

## Study on influencing parameters and long-term operation of electrocoagulation phosphorus removal from small rural domestic sewage

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### ABSTRACT

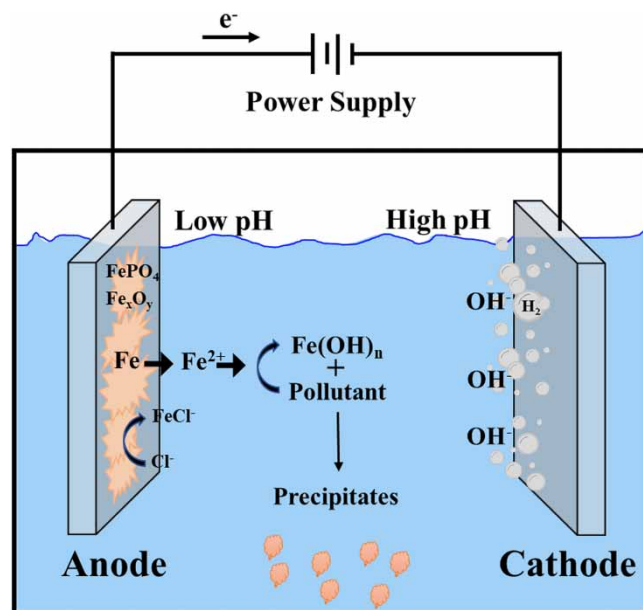
Excessive discharge of phosphorus can produce eutrophication in aquatic environments, damaging public health, the living environment, and the economy. The conventional mechanical–biological phosphorus removal methods are not suitable for small rural domestic sewage due to the features of small scale, scattered distribution, intermittent emission, and large fluctuation. This work evaluated electrocoagulation (EC) with industrial steel as electrodes on small rural domestic sewage. Results showed that the best performance was achieved at a current density of 1 mA/cm<sup>2</sup>, electrode distance of 2 cm, electrode number of 2, pH of 7, and Hydraulic Retention Time of 30 min, respectively. Under optimum conditions, the EC process removed 93.91% phosphorus while consuming around 0.25 kWh/m<sup>3</sup> of electricity. In addition, the electrode passivation of EC was further investigated; the long-term research found that the phosphorus removal efficiency only decreased by 4.34% after 10 days of continuous flow operation, and the operational energy consumption was 0.07 kWh/m<sup>3</sup> at a Cl<sup>-</sup> concentration of 500 mg/L.

**Key words:** electrocoagulation, electrode passivation, energy consumption, phosphorus removal, small rural domestic wastewater

### HIGHLIGHTS

- EC was developed to remove phosphorus from small rural domestic sewage.
- Optimization of the EC process can effectively reduce energy consumption.
- Current density and HRT were positively correlated with removal efficiency.
- Adding Cl<sup>-</sup> can inhibit electrode passivation.

### GRAPHICAL ABSTRACT



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## 1. INTRODUCTION

Phosphorus is one of the essential nutrients for living organisms and has been widely used as agricultural fertilizers and animal supplements in recent decades (Vasudevan *et al.* 2008). Excessive discharge of phosphorus into water (phosphorus concentration  $>0.03$  mg/L) will lead to eutrophication of surface water, followed by a series of environmental problems such as oxygen consumption, water quality deterioration, fish death, and biodiversity reduction (Bennett *et al.* 2001). The A<sup>2</sup>/O process has become a preferred treatment method in many urban areas. However, due to the characteristics of small discharge, scattered distribution, intermittent emission, and large quantity fluctuation (Devlin *et al.* 2017), it is impossible and uneconomical to apply in small rural domestic sewage (SRDS). Therefore, it is critical to develop a suitable means to control the phosphorus emission in small rural areas. Compared with traditional methods, the electrocoagulation (EC) technique has the advantages of simple maintenance, low infrastructure cost, short residence time, and fast settlement of flocculation (Betancor-Abreu *et al.* 2019; Tchamango *et al.* 2021). Therefore, this technique can be used for phosphorus removal.

In the EC process, the anode metal electrode is dissolved into metal ions under the action of applied potential, and electrode reactions are (Asaithambi *et al.* 2016):



When the reaction occurs in an acidic medium, Fe<sup>2+</sup> cations will oxidize very slowly when they come into contact with dissolved oxygen (Equation (3)). When the reaction occurs in a neutral or alkaline medium, Fe<sup>2+</sup> will be immediately converted to ferrous hydroxide (Equation (4)). It is rapidly oxidized by dissolved oxygen to ferric hydroxide (Equation (5)).



This process allows the phosphorus contained in the wastewater to be separated and removed by flocculation and formation of precipitation (Al-Qodah *et al.* 2020). Recent studies approved that EC can effectively remove phosphorus from wastewater. Franco *et al.* (2017) achieved 99% phosphorus removal in surface water using double Al electrodes at a current intensity of 2 A and a reaction time of 60 min. Yang *et al.* (2022) reported that the removal efficiency of PO<sub>4</sub><sup>3-</sup>-P (1.30 mg/L) could reach 98% within 2 min at a current density of 10 mA/cm<sup>2</sup>, and the final concentration was 0.01–0.07 mg/L. Devlin *et al.* (2019) found that when the pH of the Fe electrode was 4, the current density was 20 A/m<sup>2</sup>, and the EC time was 100 min, the system can reduce the concentration of phosphorus-containing wastewater from 52 mg/L to below 0.01 mg/L. Therefore, those critical achievements of the EC process in phosphorus removal from surface water provide a possibility for us to treat SRDS.

