

Cost-effective method of benzene-containing wastewater treatment using floating electro-Fenton system

Hui Qiao, Mingqi He, Qiushi Wang, Shuaishuai Han, Haiqian Zhao and Zhonghua Wang

ABSTRACT

Traditional electro-Fenton systems must continuously supply oxygen to the cathode, which leads to extensive volatilisation of benzene in solutions. In this study, we adopted a floating cathode electro-Fenton system without bubbling oxygen into the solution to treat benzene-containing wastewater. The effects of the floating cathode position and main reaction parameters on benzene degradation were analysed, and the degradation cost was estimated. The results indicated that the electro-Fenton system with floating cathode could effectively degrade benzene in solutions. For the cathode, the complete utilisation of air and oxygen released from the anode was crucial. The benzene degradation rate increased with an increase in benzene concentrations. Additionally, pH mainly affected the existing ionic state of iron and production ratio of active substances. The current intensity significantly influenced the reaction activity. Using the floating cathode electro-Fenton method, the benzene removal ratio in the solution could reach 74.80% after 60 min under the optimum reaction conditions. For the floating cathode electro-Fenton system, the cost of treating benzene-containing sewage per cubic metre was \$1.2187, which is significantly lower than that for traditional electro-Fenton technology (\$1.4000). Hence, the floating cathode electro-Fenton system is an economical and efficient method for benzene degradation in solutions.

Key words | benzene, electro-Fenton, floating cathode, wastewater

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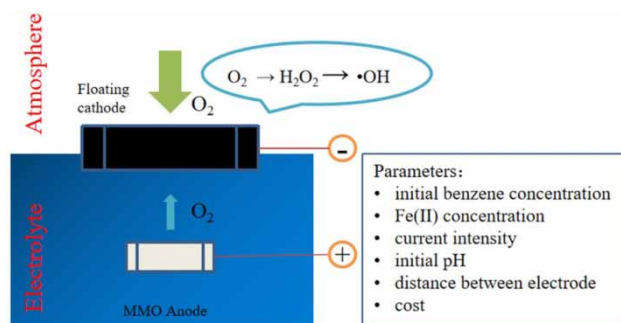
HIGHLIGHTS

- A floating cathode electro-Fenton system was used for the treatment of benzene-containing wastewater.
- The problem of benzene volatilisation was solved during the electro-Fenton degradation process.
- The floating cathode electro-Fenton is an economical method for the wastewater treatment.

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GRAPHICAL ABSTRACT

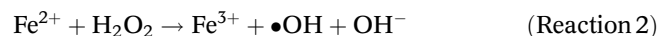


INTRODUCTION

Benzene (B), toluene (T), ethylbenzene (E), and paraxylene (X) (BTEX) are found primarily in crude oil and petroleum products. With the development of petroleum and petrochemical industries, the output and consumption of BTEX are increasing annually (Bustillo-Lecompte *et al.* 2018). BTEX compounds, represented by benzene, have become one of the most abundant chemicals worldwide. The release of large amounts of benzene poses a great threat to the health of humans. Benzene is the most toxic among all the BTEX compounds because it can be rapidly and effectively absorbed and widely distributed throughout the body. Its long-term exposure adversely affects skin health, respiratory and other senses, and the central nervous system (Anjum *et al.* 2019). Benzene also exhibits blood toxicity and genotoxicity and has been reported as one of the top 100 chemicals in the list of priority hazardous substances by the US Environmental Protection Agency (Mitra & Roy 2011). Therefore, the development of a green and efficient benzene treatment technology is urgently required.

