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Decomposition of Inorganic Nutrient Nitrogenous Matter from Lightly Polluted Wastewater Using Electrochemical System

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ABSTRACT

In this research, electrochemical system was adapted to decompose lightly polluted wastewater (LPW). The major pollutants of LPW were removed and its removal effects were investigated. Meanwhile, courses of the electrochemical reaction for nitrogen and phosphorus were discussed. Experiments results exhibited that the best removal rates of NH_3-N and TN respectively were 100% and 90.9% after dosing NaCl. In addition, under the condition of different electrolysis time, removal of pollution was better than without NaCl, namely, the pollution was removed rapidly through electrolysis when other experimental conditions remained invariable. Meanwhile, the optimal operation condition could be confirmed while the current density was 7.4mA/cm^2, the cell voltage was 6 V, electrolysis time was 30 min, the plate distance was 1cm and the ratios of concentration of NH_4^+ and Cl^- were 1:3, respectively. Besides, the removal effects for nitrogenous compounds as the change of the chloride concentration were investigated, too. And the results exhibited that higher the chloride concentration in the wastewater was, the higher the removal rate for nitrogenous compounds was. The removal rates of nitrogenous compounds increased with the increasing of cell voltage. Therefore, the influence of every factor could be obtained through considering the power consumption of per ton wastewater treatment.

INTRODUCTION

In recent years, electrochemical oxidation has developed to favourable technology for environmental pollution control, and which possessed some advantages with simple equipment, high efficiency, no secondary pollution, sterilization, etc. (Liu et al. 2013, Mook et al. 2012, Karimi-Maleh et al. 2014, Mook et al. 2014, Dos Santos et al. 2014, Zhang et al. 2013). Although this technology was applied widely, applied research on wastewater quality improvement about lightly polluted wastewater was less recently. In addition, some studies have shown that because of the presence of chloride ions in the solution, electrochemical oxidation reaction course was apt to occur (Park et al. 2013). And because of the loss of chloride ions in the anode water, a series of electron reactions produced chlorine gas, meanwhile, hypochlorous acid or hypochlorite and other active intermediate substances were generated in quantity (Sirés et al. 2014, Aquino et al. 2014, Thirugnanasambandham et al. 2014). Besides, through adding sodium chloride to the wastewater, the chloride ion concentration in the solution increased, which could enhance the effects of this electrochemical oxidation, especially a nitrogen compound treatment effect (Yang et al. 2014, Duan et al. 2014). Under the condition of different plate spacings and electrolysis time, the purpose of this experiment was to use an electrochemical oxidation method to decompose pollution from lightly polluted wastewater. Thereby, the effluent basically reached the final effluent key indicators "Surface Water Environmental Quality Standard in China" (GB3838 -2002) class III-IV water standards; meanwhile, ammonia removal rate could arrived at 50%, total nitrogen concentration was not more than 15mg/L.

MATERIALS AND METHODS

Electrolytic system: As is shown in Fig. 1, electrochemical system device consisted of a cell, the electrode plate, and the DC power supply components. It was made of plexiglass cell, with specifications as (L×W×H) 140 mm × 90 mm × 130 mm, and the effective volume of about 1.6 L. Meanwhile, the titanium anode plates were coated with Lr, Ru, Cs and other precious metals nano-coating process, and cathode plates were only titanium based plates. Each plate possessed the size of length × width as 130mm × 65mm with the thickness of 1mm. There were four plates for anode and cathode, and the plate spacing was 1cm. The maximum output voltage of DC power supply was 36V, and maximum current of 30.7A, and it could be adjusted to a constant voltage and current.

Table 1: Characteristics of the wastewater sample used in the experiments.

Parameter	Unit	Concentration
pH	-	6.5~8.0
Ammonia Nitrogen(NH_3-N)	mgL^{-1}	0.7~1.5
Total Nitrogen(TN)	mgL^{-1}	9~20
Conductivity	$mscm^{-1}$	1.5~1.8

Influent quality: In this study, the raw water was taken from the wastewater treatment plant in Tangshan. And the wastewater quality of secondary effluent is given in Table 1.