In addition, electrode passivation inevitably occurs during the long-term operation of EC. The passivation layer hinders the metal oxidation reaction and increases the resistance of the electrode surface, which raises operating costs and reduces removal efficiency (Bandaru *et al.* 2020). Previous studies on passivation problems, such as composite electrodes, have been found to be complex to operate, difficult to maintain, and expensive (Han *et al.* 2021), which may not be suitable for small rural areas where portability and economy are essential factors to consider. However, adding active ions has been shown to be effective in alleviating the passivation phenomenon (Bandaru *et al.* 2020). Chloride ions are particularly suitable for rural areas due to their low cost and ease of operation. In this study, a simple and cost-effective electrochemical treatment process using low-cost materials and a straightforward setup to remove phosphorus from wastewater in small villages was developed. This work is significant as it provides a practical and sustainable solution for rural areas. Therefore, this research used the EC to solve the phosphorus pollution problem in SRDS and explored how to alleviate electrode passivation effectively.

To date, there are no reports on phosphorus removal under continuous flow influent and long-term operating conditions in detail. However, continuous flow influent and long-term operation are the main factors determining the practical application.

Therefore, this study first discussed the feasibility of EC removal of phosphorus in SRDS, and then the effects of different process parameters (current density, electrode distance, electrode number, hydraulic retention time (HRT), and pH) on phosphorus removal were investigated. Finally, the change in removal efficiency and operation energy consumption during the long-term operation was further discussed at the optimum conditions, and a solution to the electrode passivation was provided.

## 2. METHODS

### 2.1. Material

All reagents used in this study ( $\text{KH}_2\text{PO}_4$ , NaOH, HCl, NaCl) were of analytical purity and were purchased from Sinopharm Chemical Reagent Co., China. Desired concentrations of phosphorus solutions used in the experiment were synthesized by dissolving  $\text{KH}_2\text{PO}_4$  in domestic sewage from a factory dormitory in Jiangsu, China. The chemical characterization of the domestic wastewater used is presented in Table 1. The pH was adjusted using hydrochloric acid and sodium hydroxide solutions (0.50 mol/L). Ultrapure water was used during the experiments.

### 2.2. Experiment

Industrial iron plates were used as electrodes. The reactor was made of plexiglass, and the length, width, and height of the experimental reactor were 20 cm, 20 cm, and 30 cm, and the working volume was 10 L (20 cm  $\times$  20 cm  $\times$  25 cm). The overflow port was set at 25 cm from the bottom of the reactor. The sizes of the electrodes were 15 cm  $\times$  20 cm  $\times$  0.5 cm, and the immersion area was 15 cm  $\times$  15 cm. Two parallel rods set in the reactor were used to support electrodes. Due to the significant water consumption associated with continuous flow long-term operation, the reactor size for our long-term experiment was set at 10 cm  $\times$  10 cm  $\times$  15 cm, and the working volume of the reactor was 1 L (10 cm  $\times$  10 cm  $\times$  10 cm). The overflow port was set at 10 cm from the bottom. The immersion area of the electrodes was 36 cm<sup>2</sup> (6 cm  $\times$  6 cm). The experimental setup used in this study is shown in Figure 1.

The electrodes were placed in a 36–38% hydrochloric acid bath to remove rust and oxide film. Before the experiments, the electrodes were polished with sandpaper and cleaned with ultrapure water.

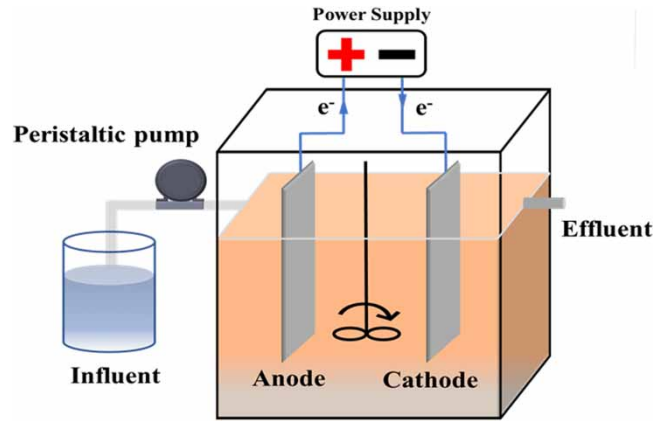
To find the optimum process parameters for phosphorus removal by EC, experiments were conducted using a continuous flow EC system with iron electrodes on both the anode and cathode. The electrical parameter, electrode parameters (electrode distance, electrode number), and hydraulic parameters (HRT and pH) were considered as the three essential parameters. Furthermore, the effect of EC on phosphorus removal during the long-term operation was investigated to solve electrode passivation.

### 2.3. Analytical methods

The reactor was initially charged with wastewater, and then the flow and electricity were concurrently initiated. Following the attainment of stabilized effluent phosphorus concentration, wastewater samples were collected from the overflow port and subsequently subjected to filtration through a 0.45- $\mu\text{m}$  cellulose acetate membrane filter (Schleicher and Schuell) for subsequent analysis. All analyses were performed within 24 h of sample collection. The analysis was repeated three times,

**Table 1** | Characteristics of the domestic wastewater used in this study

Parameters	Values
Chemical oxygen demand (mg/L)	182 $\pm$ 21
Total suspended solids (mg/L)	245 $\pm$ 28
Total phosphorus (mg/L)	4.52 $\pm$ 0.31
Total nitrogen (mg/L)	39.35 $\pm$ 7.5
Nitrate nitrogen (mg/L)	1.55 $\pm$ 0.35
Ammonium nitrogen (mg/L)	17.2 $\pm$ 1.8
Conductivity ( $\mu\text{S}/\text{cm}$ )	538 $\pm$ 21
Turbidity (nephelometric turbidity unit)	120 $\pm$ 14
pH	7.15 $\pm$ 0.25



**Figure 1** | Schematic diagram of the experimental setup.

and the results were expressed as mean values. The concentration of phosphorus was determined by the ammonium molybdate spectrophotometric method (National Standard of the People; Republic of China 1989). The voltage was recorded by a direct-current power supply (Maisheng-MS3610DS), the mass was measured by analytical balance (Tianmei-FA1204B) and the conductivity and pH were measured by a portable multiparameter analyzer (Sanxin-EC5).

