

# Process Optimization of Three-Dimensional Electro-Fenton System for Treatment of Wastewater Produced in Acrylamide Manufacturing Industry

Jian-jun Chen<sup>1</sup>, Li-ya Wang<sup>1,\*</sup>, Yue Chen<sup>2</sup>, Wei-ying Yao<sup>2</sup>

<sup>1</sup> College of Materials and Environmental Engineering, Hangzhou Dianzi University, Hangzhou 310018, PR China.

<sup>2</sup> ZheJiang Yiqing Environmental Engineering Co. Ltd., Hangzhou 310018, PR China.

\*E-mail: [13588761996@163.com](mailto:13588761996@163.com)

Received: 14 February 2022 / Accepted: 20 April 2022 / Published: 7 May 2022

Acrylamid (AM) production wastewater is biologically toxic and difficult to treat directly as it contains more ammonia and COD but a low concentration of AM monomer. The production of H<sub>2</sub>O<sub>2</sub> by electrochemical action can be controlled by the electro-Fenton method, which can degrade COD and NH<sub>3</sub>-N simultaneously while effectively improving the biodegradability of wastewater. In this study, the effects of current density, initial pH value, and iron chipping amount on the treatment efficiency of acrylamide-producing wastewater by three-dimensional electro-Fenton method were investigated. The orthogonal test explores the optimum operating parameters. The results show that the optimal process conditions for the single-factor experiment are the current density of 12.5mA/cm<sup>2</sup>, the initial pH value of 3, and the iron chipping amount of 1.25g; the optimum combined process conditions in the orthogonal experimental design are the current density of 15.0mA/cm<sup>2</sup>, the initial pH value of 4, and the iron chipping amount of 1.25g. Currently, the removal of COD and NH<sub>3</sub>-N is 85.0% and 80.1%, which is 2.9% and 13.2% higher than that under the single-factor optimal conditions, respectively. The three-dimensional electro-Fenton method can effectively break the double bond in a monomer and improve the B/C ratio of AM wastewater. The above results can effectively address the issue of NH<sub>3</sub>-N treatment of AM wastewater.

**Keywords:** acrylamide-producing wastewater; three-dimensional electro-Fenton; orthogonal test; optimization

## 1. INTRODUCTION

Acrylamide (AM) is a white crystal, and is the raw material that produce polyacrylamide. AM is widely used in many fields, such as textiles, wastewater treatment and purification, metallurgy and chemical industry, paper making, etc. The complex acrylamide is not toxic, but the monomeric form of

acrylamide is highly toxic [1]. The AM monomer contains double bonds and an acylamino, possessing the chemical generality of the double bond: the polymerization is rapidly induced under ultraviolet irradiation or at melting point temperature. Studies have shown that AM can cause the accumulation of reactive oxygen species (ROS), induce apoptosis and inflammation, and cause irritation and toxicity to humans. Due to its harmful effects, the AM pollution of wastewater must be eliminated [2-3].

It is increasingly difficult to effectively deal with toxic and harmful pollutants using conventional methods, and advanced oxidation methods have attracted considerable attention [4-7]. Electrochemical oxidation is an efficient environmental protection treatment technology, in which the electro-Fenton method is an electrochemical induced using the advanced oxidation method. In recent years, the application research of the electro-Fenton method has been increasing [8]. As the electro-Fenton method is easy to operate and has no secondary pollution, it has gradually attracted people's attention. It is primarily used in the field of wastewater treatment research [9]. The electro-Fenton method has been successfully applied to the treatment of various persistent pollutants such as dyes, halogenated hydrocarbons, and phenols [10-13]. Cathode redox in the electro Fenton system is mainly divided into two methods: (1) Oxygen loses two electrons at the cathode and is reduced to  $\text{H}_2\text{O}_2$ , and the generated  $\text{H}_2\text{O}_2$  continues to react to produce  $\text{H}_2\text{O}$ . (2) Oxygen loses four electrons at the cathode and is directly reduced to  $\text{H}_2\text{O}$ . At present, more and more high organic refractory wastewater has been primarily pretreated using the electro-Fenton method. An efficient and pollution-free method will be the critical research and development direction in the future [14-16].

AM-producing wastewater is a kind of industrial wastewater with complex components, high organic, high  $\text{NH}_3\text{-N}$  content, and specific biological toxicity. At present, there are few references on the treatment of acrylamide production wastewater by electrochemical methods. Moreover, the components of acrylamide production wastewater are complex, so treatment by the electro-Fenton method is of significant research importance. Li [17] et al. used anaerobic treatment + SBR (Sequencing Batch Reactor Activated Sludge Process) process to treat AM-producing wastewater and studied the strain AM-4 with high efficiency to degrade AM. It was found that the removal of AM, COD and  $\text{NH}_3\text{-N}$  after continuous operation for 24 hours reached 91.4%, 87.2% and 66.7%, respectively. Due to the influence of water quality fluctuation and monomer toxicity in engineering applications, it is difficult to maintain efficient and stable sludge, resulting in unacceptable ammonia concentrations by the Chinese national discharge standard (GB8978-1996). Besides, some scholars have used standard combined processes to treat AM-producing wastewater and have achieved good treatment results [18-19]. Zhuang [18] et al. added a set of iron-carbon micro-electrolysis systems and primary  $\text{A}^2/\text{O}$  treatment system on the basis of fully utilizing the original facilities to upgrade the AM wastewater project and make its effluent quality meet the standard stably. Su [19] et al. used a two-phase anaerobic +  $\text{A}/\text{O}$  process to treat acrylic fibers and acrylamide mixed wastewater. The results show that two-phase anaerobic can significantly improve the biodegradability of wastewater, and the total COD removal rate was 87%-89%. At present, there are few studies on the treatment of AM wastewater by the electro-Fenton method. This study conducts the pretreatment of AM-producing wastewater by the three-dimensional electro-Fenton method, which can remove the macromolecular refractory substances while removing COD and  $\text{NH}_3\text{-N}$ , thus improving the biodegradability of AM wastewater. This study may provide a reference for further combined process tests and the research of AM wastewater.

