Parameters optimization for eutrophic lake water treatment by a novel process of iron-carbon micro-electrolysis coupled with catalytic ozonation using response surface methodology
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ABSTRACT
A novel process of iron-carbon micro-electrolysis (ICME) coupled with catalytic ozonation (CO) for treatment of eutrophic lake water was developed. A series of batch experiments with ICME alone and CO alone was designed to investigate the effects of process parameters, such as initial pH, dose of Fe-C, time of micro-electrolysis, ozone flux, dose of TiO₂/activated carbon (TiO₂/AC), and time of ozonation, on the removal rates of total nitrogen (TN), total phosphorus (TP), COD Mn and Chl-a. The process parameters were optimized using response surface methodology. The results showed that initial pH, dose of Fe-C and ozone flux had significant effects on removal of TN, TP, COD Mn and Chl-a. Within the range of selected operating conditions, the optimized values of initial pH, dose of Fe-C, time of micro-electrolysis, ozone flux, dose of TiO₂/AC, and time of ozonation were 3.8, 13.7 g/L, 29.6 min, 3.19 L/min, 294.74 mg/L and 106.73 min, respectively. Furthermore, ICME alone had significant advantages in TP and COD Mn removal and CO alone favored TN and Chl-a. Under the optimal process conditions, the final removal rates of TN, TP, COD Mn, and Chl-a by the hybrid ICME-CO process reached 75.33%, 86.29%, 94.42% and 97.57%, respectively. The present research provides a new alternative technology with promise for treatment of eutrophic lake water.

Key words | catalytic ozone, eutrophic lake water, iron-carbon micro-electrolysis, parameters optimization, response surface methodology

HIGHLIGHTS
- A novel coupling ICME-CO process was developed for treatment of eutrophic water.
- Six process parameters were optimized using RSM.
- Initial pH, dose of Fe-C and ozone flux had significant effects on removal of various pollutants.
- The final removal rates for TN, TP, COD Mn, and Chl-a by the ICME-CO process reached 75.33%, 86.29%, 94.42% and 97.57%.

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INTRODUCTION

According to the latest survey, over 64% of the global lakes are eutrophic (Zhang et al. 2020). Lake eutrophication occurs when the abundance of potential nutrient elements (including nitrogen (N) and phosphorus (P)) required by aquatic plants are significantly increased, and the photosynthesis rate of lake aquatic organisms greatly increases (Gao et al. 2020). The eutrophication of water bodies creates many problems: (1) the rapid propagation of algae leads to insufficient light in the water column and (2) a sharp drop in dissolved oxygen. These conditions can result in an unpleasant odor emanating from water that contains widespread deadly aquatic organisms and the occurrence of algal blooms or red tides (Gao et al. 2020). These present enormous problems for the environment and human health. The ‘Taihu Lake Cyanobacteria Pollution Incident’ in China in 2007 is one example (Chen et al. 2020). Hence, the treatment of eutrophic lake water is urgently needed.

In addition to containing high levels of N and P, eutrophic lake water typically contains high concentrations of organic compounds and chlorophyll-a (Chl-a). A variety of physical (Nguyen et al. 2019), biological (Wu et al. 2019) and chemical techniques (Huang et al. 2020) has been used for eutrophic lake water treatment in order to improve the quality of the aquatic ecosystem. Physical techniques, such as adsorption (Qin et al. 2020) and aeration (Nguyen et al. 2019), have had excellent effects in pollutant removal, but the main removal targets are N/P for adsorption and organic compounds for aeration, respectively. These techniques lack the ability to remove multiple contaminants as a whole. Biological techniques, such as those involving the addition of microorganisms (Wu et al. 2019), and the cultivation of aquatic plants (Hu et al. 2008) in water, can control total nitrogen (TN) and total phosphorus (TP) within a certain range under suitable conditions, but their low removal rates of organics and Chl-a and potential risks of overpopulation of the introduced organisms are obvious disadvantages. The addition of algae removers, such as magnesium-based oxygen (Huang et al. 2020) and copper sulfate (Tsai et al. 2019), into the water body is the common chemical technique. These removers act on the algal cell wall with sulfur-containing groups to destroy algae growth. These removers have prominent removal effects on Chl-a but exhibit poor removal of nutrients and cause secondary pollution. The target compounds for removal by the above techniques for eutrophic lake water treatment are either nutrients, such as N and P, or Chl-a, and could not eliminate the harm of eutrophic water. Therefore, to realize the complete treatment of eutrophic lake water, it is necessary to identify and develop a more efficient process.