## 2.4. Calculations

- (1) Phosphorus removal efficiency

$$\eta = \frac{C_0 - C_t}{C_0} \times 100\% \quad (6)$$

where  $\eta$  is phosphorus removal efficiency (%),  $C_0$  is the initial concentration of phosphorus in the water sample (mg/L), and  $C_t$  is phosphorus concentration at time  $t$  (mg/L).

- (2) Current density calculation method:

$$\rho = \frac{1,000I}{2S} \quad (7)$$

where  $\rho$  is the current density (mA/cm<sup>2</sup>),  $I$  is current (A), and  $S$  is the electrode area (cm<sup>2</sup>).

- (3) Energy consumption calculation

Energy consumption in the EC process was calculated as follows:

$$EC = \frac{UIt}{V} \quad (8)$$

where  $EC$  refers to the energy consumption (kWh/m<sup>3</sup>),  $U$  is voltage (V),  $I$  is current (A),  $t$  is time (s), and  $V$  is water volume (m<sup>3</sup>).

- (4) Calculation of theoretical production of metal ions.

Metal ions would be expected following Faraday's law, and the current can control the total dose. Theoretical production of metal ions was calculated using the following equation:

$$\text{Theoretical production of metal ions} = \frac{ItM}{zF} \quad (9)$$

where  $I$  is current intensity (A),  $F$  is the Faraday constant (96,485 C/mol),  $t$  is electrolytic time (s),  $M$  is the molar mass of metal (g/mol), and  $z$  is the charge transfer number.

(5) The amount of sludge

$$\text{The amount of sludge} = \frac{m_1 - m_0}{V} \quad (10)$$

where  $m_1$  is the total mass after drying (mg),  $m_0$  is the mass of filter paper after drying (mg), and  $V$  is the volume of filtered water sample (L).

## 2.5. Statistics

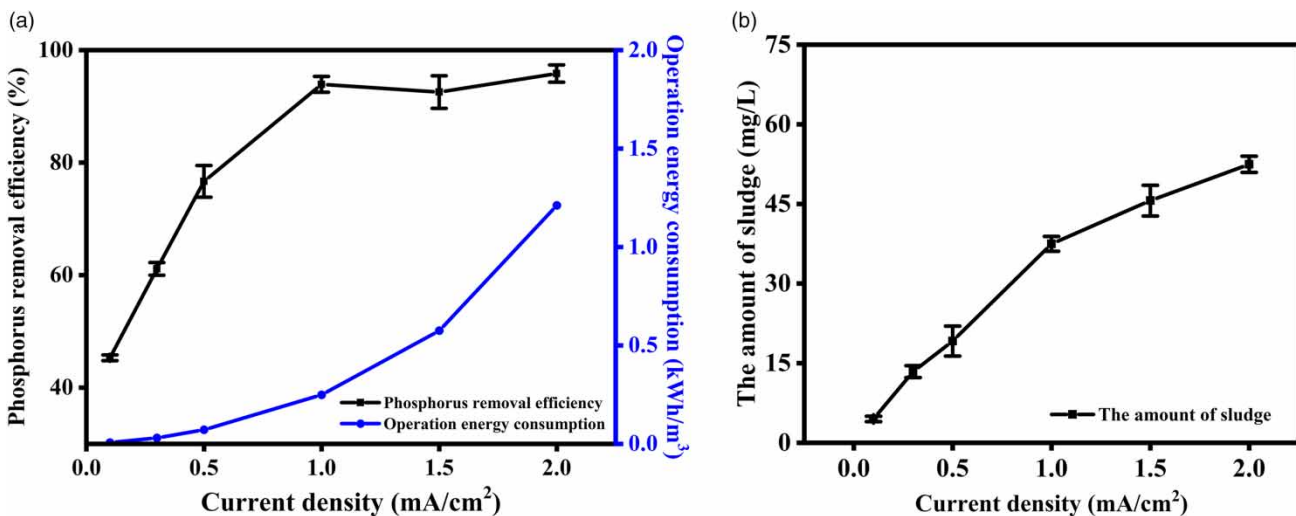
All statistical analyses were performed through the statistical program SPSS, including analysis of variance, and correlation analysis using the Spearman algorithm. All of the data were obtained by repeating the experiments three times.

## 3. RESULTS AND DISCUSSION

### 3.1. Effect of current density

The current intensity is a crucial parameter affecting phosphorus removal and operation energy consumption in the EC process. Figure 2(a) illustrates the effect of current intensity on phosphorus removal efficiency and energy consumption. When the current density was 0.1, 0.3, 0.5, 1, 1.5, and 2 mA/cm<sup>2</sup>, the phosphorus removal efficiency reached 45.28, 61.11, 76.67, 93.91, 92.55, and 95.83%, respectively. This result indicates that the phosphorus removal efficiency improved with the increase of current intensity from 0.1 to 1 mA/cm<sup>2</sup>, which was consistent with Faraday's law. It implies that the increase of current density increases the number of iron ions, thereby enhancing the production of flocculants. Meanwhile, the operation energy consumption of the EC process increased significantly with current density, and the operation energy consumption was 0.01, 0.03, 0.07, 0.25, 0.58, and 1.21 kWh/m<sup>3</sup>, respectively. When the current density was 1 mA/cm<sup>2</sup>, the phosphorus removal efficiency of EC reached 93.91%, and the energy consumption was 0.25 kWh/m<sup>3</sup>. However, when the current density was further increased, the phosphorus removal efficiency had almost no significant change with the increase in energy consumption. This result indicated that there was a threshold in the increase of current density on phosphorus removal efficiency. To prevent the potential rise leading to the anode side reaction and high operation energy consumption, the current intensity used in subsequent experiments was 0.5 mA/cm<sup>2</sup>.