## 2. MATERIALS AND METHODS

### 2.1. Experimental materials

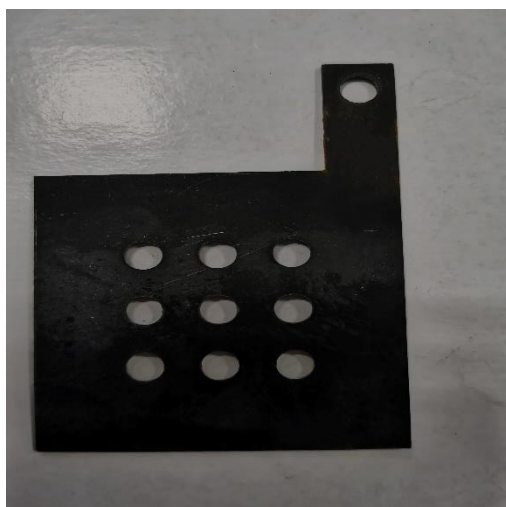
The reagents used in this experiment are as follows: sodium hydroxide (NaOH); Tin tetrachloride pentahydrate ( $\text{SnCl}_4 \cdot 5\text{H}_2\text{O}$ ); Hydrochloric acid (HCl); Antimony trichloride ( $\text{SbCl}_3$ ); N-butanol ( $\text{C}_4\text{H}_{10}\text{O}$ ); Sulfuric acid ( $\text{H}_2\text{SO}_4$ ). The above reagents are A.R.

The instruments and equipment used in this experiment are as follows: COD rapid tester (5B-3A); Field emission scanning electron microscope (Gemini500); pH meter (pHS-3C); Magnetic heating agitator (CJJ78-1); DC power supply (MS-3050); Fourier micro infrared spectrometer (IS20-IN5).

### 2.2. Electrode preparation

The titanium plate is polished with sandpaper and washed with deionized water. Then the plate is placed in a beaker containing 40% NaOH solution, then placed into a thermostat water bath, and heated in 95 °C water bath for 2 hours. Next, the plate is washed with deionized water and dry using filter paper. The plate is then placed in a beaker containing 1:1 HCl solution and heated in 95 °C water bath for 1 hour. Finally, a flat grey-white titanium plate was obtained.

26.6 g of  $\text{SnCl}_4 \cdot 5\text{H}_2\text{O}$  and 1.9 g of  $\text{SbCl}_3$  are accurately weighed and dissolved in a solution containing 100mL N-butanol and 1mL HCl. Next, the solution is stirred evenly by a glass rod, dipped with a brush, and evenly painted to the plate. This plate is placed in an oven at 100 °C for approximately 10 minutes and then placed in a 500 °C muffle furnace to oxidize the electrode plate for 15 minutes. This process is repeated 15 times. Finally, the plate is annealed in a muffle furnace at 500 °C for 1 hour [20-22]. The Ti-based Dimensionally Stable Anode (DSA) electrode with  $\text{SnO}_2\text{-Sb}_2\text{O}_5$  coating is completed. Fig. 1 shows the self-made DSA electrode plate.



**Figure 1.** Self-made Ti-based DSA electrode plate with  $\text{SnO}_2\text{-Sb}_2\text{O}_5$  coating

### 2.3. Wastewater source

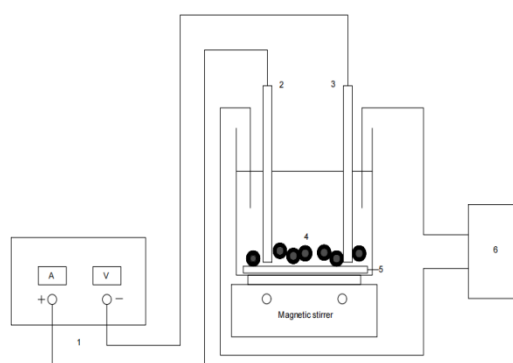
This paper takes AM wastewater produced by a biochemical enterprise in Ningbo as the research object. The water quality characteristics of wastewater are shown in Table 1.

**Table 1.** water quality characteristics of AM-producing wastewater

Measure s	COD <sub>Cr</sub> mg/L	SS mg/L	NH <sub>3</sub> -N mg/L	TN mg/L	AM mg/L	Salt content mg/L	pH
Value	≤8000	≤500	≤1000	≤120 0	≤20	10000- 12000	5.0-6.0

### 2.4. Experimental methods

The electro-Fenton device is improved in this paper[20-22], and the double cathode is selected to improve the removal efficiency of the reaction. A 250 mL beaker is used as a container for wastewater treatment, a 2 mm carbon felt is placed at the bottom of the beaker, the self-made Ti-based DSA plate acts as the anode and a stainless steel plate as the cathode, an aeration device is connected to aerate the beaker.