In iron-carbon micro-electrolysis (ICME), also known as internal electrolysis, microprimary cells are formed by iron (Fe) and carbon (C) contained in iron filings under acidic conditions, allowing degradation-resistant organics in water to be converted into easily degradable organics (Zheng et al. 2019). At present, this method is widely used for the treatment of wastewater that contains high concentrations of organics, such as dye wastewater (Zhu et al. 2018), pharmaceutical wastewater (Malakootian et al. 2019), papermaking wastewater (Fan et al. 2020), and mining wastewater (Lin et al. 2016), due to its advantages of low complexity, ease of operation, low raw material cost, short processing time, and low secondary pollution from the reaction products, among others. Furthermore, the Fe in iron filings can react with dissolved phosphate to form iron phosphate precipitates, such as Fe₃(PO₄)₂ and FePO₄, indicating that ICME should be a suitable technology for eutrophic water treatment. The main advantage of ICME is to convert refractory organic matter into easy degradable organic matter, but it does not provide the extensive and complete removal of organic matter. In contrast, ozone oxidation is more effective for deep removal of organic matter, but it typically exhibits low ozone utilization (Lu et al. 2019). To overcome this drawback, ozone oxidation can be used in combination with a catalytic material to generate a strong oxidizing hydroxyl radical to enhance the method’s oxidation efficiency, a process known as catalytic ozonation (CO) (Wang & Chen 2020). Therefore, following pretreatment with ICME the addition of CO can theoretically achieve complete removal of multiple contaminants in eutrophic water. However, to date, little research on eutrophic water treatment via the coupling of ICME with CO has been reported. Hence, investigation of the optimal parameters for this novel combination process for the complete treatment of pollutants in eutrophic water is warranted.
In this study, a novel process of ICME coupled with CO (ICME-CO) was constructed for treatment of eutrophic lake water, ICME and CO were used as the pretreatment and advanced treatment of ICME-CO, respectively. A series of batch experiments with ICME alone and CO alone was designed to investigate the effects of process parameters, such as initial pH, dose of Fe-C, time of micro-electrolysis, ozone flux, dose of TiO₂/AC, and time of ozonation, on the removal rates of TN, TP, CODₘₙ and Chl-α, respectively. Moreover, the above process parameters were further optimized using response surface methodology (RSM) constructed by central composite design. Moreover, the hybrid ICME-CO process was further used in actual eutrophic lake water treatment based on the optimized process parameters. The present research provides a new alternative technology with promise for the treatment of eutrophic lake water.

**MATERIALS AND METHODS**

**Materials**

In this study, simulated and actual eutrophic lake water were prepared and collected, respectively. The simulated eutrophic lake water was taken from Ruoyu Lake in Changzhou University, and ammonium chloride (National Pharmaceutical Group, China), potassium phosphate (National Pharmaceutical, China), and self-cultivated algae were added into it to control the concentrations of TN, TP, CODₘₙ and Chl-α within a certain range, which were used for single-factor and response surface analysis with the purpose of determining the optimal process parameters. The actual eutrophic lake water was taken from Taihu Lake in China (latitude 30°55′40″–31°32′58″N, longitude 119°52′32″–120°36′10″E), which was used for the comparative analysis of individual ICME, individual CO and ICME-CO to judge whether the selected conditions had the desired effect of dealing with the eutrophic water. The detail water quality indexes of simulated and actual eutrophic lake water are listed in Table 1. The iron filings (Yangzhou Changpu Chemical Reagent Co., Ltd, China) and granular activated carbon (National Pharmaceutical Group, China) used in the experiment were of analytical grade. The composite catalyst used for CO was TiO₂/activated carbon (TiO₂/AC), and the preparation method for TiO₂/AC was referenced in the study by Telegang Chekem (Telegang Chekem et al. 2019).