The sludge generation during the EC process was analyzed, and the results are presented in Figure 2(b). It was observed that the amount of sludge increased linearly from 4.49 to 52.46 mg/L as the current density increased from 0.1 to 2 mA/



**Figure 2** | Effect of current density on phosphorus removal and operation energy consumption (a) and the amount of sludge (b) (phosphorus concentration = 5 mg/L, electrode distance = 2 cm, electrode number = 2, HRT = 30 min, pH 7).

cm<sup>2</sup>. Due to the fact that the sludge was mainly composed of flocculants generated through the hydrolysis and adsorption of iron ions during the EC process, at high current densities, part of the flocs settled at the bottom of the reactor. Therefore, with increasing generation of Fe<sup>2+</sup> at high current densities, pollutants in the reactor would combine with Fe<sup>2+</sup> to form flocculants that would eventually lead to an increase in sludge production (Castaneda *et al.* 2020).

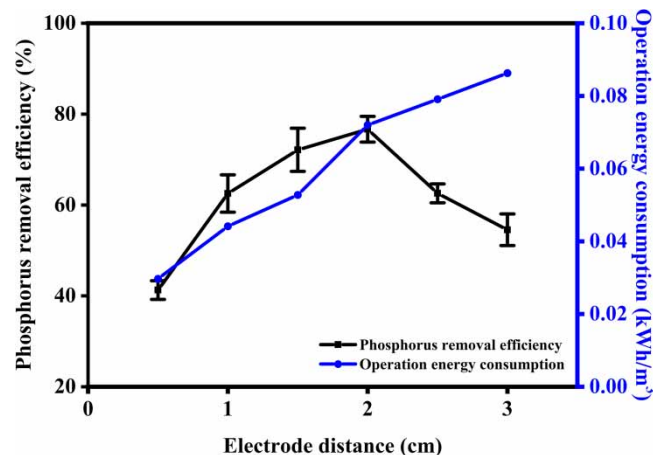
### 3.2. Effect of electrode parameters

#### 3.2.1. Effect of electrode distance

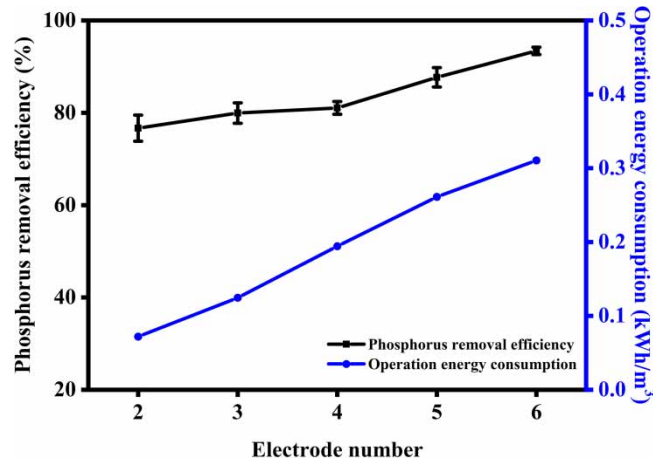
In the EC process, the electrode distance is associated with the resistance and the solution mass transfer rate, which may affect the phosphorus removal efficiency (Zeng *et al.* 2021). The effect of electrode distance on phosphorus removal and operation energy consumption are shown in Figure 3. When the electrode distance was 0.5, 1, 1.5, 2, 2.5, and 3 cm, the phosphorus removal efficiency was 41.26, 62.52, 72.14, 76.67, 62.56, and 54.54%, respectively, and the respective operation energy consumption was 0.03, 0.04, 0.05, 0.07, 0.08, and 0.09 kWh/m<sup>3</sup>. It was noteworthy that EC achieved 76.67% of maximum phosphate removal efficiency at 2.0 cm. The results of this experiment were consistent with the conclusions of Zhang *et al.* (2013). The removal efficiency of phosphorus was the lowest when the distance was 0.5 cm, and this is because a small electrode distance results in a concentration polarization effect, which reduces the electrode reaction rate. In contrast, a large electrode distance reduces the migration rate of ions between the electrodes, leading to poor flocculation. In addition, when the distance expands, the cathodic hydrogen evolution is weakened, which does not facilitate the shedding of flocs on the anode surface, thus hindering the electrode reaction. Omwene & Kobya (2018) observed that the sizes of flocs formed at different distances were also different. The larger the distance, the smaller the size of the flocs. Meanwhile, the operation energy consumption increased rapidly from 0.030 to 0.086 kWh/m<sup>3</sup> with the increase in electrode distance. Consequently, considering the removal efficiency of phosphorus and energy consumption, 2 cm was selected as the best electrode distance.

