Experimental study on treatment of copper Ions in water by circulating electric-flocculation

Zhijie Zhang  Quan Wang  Linlin Xu  Yong Chen*
(College of Petrochemical and Energy Engineering Zhejiang Ocean University, Zhoushan, Zhejiang, China, 316022)

* Corresponding author: Chen Yong (1984-), E-mail: Chenyong@zjou.edu.cn

Abstract: This study is to change the treatment method of electric-flocculation technology, and the water to be treated enters the electrolysis tank in the form of circulation. Comparison of efficiency and energy consumption by circulating electric-flocculation and ordinary electric-flocculation. Explore the feasibility and energy consumption of the cyclic electric-flocculation method. It is found that the common electric-flocculation method and the cyclic electric-flocculation method have the same removal effect when the copper ion concentration is high, and the effect of voltage on the removal effect is very small; when the copper ion concentration is low, the ordinary electric-flocculation method has better removal effect than the cyclic type. The effect of the removal is good, the voltage has little effect on the removal effect of the common electric-flocculation method, and the effect on the circulation type is greatly affected; as the voltage rises, the limit of the copper ion concentration which is different between the removal effect of the ordinary electric-flocculation method and the circulation type is lowered; Under the same conditions, the energy consumption of ordinary electric-flocculation is always higher than that of the circulating type.

1. Foreword
In recent years, the problem of heavy metal pollution of water resources has become increasingly prominent and heavy metal ions with excessive concentrations have serious toxic effects on aquatic animals, often affecting the normal life activities of aquatic animals, causing their poisoning and death, which seriously affects aquatic products. This is only a small part of the harm to the environment and people. Therefore, the problem of heavy metal pollution and its control has aroused widespread concern. The electric-flocculation is a new technology in the field of water treatment, which is widely used, mainly in the treatment of heavy metal waste-water and dye wastewater. In the treatment of heavy metal wastewater, chromium and copper are treated more. For example: Liu Yu ling et al use electric-flocculation and activated carbon technology to treat chromium in industrial wastewater and reduce it to a certain concentration. In the treatment of dye wastewater, the treatment of acid dyes is mostly. The technology began to be applied to the treatment of seawater pollution. For instance: Wang uses electric-flocculation technology to treat polluted seawater. In order to study the electric flocculation technology in depth, the electric flocculation device is usually changed or used in conjunction with other ways. For examples: Tan Zhu uses aluminum-iron electricide combined with electric-flocculation technology to treat Cu-EDTA wastewater; Liu Yan[7] aluminum-titanium electric-flocculation process for papermaking wastewater treatment and mechanism; Sun Flocculation-biochemical method for the treatment of dye waste water. The mechanism of electric flocculation has three aspect: flocculation, air flotation, and oxidation-reduction. It can be known
from the principle of electric-flocculation technology that the technology has obvious characteristics compared with other technologies and its characteristics are obvious but also have a disadvantage at the present stage: industrial intermittent operation, high energy consumption.

The research on the treatment stage of water by circulating electric-flocculation technology is still relatively small and the efficiency of dealing with heavy metals is still unknown compared with ordinary electric-flocculation, so this research is still of great significance. In this paper, the removal of copper ions in water is taken as an example. When the water bodies are respectively entered into the electrolytic cell in the form of circulation and direct storage, the copper ions are removed by electric-flocculation, the effects of various factors and the energy consumption changes are compared. The results can guide the improvement of the electrical flocculation device so that it can operate continuously and reduce energy consumption. Provide guidance and basic support for the industrial and practical application of cyclic electric-flocculation heavy metal treatment technology.

2. Experimental Design

2.1. Experimental device and analytical instrument

2.1.1. Electric-flocculation experimental device

The schematic diagram of the experimental device is shown in figure 1; the diagram is divided into three parts: the first part is the electrolysis cell; the second part is the electrolysis unit; the third part is the material tank and the fourth part is the other parts. The electrolyze cell is made of Alec sheet and its length, width and height are 20cm, 20cm and 15cm respectively. Its effective volume is 4L. The raw material tank is made of a pressure plate likewise, and its length, width and height are 10cm, 10cm and 20cm respectively. The electrolysis device of the second part is an electrode made of an aluminum plate, a carbon rod. The specification of the aluminum plate is 100 x 100 x 10 mm. The specification of the carbon rod is Ø20mm. Other components include the power supply, the voltage range is 0~31V, and the output of different voltages can be set; the two small pumps have a specification of 12V/A. The electrolysis unit was placed in an electrolysis cell. When the valve 1 is opened and the valve 2 is closed, the pump 1 can pump all the prepared solution in the raw material tank into the electrolytic tank. At this time, an ordinary electric-flocculation experiment can be performed; when the valve 1 and the valve 2 are opened and the flow rate is adjusted The liquid level in the electrolytic cell and the transition tank is the same, and circulating electric-flocculation can be performed at this time.

