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REVIEW ARTICLE

# Analysis of factors influencing on Electro-Fenton and research on combination technology (II): a review

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**Abstract**

The principle of Fenton reagent is to produce ·OH by mixing $H_2O_2$ and $Fe^{2+}$ to realize the oxidation of organic pollutants, although Fenton reagent has the advantages of non-toxicity and short reaction time, but there are its related defects. The Fenton-like technology has been widely studied because of its various forms and better results than the traditional Fenton technology in terms of pollutant degradation efficiency. This paper reviews the electro-Fenton technology among the Fenton-like technologies and provides an overview of the homogeneous electro-Fenton. It also focuses on summarizing the effects of factors such as $H_2O_2$, reactant concentration, reactor volume and electrode quality, reaction time and voltage (potential) on the efficiency of electro-Fenton process. It is shown that appropriate enhancement of $H_2O_2$ concentration, voltage (potential) and reaction volume can help to improve the process efficiency; the process efficiency also can be improved by increasing the reaction time and electrode quality. Feeding modes of $H_2O_2$ have different effects on process efficiency. Finally, a considerable number of experimental studies have shown that the combination of electro-Fenton with ultrasound, anodic oxidation and electrocoagulation technologies is superior to the single electro-Fenton process in terms of pollutant degradation.

**Keywords** Homogeneous electro-Fenton · Reactant concentration · $H_2O_2$ · Anodic oxidation · Electrocoagulation technology · Ultrasound technology

## Introduction

Fenton's reagent is a mixture of $H_2O_2$ and $Fe^{2+}$, through the combination of these two substances produces ·OH in order to achieve the oxidation of organic pollutants for degradation and removal. The use of this reagent dates back to the late 19 century when Fenton (1894, 1896) catalyzed the oxidation of $H_2O_2$ through the use of $FeSO_4$ salt and was used to degrade tartaric acid. In 1934, Haber and Weiss (1934) formalized the reaction mechanism of Fenton's reagent, which included the involvement of hydroxyl radicals (·OH). The ·OH is the most reactive oxygen species (ROS) known, with a standard oxidation potential of up to 2.8 V, and therefore has a low selectivity for substances. From the reaction mechanism, Fenton technology is in acidic conditions, $H_2O_2$ in the presence of $Fe^{2+}$ to generate a strong oxidizing capacity of the hydroxyl radical and trigger more other reactive oxygen, in order to achieve the degradation of organic matter, the oxidation process for the chain reaction. The relevant reaction equations are as follows:

Chain reaction initiation phase:

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + \cdot OH \tag{1}$$

Chain reaction delivery phase:

$$\cdot OH + Fe^{2+} \longrightarrow Fe^{3+} + OH^- \tag{2}$$

$$\cdot OH + H_2O_2 \longrightarrow \cdot HO_2 + H_2O \tag{3}$$

$$Fe^{3+} + H_2O_2 \longrightarrow Fe^{2+} + \cdot HO_2 + H^+ \tag{4}$$

$$\cdot HO_2 + Fe^{3+} \longrightarrow Fe^{2+} + \cdot O_2 + H^+ \tag{5}$$

Chain reaction termination phase:

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$$\cdot HO_2 + Fe^{2+} + H^+ \longrightarrow Fe^{3+} + H_2O_2 \quad (6)$$

$$\cdot HO_2 + \cdot O_2 - + H^+ \longrightarrow H_2O_2 + O_2 \quad (7)$$

$$\cdot O_2 - + Fe^{2+} + 2H^+ \longrightarrow Fe^{3+} + H_2O_2 \quad (8)$$

In the above reaction, the reaction of $Fe^{3+}$ with $H_2O_2$ produces perhydroxyl radicals ($\cdot HO_2$), which is also known as a "Fenton-like" reaction. The oxidizing ability of $\cdot HO_2$ is lower than that of other ROS, but $\cdot HO_2$ is involved in the reduction of $Fe^{3+}$ to $Fe^{2+}$ (Equation (5)), which maintains the generation of ·OH and thus the reaction (Equation (2)). (Salgado et al. 2013).

Although the Fenton reagent has the advantages of non-toxicity, short reaction time and low cost, it also has the disadvantages of adding too much reagent and easy to produce secondary pollution. Fenton-like technology is a method of adding other technologies on the basis of traditional Fenton technology to achieve efficient degradation of pollutants. Since the Fenton-like technology has obvious improvement compared with the traditional Fenton technology, the research on Fenton-like technology has gradually become a hot spot. For example, Possetto et al. (2018) investigated the effect of degradation of Bioallethrin insecticide by using modified photo-Fenton process, and the experimental results showed that modified photo-Fenton process could achieve the complete mineralization of this insecticide in less than one hour. Hung et al. (2019) used a bacterial enzyme, dihydrolipoamide dehydrogenase, extracted from strain TX1 to form a Fenton's reagent for the degradation of the organic matter octylphenol polyethoxylate ($OPEO_n$) by the bio-Fenton process in conjunction with its bacterial enzyme in the presence of excess metal mixtures, and the experimental results showed that the process combined with its bacterial enzyme allowed for the breakage of ethoxylate chains, which led to the efficient degradation of ethoxylates. Wang et al. (2020) carried out a study related to the de-complexation of the organic substance Ni-EDTA by microwave-assisted Fenton process, and the experimental results showed that microwave irradiation could promote the cleavage of refractory bonds in the substance under certain conditions, and the de-complexation efficiency of 94% could be achieved in 10 minutes of the reaction. Li et al. (2013b) carried out a study related to the treatment of wastewater collected from the munitions processing plant by using the ultrasound and Fenton process, and the results showed that: under certain conditions of pH value, temperature, and reaction time, increasing the ultrasonic intensity can accelerate the removal rate of TOC, COD and colored substances in the wastewater.