#### 3.2.2 Effect of electrode number

In the EC process, multiple electrode configurations can be divided into series and parallel according to the connection method, and can be divided into bipolar and monopolar by the polarity of the electrode surface. To compare the energy consumption and efficiency between multi-stage series connection and a single set of electrodes, the series bipolar (BP-S) configuration was chosen as it has been demonstrated to be effective for phosphorus removal. As shown in Figure 4, when the electrode number was 2, 3, 4, 5, and 6, the phosphorus removal efficiency was 76.67, 79.95, 81.06, 87.68, and 93.41%, and the operation energy consumption was 0.07, 0.13, 0.19, 0.26, and 0.31 kWh/m<sup>3</sup>, respectively. The more electrodes used in the EC process, the better the phosphorus removal. Because the removal efficiency has a direct relationship with the total electrode area, with the increase of the electrode area, the iron ions released from the anode increased, the reaction rate with phosphorus ions was accelerated, and the removal efficiency was improved. In addition, the multiple electrodes simultaneously generated flocculants to make the density distribution more uniform and effective so that more phosphorus



**Figure 3** | Effect of electrode distance on phosphorus removal and operation energy consumption (phosphorus concentration = 5 mg/L, current density = 0.5 mA/cm<sup>2</sup>, electrode number = 2, HRT = 30 min, pH 7).



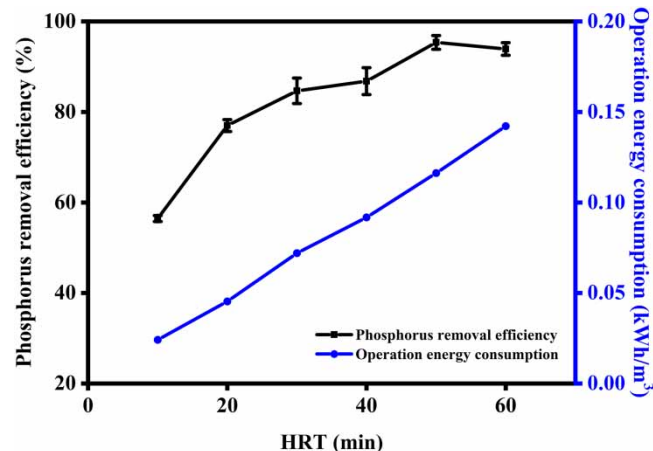
**Figure 4** | Effect of electrode number on phosphorus removal and operation energy consumption (phosphorus concentration = 5 mg/L, current density = 0.5 mA/cm<sup>2</sup>, electrode distance = 2 cm, HRT = 30 min, pH 7).

was precipitated and removed (Daneshvar *et al.* 2003). Furthermore, multiple electrodes can reduce the concentration polarization, so the effect of electrode passivation was alleviated. Consequently, the operation was more stable. Nevertheless, the operation energy consumption of EC increased fast with the increase of the electrode number. Considering that adding electrodes was less effective than increasing the current density under the same conditions, two electrodes were used for further experiments.

### 3.3. Effect of hydraulic parameters

#### 3.3.1. Effect of HRT

HRT affects the reaction time between flocculants and pollutants, determining the volume of the reactor. Therefore, the effect of HRT on the phosphorus removal process is worth exploring. As shown in Figure 5, when the HRT was 10, 20, 30, 40, 50, and 60 min, the phosphorus removal efficiency was 56.46, 77.03, 84.67, 87.80, 95.36, and 93.91%, respectively. It revealed that HRT influenced the phosphorus removal efficiency in the process. It was found that the removal efficiency of phosphorus increased gradually as the HRT increased from 10 to 50 min. In particular, when the HRT was 50 min, the phosphorus removal efficiency reached 95.36%, indicating that the phosphorus removal reached the limit. There was no apparent



**Figure 5** | Effect of HRT on phosphorus removal and operation energy consumption (phosphorus concentration = 5 mg/L, current density = 0.5 mA/cm<sup>2</sup>, electrode distance = 2 cm, electrode number = 2, pH 7).

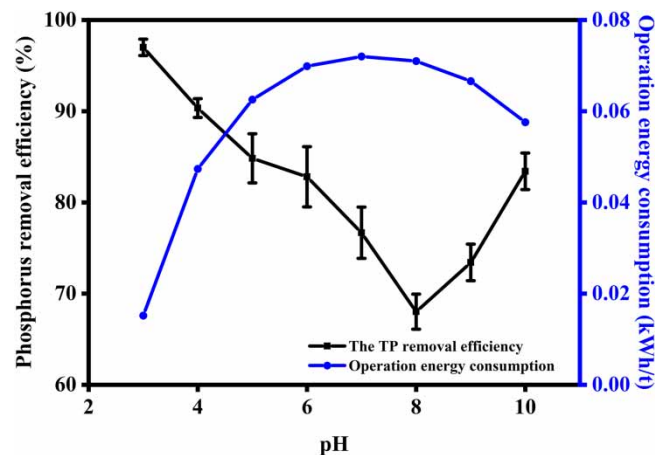
change with HRT further increasing. Meanwhile, the operation energy consumption was 0.02, 0.05, 0.07, 0.09, 0.11, and 0.14 kWh/m<sup>3</sup>, respectively. The longer the HRT, the higher the energy consumption. An HRT of 30 min was used in this study to achieve an efficient and energy-saving operation.