For the treatment of benzene-containing wastewater, physical, biodegradation, and advanced oxidation methods are mainly used. In the physical method, benzene present in water is allowed to be adsorbed and enriched onto adsorbent materials to minimise its concentration in water. However, the selective adsorption effect of most adsorbent materials on benzene is general (Seifi *et al.* 2011), and benzene is transferred from solution to adsorbent materials, but benzene is not generally converted into a harmless substance. In the biodegradation method, microorganisms are employed to metabolise organic matter into inorganic matter to treat wastewater; however, this method requires favourable environmental conditions and the treatment time is long. Electro-Fenton technology, a new Fenton

derivative system, is a promising advanced oxidation technology (Nakajima *et al.* 1997). In this method, first, H_2O_2 is generated *in situ* at the cathode through the oxygen reduction reaction (ORR) (Reaction 1). Subsequently, H_2O_2 reacts with Fe^{2+} in the system to generate Fe^{3+} and a hydroxyl radical ($\bullet\text{OH}$) with strong oxidisability (Reaction 2) (Zhao *et al.* 2018). Simultaneously, Fe^{3+} at the cathode undergoes reduction reaction to generate Fe^{2+} (Reaction 3). This method has advantages such as less secondary pollution, low storage and transportation costs, and mild process conditions. The reaction ratio can be controlled by adjusting the reaction conditions, which facilitates automation realisation (Deng *et al.* 2019). Studies have previously been conducted on benzene degradation using the electro-Fenton process (Mohammad *et al.* 2019; Mahmoud *et al.* 2019):



The electro-Fenton performance for pollutant degradation depends on the rate of H_2O_2 production (Yu *et al.* 2015). However, H_2O_2 generation, relying on the 2e-ORR reaction, is limited by the solubility of oxygen in water (Zhou *et al.* 2018). In general, oxygen is supplied to the cathode surface through aeration or an external source; however, this approach is associated with an extremely low oxygen utilisation rate and a high cost (Perez *et al.* 2016). Moreover, because benzene has strong volatility, the traditional oxygen supply method leads to strong benzene volatilisation.

To solve these key problems, we proposed a floating cathode electro-Fenton process, which can result in the formation of a three-phase interface, synergistic utilisation of oxygen generated using air and anode, and improvement in the oxygen utilisation rate. For the degradation of benzene-containing wastewater, this method is highly efficient and energy saving. We investigated the effects of reaction conditions, such as initial benzene concentration, Fe(II) concentration, pH, current intensity, and plate distance, on benzene degradation and estimated the process cost. The experimental results revealed a cost-effective and efficient treatment process for benzene-containing wastewater degradation.

MATERIALS AND METHODS

Materials

All reagents in the experiments were of analytical grade. Benzene, sodium sulphate (Na_2SO_4), ferrous sulphate (FeSO_4), dilute sulphuric acid (H_2SO_4), petroleum ether, and sodium hydroxide (NaOH) were acquired from Aladdin (Shanghai, China).

Experimental method

The primary equipment and the model are shown in Table 1. All the experiments were performed in a 200-mL electrolytic cell with positive and negative electrodes. To the cell, 150 mL of benzene solution of a specific concentration was added. The cathode was a 2 cm × 4 cm × 0.5 cm graphite felt, and the anode was a 2 cm × 2 cm × 0.2 cm mixed metal oxide (MMO) placed vertically at the centre of the cell opposite to the cathode. We used a screw with nuts at both ends to adjust the distance between the cathode and anode and to fix the cathode so that it could float above the liquid surface

Table 1 | The primary equipment and the model

Equipment name	Model	Manufacturer
pH meter	pHS-25	Rex Electric Chemical Instrument Factory, China
Digital DC Power Supply	MS1003D	MaiSheng Power Supply Co., China
High-performance gas chromatograph	GC7980	Techcomp Co., China
Magnetic stirrer	C-MAG MS 4	IKA, Germany

during the experiment. The solution was stirred with a magnetic stirrer at a speed of 400 r/min to ensure the uniform concentration of the electrolyte solution and to maintain the solution at specific temperature. To prevent the strong volatilisation of benzene, we employed the constant temperature water bath method to maintain temperature at 25 °C. Before the initiation of the chemical reaction, solution pH was adjusted with NaOH and dilute H_2SO_4 . A pH meter (pHS-25, Rex Electric Chemical Instrument Factory, China) was used to record solution pH. Electrodes were connected to the digital DC power supply (MS1003D, MaiSheng Power Supply Co., China), with two modes of constant current and voltage. Because our objective was to determine the influence of current intensity, the power supply was set to a constant current state. A desired amount of FeSO_4 was added while switching on the DC power supply. To obtain the desired conductivity required for the electro-Fenton process, a certain amount of Na_2SO_4 was dissolved in the solution. In this process, the samples were transferred in test tubes at 0, 10, 20, 30, 40, 50, and 60 min and were mixed with the same amount of petroleum ether; the mixture was shaken. Subsequently, the solution was analysed through gas chromatography. After each operation, electrodes were washed with deionised water and dried at room temperature.