Electro-Fenton, as a kind of Fenton-like technology, its principle is to add an external power source on the basis of its Fenton technology to continuously generate $H_2O_2$ through electrochemical reaction, so as to degrade organic substances and pollutants. The principle of electro-Fenton technology in the electric field and acidic solution conditions, electro-Fenton system of the cathode surface of the electronic reaction of oxygen to generate $H_2O_2$, at the same time $Fe^{3+}$ get electrons to reduce to $Fe^{2+}$, through the strength of the electric field to control the rate of generation of $H_2O_2$ and $Fe^{2+}$; in the anodic surface, the water loses electrons to be oxidized, generating oxygen and $H^+$; the solution of the $H_2O_2$ and $Fe^{2+}$ reaction to generate ·OH, used for oxidative degradation of organic matter, pollutants. Electro-Fenton does not require or only need to add a small amount of chemical reagents, and does not produce secondary pollution, in the electrolysis process parameters can be freely controlled, electro-Fenton process compared to the traditional Fenton reagent an advantage is that the reaction (Equation (1)) is electrocatalytic, because $Fe^{2+}$ can be reduced by $Fe^{3+}$ at the cathode and quickly generate (Brillas et al. 2009; Labiadh et al. 2015).

This review outlines the study of electro-Fenton related influencing factors and their related combined technologies, with the first part outlining the principle of homogeneous electro-Fenton reaction and their reaction forms. In the second part, the literature analysis is summarized for $H_2O_2$, reactant concentration, reaction volume and electrode quality, reaction time and voltage (potential) among the electro-Fenton influencing factors. The third part describes the combined application of electro-Fenton with ultrasound, anodic oxidation and electrocoagulation technology.

## Homogeneous electro-fenton

The homogeneous electro-Fenton reaction, as a form of electro-Fenton reaction, occurs in the same phase, i.e., all chemical reactions take place in the liquid phase; oxidation and mineralization of the target pollutants are achieved through the use of $H_2O_2$ generated at the cathode, and catalyzed by soluble $Fe^{2+}$ in solution. The advantages of this method, in addition to the fact that the $Fe^{3+}$ produced after the reaction can be reduced to $Fe^{2+}$ at the cathode, are as follows (Martínez-Huitle and Brillas 2009):

(i) Since $H_2O_2$ is generated in situ, it effectively avoids transportation and storage risks;
(ii) The addition of $Fe^{2+}$ is much smaller than conventional Fenton reagents and the degradation rate of organic matter is faster than that of conventional Fenton;
(iii) Potential total mineralization with relatively low power consumption.

However, there are corresponding disadvantages to the homogeneous electro-Fenton (Brillas 2021):

(i) There is a great limitation of the reaction pH;
(ii) Acidification of wastewater with large quantities of chemicals prior to treatment or neutralization of treated solutions prior to treatment;
(iii) Sludge treatment after neutralization will increase the operating cost.

In disadvantage (i), since the application of homogeneous electro-Fenton is limited by strongly acidic pH conditions (pH: 2.8-3.5), post-treatment neutralization of the treated water is required from an environmental point of view (Ganiyu et al. 2018; Poza-Nogueiras et al. 2018). Figure 1 shows the mechanism of pollutant degradation by homogeneous electro-Fenton.

The homogeneous electro-Fenton process can have two different configurations. In the first configuration the Fenton reagent is added externally to the reactor and an inert electrode with high catalytic activity is used as an anode material, while in the second configuration only hydrogen peroxide is added externally and $Fe^{2+}$ is supplied by a sacrificial cast iron anode (Govindan et al. 2014b, 2015a, 2020b). Depending on the addition or formation of Fenton reagents, the homogeneous electro-Fenton process can be divided into four electro-Fenton processes: Cathode-$Fe^{2+}$ cycle method (EF-$H_2O_2$-Fere), Cathode-electro-Fenton (EF-$H_2O_2$), Sacrificial anode electro-Fenton (EF-Feox), $Fe^{2+}$ cyclic method (EF-Fere) (Jiang and Zhang 2007; Kumar et al. 2019):

(1) Cathode-$Fe^{2+}$ cycle method (EF-$H_2O_2$-Fere)