**Experimental design**

**Experimental device of ICME-CO**

Batch experiments of ICME alone and CO alone were used for eutrophic lake water treatment. The hybrid ICME-CO process used ICME as a pretreatment process and CO as an advanced treatment process. Figure 1 shows the experimental diagram set-up.

For the ICME process alone (Figure 1(a)), 1,000 mL of simulated eutrophic lake water was accurately measured in a large beaker, and the pH adjusted to the required pH value. The appropriate amounts of iron filings and granular activated carbon (volume ratio is 1:1) were added into it to control the concentrations of TN, TP, CODₘₙ and Chl-α within a certain range, which were used for single-factor and response surface analysis with the purpose of determining the optimal process parameters. After the required reaction time for micro-electrolysis, the water sample was left to undergo static settlement, and the TN, TP, CODₘₙ and Chl-α contents in the beaker were determined. The removal rate of each component was calculated using Equation (1):

\[
\text{removal rate (\%) } = \frac{C_t - C_0}{C_0} \times 100 \quad (1)
\]

where \(C_t\) was the pollutant concentration after the reaction, mg/L; and \(C_0\) was the initial concentration of pollutant, mg/L. For the individual CO process (Figure 1(b)), the target flux ozone (30 mg O₃/L) prepared by an ozone generator was added into the simulated eutrophic lake water by an aerator.
Furthermore, a certain amount of TiO$_2$/AC composite catalyst was also added into the water. After a given ozonation reaction time, the TiO$_2$/AC in the water settled out and TN, TP, COD$_{Mn}$ and Chl-$a$ contents were determined as in the above method. Conversely, the CO inflow was the ICME effluent for the treatment of actual eutrophic lake water by the hybrid ICME-CO process, and the other conditions were the same.

**Single-factor experiment**

To optimize the reaction conditions of the hybrid ICME-CO process for the treatment of eutrophic lake water, single-factor experiments with ICME alone and CO alone were designed to investigate their effects on the removal rates of TN, TP, COD$_{Mn}$ and Chl-$a$ in eutrophic lake water. For the ICME process, the initial pH, dose of Fe-C and time of micro-electrolysis were the main factors; for CO, ozone flux, dose of TiO$_2$/AC and time of ozonation were investigated. The detailed experimental conditions for the treatment of simulated eutrophic lake water by the above two processes are listed in Table 2.

**Experimental design using RSM**

RSM was used to optimize the parameters of the ICME and CO processes and to determine the final optimal conditions for the hybrid ICME-CO process. Based on the single-factor experiments with ICME and CO, a set of $3 \times 3$ response surface analysis experiments was designed using Design-Expert 11.0 software. Details of the test factors and level design of the individual ICME and CO processes are presented in Table 3. Furthermore, according to the above scheme, 34 combinations of initial conditions were used for the individual ICME and CO processes. Additionally, analysis of variance (ANOVA) was used to analyze the suitability of the regression models at the 95% confidence level. The $P$-value and $F$-value were used to assess the significance of the variables, and a model with a $P$-value less than 0.05 and a large $F$-value was considered significant (Nayak & Vyas 2019).

**Analysis methods**

The TN, TP, COD$_{Mn}$ and Chl-$a$ concentrations of all water samples in this study were determined using a UV-visible
spectrophotometer (UV-vis; Pharo300, Merck, Germany) using the potassium persulfate oxidation-ultraviolet spectroscopic, ammonium molybdate spectrophotometry and acidic potassium permanganate, ethanol-spectrophotometry methods, respectively (Jiang et al. 2017; Kim et al. 2019). All experiments were conducted in triplicate, and the data were represented by the mean of measured values.