### 3.3.2 Effect of pH

The pH of wastewater determines the speciation of iron in the solution, which influences the performance of EC and significantly affects the treatment of phosphorus removal. When iron is used to remove phosphorus, it can either combine with phosphorus ions to form insoluble precipitates or form polynuclear hydroxyl complexes with long linear structures to absorb phosphorus through the hydrolysis reaction (Franco *et al.* 2017). As presented in Figure 6, when the pH was 3, 4, 5, 6, 7, 8, 9, and 10, the phosphorus removal efficiency was 97.00, 90.34, 84.83, 82.81, 76.67, 68.01, 73.40, and 83.40%, respectively, while the operation energy consumption was 0.02, 0.05, 0.06, 0.07, 0.07, 0.07, 0.07 and 0.06 kWh/m<sup>3</sup>. With the increase in pH, the removal efficiency of phosphorus gradually decreased. However, the phosphorus removal efficiency slightly increased when the wastewater was alkaline. At pH 3, the phosphorus removal efficiency of EC was the highest, reaching more than 97%. It demonstrated that the acidic environment was suitable for the phosphorus removal reaction. At pH < 3, iron mainly existed in Fe<sup>3+</sup> and Fe<sup>2+</sup>, which could be fully combined with phosphorus in the solution to form precipitation. As the pH increased from 3 to 8, more iron existed in the form of hydroxyl compounds, so phosphorus removal efficiency could not be further improved. Distinctly, when the pH was 8, the phosphorus removal efficiency was only 68.01% because the iron in the form of hydroxyl compounds could not react directly but removed phosphorus through charge adsorption. When the pH continued to rise to 10, part of the iron existed in the form of Fe(OH)<sub>3</sub>, and the phosphorus removal efficiency improved from 68.01 to 83.40%, indicating that the Fe(OH)<sub>3</sub> could facilitate to form the flocculation and sedimentation of phosphorus. Correspondingly, Table 2 suggests that the acidity or alkalinity increased the conductivity of the solution, resulting in the energy consumption decreasing. Nevertheless, EC had the highest phosphorus removal efficiency in acidic, alkaline, and neutral conditions, indicating that EC could be suitable for various water quality.

### 3.4. Correlation analysis

Figure 7 shows the correlation analysis of current density, electrode distance, electrode number, HRT, and pH on phosphorus removal efficiency and operation energy consumption. The correlation coefficients with the removal efficiency were that the current density was 0.54, HRT was 0.40, pH was 0.38, the electrode number was 0.21, and the electrode distance was 0.1. The current density and HRT were positively correlated with the removal efficiency. In contrast, the electrode distance was weakly correlated with the removal efficiency, and the pH was negatively correlated with the removal efficiency, consistent with the experimental results. The correlation coefficients between the five parameters of current density, electrode distance, electrode number, HRT, and pH, and operation energy consumption were 0.55, 0.48, 0.36, 0.31, and 0.13, respectively, indicating that current density was more positive than other parameters. In contrast, the correlation between pH and

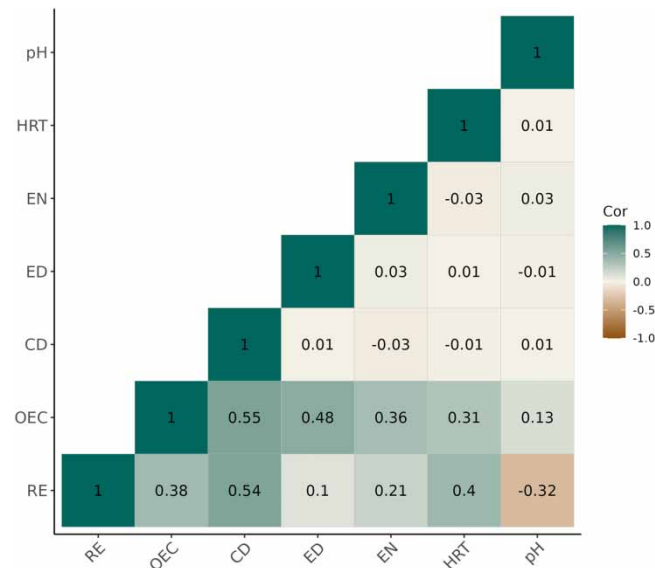


**Figure 6** | Effect of pH on phosphorus removal and operation energy consumption (phosphorus concentration = 5 mg/L, current density = 0.5 mA/cm<sup>2</sup>, electrode distance = 2 cm, electrode number = 2, HRT = 30 min, pH 7).



**Table 2** | pH and total dissolved solids of influent and effluent

Influent pH	Influent conductivity ( $\mu\text{S/cm}$ )	Effluent pH	Effluent conductivity ( $\mu\text{S/cm}$ )
3	1,940	3.05	1,972
4	479	4.76	503
5	320	8.86	382
6	287	8.91	307
7	265	8.92	299
8	271	9.23	302
9	284	9.36	314
10	365	10.26	410