Analytical method

High-performance gas chromatography (GC7980, Techcomp Co., China) was used to quantify the benzene concentration. Pre-column pressure was 9 psi. Samples (2 μL) were automatically transferred into a 30 m × 0.32 mm × 0.25 μm SCION-WAXMS capillary column. Inlet, detector, and column box temperatures were set to 200, 250, and 80 °C, respectively. The split-flow ratio and purge-flow rate were 10:1 and 2 mL/min, respectively. High-purity nitrogen was used as carrier gas at a flow rate of 21.14 mL/min.

RESULTS AND DISCUSSION

Volatilisation characteristics of benzene

Figure 1(a) shows the volatilisation characteristics of 1,000 mg/L benzene at 25 °C and with oxygen bubbling into the solution.

At the oxygen flow rate of 1.2 L/min, benzene volatilisation was considerably intense, and 98.2% benzene volatilised in 15 min. The oxygen flow rate used in the

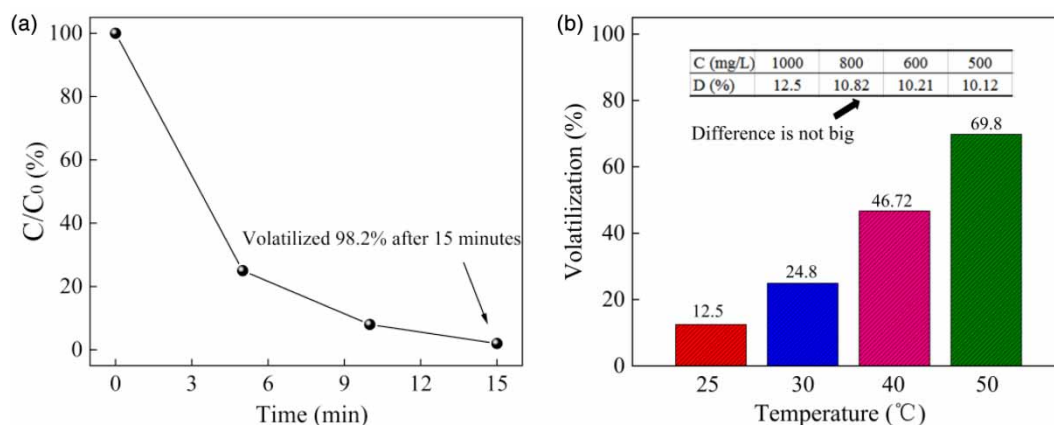


Figure 1 | (a) Volatilisation characteristics of benzene with 1.2 L/min oxygen at 25 °C. (b) Volatilisation characteristics of 1,000 mg/L benzene at different temperatures and those of different benzene concentrations at 25 °C.

experiment was similar to the ventilation flow commonly used in the traditional electro-Fenton system. If the traditional electro-Fenton system is used for benzene degradation under the bubbling oxygen condition, benzene volatilisation inevitably leads to substantial errors.

Figure 1(b) illustrates the effects of temperature on benzene volatilisation, with no oxygen bubbling into the solution. The initial benzene concentration was 1,000 mg/L. The experimental results indicated that the higher is the temperature, the higher is the benzene volatilisation rate. The benzene volatilisation ratios at 25 and 30 °C were 12.5% and 24.8%, respectively, in 60 min. At a temperature of >30 °C, the volatilisation rate further increased. Volatilisation ratios at 40 and 50 °C were 46.72% and 69.8%, respectively. These results indicated that benzene volatilisation is intense at high temperatures. To minimise the influence of benzene volatilisation on benzene degradation using the electro-Fenton system, the solution temperature was set to 25 °C.