This reaction configuration does not require the addition of $H_2O_2$ solution and $Fe^{2+}$ solution, and $H_2O_2$ is produced by exposing air or $O_2$ near the cathode, and under acidic conditions $O_2$ undergoes a 2-electron reduction reaction at the cathode. While the anode material is iron, Fe loses electrons by oxidation to produce $Fe^{2+}$. In this configuration, the electrode reaction is as shown in Fig. 2, and the related electrode reaction equation is as follows:

Anodic reaction:

$$Fe - 2e^- \longrightarrow Fe^{2+} \tag{9}$$

$$2H_2O - 4e^- \longrightarrow O_2 + 4H^+ \tag{10}$$

Cathodic reaction:

$$O_2 + 2H^+ + 2e^- \longrightarrow H_2O_2 \tag{11}$$

$$2H_2O + 2e^- \longrightarrow H_2 + 2OH^- \tag{12}$$

$$Fe^{3+} + e^- \longrightarrow Fe^{2+} \tag{13}$$

(2) Cathode-electro-Fenton method (EF-$H_2O_2$)

In this reaction configuration, $H_2O_2$ is still produced in situ by an electrode reaction, but $Fe^{2+}$ needs to be added externally to participate in the system reaction to form the Fenton reagent. The reaction mechanism is that $H_2O_2$ is generated by exposing $O_2$ or air near the cathode under acidic solution conditions and the reduction reaction occurs at the cathode, and $H_2O_2$ undergoes a Fenton reaction with the added $Fe^{2+}$ to generate ·OH, and at the same time, oxidized

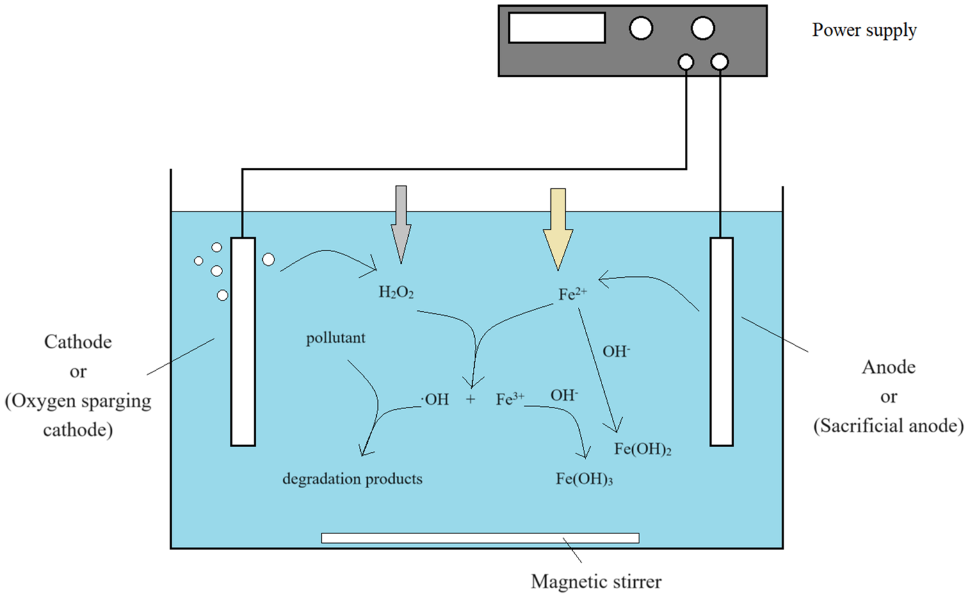


**Fig. 1** Mechanism diagram of homogeneous electro-Fenton degradation of pollutants

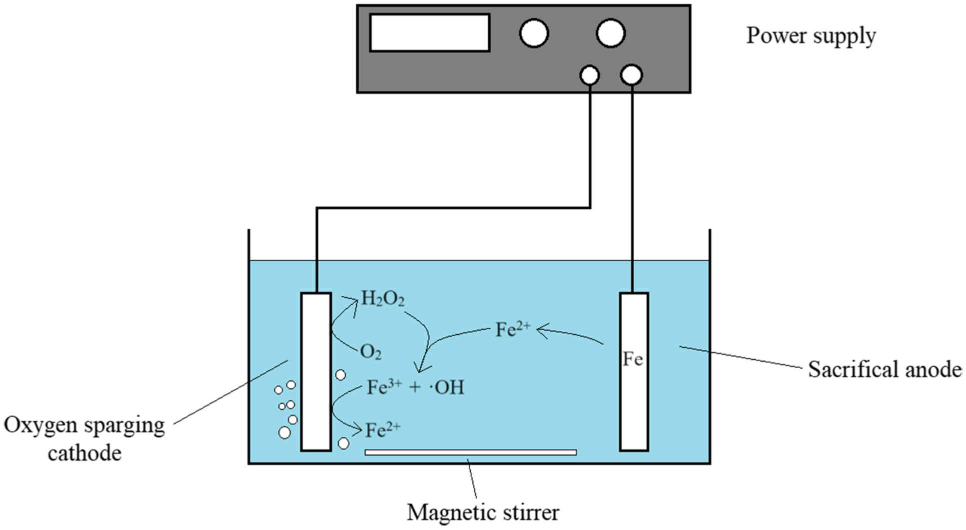


**Fig. 2** Cathode-$Fe^{2+}$ cycle method

$Fe^{3+}$ is obtained, and $Fe^{3+}$ can be reduced again at the cathode to form $Fe^{2+}$. The reaction schematic diagram of the present configuration is shown in Fig. 3, and the relevant electrode reactions of this reaction configuration are as follows:

Anodic reaction:

$$2H_2O{-}4e^- \longrightarrow O_2 + 4H^+ \tag{14}$$

Cathodic reaction:

$$O_2 + 2H^+ + 2e^- \longrightarrow H_2O_2 \tag{15}$$

$$2H_2O + 2e^- \longrightarrow H_2 + 2OH^- \tag{16}$$

$$Fe^{3+} + e^- \longrightarrow Fe^{2+} \tag{17}$$

Sometimes this reaction configuration can also be achieved by direct addition of $Fe^{3+}$, which is reduced to $Fe^{2+}$ by gaining electrons at the cathode, resulting in a Fenton's reagent reaction with $H_2O_2$.

### (3) Sacrificial anode electro-Fenton method (EF-Feox)

In this reaction configuration, the anode material is iron electrode, $Fe^{2+}$ is generated by Fe oxidation reaction at the anode, i.e., Fe loses two electrons to generate $Fe^{2+}$, then $Fe^{2+}$ reacts with the externally added $H_2O_2$ to generate ·OH, and $Fe^{2+}$ generates $Fe^{3+}$ through oxidation. In addition to the factor of ·OH in the process of degradation of organics and pollutants in the system, there are also sometimes flocculation effects of $Fe(OH)_2$ and $Fe(OH)_3$. Figure 4 shows

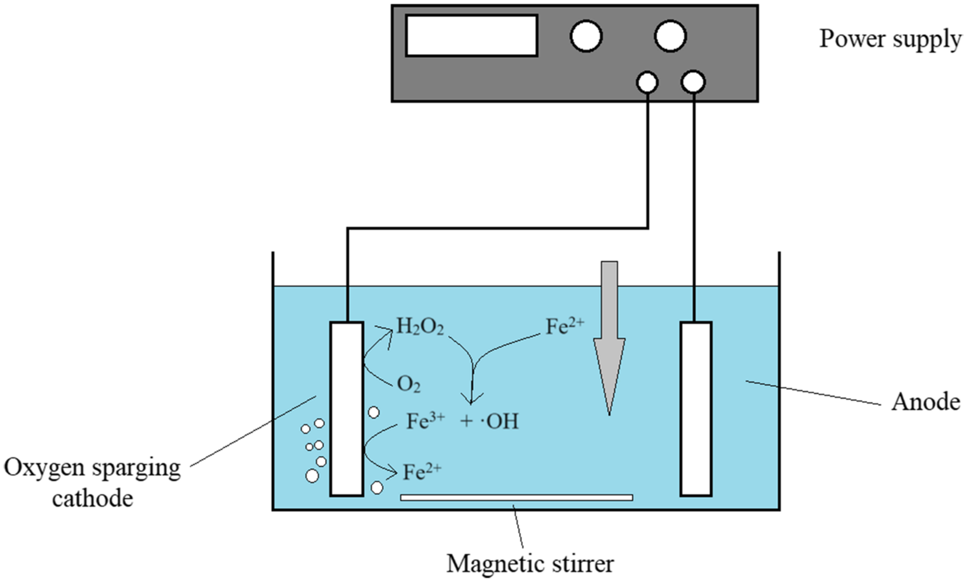


**Fig. 3** Cathode-electro-Fenton method

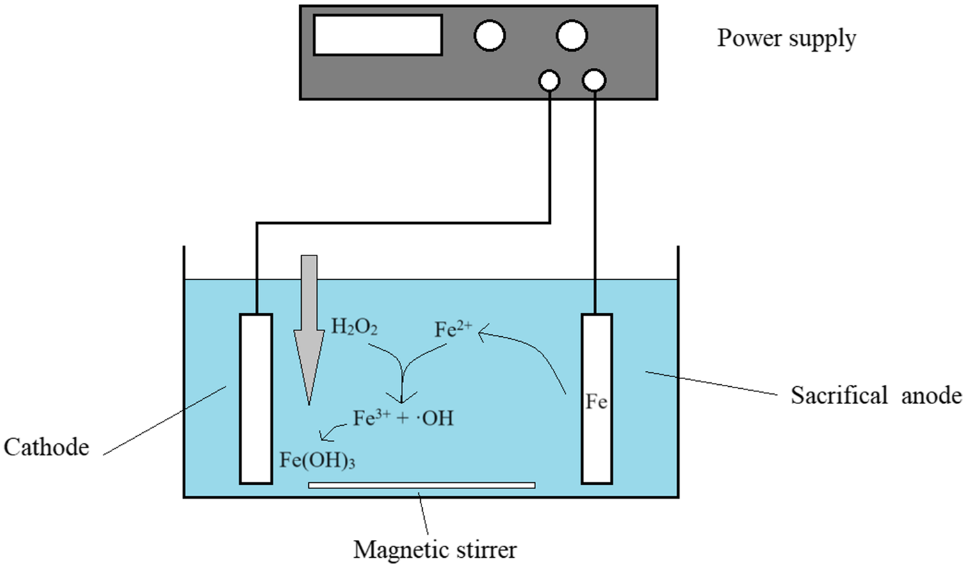


**Fig. 4** Sacrificial anode electro-Fenton method

the schematic diagram of this reaction configuration, and its related electrode reaction formula is as follows:

Anodic reaction:

$$Fe - 2e^- \longrightarrow Fe^{2+} \tag{18}$$

$$2H_2O - 4e^- \longrightarrow O_2 + 4H^+ \tag{19}$$

Cathodic reaction:

$$2H_2O + 2e^- \longrightarrow H_2 + 2OH^- \tag{20}$$

(4) $Fe^{2+}$ cyclic Method (EF-Fere)

In this reaction configuration, $H_2O_2$ and $Fe^{2+}$ are added externally to participate in the electro-Fenton system reaction, and there is no need to expose air or $O_2$. During the electro-Fenton reaction, $Fe^{2+}$ can be oxidized to produce $Fe^{3+}$, and $Fe^{3+}$ can be formed through the cathode reduction reaction to form $Fe^{2+}$, which realizes the reuse of $Fe^{2+}$. Moreover, this reaction does not produce a large amount of sludge, which causes secondary pollution. In addition, the reaction will not produce a large amount of sludge and cause secondary pollution. Figure 5 shows the schematic diagram of the reaction configuration, and its related electrode reaction equation is as follows:

Anodic reaction:

$$2H_2O - 4e^- \longrightarrow O_2 + 4H^+ \tag{21}$$

Cathodic reaction:

$$2H_2O + 2e^- \longrightarrow H_2 + 2OH^- \tag{22}$$

$$Fe^{3+} + e^- \longrightarrow Fe^{2+} \tag{23}$$

Similar to the reaction configuration in (2), sometimes this reaction configuration can be furthered by direct addition of $Fe^{3+}$, by cathodic reduction, which results in a Fenton's reagent reaction.

## Influencing factor on electro-Fenton

### Effect of $H_2O_2$

$H_2O_2$ as an important component of the Fenton reagent in the electro-Fenton reaction has a significant impact on the degradation of pollutants or organic matter. $H_2O_2$ reagent can provide ·OH for the Fenton reaction and is used to perform the mineralization mechanism of the generated sludge (Rahmani et al. 2015; Moussavi et al. 2012). Related studies have shown that the concentration (dosage) and method of addition of $H_2O_2$ reagents can have an impact on the electro-Fenton process; the appropriate method of addition of $H_2O_2$ and the concentration (dosage) of the reagents can optimize the degradation efficiency of the electro-Fenton process to achieve efficient degradation of organic matter or pollutants, but if they are added inappropriately, the electro-Fenton reaction will be negatively impacted, especially for the economic cost of the $H_2O_2$ and the possible negative impact on the performance of the electro-Fenton (Akyol et al. 2013).

(1) Additive concentration (dosage)

In the electro-Fenton reaction, the addition of $H_2O_2$ plays an important role in the degradation of substances. However, the concentration (dosage) of $H_2O_2$

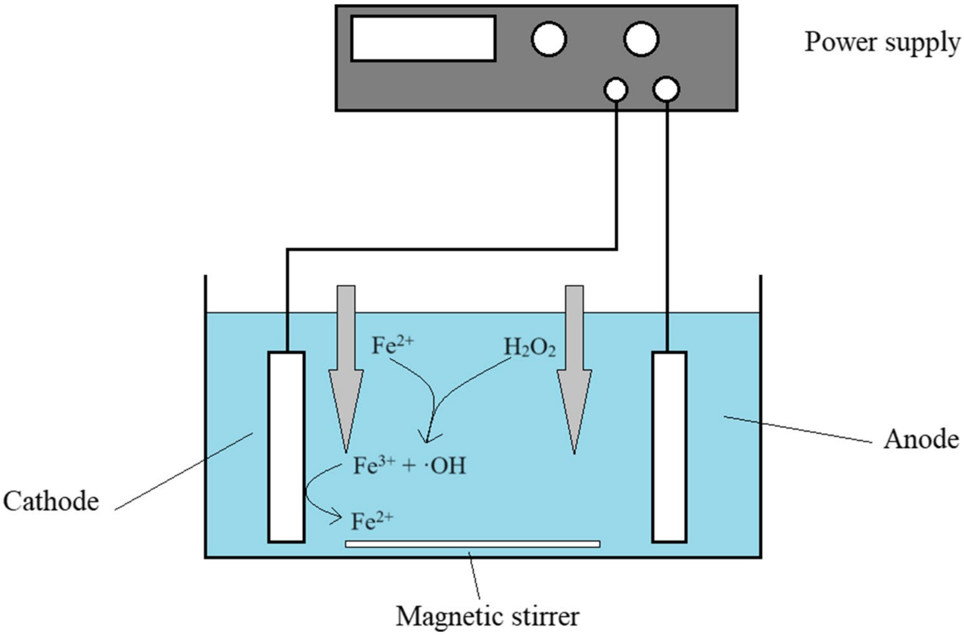


**Fig. 5** $Fe^{2+}$ cycling method

does not have a positive relationship with the removal of substances in water. Related studies have shown that too high concentration (dosage) of $H_2O_2$ does not result in a higher removal efficiency of COD, and at the same time reduces the efficiency of the electro-Fenton in removing organic matter or pollutants (Zhang et al. 2021; Ghosh and Samanta 2011; Dolatabadi and Ahmadzadeh 2019).