1. DC power supply; 2. Self-made DSA anode; 3. Stainless steel cathode; 4. Iron chipping; 5. Carbon felt; 6. Aeration device

**Figure 2.** Improved electro-Fenton device

The third electrode (the particle electrode) is usually filled between the two electrodes with debris or granular electrode material, and the surface of the particle electrode is charged and polarized under the action of electrostatic induction to become a new third electrode[23]. Compared with the traditional two-dimensional electrode, the specific surface area of the three-dimensional electrode is significantly increased, the space utilization is greatly improved, and the spacing between the filled particle electrodes is small, which significantly improves the mass transfer process and effect[24]. Activated carbon as a filler can improve the removal effect of traditional electro-Fenton, and the specific surface area of carbon

felt is larger than activated carbon. Carbon felt and a part of iron chipping in the reaction system will also form an iron carbon micro electrolysis system. It is of significance to explore the degradation effect of electro-Fenton device with carbon felt as a filler on acrylamide production wastewater. The device connection is shown in Fig. 2.

150 mL of pH adjusted AM wastewater and some iron chippings are added into the beaker with a 2 mm carbon felt lying on the bottom. The power switch is turned on and the appropriate current is adjusted. The whole process is timed and a water sample is taken every hour during the reaction. This water sample is adjusted to the pH value to about 10.0. A high-speed centrifuge is used to separate the solution. Finally, the supernatant is taken and the values of COD and  $\text{NH}_3\text{-N}$  are measured. Each water sample is measured three times and the average value is taken.

### 2.5. Orthogonal experimental design

This study referred to the research results of other scholars[25-27]. The effects of current density, initial pH value, and iron chipping amount (solid to liquid ratio, fixed 150mL waste water volume ) were mainly researched in the AM wastewater treatment under the condition determining the thickness of carbon felt (2 mm), electrode plate spacing (1 cm), aeration volume (1 L/min), and electrolysis time (6 h). The solid to liquid ratio refers to the quality of iron chipping to the quality of wastewater. The orthogonal experimental design method is used to find the best conditions, and a total of nine tests were carried out. The factor level of the orthogonal test is shown in Table 2.

**Table 2.** factor level of orthogonal test

Level/Factor	A-current density (mA/cm <sup>2</sup> )	B-initial pH	C-iron chipping amount (g)
1	10.0	2	0.75
2	12.5	3	1.25
3	15.0	4	1.50

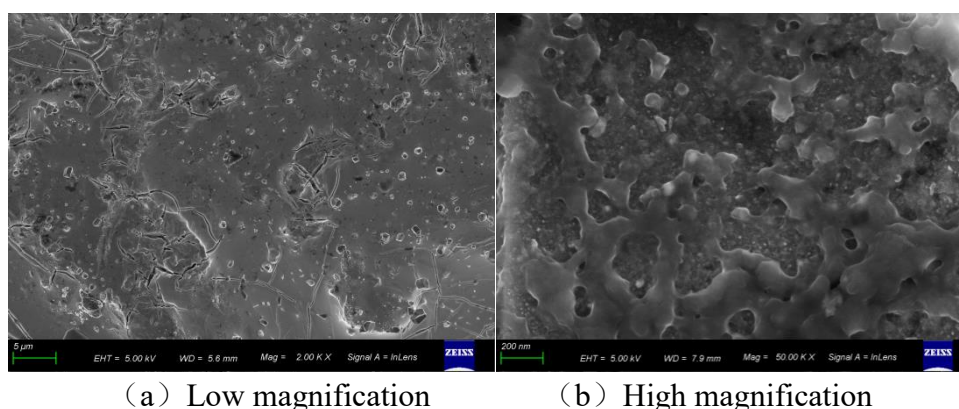
### 2.6 Infrared spectrum detection

Untreated AM wastewater and AM wastewater after three-dimensional electro-Fenton treatment under optimal conditions were obtained. After appropriate treatment, these water samples underwent infrared characterization with a Fourier infrared spectrometer. Meanwhile, the transformation of wastewater components in the electrolysis process by Fourier infrared spectroscopy is explored.

### 3. RESULTS AND DISCUSSION

#### 3.1. Electrode surface characteristics

Fig. 3 are the surface shape appearance figures of the self-made electrode plate obtained with different magnifications using the scanning electron microscope. Fig. 3(a) shows that the DSA electrode plate coating belongs to the "mud crack" shape. The cracks are created during drying and high-temperature oxidation, which will facilitate the electrolyte to flow deep and increase the contact area of the reaction. Fig. 3(b) shows that the surface of the electrode plate is an uneven structure, which has a more extensive specific surface area that is conducive to increasing the reaction contact between the electrode plate and the electrolyte.