(a) power source, (b) electrolyzer, (c) valve 2, (d) strainer, (e) pump 2, (f) raw material tank, (g) valve 1, (h) pump 1, (i) sample connection

Figure 1. Experimental device schematic.
2.1.2. Experimental water body and analytical method
The tested water samples are prepared by laboratory containing in Cu$^{2+}$ water. The water sample is configured by using the laboratory tap water, and every time to experiment with five water copper sulfate prepared 4 L of Cu containing 200 mg/L Cu$^{2+}$ is introduced. And sodium chloride is used to adjust the conductivity to stabilize at about 2ms/cm. The pH was measured at 7-8. Analytical. Projects and analytical methods during the experiment Table 1 Shown

| Serial number | Analysis Project        | Analytical Instruments               |
|---------------|-------------------------|--------------------------------------|
| 1             | PH value, conductivity  | Ct-6322ph meter / conductivity meter |
| 2             | Voltage and current     | Student power                        |
| 3             | Cu$^{2+}$ concentration | Atomic absorption spectrophotometer   |

2.1.3. Experimental Materials
(1) Electrode material

| material          | specification |
|-------------------|---------------|
| Aluminum plate    | 100x100x10mm  |
| Carbon rod        | 20mm x10mm    |
| Pressure plate    |               |
| Copper wire       |               |

Laboratory reagents

(2) The reagent

| Drug Name          | level       | Manufacturer                                   |
|--------------------|-------------|-----------------------------------------------|
| Copper sulfate penta hydrate | Analytical purity | Sinopharm Chemical Reagent Co., Ltd. |
| Sodium chloride    | Analytical purity | Sinopharm Chemical Reagent Co., Ltd. |
| Nitric acid        | Analytical purity | Shanghai Susong Chemical Reagent Co., Ltd. |

2.2. Experimental steps and data analysis methods

2.2.1. Experimental procedure
(1) Preparation: That prepare a concentration of 200mg / L Cu$^{2+}$ the solution was 4 L and about 20 g of NaCl was used to adjust the conductivity to 2ms / cm. Install the experimental device, connect the positive electrode of the power supply to the aluminum plate, and connect the negative electrode to the graphite rod. Turn on the 1 and 2 pumps for flow balance adjustment to balance the flow.

(2) Circulating electric-flocculation at different voltages: Adjust the regulated power to supply for regulated output. Then collect the sample according to the time node, collect the water sample at the bottom of the raw material tank, and collect the sample every 10 minutes: 0, 10, 20... 60min. A total of 7 samples were collected. And record the current and voltage of each node.

(3) Perform common electric-flocculation at different voltages: adjust the regulated power to supply for regulated output. Then collect the sample according to the time node, collect the water sample at the bottom of the electrolytic cell and collect the sample every ten minutes: 0, 10, 20... 60min. A total of 7 samples were collected. And record the current and voltage of each node.

(4) Data processing: The collected sample is filtered, and after filtration, the sample is measured by
an atomic absorption spectrophotometer. Cu$^{2+}$ The concentration is then calculated by calculating the copper ion removal rate, and the current and voltage are used to calculate the consumed electric power.

2.2.2. Data analysis method

(1) The method for determining copper ions in this experiment is to refer to the relevant national standards. The lowest concentration that can be detected by this method is 0.5 mg/L, and the highest can be measured at 5 mg/L. The measured concentration was determined by the following formula to determine the copper ion concentration for the corresponding period of time.

Copper ion removal rate calculation formula: 

$$ R = \frac{C_0 - C_f}{C_0} \times 100\% $$

- $R$ - Cu$^{2+}$ Removal rate%;
- $C_0$ - Experimental water Cu$^{2+}$ Initial concentration mg/L;
- $C_f$ - After treatment in the water body Cu$^{2+}$ Initial concentration mg/L

(2) The voltage and current of this experiment were measured directly on the power supply. Calculation formula for power consumption: $P=U\times I$

- P - electric power W;
- U - voltage V;
- I - current A;

3. Experimental results and discussion

3.1. Different voltages Cu$^{2+}$ Remove impact

![Figure 2. Ordinary and cyclic electric-flocculation at different voltages Cu$^{2+}$ Removal rate as a function of processing time.](image)

The experimental conditions of ordinary electric-flocculation and circulating electric-flocculation are Cu$^{2+}$ which initial concentration is 200 mg/L, the electrolyte NaCl mass fraction is 0.9%, and the initial pH is 7-8. To adjust the different voltages, the corresponding voltage is 6V, 9V, and then power on for 60 minutes.