Figure 1(b) illustrates benzene volatilisation at different initial benzene concentrations at 25 °C observed without blowing oxygen. Under these conditions, the trends of benzene volatilisation were similar when using different concentrations. The volatilisation rate was positively correlated with the concentration; however, the overall difference in the volatilisation rate was not obvious. The traditional electro-Fenton system must continuously supply oxygen to the cathode to generate H_2O_2 (Reaction 1) and then must generate hydroxyl radicals ($\cdot OH$) to degrade pollutants. When the traditional electro-Fenton system was used to degrade benzene, supplying oxygen to the cathode inevitably leads to the volatilisation of a large amount of benzene. The use of the electro-Fenton system with floating cathode to

degrade pollutants yielded favourable experimental results (Zhou *et al.* 2018). In this study, the electro-Fenton system with floating cathode was observed to degrade benzene at 25 °C, at which the cathode can directly come in contact with the air. Oxygen in the air can replace oxygen supplied to the cathode to correct the volatilisation interference.

Oxygen source of floating cathode

A graphite felt cathode with a thickness of 0.5 cm was selected to determine the influence of different cathode positions (Figure 2) on benzene degradation. Figure 3 presents the results. The degradation effect was optimal when the cathode was located 0.25 cm above the water surface; the degradation ratio reached 72.7% after 60 min. However, the degradation effect was the worst when the cathode was submerged into the solution, with the degradation ratio reaching only 39.8% after 60 min. The position of the floating cathode significantly influences benzene degradation.

This is because when oxygen is not blown in, there are two sources of oxygen, which are oxygen from the air and oxygen generate from the MMO anode (Zhang *et al.* 2019). When the cathode was placed 0.25 cm above the water surface, it could simultaneously utilise both the oxygen sources to achieve the optimal degradation effect, whereas the utilisation of oxygen present in the air was difficult for the cathode submerged in the solution, and relying only on the oxygen precipitated from the anode leads to the worst degradation effect. For the cathode placed on the water surface, the utilisation of oxygen precipitated from the anode was difficult, which resulted in a degradation ratio of only 39.8%.

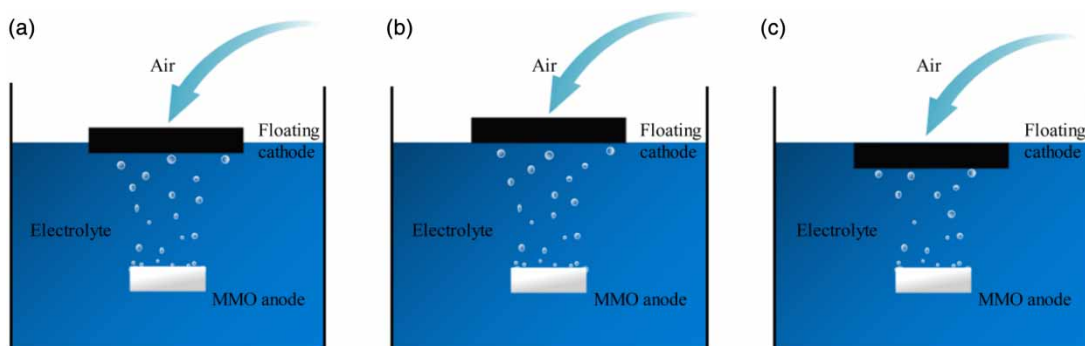


Figure 2 | Different cathode placement positions. (a) 0.25 cm above the water surface; (b) on the water surface; (c) submerged into the solution.