The main reason why the addition of $H_2O_2$ enhances COD removal is the possible production of ·OH, which plays an important role in the decomposition of organic matter (Babuponnusami and Muthukumar 2012; Moayerikashani and Soltani 2013). The concentration of ·OH is determined by the concentration (dosage) of $H_2O_2$ added until the optimal $H_2O_2$ concentration (dosage) of the target pollutant is reached. However, when the concentration (dosage) of $H_2O_2$ exceeds the optimal value for the degradation of the pollutant, the degradation rate decreases. Relevant studies have analyzed that at too high concentration (dosage) of $H_2O_2$, the oxidation potential of the electro-Fenton process is lowered due to the scavenging effect of $H_2O_2$ on ·OH (Equation (24)(25)) (Sevimli et al. 2014; Mohajeri et al. 2010); in addition, at too high concentration (dosage) of $H_2O_2$, the combination of the two ·OHs to form $H_2O_2$ also results in the decrease in the efficiency of pollutant's degradation (Equation (26)) (Muruganandham and Swaminathan 2004), and the relevant reaction equations are as follows. In addition to the above reasons, it has been suggested that excess $H_2O_2$ leads to the generation of short-chain organic acids during pollutant degradation, which hinders further oxidation of the pollutant (Umar et al. 2010; Dargahi et al. 2021b).

$$H_2O_2 + \cdot OH \longrightarrow H_2O + \cdot HO_2 \tag{24}$$

$$\cdot HO_2 + \cdot OH \longrightarrow H_2O + O_2 \tag{25}$$

$$2 \cdot OH \longrightarrow H_2O_2 \tag{26}$$

In addition to this, the addition of $H_2O_2$ also increases the pH of the reaction solution (Zhao et al. 2012). Although electro-Fenton has a high removal efficiency, the consumption of $H_2O_2$ also increases the cost of consumption in addition to the electrode consumption (Can 2013). Therefore, the initial concentration (dosage) of $H_2O_2$ plays a very important role in the electro-Fenton reaction (Ting et al. 2009). Figure 6 also shows the influence of $H_2O_2$ concentration (dosage) on the electro-Fenton process in some studies.

(2) Feeding mode

The feeding mode of $H_2O_2$ is an important parameter for the performance of an electro-Fenton system, and increasing $H_2O_2$ during the initial or reaction process will result in changes in the ratios of $[H_2O_2]/[COD]$ and $[H_2O_2]/[Fe^{2+}]$to achieve the final removal of COD (Primo et al. 2008). Back in 2005, Zhang et al. (2005) found that increasing the concentration of $H_2O_2$ in multiple steps in the Fenton system improved the treatment of waste leachate compared to increasing it in a single stage. In a subsequent study, they applied the electro-Fenton method to the treatment of waste leachate by using an electrolytic batch reactor containing 200 mL of solution; the results showed that COD removal increased with the gradation of dosage, with the highest removal (79%) when using a continuous addition mode (Zhang et al. 2006) (Fig. 7a). In recent years, Alavi et al. (2019) (Fig. 7b) compared the effect of one-step injection $H_2O_2$ versus four-step injection $H_2O_2$ on COD removal efficiency in the electro-Fenton process when treating leachate from a composting plant through the electro-Fenton process; the results showed that the minimum value of COD removal was associated with one-step injection and the maximum value was associated with four-step injection, and this finding exists in agreement with the trend of the results of the study conducted by Zhang et al. (2005, 2006).