Experimental methods: During the experiment, under the condition of constant current density, plate spacing of 1cm and different electrolysis time, electrochemical oxidation course was analysed and evaluated.

RESULTS AND DISCUSSION

In this experiment, when the current density was of 7.4mA/cm^2, adding a certain amount of NaCl to the solution at different sampling times, electrolysis effect was measured under the same conditions.

Nitrogenous compounds removal: At this stage, under the condition of constant current with 5A, and current density of 7.4mA/cm^2, direct electrolysis was carried out using the electrode plate. And under the same conditions, adding a certain concentration of NaCl solution to the solution, the treatment effect for wastewater was measured with different electrolysis times, and the removal effects of ammonia and total nitrogen are shown in Fig. 2.

As shown in Fig. 2, the electrode plates exhibited favourable removal effects for ammonia and total nitrogen. Because the initial ammonia concentration was very low, the removal rate could reach the maximum of 100% after dosing the NaCl under the electrolysis reaction. In addition, electrode electrolysis wastewater directly could attain highest removal rate of 90.3% for total nitrogen, while the 90.9% after dosing the NaCl, and total nitrogen concentration was of 1.16mg/L. The removal effect of nitrogen compounds could increase with the electrolysis time. In this experiment, because the ammonia content in wastewater was small, so ammonia was removed obviously when the electrolysis time was 20min, but in the 10min while comparing the treatment effect, the removal effect was significantly higher than no NaCl. And it showed after dosing NaCl that electrolytic conductivity increased significantly, which speeded up the rate of •OH generating on the electrode plate and other strong oxidizing substances. Therefore, ammonia could be removed obviously after dosing NaCl. In addition, when the electrolysis time was 30min, the removal effect of total nitrogen could reach the optimal

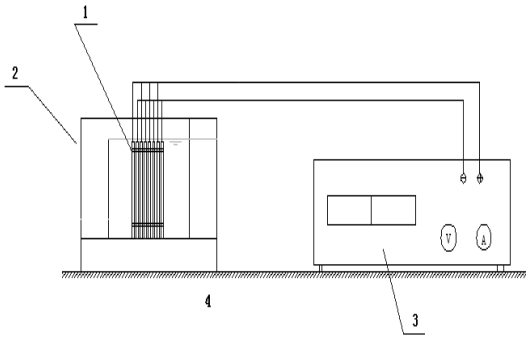

Fig. 1: Schematic diagram of the experimental apparatus (1. electrode plates, 2. electrolytic cell, 3. electric power source, 4. experiment table).

value under dosing NaCl, meanwhile, the removal rates changed significantly during the first 30min, while slowly increasing during the next 30min. This showed that activated ion conductivity relative limit reached during the first 30min, the treatment rate of nitrogen compounds also reached the relative limits, and it could be seen from the experimental data, under constant current conditions, that the electrochemical method with removal of nitrogen compounds was still relatively high.

As shown in Fig. 3 and Fig. 4, ammonia and total nitrogen removal rates were significantly improved with the increase of the voltage and NaCl dosing. Because when the voltage increased, the current density increased, and electron transfer speed was directly oxidized at the anode surface reactions, not in the case of dosing NaCl, ammonia nitrogen and total nitrogen removal rates increased from 3.1% and 4.1% to 69.6% and 53.1% when the cell voltage increased from 6V to 11V. However, it was clear that with the increasing of the ratio of dosing, even at the same voltage, an increase of ammonia and total nitrogen removal was equally obvious. When the cell voltage was 6V, ammonia nitrogen and total nitrogen removal were never increased obviously when dosing NaCl solution, and up to ratio of 1:3 at 50.2%. With the cell voltage increasing, it increased gradually when the cell voltage was 11V, the ammonia and total nitrogen removal rates also increased with dosing NaCl solution from 53.1% to 73.5%. This was due to the dosing of sodium chloride, so that the chloride ion concentration in the solution increased, chloride ions were oxidized to chlorine gas at the anode, such as hypochlorous acid or hypochlorite intermediate active substances. And it had strong oxidation resistance, thereby speeding up the rate of oxidation of the anode indirectly.