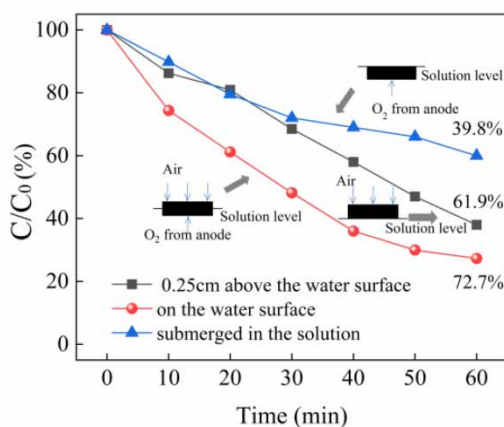


Figure 3 | Influence of different positions of the cathode ([Benzene] = 1,000 mg/L, [Na₂SO₄] = 50 mmol/L, T = 25 °C, and pH = 3).

The comparison of degradation effects showed that the contribution of oxygen available in the air was obviously higher than that of oxygen precipitated from the anode. In addition, air humidity and water pressure were other limiting factors (Yongtae *et al.* 2014). When these two parameters are considerably high, the diffusion layer is easily blocked, affecting oxygen diffusion and electron transfer ratios, and leads to a decrease in cathode performance. Therefore, reasonably determining the position of the floating cathode and ensuring the complete utilisation of the two oxygen sources are essential to ensure desirable oxygen diffusion and electron transfer rates.

Influence of reaction conditions on the degradation effect

Effect of the initial benzene concentration

Figure 4 presents the results of benzene degradation through the floating cathode electro-Fenton system under different

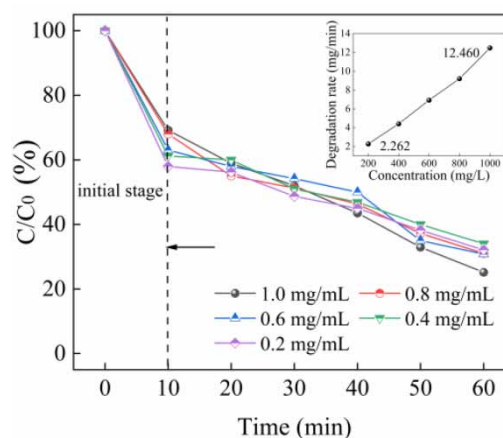


Figure 4 | Effect of the initial benzene concentration on the benzene degradation rate ([Na₂SO₄] = 50 mmol/L, [FeSO₄] = 60 mg/L, I = 200 mA, T = 25 °C, and pH = 3).

initial benzene concentrations. The benzene degradation rate varied with different initial benzene concentrations. The degradation rate of benzene continued to increase with an increase in the initial concentration of benzene (Figure 4).

In the experiment, the reaction temperature and initial concentration of Fe(II) were the same, and the generation ratio of •OH at the initial time was equal. The higher was the initial concentration of benzene, the faster was the reaction between •OH and benzene. Therefore, the benzene degradation rate increased with an increase in its initial concentration. By contrast, the benzene degradation ratio decreased with an increase in its initial concentration. At a high initial concentration of benzene, the degradation ratio decreased because the oxidation of a large number of benzene molecules requires the same amount of oxidant, and many Fe(II) may be required to minimise the reaction time for complete degradation (Babuponnusami & Muthukumar 2012). The results indicated that benzene

degradation was rapid at the beginning of the reaction, that is, at the predominant stage, and then this degradation became slower with time. This phenomenon may result from the rapid generation of hydroxyl radicals within a short time period. However, $\text{Fe}(\text{OH})_3$ begins to participate and leads to a decrease in the amount of the hydroxyl radicals generated (Brillas *et al.* 2009).

Effect of the Fe(II) concentration

Figure 5 illustrates the influence of different Fe(II) concentrations on benzene degradation through floating cathode electro-Fenton system.