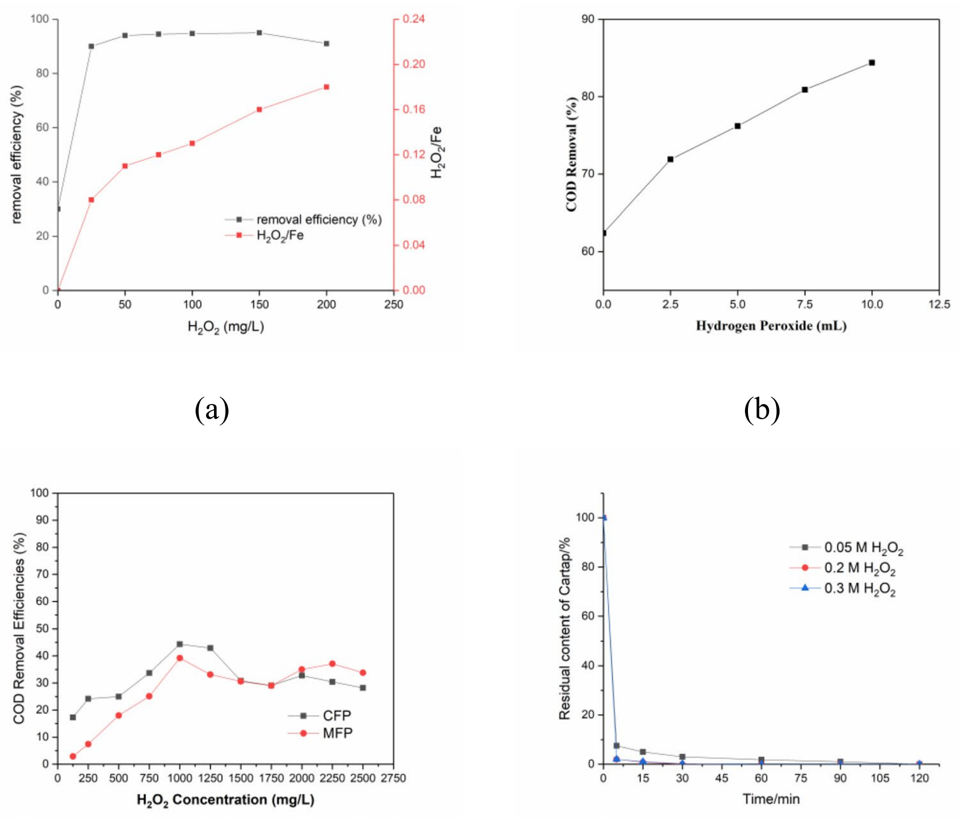
The reason for this experimental result is that the gradual addition of $H_2O_2$ keeps $H_2O_2$ at a low value, minimizing the negative effect of ·OH removal (Zhang et al. 2006). On the other hand, the addition of $H_2O_2$ in the initial steps of each stage of the experiment leads to a rapid production of ·OH, which makes the reaction of ·OH with $H_2O_2$ and $Fe^{2+}$ more prominent; the relevant reaction equation is as follows (Alavi et al. 2019):

$$\cdot HO + H_2O_2 \longrightarrow \cdot HO_2 + H_2O \tag{27}$$

$$\cdot HO + Fe^{2+} \longrightarrow Fe^{3+} + OH^- \tag{28}$$

In addition, Zhang et al. (2007) further investigated the effect of gradually shortening the feeding time on the degradation rate of pollutants on the basis of multiple dosing of $H_2O_2$. The results showed that appropriately shortening the feeding time can increase the content of $H_2O_2$, which can produce more ·OH by reacting with $Fe^{2+}$, so as to enhance the pollutant removal efficiency; however, the further shortening of the feeding time will lead to the scavenging effect of the ·OH by the excess $H_2O_2$, resulting in a lower removal efficiency of the pollutants. However, Anotai et al. (2010) (Fig. 7c) found that the one-step and two-step additions of $H_2O_2$ resulted in similar degradation rates without significant differences in the degradation of aniline by the electro-Fenton process. During the degradation process, aniline was rapidly degraded in the first two minutes of the reaction, while the rate of aniline degradation slowed down in 2-30 minutes.

**Fig. 6** Experimental research on the influence of $H_2O_2$ concentration (dosage) (presented from (**a**) Teymori et al. 2019; (**b**) Can 2013; (**c**) Sevimli et al. 2014; (**d**) Hoang and Holze 2020)

