suitably utilized as the wastewater pretreatment and biodegradability of the wastewater could be greatly improved.
(2) Utilization of UASB and A/O for the secondary treatment of the wastewater was satisfactory, most of the CODcr and NH_3–N could be removed by these two systems.

(3) BAF was suitably used for the wastewater advanced treatment, the final effluent (CODcr of 275.3 mg L^{-1}, NH_3–N of 19.8 mg L^{-1}) could reach the requirement of the national discharge standards (CODcr ≤ 300 mg L^{-1}, NH_3–N ≤ 25 mg L^{-1}) (CJ 343-2010).

(4) Especially, about 80% of penicillin G residue in the wastewater could be removed or converted by ME reactor, and the wastewater could be effectively deposed in this system.

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