The results indicated that the initial degradation rate (within 0–10 min of the reaction) of benzene increased with an increase in the Fe(II) concentration and gradually decreased with reaction progression. This trend was observed because Fe(II) plays an important role in $\cdot\text{OH}$ generation. In the beginning of the experiment, the high Fe(II) concentration resulted in the strong catalysis of H_2O_2 to generate $\cdot\text{OH}$, and the degradation ratio was high (Reaction 2). However, with the gradual consumption of Fe(II), the benzene degradation ratio decreased. After 50–60 min (the end of the reaction), the degradation ratio at the Fe(II) concentrations of 15 and 30 mg/L was extremely low. However, the degradation ratios of other three groups were similar and did not increase significantly with the increase in the Fe(II) concentration because the low concentration of Fe(II) is likely to be consumed at the end of the reaction, resulting in limited degradation. But too high the concentration of Fe(II) is also bad. A large amount of iron sludge precipitates within 60 min of the reaction at Fe(II) concentrations of 120 and 240 mg/L, which increases the post-treatment difficulty.

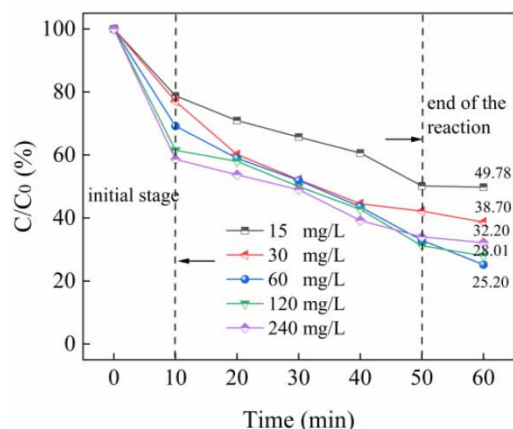


Figure 5 | Effect of the Fe(II) concentration ([Benzene] = 1,000 mg/L, $[\text{Na}_2\text{SO}_4]$ = 50 mmol/L, I = 200 mA, T = 25 °C, and pH = 3).

Excessive Fe(II) levels also result in side reactions, which can affect the treatment effect and are not conducive to cost saving. Therefore, a Fe(II) concentration of 60 mg/L was selected for the experiment.

Effect of the current intensity

Current intensity is substantially important in electrochemistry. Figure 6 illustrates the effect of different current intensities on the degradation ratio of the electro-Fenton system. The benzene degradation ratio was the highest in the initial stage and gradually decreased. Under current intensity of the range 100–300 mA, the degradation ratio increased significantly with an increase in the current intensity. However, when the current intensity increased to 400 mA, the degradation ratio began to decrease.

The current intensity is a key parameter in the electro-Fenton process (Chen *et al.* 2016) and plays an important role in promoting H_2O_2 production near the cathode (Reaction 1) (Qiu *et al.* 2018). The increase in H_2O_2 improves the reaction activity and provides relatively more $\cdot\text{OH}$, which is essential for degradation (Reaction 2) (Babaei-Sati & Parsa 2017). Moreover, a high current intensity is beneficial to the oxygen evolution at the MMO anode. Finally, the current intensity can enhance Fe^{2+} regeneration (Reaction 3) to improve the degradation effect. However, when the current is semantically large, O_2 electroreduction is limited by the mass transfer of oxygen on the cathode surface; hence, a large amount of charge transferred by the system is not conducive to degradation (Ma *et al.* 2019). In addition, the high action potential is conducive to reduce H_2O_2 into H_2O and the hydrogen evolution reaction (Khataee *et al.* 2017), which is not conducive to free radical generation.

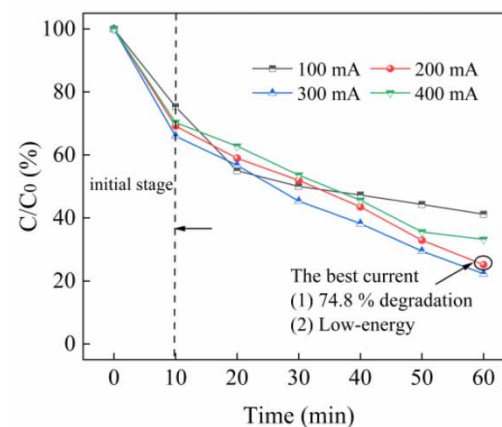


Figure 6 | Effect of current intensity ([Benzene] = 1,000 mg/L, $[\text{Na}_2\text{SO}_4]$ = 50 mmol/L, $[\text{FeSO}_4]$ = 60 mg/L, and T = 25 °C).