Nitrite nitrogen removal: As is shown in Fig. 5, when no NaCl dosing, as the voltage was increased nitrite significantly from 6V to 11V at 6.8% and 82.6%, and the ratio of NaCl solution increased dosing largely, nitrite nitrogen re-

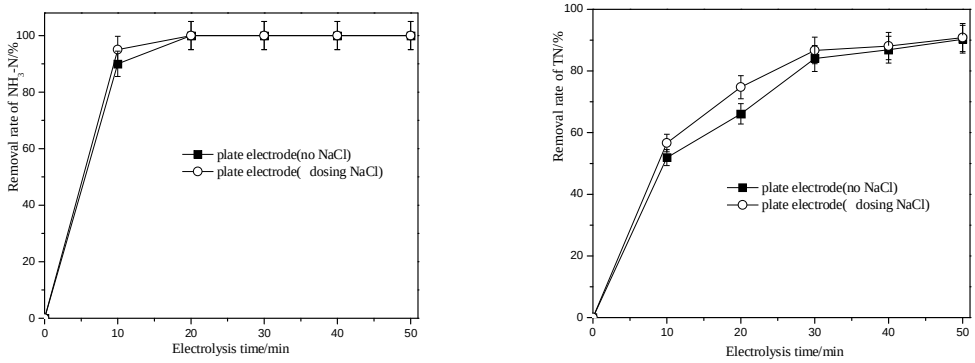

Fig. 2: The removal effect of nitrogenous compounds is shown when the electric current was constant with the electrolysis time prolonging.

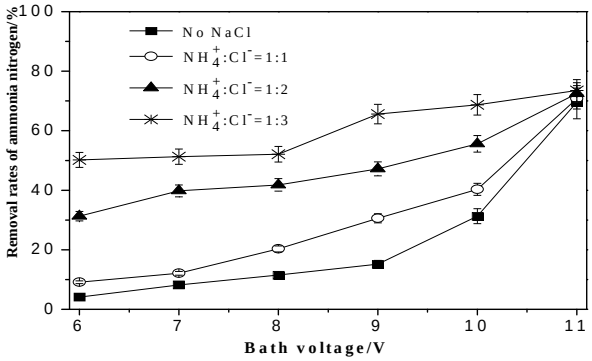

Fig. 3: Influence of ammonia nitrogen removal by dosing under different voltages.

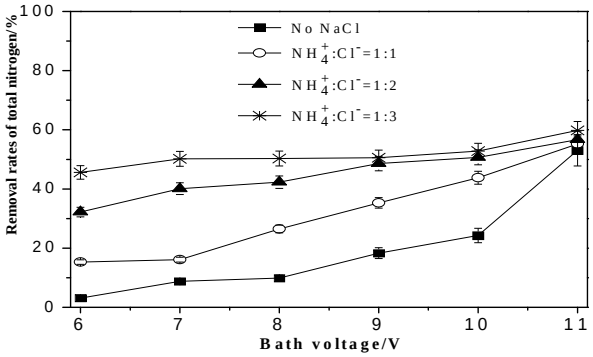

Fig. 4: Influence of total nitrogen removal by dosing under different voltages.

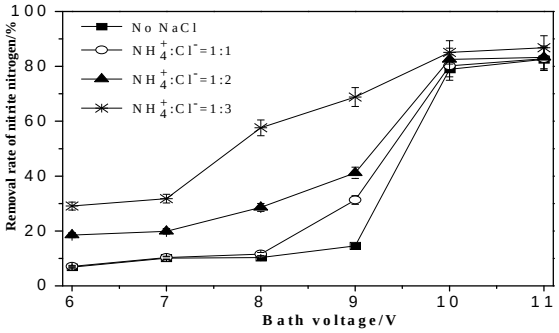

Fig. 5: Influence of nitrous nitrogen removal by voltage under different additive amounts.