RESULTS AND DISCUSSION

Single-factor analysis

Figure 2 demonstrates the influences of individual variables on the removal of TN, TP, CODMn and Chl-α in simulated eutrophic lake water by ICME and CO alone. As shown in Figure 2(a), as pH was increased from 2 to 4, the removal rates of TN, TP and Chl-α increased but CODMn decreased. At pH 4, the removal rates of TN, CODMn and Chl-α reached maximum levels, decreasing at pH > 4, whereas TP remained stable at pH > 4. These results can be attributed to a reduction in the number of H⁺ participating in the cathode reaction as pH increases; the reaction of Fe oxidation to Fe²⁺ was inhibited, which affected the removal of CODMn (Huang et al. 2018). Conversely, the stronger the acidity of the reaction system, the larger the potential difference of the microbattery, which facilitated the electrode reaction. In contrast, the concentration of H⁺ participating in the cathodic reaction was low in the high-pH environment, and the anode reaction was inhibited (Zheng et al. 2019). When the pH was low enough, the Fe passivation caused the formation of a hard and dense layer on the surface, which affected the production of Fe²⁺ (Zheng et al. 2019). With increasing dose of Fe-C, the removal rates of TN, TP, CODMn and Chl-α increased (Figure 2(b)), reaching maximum values at 12 g/L. At higher doses, the removal rates of TP, CODMn and Chl-α became stable whereas TN decreased significantly. The reduction of TN removal rate may have been related to a reduction in the contact area of Fe with H₂O by the presence of excessive iron filings, as reported by Zhu et al. (Zhu et al. 2018). As the reaction time of micro-electrolysis increased from 10 to 30 min, the removal rates of TN, TP, CODMn and Chl-α decreased and, beyond 30 min, TN, CODMn and Chl-α decreased whereas TP continued to increase (Figure 2(c)). The above phenomena were attributed to the large amounts of Fe²⁺ and Fe³⁺ that were generated and passivated the surface of the iron powder, which had a flocculation and sedimentation effect and thereby inhibited the ICME reaction (Zhang et al. 2018).

However, the removal of TP in this type of system depends mainly on the adsorption of granular Fe/C.

Figure 2(d)–2(f) show the effects of ozone flux, dose of TiO₂/AC, and time of ozonation on removal rate of TN, TP, CODMn and Chl-α by the CO process. As ozone flux increased, the removal rate of each index first increased and then decreased (Figure 2(d)). At ozone flux values less than 3 L/min, the removal rates of TN, TP, CODMn and Chl-α increased and, beyond 30 min, TN, CODMn and Chl-α decreased whereas TP continued to increase (Figure 2(c)). The above phenomena were attributed to the large amounts of Fe²⁺ and Fe³⁺ that were generated and passivated the surface of the iron powder, which had a flocculation and sedimentation effect and thereby inhibited the ICME reaction (Zhang et al. 2018).

However, the removal of TP in this type of system depends mainly on the adsorption of granular Fe/C.

Table 3 | Test factors and level design of RSM for the process of ICME alone and CO alone

| Type of process | Factors                        | Code rule |
|----------------|--------------------------------|-----------|
| ICME           | Initial pH                     | -1 4 6    |
|                | Dose of Fe-C (g/L)             | 2 4 6     |
|                | Time of micro-electrolysis (min)| 10 30 50  |
| CO             | Ozone flux (L/min)             | 1 3 5     |
|                | Dose of TiO₂/AC (mg/L)         | 50 175 300|
|                | Time of ozonation (min)        | 30 90 150 |
increased amounts of the catalyst is associated with increased processing cost. Thus, it was necessary to identify a reasonable amount of catalyst. The effects of time of ozonation were similar to those of ozone flux, and the removal rate of each index was maximized at a treatment time of 90 min (Figure 2(f)). However, when the total amount of pollutants was held constant, extending the reaction time beyond this point did not continuously enhance the reaction effect.
Regression model development and response surface analysis