Therefore, the considerably high current intensity leads to a decrease in the electro-Fenton system efficiency and degradation ratio. The degradation ratio was the highest (74.8% and 77.7%) at 200 and 300 mA, respectively, indicating that the gap in these ratios was not large. Therefore, considering the cost and degradation ratio, 200 mA was selected as the optimal current.

Effect of initial pH

The effect of different pH values of the solution on the ratio of benzene degradation using the floating cathode electro-Fenton system was also investigated. Figure 7 shows the results.

The pH of the solution mainly affected the initial stage of the reaction (0–10 min) (Figure 7). A pH of 3 was considered the optimal pH of the system at the initial stage because at this pH, the amount of $\cdot\text{OH}$ produced in the Fenton reaction was the largest and the pollutant removal effect was optimal (Ruiz *et al.* 2011). Moreover, 2e-ORR in the electro-Fenton process is particularly important to improve the reaction efficiency and degradation effect (Narayanan *et al.* 2003) and can be promoted when the solution is suitably acidic (Reaction 1). However, at a pH of >4 , Fe^{2+} is easily oxidised to produce a large amount of iron precipitate, which affects the Fenton reaction and leads to a decrease in the degradation ratio. As the reaction progresses, solution pH becomes steady after 60 min (nearly 5–6) (Figure 7). Therefore, the influence of pH on the electro-Fenton process is considerably less than that of current intensity and other factors, and the degradation ratio almost stabilises in 60 min (the end of the reaction) when other factors remain constant.

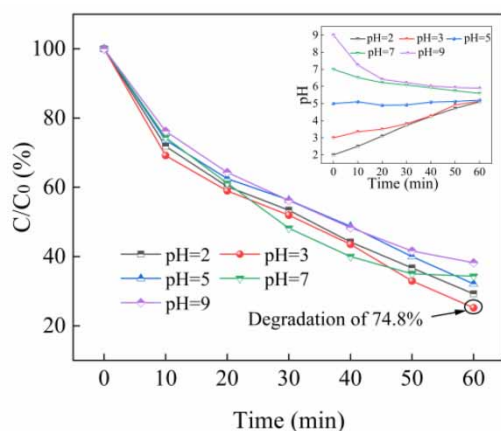


Figure 7 | Effect of initial pH and the change in pH during the experiment. ([Benzene] = 1,000 mg/L, $[\text{Na}_2\text{SO}_4]$ = 50 mmol/L, $[\text{FeSO}_4]$ = 60 mg/L, and $T = 25^\circ\text{C}$).

Effect of the distance between the cathode and anode

The change in the distance between the cathode and anode greatly influences the wastewater treatment and electrochemical reaction rate. We analysed the effect of the distance between the cathode and anode on the ratio of benzene degradation through the floating cathode electro-Fenton system. The effect of the distance between cathode and anode of 3, 4, 5, and 6 cm was analysed. Figure 8 presents the results.

With an increase in the distance between the cathode and anode, the benzene degradation ratio first increased and then decreased within the reaction time of 60 min. The degradation ratio of benzene was the highest (74.56%) when the distance was 4 cm. This result indicated that considerably small or large distances between the cathode and anode can adversely affect the reaction.

When the distance between the cathode and anode is considerably large, the system resistance increases, leading to an increase in the input energy. Most electricity is wasted on side reactions. Thus, the considerably large distances not only worsen the wastewater treatment effect but also increase the costs. Similarly, when the distance between the cathode and anode is substantially small, large amounts of ferrous ions dissociate outside the cathode and anode, thereby decreasing the pollutant removal ratio. Therefore, the most suitable distance between the cathode and anode is 4 cm.