**Figure 3.** SEM images of the electrode materials under different magnification

#### 3.2 Single factor impact analysis

##### 3.2.1 Current density

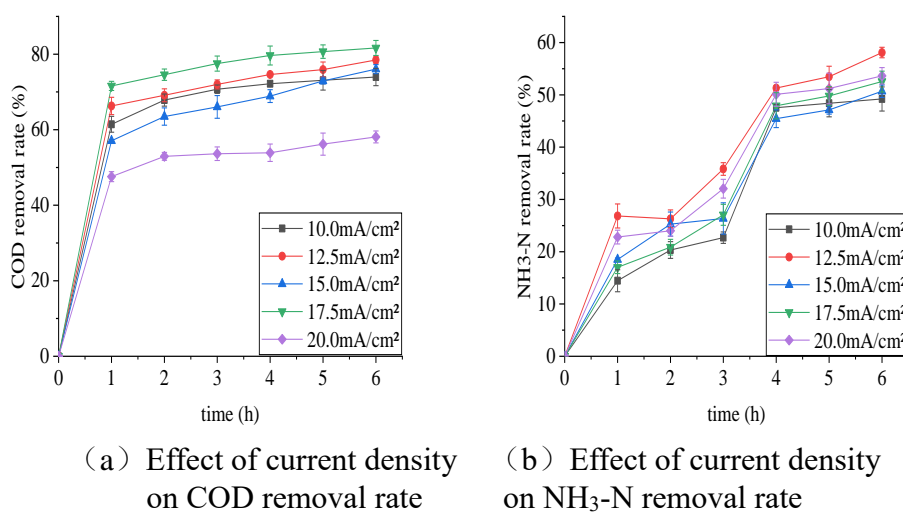
The imposed current density has a significant effect on the degradation of organic compounds in the electro-Fenton reaction. The current density of the reaction was set to 10.0, 12.5, 15.0, 17.5 and 20.0 mA/cm<sup>2</sup> respectively, and the initial pH value was adjusted to about three, then 1.25g of iron chippings were added into the device. The experimental results are shown in Fig. 4.

Fig. 4 (a) shows the COD removal effect is significant in the first 1 h. With the extension of the reaction time, the COD removal efficiency under different current densities increases slowly. The COD removal effect can conclude that the current density is 17.5 mA/cm<sup>2</sup>, 12.5 mA/cm<sup>2</sup>, 10.0 mA/cm<sup>2</sup>, 15.0 mA/cm<sup>2</sup>, 20.0 mA/cm<sup>2</sup> in sequence. When the current density is 17.5 mA/cm<sup>2</sup>, the COD removal effect is the highest, reaching 81.19%. Relatively high current density can promote the anode surface oxidation reaction, thus helping to produce more hydroxyl radicals, and accelerate the reaction rate. However, when the current density increases to 20.0 mA/cm<sup>2</sup>, the COD removal rate reduces. This result is consistent with results obtained by some scholars when using electrolysis to treat municipal solid waste leachate [28-29]. The applied current is the driving force of H<sub>2</sub>O<sub>2</sub> production by electrocatalysis.

Obviously, with the increase of current intensity, the electro-Fenton degradation rate increases gradually, which is due to stronger anodic oxidation, more  $\text{H}_2\text{O}_2$  generation and faster  $\text{Fe}^{3+}$  reduction. Too high current density will cause over heating in the reaction device and may also accelerate the rate of some side reactions (Hydrogen evolution reaction and oxygen evolution reaction), resulting in a low COD removal rate. The experiment uses high current density for reaction, which will also increase the economic cost of wastewater treatment.

Fig. 4(b) shows the  $\text{NH}_3\text{-N}$  removal trend is relatively flat in the first 2 h and 4-6 h, and the increase of  $\text{NH}_3\text{-N}$  removal rate in 2-4 h is significantly increased. From the  $\text{NH}_3\text{-N}$  removal effect, it can be concluded that the current density is  $12.5 \text{ mA/cm}^2$ ,  $20.0 \text{ mA/cm}^2$ ,  $17.5 \text{ mA/cm}^2$ ,  $15.0 \text{ mA/cm}^2$ ,  $10.0 \text{ mA/cm}^2$  in sequence. When the current density is  $12.5 \text{ mA/cm}^2$ , the  $\text{NH}_3\text{-N}$  removal effect is the highest, reaching 58.90%. When the current density is  $12.5 \text{ mA/cm}^2$ , few bubbles can be observed, which is more conducive to the direct oxidation of organic matters on the electrode plate. When the current density is high, many bubbles gather near the electrode plate. The productivity of  $\text{H}_2\text{O}_2$  will be controlled by mass transfer, and the effect of current density on the degradation rate of  $\text{NH}_3\text{-N}$  will no longer be significant. There are two ways to remove  $\text{NH}_3\text{-N}$  by electrochemical oxidation: (1) Direct oxidation:  $\text{NH}_3\text{-N}$  can directly lose three electrons on Pt anode and be oxidized to nitrogen and water[30]; (2) indirect oxidation[31]: chloride ions are first oxidized to free chlorine on the anode, and then dissolved in aqueous solution to form “active chlorine”, which reacts with  $\text{NH}_3\text{-N}$  as a strong oxidant to produce nitrogen, and finally remove  $\text{NH}_3\text{-N}$ . When the current density ( $=10.0 \text{ mA/cm}^2$ ) is too low, the production efficiency of cathode hydrogen peroxide and the reduction rate of  $\text{Fe}^{3+}$  are affected, resulting in poor  $\text{NH}_3\text{-N}$  removal effect[32].

Therefore, considering the economic feasibility and the removal rate of characteristic indexes, the ideal current density is  $12.5 \text{ mA/cm}^2$ .



**Figure 4.** Effect of current density on COD and  $\text{NH}_3\text{-N}$  removal from the wastewater. Conditions: Initial pH 3; aeration volume 1 L/min; iron chipping 1.25g.