The relationships between dependent parameters (removal rate of TN, TP, COD\(_{\text{Mn}}\) and Chl-a) and independent parameters (ICME: initial pH (A), dosage of Fe-C (B), and time of micro-electrolysis (C); CO: ozone flux (D), dosage of TiO\(_2\)/AC (E), time of ozonation (F)) obtained in this study are expressed by Equations (2)-(9), respectively. The optimal reaction conditions of ICME and CO were determined to be: ICME: initial pH = 0.4, dosage of Fe-C = 3.00 g/L, and time of micro-electrolysis = 14 hours; CO: ozone flux = 8.45 g/m\(^3\)-hr, dosage of TiO\(_2\)/AC = 0.05 g/L, and time of ozonation = 8 hours. The relationships between actual removal rate in the experiment and predicted removal rate (calculated according to regression model of Equations (2)-(9)) of TN, TP, COD\(_{\text{Mn}}\) and Chl-a are shown in Table 4. The correlation coefficient (\(R^2\)) values for TN, TP, COD\(_{\text{Mn}}\) and Chl-a were found to be 0.9735, 0.9870, 0.9349, and 0.9735, respectively, for the ICME process and 0.9714, 0.9897, 0.9745, and 0.9839, respectively, for the CO process, revealing strong correlations between the independent variables and dependent variables for both processes. The three-dimensional (3D) response surface plots are established in Figs. S1 and S2 to verify the dependence of the removal rates of TN, TP, COD\(_{\text{Mn}}\) and Chl-a by the ICME and CO process with different process parameters, respectively (see supplementary material).

| Parameter       | Equation          |
|-----------------|-------------------|
| TN (%)          | \( Y_{\text{TN}} = 46.80 + 0.025A + 10.08B + 1.0C + 0.32AB - 6.38AC + 1.63BC - 13.21A^2 - 15.11B^2 - 8.36C^2 \) |
| TP (%)          | \( Y_{\text{TP}} = 81.80 + 10.50A + 16.24B + 4.81C - 3.00AB - 5.80AC + 6.77BC - 18.51A^2 - 21.79B^2 - 10.94C^2 \) |
| COD\(_{\text{Mn}}\) (%) | \( Y_{\text{CODMn}} = 76.30 - 21.28A + 0.73B - 1.52C + 7.03AB - 1.12AC + 2.23BC - 8.11A^2 - 8.11B^2 - 11.11C^2 \) |
| Chl-a (%)       | \( Y_{\text{Chl-a}} = 72.70 + 4.60A - 2.23B - 8.45C + 4.45AB + 10.95AC - 2.65BC - 12.85A^2 - 10.80B^2 - 29.15C^2 \) |

CO process:

| Parameter       | Equation          |
|-----------------|-------------------|
| TN (%)          | \( Y_{\text{TN}} = 48.00 + 4.51D + 4.99E + 6.08F + 0.83DE - 2.75DF + 1.80EF - 16.91D^2 - 3.46E^2 - 11.39F^2 \) |
| TP (%)          | \( Y_{\text{TP}} = 62.30 + 2.54D + 5.54E + 7.08F - 1.13DE + 0.35DF + 1.25EF - 22.54D^2 - 2.54E^2 - 10.71F^2 \) |
| COD\(_{\text{Mn}}\) (%) | \( Y_{\text{CODMn}} = 36.00 + 3.46D + 1.84E + 2.73F + 0.40DE + 1.35DF + 2.68EF - 9.25D^2 - 0.25E^2 - 6.53F^2 \) |
| Chl-a (%)       | \( Y_{\text{Chl-a}} = 94.10 + 2.53D + 2.29E + 3.41F - 0.50DE - 2.55DF - 1.53EF - 26.31D^2 - 3.14E^2 - 15.94F^2 \) |

The relationships between actual removal rate in the experiment and predicted removal rate (calculated according to regression model of Equations (2)-(9)) of TN, TP, COD\(_{\text{Mn}}\) and Chl-a are shown in Table 4. The correlation coefficient (\(R^2\)) values for TN, TP, COD\(_{\text{Mn}}\) and Chl-a were found to be 0.9735, 0.9870, 0.9349, and 0.9735, respectively, for the ICME process and 0.9714, 0.9897, 0.9745, and 0.9839, respectively, for the CO process, revealing strong correlations between the independent variables and dependent variables for both processes. The three-dimensional (3D) response surface plots are established in Figs. S1 and S2 to verify the dependence of the removal rates of TN, TP, COD\(_{\text{Mn}}\) and Chl-a by the ICME and CO process with different process parameters, respectively (see supplementary material).