Cost estimation

The process cost of the degradation of benzene-containing wastewater using the new floating cathode electro-Fenton

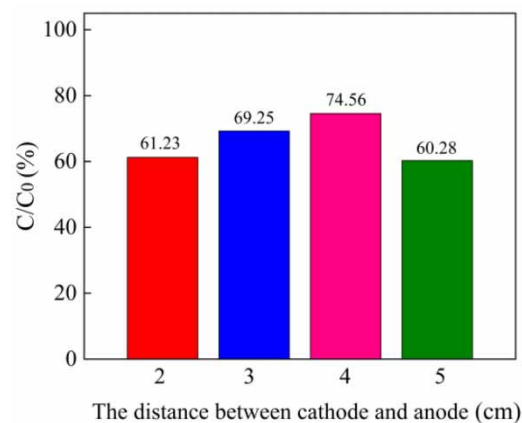


Figure 8 | Effect of the distance between the cathode and anode. ([Benzene] = 1,000 mg/L, $[\text{Na}_2\text{SO}_4]$ = 50 mmol/L, $[\text{FeSO}_4]$ = 60 mg/L, $T = 25^\circ\text{C}$, pH = 3, and $I = 200\text{ mA}$).

method was experimentally estimated. The total cost (TC) of the treatment process was determined using the sum of the amortisation cost (AC) and operation cost (OC); $TC = AC + OC$. AC is calculated by adding the costs of all devices and equipment, such as reinforced concrete main body, mixing motor, DC supplier, and other miscellaneous equipment. We used the following formulae for AC calculation:

$$C_p = C_{p0} + n \times A$$

$$V_t = \frac{V_c \times D \times T_w}{T_b}$$

$$AC = \frac{C_p \times V_c}{n \times V_t}$$

$$A = \frac{C_{p0}(i(1+i^n))}{(1+i)^n - 1}$$

where C_{p0} is the present AC per one cubic metre of the reactor volume, A is the annual investment cost of C_{p0} , C_p is the net cost per one cubic metre of the volume, i is the interest rate, V_t is the total volume of treated effluent per year, D is the working days per year, n is the life time, T_w is the reactor working time, V_t is the total annual treatment amount, and V_c is the reactor volume. In this paper, $n = 25$ (years), $D = 300$ (days), $T_w = 14$ (h), $C_{p0} = 1,830$ (\$), $i = 6\%$, and $V_c = 10$ (m^3).

The estimated AC is 1.1745 \$/m³.

To calculate the OC, the following formulae were used:

$$OC = C_{ch} + C_e + EC + 0.002AC$$

$$EC = \frac{E \times P_E \times T_w \times D}{V_t}$$

where E is the consumed power, P_E is the unit price of power energy, C_{ch} is the cost of chemicals, and C_e is the cost of replacement electrodes. In this paper, $P_E = 1$ (yuan), $C_{ch} = 0.3658$ (\$/m³), and $C_e = 0.1677$ (\$/m³).

The estimated OC is 0.0442 \$/m³, and thus the estimated TC is 1.2187 \$/m³. However, the cost of the traditional electro-Fenton system for benzene series degradation is 1.4 \$/m³ (Mahmoud *et al.* 2019). The results indicated that the benzene degradation cost can be considerably decreased by using the floating cathode electro-Fenton process.

CONCLUSIONS

The volatilisation characteristics of benzene at different temperatures and under conditions of oxygen bubbling were investigated. Benzene volatilisation severely influences the judgement of the effect of the traditional electro-Fenton system. An improved electro-Fenton technology-floating cathode electro-Fenton method was developed to degrade benzene-containing wastewater, and the influence of different experimental conditions on the degradation effect was analysed. The benzene removal ratio was 74.80% under the following conditions: [benzene] = 1,000 mg/L; pH = 3; $T = 25$ °C; [Na₂SO₄] = 50 mmol/L; [FeSO₄] = 60 mg/L; and $I = 200$ mA. Finally, the cost of treating benzene-containing wastewater per cubic metre estimated using the floating cathode electro-Fenton method is \$1.2187. The proposed method can thus help to realise the degradation of benzene-containing wastewater with high efficiency and energy saving.

DATA AVAILABILITY STATEMENT

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

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