### 3.2.2 Initial pH value

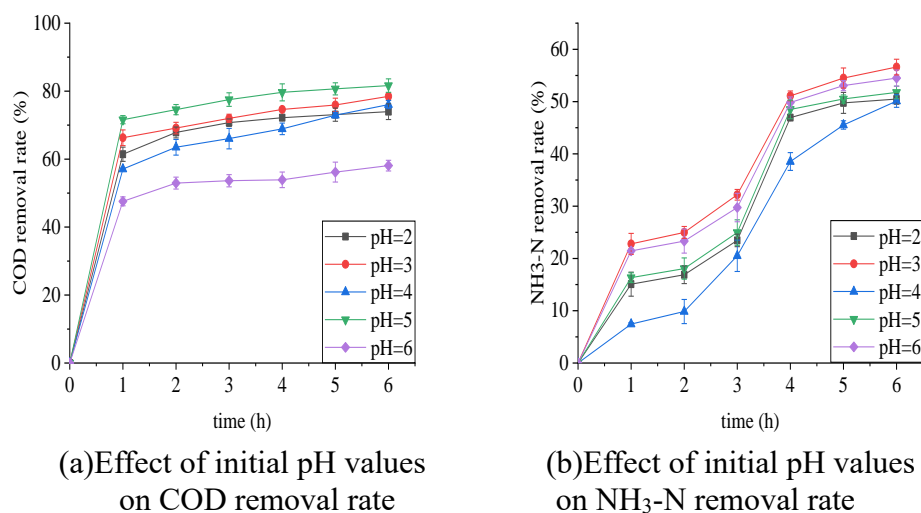
The pH value in the solution will change the charged state in the liquid, so the initial pH value will also affect the electrolytic efficiency of the reaction. The initial pH values of the reaction were set to 2, 3, 4, 5, and 6 respectively, the current density were adjusted to  $12.5 \text{ mA/cm}^2$  and then 1.25g iron chippings were added into the device. The experimental results are shown in Fig. 5.

Fig. 5(a) shows the COD removal effect is significant in the first 1h. With the extension of reaction time, the COD removal efficiency under different pH increased slowly. The COD removal effect can conclude that the initial pH value is 4, 3, 2, 6, 5 in sequence. When the initial pH value is 4, the highest COD removal rate reaches 80.00%. As the chloride ion ( $\text{Cl}^-$ ) in the solution will generate chlorine ( $\text{Cl}_2$ ) during the electrolysis when the pH value is between 2 to 4, more  $\text{Cl}_2$  in the solution will be transformed into oxidizing  $\text{HOCl} \cdot$ . The hydroxyl radical generation will also increase with the pH value so that higher degradation efficiency can be obtained. When the initial pH of the electro-Fenton system is 4, the production of  $\text{HOCl} \cdot$  and  $\text{OH} \cdot$  in this system reaches the maximum, so the COD removal efficiency of acrylamide production wastewater is highest when  $\text{pH} = 4$ . Liu[33] et al used three-dimensional electrode and electro-Fenton coupling method to degrade when drilling petroleum the wastewater produced. The experiment found that when the pH of the reaction system was 4, the  $\text{H}_2\text{O}_2$  and  $\text{OH} \cdot$  concentration was highest.

Fig. 5(b) shows the  $\text{NH}_3\text{-N}$  removal trend is relatively flat in the first 2 h and 4-6 h, and the increase of  $\text{NH}_3\text{-N}$  removal rate in 2-4 h is significantly increased. The  $\text{NH}_3\text{-N}$  removal effect can conclude that the initial pH value is 3, 6, 5, 2, 4 in sequence. When the initial pH value is 3, the  $\text{NH}_3\text{-N}$  removal rate is the highest, reaching 57.00%. The reason is that the acidic condition is conducive to accelerating the dissolution of  $\text{Fe}^{2+}$  from iron filings and reacts with  $\text{H}_2\text{O}_2$  to form hydroxyl radical ( $\text{HO} \cdot$ ). Besides, the acidic condition is more conducive to the forward reaction of resolving  $\text{H}_2\text{O}_2$  to  $\text{HO} \cdot$ . Some scholars believe that the presence carbonate and bicarbonate will also eliminate  $\text{HO} \cdot$  under alkaline conditions[34]. Ammonia nitrogen can be degraded faster in a lower pH environment, which may be due to the hydrolysis of  $\text{Cl}_2$  produced by electrolysis to form hypochlorous acid. The oxidation of  $\text{NH}_3\text{-N}$  mainly depends on the active chlorine in the solution[35]. The phenomenon of "the optimal initial pH value of  $\text{NH}_3\text{-N}$  is different from COD" obtained from the experimental results can not be reasonably explained after consulting the literature. This paper will further study this phenomenon.

Because the initial pH value of AM-producing wastewater is approximately 5.4, and considering the removal effectiveness and cost, the best initial pH value is 3.