Table 4 also presents the ANOVA results regarding the removal rates of TN, TP, COD\(_{\text{Mn}}\) and Chl-a by the ICME and CO processes individually. It is clear to see that the \(P\)-values of eight models established in the present study were less than 0.05, indicating that the above models were significant at 95\% confidence level. Moreover, for the removal rate of COD\(_{\text{Mn}}\) during the ICME process, the \(P\)-values of A, B and C and \(A^2\) were less than 0.0001, 0.8, 0.6 and 0.05, and the \(F\)-values of A, B, C, and \(A^2\) were 71.9, 0.08, 0.4, and 0.5, respectively. It can be concluded that initial pH has a more significant effect than dose of Fe-C and reaction time of micro-electrolysis on the removal rate of COD\(_{\text{Mn}}\) (Singh et al. 2019). Furthermore, it can be concluded that the removal rates of TN and TP were mainly affected by dose of Fe-C and that the removal of Chl-a was related to the time of micro-electrolysis based on the low associated \(P\)-values and high \(F\)-values, and was consistent with the analysis described in Single-factor analysis. In addition, and based on the above analysis, it can be concluded that for the removal of TN and COD\(_{\text{Mn}}\) by CO alone, ozone flux had a larger effect than dose of TiO\(_2\)/AC and time of ozonation and that ozone flux and time of ozonation had similar significant influences on the removal of TP and Chl-a.

Optimization of process parameters for ICME-CO

Based on the analyses described in the previous sections, the optimal reaction conditions of ICME and CO were
Table 4 | The ANOVA of the removal rate of TN, TP, COD<sub>Mn</sub> and Chl-a by ICME and CO processes for quadratic model

| Source | TN | | | TP | | | COD<sub>Mn</sub> | | | Chl-a | |
|--------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|
|        | Sum of squares | Mean square | F-value | P-value | Sum of squares | Mean square | F-value | P-value | Sum of squares | Mean square | F-value | P-value | Sum of squares | Mean square | F-value | P-value |
| Model  | 2786.8          | 309.7          | 7.9     | 0.006   | 6309.3         | 701.0          | 15.0    | 0.0009  | 5062.4         | 562.5          | 11.2    | 0.002   | 6548.9          | 727.7          | 28.58   | 0.0001  |
| A      | 9.2             | 9.2            | 0.2     | 0.6     | 242.0          | 242.0          | 5.2     | 0.06    | 3621.0         | 3621.0         | 71.9    | <0.0001 | 169.3           | 169.3          | 6.65    | 0.0366  |
| B      | 588.3           | 588.3          | 15.0    | 0.006   | 7.2            | 7.2            | 0.2     | 0.7     | 4.2             | 4.2            | 0.8     | 0.08    | 39.6            | 39.6           | 1.56    | 0.2525  |
| C      | 14.6            | 14.6           | 0.4     | 0.6     | 521.6          | 521.7          | 11.2    | 0.01    | 18.6            | 18.6           | 0.4     | 0.6     | 571.2           | 571.2          | 22.43   | 0.0021  |
| AB     | 21.6            | 21.6           | 0.6     | 0.5     | 14.4           | 14.1           | 0.3     | 0.6     | 197.4           | 197.4          | 3.9     | 0.08    | 79.2            | 79.2           | 3.11    | 0.1211  |
| AC     | 124.3           | 124.3          | 3.2     | 0.1     | 416.2          | 416.2          | 8.9     | 0.02    | 5.1             | 5.1            | 0.1     | 0.8     | 479.6           | 479.6          | 18.84   | 0.0034  |
| BC     | 10.6            | 10.6           | 0.3     | 0.6     | 28.1           | 28.1           | 0.6     | 0.5     | 19.8            | 19.8           | 0.4     | 0.5     | 491.1           | 491.1          | 19.29   | 0.0002  |
| A<sup>2</sup> | 592.5          | 592.5          | 15.1    | 0.006   | 796.1          | 796.1          | 17.0    | 0.004   | 277.1           | 277.1          | 5.5     | 0.05    | 695.3           | 695.3          | 27.30   | 0.0012  |
| B<sup>2</sup> | 880.7          | 880.7          | 25.2    | 0.002   | 652.6          | 652.6          | 14.0    | 0.007   | 277.1           | 277.1          | 5.5     | 0.05    | 491.1           | 491.1          | 19.29   | 0.0003  |
| C<sup>2</sup> | 342.0          | 342.0          | 8.7     | 0.02    | 3184.2         | 3184.2         | 68.1    | <0.0001 | 519.9           | 519.9          | 10.3    | 0.01    | 3577.8          | 3577.8         | 140.51  | <0.0001 |
| Residual | 274.0          | 39.2           |         |         | 327.2          | 46.8           |         |         | 352.60          | 50.4           |         |         | 178.2            | 25.5           |         |         |
| Lack of fit | 274.0          | 91.5           |         |         | 327.2          | 109.1          |         |         | 352.60          | 117.5          |         |         | 178.2            | 94.4           |         |         |
| Pure error | 4.0            | 0.0            |         |         | 0.0            | 0.0            |         |         | 0.0             | 0.0            |         |         | 0.0              | 0.0            |         |         |