As shown in figure 2, when the concentration is higher than 120 mg/L, the common electric-flocculation method and the cyclic electric-flocculation method have the same removal
effect, and the voltage has a very small effect on the removal effect; at a concentration lower than 120 mg/L, the voltage is 6V. The removal rate of the electric-flocculation method began to decrease. At a concentration lower than 40 mg/L, the removal rate of the cyclic electric-flocculation method with a voltage of 9 V began to decrease. Therefore, for different voltages, there is a limit concentration of heavy metals in the circulating electric-flocculation method. When the limit concentration is higher, the removal effect of the ordinary electric-flocculation method is the same as that of the circulating electric-flocculation method. When the limit concentration is lower, the removal effect of the ordinary electric-flocculation method is better than that of the circulating electric-flocculation method. The limit concentration is related to the magnitude of the voltage, and as the voltage rises, the limit concentration decreases.

The main function of the ordinary electric flocculation method is realized by an electric field. The higher the voltage, the higher the current density between the plates; the reaction rate between the anode and the cathode is related to the current density \([10]\); the greater the current density, the reaction rate becomes, the greater the anode of \(\text{Al}^{3+}\) quantity will increase, increases the floc \(\text{Cu}^{2+}\) removal rate increased; at the same time, the cathode also generates more bubbles, which increases the air flotation effect, making the treatment effect better. However, the increase of the current density will also accelerate the passivation of the electrode, so the effect of the voltage on the ordinary electric-flocculation method is very small. However, circulating electric-flocculation method because of the way the water flow will affect the air flotation effect, which role in the electric field, flow field, there is the effect of flow field in low concentrations \(\text{Cu}^{2+}\) can not be good at the plate gathered themselves together, and so lead to just below the limit concentration, removal rate began to reduce. The increase in voltage enhances the effect of the electric field and enhances this aggregation ability, so there was a significant difference between 9V and 6V, and the limit concentration decreases with the increase of voltage.

### 3.2. Comparison of electric power consumption between circulating electric flocculation and ordinary electric flocculation

![Figure 3](image_url)

Figure 3. the electric power of ordinary electric flocculation and circulating electric flocculation with time under 6v voltage.
The experimental conditions for the two methods of electric-flocculation: Cu^{2+} which the initial concentration is 200 mg/L solution 4 L, the electrolyte NaCl mass fraction is 0.9%, and the initial pH is 7-8. The output voltage is 6V and 9V.

As shown in Figure 3. It is shown that at the same voltage, the electric power consumed by ordinary electric flocculation is always higher than the electric power of the circulating electric flocculation. The reason why the power of ordinary electric-flocculation is higher than that of the circulating electric-flocculation is that the volume of the solution in the electrolytic cell of the circulating electric-flocculation is about half that of the ordinary electric-flocculation when the solution of the same volume is processed, which will greatly reduce the current density in the circulating electric-flocculation electrolytic cell compared with the ordinary electric-flocculation. The second reason is the polarization of the electrode. There are two polarization forms in the electrolyte, concentration polarization and electrochemical polarization. The circulating electric flocculating water turbulence can greatly reduce concentration polarization, thus reducing the electric power. The polarization effect of common electric flocculation is strong, and the terminal voltage of the electrolytic cell will continue to increase. Since the electrolytic cell is a stable voltage input, the current density will increase with time, and the energy consumed at this time will also increase, so the power consumed by common electric flocculation is higher than that of the circulating electric flocculation.

4. Experimental conclusions and prospects

There is a limit concentration of heavy metals in the circulating electric-flocculation method. When the limit concentration is higher, the removal effect of the ordinary electric-flocculation method is the same as that of the circulating electric-flocculation method. When the limit concentration is lower, the removal effect of the ordinary electric-flocculation method is better than that of the circulating electric-flocculation method. The reason is that the circulating electric flocculation has the effect of flow field in addition to the electric field. Under the limit concentration, the effect of the flow field is greater than that of the electric field, which destroys the accumulation of heavy metal ions on the plate.

The limit concentration is related to the magnitude of the voltage, and as the voltage rises, the limit concentration decreases. When the voltage is increased, the electric field is enhanced and the gathering ability is enhanced.

The power consumed by the circulating electric-flocculation is much lower than the power consumed by ordinary electric-flocculation in the same situation. By selecting the voltage, the removal effect can be made the required range, but the ordinary electric flocculation energy consumption is high, and the circulating electric flocculation energy consumption is relatively low.

Outlook: There is only electric field in ordinary electric flocculation, but there are flow field and electric field in circulating electric flocculation. Under the same flow rate, the influence of flow field on electric field and removal effect is not explored in this experiment, but it will be explored in the next experiment.

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