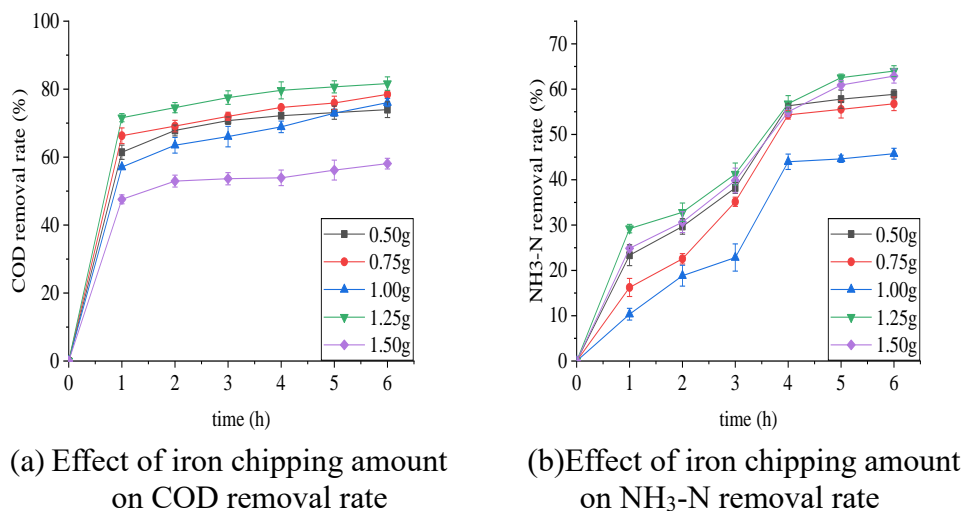
**Figure 5.** Effect of initial pH values on COD and NH<sub>3</sub>-N removal from the wastewater. Conditions: Current density 12.5 mA/cm<sup>2</sup>; aeration volume 1 L/min; iron chipping 1.25g.

### 3.2.3 Effect of iron chipping

The Fe<sup>2+</sup> ion concentration in the solution can affect the formation rate of hydroxyl radical and further affect the electrolysis efficiency in the electro-Fenton reaction process. The iron chipping amount of the reaction was set to 0.5g(0.44%), 0.75g(0.66%), 1.00g(0.88%), 1.25g(1.11%), and 1.50g(1.33%) respectively, and the current density adjusted to 12.5 mA/cm<sup>2</sup> and the solution initial pH value adjusted to about 3. The experimental results are shown in Fig. 6.

Fig. 6(a) shows the COD removal effect is significant in the first 1 h. With the extension of the reaction time, the COD removal efficiency under different iron filings dosages increased slowly. The COD removal effect can conclude that the iron chipping amount is 1.25g(1.11%), 0.75g(0.66%), 0.5g(0.44%), 1.50g(1.33%), 1.00g(0.88%) in sequence. When the iron chipping amount is 1.25g, the COD removal rate is the highest, reaching 81.00%. The solution pH value should be maintained at about 3.0 to make the iron filings precipitate Fe<sup>2+</sup> ions stably in the electro-Fenton device. When the iron filings amount increases, the COD removal rate will decrease. This reason may be that a large amount of Fe<sup>2+</sup> will react with HO• to generate OH<sup>-</sup>, so that part of HO• will lose oxidation, resulting in the removal rate reduction[36]. It may also be that excessive iron filings need a stricter acidic environment.

Fig. 6(b) shows the NH<sub>3</sub>-N removal effect is significant in 1-4 h and increased slowly in 4-6 h. From the NH<sub>3</sub>-N removal effect can be concluded that the iron chipping amount is 1.25g(1.11%), 1.50g(1.33%), 0.5g(0.44%), 0.75g(0.66%), 1.00g(0.88%) in sequence. When the iron chipping amount is 1.25g, the NH<sub>3</sub>-N removal rate is the highest, reaching 64.90%; Similarly, when the iron chipping amount is 1.00g, the removal efficiency is the worst. When the addition of iron filings is less than 1.25g, the NH<sub>3</sub>-N removal efficiency is not high, indicating that too low Fe<sup>2+</sup> is not conducive to Fenton reaction and reduces HO• production efficiency[37,38]. Considering the removal effectiveness and cost, the best iron chipping amount is 1.25g(The solid to liquid ratio is 1.11%).



**Figure 6.** Effect of iron chipping amount on COD and NH<sub>3</sub>-N removal from the wastewater. Conditions: Current density 12.5mA/cm<sup>2</sup>; aeration volume 1 L/min; initial pH 3.

### 3.3. Multi factor interaction analysis

The orthogonal test result is shown in Table 3, and the error analysis is shown in Table 4.

Table 3 shows the optimum process conditions of the three-dimensional electro-Fenton are the current density of 15 mA/cm<sup>2</sup>, the initial pH value of 4, and the iron chipping amount of 1.25 g. Combined with the above single-factor analysis and figures 4-6, the removal rate of COD, especially NH<sub>3</sub>-N, tends to be flat after the 4 h electrolysis time. In addition, the ratio of BOD to COD(B/C) of wastewater after electrolysis for 4 h is 0.40. Given the economic costs, it is finally determined that the optimal operating conditions of the three-dimensional electro-Fenton are: electrolysis for 4 h under the process conditions of the current density of 12.5 mA/cm<sup>2</sup>, the initial pH value of 3, and the iron chipping amount of 1.25 g.

**Table 3.** orthogonal test results

Number/Factor	A (mA/cm <sup>2</sup> )	B	C (g)	COD removal (%)	NH <sub>3</sub> -N removal (%)
1	10.0	2	0.75	56.31	59.98
2	10.0	3	1.25	60.62	62.67
3	10.0	4	1.50	62.89	64.81
4	12.5	2	1.25	64.73	68.98
5	12.5	3	1.50	68.83	72.98
6	12.5	4	0.75	65.21	70.54
7	15.0	2	1.50	83.11	77.52
8	15.0	3	0.75	79.95	74.91
9	15.0	4	1.25	85.00	80.10