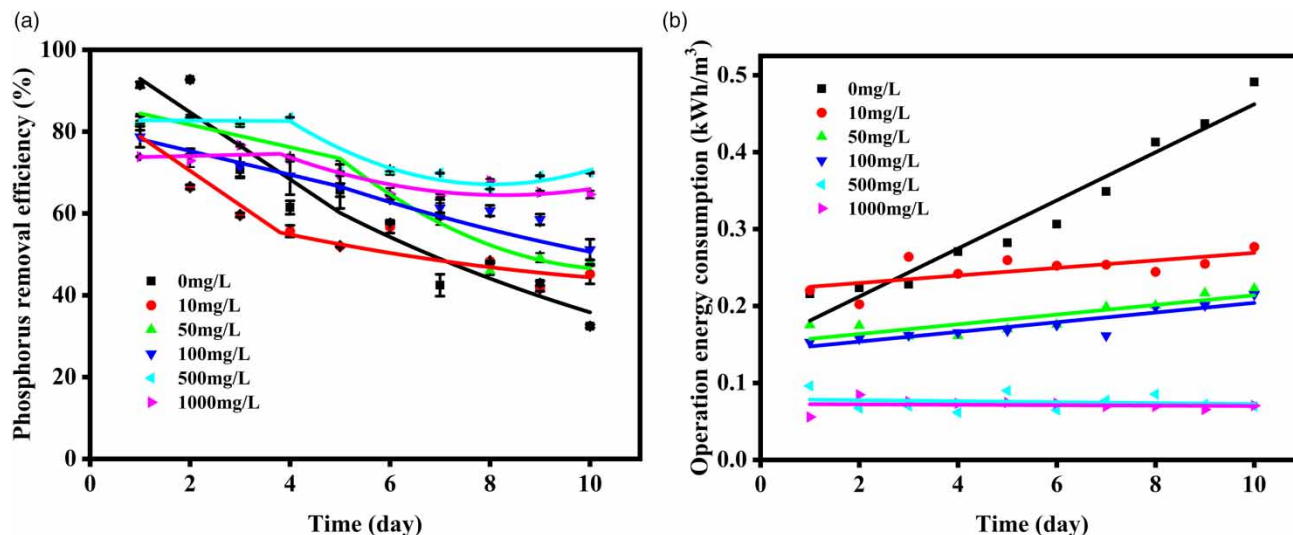


**Figure 7** | Correlation coefficients of different parameters and phosphorus removal efficiency and operation energy consumption (herein, CD, ED, EN, RE, and OEC represent current density, electrode distance, electrode number, phosphorus removal efficiency, and operation energy consumption, respectively).

operation energy consumption was not significant. Therefore, the effects of current density and HRT on removal efficiency, as well as the effects of current density and electrode distance on energy consumption should be fully considered in EC applications.

### 3.5. Effect of long-term operation

In the short-term experiment of the 10-L reactor, electrode passivation was found to be a key factor affecting the long-term stable operation of EC. In order to investigate the defense ability of the constructed device against electrode passivation under the optimum condition, a long-term experiment was conducted by only changing the reactor volume while keeping the water quality and HRT the same. As the reaction progresses, the precipitation of solids on the electrodes forms a passivation layer that hinders metal dissolution and increases the electrode–electrolyte interface resistance. Electrode passivation leads to an over-potential phenomenon and side reactions that result in higher energy consumption and lower phosphorus removal efficiency (Eyvaz 2016). The long-term operation of the EC was investigated using a 1-L reactor and bottom precipitation was removed after daily sampling. The results are shown in Figure 8. Adding 0, 50, 100, 500, and 1,000 mg/L of  $\text{Cl}^-$  to the influent water, the phosphorus removal efficiency of the initial operation of EC was 91.44, 82.37, 78.89, 82.31, and 73.82%, and the



**Figure 8** | Effect of  $\text{Cl}^-$  on phosphorus removal efficiency (a) and operation energy consumption (b) (phosphorus concentration = 5 mg/L, current density = 0.5 mA/cm<sup>2</sup>, electrode distance = 2 cm, electrode number = 2, pH = 7).

removal efficiency after 10 days was 47.19, 47.97, 51.15, 77.97, and 64.63%, respectively. The energy consumption of the initial operation of EC was 0.22, 0.22, 0.18, 0.15, 0.10, and 0.06 kWh/m<sup>3</sup>, respectively. The energy consumption after 10 days of operation was 0.50, 0.28, 0.22, 0.21, 0.07, and 0.07 kWh/m<sup>3</sup>, respectively.

Figure 8(a) indicates that the phosphorus removal efficiency first decreased linearly in the long-term operation of EC and then gradually decreased and stabilized. The initial phosphorus removal efficiency of the control group could reach more than 90%. With the operation of EC, the phosphorus removal performance continuously deteriorated. On the 10th day, the phosphorus removal efficiency was only 32.41%. However, the energy consumption continued to rise to 0.50 kWh/m<sup>3</sup> on the 10th day compared to the initial 0.22 kWh/m<sup>3</sup>.

The addition of  $\text{Cl}^-$  can effectively inhibit the passivation of the electrode, which leads to the reduction of the removal efficiency during operation. The phosphorus removal efficiency of the addition of 500 mg/L  $\text{Cl}^-$  decreased by only 4.34% within 10 days, while the phosphorus removal efficiency of the control group dropped by 59.03%. However, there was a threshold for the effect of  $\text{Cl}^-$ . When the initial phosphorus concentration was raised to 1,000 mg/L, the removal efficiency decreased to only 73.82% due to the occurrence of a side reaction ( $\text{Cl}^- \rightarrow \text{Cl}_2$ ) in the anode area (Zhou *et al.* 2020). The occurrence of anode side reactions can lead to a decrease in current efficiency, further affecting the phosphorus removal efficiency.

In addition, the addition of  $\text{Cl}^-$  increased the conductivity of the solution, thereby effectively reducing the operation energy consumption of EC. Figure 8(b) implies that when  $\text{Cl}^-$  was added to 500 mg/L, the operation energy consumption reached the lowest level; with further addition of  $\text{Cl}^-$ , the energy consumption showed no apparent changes. The addition of  $\text{Cl}^-$  could also effectively suppress the increase in energy consumption during the operation, and the  $\text{Cl}^-$  at a low concentration of 10 mg/L could be effective.

In conclusion, the addition of 500 mg/L  $\text{Cl}^-$  effectively inhibits electrode passivation, prevents the occurrence of anode side reactions, and enables a long-term stable operation during EC process.