| R<sup>2</sup> | 0.9735 | 0.9780 | 0.9349 | 0.9735 |

A, initial pH; B, dose of Fe-C; C, time of micro-electrolysis; D, ozone flux; E, dose of TiO<sub>2</sub>/AC; F, time of ozonation.
obtained, as follows: (1) ICME process: initial pH = 3.8, dose of Fe-C = 13.7 g/L, reaction time = 29.6 min; (2) CO process: ozone flux = 3.19 L/min, reaction time = 106.73 min, and dose of TiO2/AC = 294.74 mg/L. Under the above optimal reaction conditions of the two stages, the hybrid ICME-CO process was used for the actual eutrophic lake water treatment, and the results are shown in Table 5.

The removal rates of TN, TP, CODMn, and Chl-a by ICME alone were 48.7%, 84.21%, 77.73%, and 77.81%, removal rates of TN, TP, CODMn, and Chl-a by CO alone were 52.14%, 66.32%, 38.19%, and 92.23%, and the final removal rates of TN, TP, CODMn, and Chl-a by the ICME-CO process reached 75.33%, 86.29%, 94.42%, and 97.57%, respectively, demonstrating that the method has excellent performance in the removal of contaminants from eutrophic lake water.

**CONCLUSIONS**

A new hybrid ICME-CO process was constructed for the treatment of eutrophic lake water. Experimental investigation of parameters optimization of ICME-CO using RSM. The polynomial equation with $R^2$ for TN, TP, CODMn, and Chl-a removal rates for the ICME and CO processes were greater than 0.95 suggesting that the RSM could predict the experimental results with high accuracy. The optimized value of initial pH, dosage of Fe-C, time of micro-electrolysis, ozone flux, dosage of TiO2/AC, and time of ozonation were 3.8, 13.7 g/L, 29.6 min, 3.19 L/min, 294.74 mg/L and 106.73 min, respectively. Based on the optimal process conditions, the final removal rates of TN, TP, CODMn, and Chl-a by the ICME-CO process could reached 75.33%, 86.29%, 94.42%, and 97.57%, respectively. Thus, it has excellent performance for complete removal of contaminants from eutrophic lake water. The present research provides a new alternative technology and has a good guiding value for the treatment of actual eutrophic lake water.

**ETHICS APPROVAL AND CONSENT TO PARTICIPATE**

Not applicable

**CONSENT FOR PUBLICATION**

Written informed consent for publication was obtained from all participants.

**CONFLICT OF INTEREST**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

**CREDIT AUTHORSHIP CONTRIBUTION STATEMENT**

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Yuanyuan Zhou: Data curation.

Liping Wang: Project administration.

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DATA AVAILABILITY STATEMENT

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

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