$$R^2_{\text{COD}}=0.998, R^2_{\text{NH}_3\text{-N}}=0.988$$

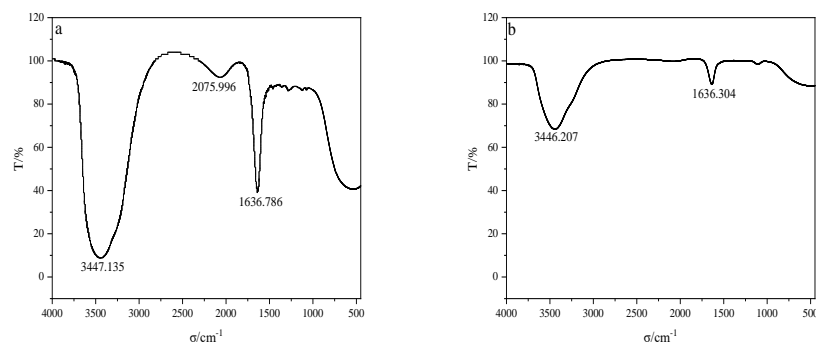
**Table 4.** Analysis of variance of orthogonal test Dependent Variable: COD removal and NH<sub>3</sub>-N removal

Source	Type III Sum of Squares	df	Mean Square	F	Sig.
Corrected Model	0.176 <sup>a</sup>	6	0.029	1634.070	.000
C	0.006	2	0.003	173.615	.000
B	0.003	2	0.001	75.686	.000
A	0.167	2	0.084	4652.909	.000
Error	0.000	11	1.799E-5		
a. R Squared=0.999 (Adjusted R Squared=0.998)					
Dependent Variable: NH <sub>3</sub> -N removal rate					
Source	Type III Sum of Squares	df	Mean Square	F	Sig.
Corrected Model	0.074 <sup>a</sup>	6	0.012	288.654	.000
C	0.003	2	0.002	28.755	.000
B	0.003	2	0.001	25.150	.000
A	0.068	2	0.034	632.056	.000
Error	0.001	11	5.367E-5		
a. R Squared=0.992 (Adjusted R Squared=0.988)					

Table 4 shows that these three factors significantly impact the COD removal rate and the NH<sub>3</sub>-N removal rate. The primary and secondary relationship of the three factors is current density > iron chipping amount > initial pH value on the COD removal rate influence. The primary and secondary relationship of the three factors is current density > iron chipping amount > initial pH value on the NH<sub>3</sub>-N removal rate influence. The experimental results in this paper are consistent with the results of other scholars' electro-Fenton treatment of other wastewater[39].

### 3.4 Conversion analysis of AM monomer

Fig. 7 are two Fourier infrared spectrograms of AM wastewater before and after treatment. Fig. 7 shows the intensity of the peak changes significantly before and after the three-dimensional electro-Fenton treatment. After electro-Fenton treatment, the original peak at 2075.996 cm<sup>-1</sup> disappeared, and the intensity of the peaks at 3500-3400 cm<sup>-1</sup> and 1650-1600 cm<sup>-1</sup> also decreased significantly. The 2075.996 cm<sup>-1</sup> is in the triple bond and cumulative double bond region (2500-1900 cm<sup>-1</sup>), indicating that polymers and other substances contain carbon-carbon double bonds in the wastewater except for AM[40]; the 1638cm<sup>-1</sup> is the characteristic peak of carbon-oxygen double bond in the double bond stretching-vibration region (1900-1200 cm<sup>-1</sup>). Fig. 7 shows the degradation of organic matter in wastewater mainly occurs on unsaturated bonds. After many double bonds are broken by electro-Fenton reaction in wastewater, they react with free radicals in solution to form organic compounds with small molecular formulas and CO<sub>2</sub>. The amino group is also broken in the electro-Fenton process, resulting in the formation of NH<sub>3</sub>, which dramatically weakens the intensity of the peak.



a. Untreated wastewater; b. Wastewater after three-dimensional electro-Fenton treatment

**Figure 7.** Infrared spectrograms of AM wastewater

#### 4. CONCLUSIONS

As an important electrochemical advanced oxidation technology, electro-Fenton technology has application prospects in the treatment of acrylamide production wastewater. The three-dimensional electro-Fenton pretreatment can effectively reduce the COD and  $\text{NH}_3\text{-N}$  in AM-producing wastewater. After the electro-Fenton pretreatment, the double bond in a monomer can be well broken and the B/C ( $\text{BOD}_5/\text{COD}_{\text{Cr}}$ ) ratio of AM-producing wastewater can be effectively improved. Three-dimensional electro-Fenton treatment of AM-producing wastewater has excellent application prospects.