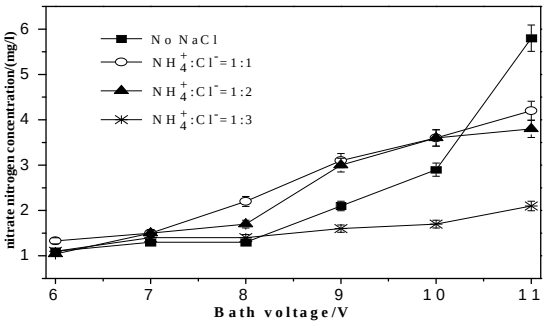

Fig. 6: Influence of nitrate nitrogen concentration by voltage under different additive amounts.

moval was also significantly improved. As the voltage was 6V, dosing ratio of 1:1 , nitrite nitrogen removal rate was of 7.1%. When the dosing ratio was of 1:2 , nitrite removal rate increased to 18.5%. And when dosing ratio was of 1:3 , nitrite nitrogen removal rate increased to 29.1%; when the voltage increased to 11V, the dosing ratio of 1:1, nitrite nitrogen removal rate was 82.8%, when the dosing ratio of 1:2, nitrite nitrogen removal rate increased to 83.3%, when dosing ratio of 1:3, nitrite nitrogen removal rate was of 86.8%. At lower voltages with increasing dosage ratio increased nitrite removal, when the voltage was increased to a large, nitrite removal increased, but relatively slow, and the removal rates of ammonia nitrogen and total nitrogen still maintained favourable effects. It could be inferred that during the electrolysis, the chloride ions could be added to produce a strong oxidizing agent such as OCl^-, and wastewater with nitrite on the anode was indirectly oxidated into other forms of nitrogen, the wastewater was obtained with representing nitrite completely removed.

Nitrate nitrogen removal: As shown in Fig. 6, when no

NaCl dosing, voltage increased as the concentration of nitrate nitrogen, which significantly increased from 1.1mg/L to 5.89mg/L when the cell voltage increased from 6V to 11V. With the ratio of dosing increasing, the concentration of nitrate was in the overall downward trend, such as at the voltage was of 6V, dosing ratio of 1:1, and the nitrate concentration of 1.33mg/L. When the dosing ratio was of 1:2, the concentration of nitrate nitrogen was 1.04mg/L, and when the dosing ratio was of 1:3, the concentration of nitrate was 1.1mg/L. When the voltage increased to 11V, dosing ratio of 1:1, the concentration of nitrate was 5.07mg/L, and the dosing ratio was of 1:2, the concentration of nitrate decreased to 4.46mg/L. When the dosing ratio was of 1:3, the concentration of nitrate reduced to 3.03mg/L. It could be concluded comparing with the increase of dosing, the change was not obvious for nitrate concentration under the condition of the low voltage.

CONCLUSION

According to the experimental results, the optimal operating conditions can be determined from this experiment: when current density was of 7.4mA/cm^2, the cell voltage was of 6 V, electrolysis time was of 30 min, the plate distance was of 1cm and the ratios of concentration of NH_4^+ and Cl^- were 1:3, respectively. Experimental results showed that the treatment efficiency with the NaCl was not obvious when electrode reaction for all major pollutants was higher than dosing NaCl. In the electrochemical oxidation process, NaCl solutions with different concentrations had a significant impact on the removal of major nitrogenous compounds. In the case of maintaining a certain voltage, when NH_4^+ and Cl^- ratio was of 1:3, the lightly polluted wastewater for each major pollutant removal was more favourable. In addition, when NH_4^+ and Cl^- ratio was of 1:3, ammonia nitrogen and total nitrogen removal was also increasing with increasing voltage. And under the condition of electrolysis voltage of 6V, ammonia and total nitrogen removal rates could attain the 50.2% and 45.6%, respectively.

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