### 3.6. Comparison with other EC phosphorus removal studies

In the current literature, many metal electrodes such as Fe, Al, and Al/Fe mixed electrodes have been used to remove phosphorus from water using the EC process. The results of phosphorus removal in synthetic aqueous solution and wastewater are shown in Table 3. The removal efficiency of phosphorus-containing wastewater by the EC process with Fe and Al electrodes can reach 45–100%. According to these results, the conditions for removing phosphorus were the initial pH value of 3–11, the current density of 0.6–150 mA/cm<sup>2</sup>, and the reaction time of 1–100 min, respectively. These studies have shown that EC can effectively remove phosphorus under various water quality conditions. Compared with other studies, the removal object of this study was SRDS, and the phosphorus concentration was about 5 mg/L. When the HRT was 30 min and the current

**Table 3** | Comparative effectiveness of EC on phosphorus removal

Ref.	Experimental conditions	Electrode material	Phosphorus removal efficiency	Energy consumption
Bouamra <i>et al.</i> (2012)	CD: 0.6–2 mA/cm <sup>2</sup> Initial PO <sub>4</sub> <sup>3-</sup> : 10 mg/L Initial pH: 3–9 Duration time: 10–90 min	Fe	90%	–
Mores <i>et al.</i> (2016)	CD: 16.3–57.5 mA/cm <sup>2</sup> Initial PO <sub>4</sub> <sup>3-</sup> : 73.41 ± 3.30 mg/L Initial pH: 6–7 Duration time: 31.8–118.2 min	Fe/Al	64.93% ± 19.57%	2.10–15.30 kWh/L
Stafford <i>et al.</i> (2014)	CD: 1.1–5.4 mA/cm <sup>2</sup> Initial PO <sub>4</sub> <sup>3-</sup> : 5.2 ± 0.8 mg/L Initial pH: 7.1–7.8 Duration time: 35–85 S	Fe	82%	0.43 kWh/m <sup>3</sup>
Oztekin <i>et al.</i> (2022)	CD: 12–40 mA/cm <sup>2</sup> Initial PO <sub>4</sub> <sup>3-</sup> : 600–800 mg/L Initial pH: 5–9 HRT: 0–60 min	Fe	90%	–
Zeng <i>et al.</i> (2021)	CD: 1.5–3.5 mA/cm <sup>2</sup> Initial PO <sub>4</sub> <sup>3-</sup> : 8.8–9.8 mg/L Initial pH: 6.57–7.59 HRT: 34–102 min	Fe	90.24%	0.26 kWh/g PO <sub>4</sub> <sup>3-</sup>
Dian <i>et al.</i> (2013)	CD: 0.8 mA/cm <sup>2</sup> Initial PO <sub>4</sub> <sup>3-</sup> : 5 mg/L Initial pH: 5.2 HRT: 1–5 min	Fe	89.2%	–
Irdemez <i>et al.</i> (2006a)	CD: 1 mA/cm <sup>2</sup> Initial PO <sub>4</sub> <sup>3-</sup> : 100 mg/L Initial pH: 3–10 HRT: 4–36 min	Fe	86%	1.14 kWh/m <sup>3</sup>
Irdemez <i>et al.</i> (2006b)	CD: 0.2–2 mA/cm <sup>2</sup> Initial PO <sub>4</sub> <sup>3-</sup> : 25–100 mg/L Initial pH: 3–10 HRT: 5–50 min	Fe/Al	90%	14.87 kWh/m <sup>3</sup>
Makwana & Ahammed (2016)	CD: 16.3–57.5–2 mA/cm <sup>2</sup> Initial PO <sub>4</sub> <sup>3-</sup> : 4.5–6.4 mg/L Initial pH: 7.1–7.9 HRT: 31.8–188.2 min	Fe/Al	87.1%	4,400 kWh/m <sup>3</sup>
	CD: 8 mA/cm <sup>2</sup> Initial PO <sub>4</sub> <sup>3-</sup> : 60 mg/L Initial pH: 4–10 HRT: 10–60 min	Fe	71%	0.21 kWh/m <sup>3</sup>

density was 1 mA/cm<sup>2</sup>, the removal efficiency of phosphorus could reach 93.91%. At the same time, the energy consumption was only 0.25 kWh/m<sup>3</sup>, which was better than the related research because of the achievement of effective phosphorus removal at a much lower operation energy consumption. There was still a lack of studies on the long-term operation. To achieve a long-term stable operation, the process needs to be optimized. This study found that adding 500 mg/L of Cl<sup>-</sup> into the wastewater could effectively inhibit electrode passivation.

#### 4. CONCLUSION

In this study, experiments were carried out to remove phosphorus from SRDS. Different operating parameters such as current density, electrode distance, electrode number, HRT, and pH were investigated in detail. Correlation analysis showed that

current density was significantly correlated with HRT, and pH was negatively correlated with removal rate. The current density between operation energy consumption was more positive than other parameters.

The EC process removed 93.91% phosphorus while consuming around 0.25 kWh/m<sup>3</sup> of electrical energy at the current density of 1 mA/cm<sup>2</sup>, phosphorus of 5 mg/L, electrode distance of 2 cm, electrode number of 2, pH of 7, and HRT of 30 min, respectively.

In the long-term operation, the addition of Cl<sup>-</sup> can effectively inhibit the passivation of the electrode and improve the conductivity, thus keeping the operation stable and reducing the operation energy consumption of EC. However, the high concentration of Cl<sup>-</sup> led to the occurrence of side reactions and reduced the phosphorus removal efficiency; thus, the optimum Cl<sup>-</sup> concentration was 500 mg/L.

## ACKNOWLEDGEMENTS

This work was financially supported by the National Natural Science Foundation of China (42077108).

## DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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First received 18 October 2022; accepted in revised form 3 April 2023. Available online 12 April 2023