#### References

1. K. Buranasilp, J. Charoenpanich, *J. Environ. Sci. (China)*, 23(2011)396.
2. D. Song, C. Xu, A.L. Holck, R. Liu, *Ecotox. Environ. Safe.*, 208(2021)14.
3. R. Madmanang, S. Jangkorn, J. Charoenpanich, T. Sriwiriya, *Environ. Eng. Res.*, 24(2019)33.
4. Z. Es'haghzade, H. Bahrami, M. Arami, E. Pajootan, *J. Taiwan Inst. Chem. E.*, 71(2017)91.
5. T.X.H. Le, T.V. Nguyen, Z. A. Yacouba, L. Zoungrana, F. Avril, D.L. Nguyen, E. Petit, J. M Eendret, V. Bonniol, M. Bechelany, S. Lacour, G. Lesage and M. Cretin, *Chemosphere*, 172(2017)1.
6. O.T. Can, M. Kobya, E. Demirbas and M. Bayramoglu, *Chemosphere*, 62(2006)181.
7. E. Brillas, *Chemosphere*, 250(2020)126198.
8. J. Chen, *Nanchang Hangkong Univ.*, 2019.  
<https://kns.cnki.net/KCMS/detail/detail.aspx?dbname=CMFD201902&filename=1019654493.nh>
9. W.L. Yang, M.H. Zhou, N. Oturan, Y.W. Li, P. Su, M.A. Oturan, *Electrochimica. Acta.*, 297(2019)582.
10. E. Isarain-Chávez, J.A. Garrido, R.M. Rodríguez, F. Centellas, C. Arias, P.L. Cabot and E. Brillas, *J. Phys. Chem. A*, 115(2011)1234.
11. S.C. Elaoud, M. Panizza, G. Cerisola and T.Mhiri, *J. Electroanal. Chem.*, 667(2012)19.
12. S.Y. Yuan, Y. Fan, Y.C. Zhang, M. Tong and P. Liao, *Environ. Sci. Technol.*, 45(2011)8514.
13. H. Qiao, M.Q. He, Q.S. Wang, S.S. Han, H.Q. Zhao and Z.H. WANG, *Water Sci. Technol.*, 83(2021)20.
14. I.A. Talalaj and P. Biedka, *Ecol. Eng.*, 85(2015)185.

15. G. Buftia, E. Rosales, M. Pazos, G. Lazar and M.A. Sanroman, *Sci. Total Environ.*, 635(2018)200.
16. L.D. Lei, Z.W. Zhou, Y. Gao, Y. Huang, X.W. Duo, Y.P. Cui and Z.Q. Huang, *Safety and Environ. Eng.*, 28(2021)108.
17. J.Y. Li, Z.W. Ji and C. Kang, *Water Purification Technology*, 39(2020)103.
18. X.J. Zhuang, W.Y. Yao and L.M. Liu, *Guangdong Chemical Industry*, 44(2017)173.
19. S.G. Su, J. Wang, L.L. Zhang and Z.X. Pei, *Environmental Protection of Chemical Industry*, 34(2014)443.
20. Q. Jiang, *Hangzhou Dianzi Univ.*, 2019.  
<https://kns.cnki.net/KCMS/detail/detail.aspx?dbname=CMFD202001&filename=1019023054.nh>
21. Y. Zhang, L. Liu, X.D. Sun, R.Q. Zhang and L. Wang, *Materials protection*, 41(2008)31.
22. Y.H. Cui, Y.J. Feng and J.F. Liu, *Journal of Functional Materials*, 36(2005)234.
23. H. Liu, C. Wang, X.Z. Li, X.L. Xuan, C.C. Jiang and H.N. Cui, *Environ. Sci. Technol.*, 41(2007)2937.
24. S. Liu, Z.Y. Wang, J.F. Li, C. Zhao, X.L. He and G. Yang, *Chemosphere*, 213(2018)377.
25. J.K. Xiong, G.Y. Li, P.A. Peng, F. Gelman, Z. Ronen and T.C. An, *Chemosphere*, 258(2020)127378.
26. H.Z. Zhao, Y. Sun, L.N. Xu and J.R. Ni, *Chemosphere*, 78(2009)46.
27. F. Zhang, J.G. Zhan, X.W. Li, M. Qiao, R. Mao, H. Wang and X. Zhao, *Chinese J. Environ. Eng.*, 14(2020)2428.
28. X.L. Yao, X. Qin and W. Liu, *Sichuan Environment*, 3(2008)6.  
DOI:10.14034/j.cnki.schj.2008.03.002.
29. Y.Q. Liu, P.F. Yuan, H.X. Zhang and Z.G. He, *Technology of Water Treatment*, 47(2021)57.
30. Y.W. Yao, M.Y. Li, Y. Yang, L.L. Cui and G. Lin, 216(2019)812.
31. M.A. Oturan, *J. Appl. Electrochem.*, 30(2000)475.
32. S.H. Kim and K.Y. Ann, *Int. J. Environ. Sci. Te.*, 5(2010)302.
33. S.H. Tong, Shenyang Jianzhu University, 2016.  
<https://kns.cnki.net/KCMS/detail/detail.aspx?dbname=CMFD201902&filename=1018125312.nh>
34. W.B. Li, G.H. Wang, D. Wan, K. Chen, Q. Hu, L.L. Lu and L.U. Lu, *Korean J. Chem. Eng.*, 33(2016)1557.
35. J.L. Chen, H.C. Shi and L.L. Xu, *Environmental Science*, 8(2008)2277.  
DOI:10.13227/j.hjlx.2008.08.035.
36. S.O. Fatin, H.N. Lin, W.T. Tan and N.M. Huang, *Int. J. Electrochem. Sci.*, 7(2012)9074.
37. Z. Wu, W. Zhu, Y. Liu, P. Peng, X. Li, X. Zhou and J. Xu, *Chemosphere*, 246(2020)125703.
38. C.K. Sim, S.R. Majid and N.Z. Mahmood., *Int. J. Electrochem. Sci.*, 10(2015)10157.
39. H.R. Zakeri, M. Yousefi, A. Mohammadi, M. Baziar, S.A. Mojiri, S. Salehnia, A. Hosseinzadeh, *Int. J. Environ. Sci. Te.*, 18(2021)12.
40. Z.S. Iro, C. Subramani, S.S. Dash, *Int. J. Electrochem. Sci.*, 11(2016)10628.