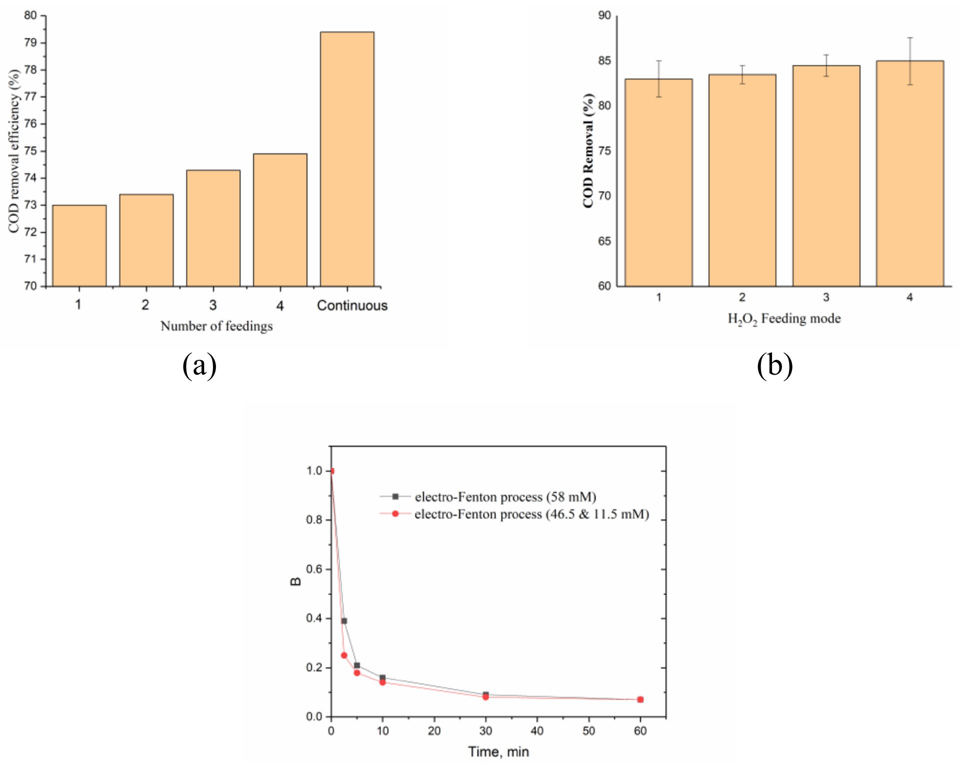


(a)

(b)

(c)

(d)



(a) (b)

(c)

**Fig. 7** Experimental research on the influence of $H_2O_2$ feeding mode (presented from (**a**) Zhang et al. 2006; (**b**) Alavi et al. 2019; (**c**) Anotai et al. 2010)

Finally, Table 1 lists relevant literature studies on the effect of $H_2O_2$ on the electro-Fenton process.

Table 1 Examples of effect of $H_2O_2$ by Electro-Fenton reaction process

## Effect of reactant concentration

The concentration of reactive substances, as an important variable in the electro-Fenton reaction, can be analyzed to elucidate where the ability of the oxidation of substances in the electro-Fenton process lies. And the change of reaction concentration will also affect the degradation efficiency and degradation process of electro-Fenton reaction.

Bedolla-Guzman et al. (2016) in investigating the effect of azo dye concentration on the degradation process found that the decolorization rate decreased as the dye concentration increased; Guelfi et al. (2018) in investigating the effect of herbicide concentration on the rate of degradation under certain conditions found that the time to reach complete removal increased when the concentration of herbicide increased. The reason for such an experimental phenomenon is that under certain reaction conditions, a certain amount of ·OH is produced per unit time. When the concentration of pollutant increases, the amount of ·OH is not sufficient to degrade the high concentration of pollutant, so the removal efficiency of pollutant decreases (Teymori et al. 2019). In contrast, the increase of organic molecules in solution requires more hydroxyl radicals for degradation (Annabi et al. 2016), so the time required to achieve a higher degradation rate increases when the concentration of reactants is higher. In addition, for the degradation of some organic substances or pollutants, higher reactive pollutants produce intermediates that are difficult to degrade during the degradation process, and this intermediate product is an obstacle to the degradation of the original target substance, and the intermediate product competes with the target substance for ·OH, which leads to a decrease in the rate of the reaction, which can be deduced from the experimental studies of El-Desoky et al. (2010) and Kadji et al. (2020) analysis.

On the other hand, changes in the concentration of reacting substances are also related to kinetics, and pseudo first-order and pseudo second-order reaction kinetics are commonly used to explore the relationship between the concentration of reacting substances and the rate of degradation with the following correlation equations (Equation (29)(30)):

Pseudo first-order reaction kinetics:

$$\ln C_t = \ln C_0 - K_1 t \tag{29}$$

Pseudo second-order reaction kinetics:

$$\frac{1}{C_t} = K_2 t + \frac{1}{C_0} \tag{30}$$

**Table 1** Examples of effect of $H_2O_2$ by Electro-Fenton reaction process

| Effect of adding concentration (dosage) | | | | |
|---|---|---|---|---|
| Organics/Pollutants | Reaction conditions | $H_2O_2$ conditions | Results | References |
| Fruit-juice Production Wastewater | pH:3.5,<br>time: 25min,<br>stirred under 200rpm after reaction in 120min | dosage: 0, 2.5, 5, 7.5, 10 mL | COD removal increased to 84.4% under 10mL | Can 2013 |
| White Liquor | pH:3, I=1.0A, time=45min | dosage: 250-5000 mg/L | COD removal increased to 54.3 under 1500mg/L | Sevimli et al. 2014 |
| Rayon Industry Wastewater | pH:3, I=0.2A, time: 50min | dosage: 765, 1530, 3825 mg/L | The maximum reduction of COD was 88% (1530 mg/L) | Ghosh and Samanta 2011 |
| Landfill Leachate | pH:3.5,<br>electrodes distance: 3cm,<br>time: 30min,<br>voltage: 30V, | initial concentration: 7.5, 15, 100, 200 mg/L | COD removal: 85%<br>$NH_3$-N removal: 30.9%;<br>Phosphat removal:78.2% (the best condition in 15 mg/L) | Aval et al. 2017 |
| Scarlet Red Dye | dye:0.4 g/L,<br>pH:3,<br>$Fe^{2+}$=0.025 g/L,<br>CD=0.4 mA $cm^{-2}$ | initial concentration: 0.1-0.5 g/L | the maximum removal efficiency is about 90% under 0.5 g/L | Gomathi et al. 2018 |
| Composting plant leachate | $COD_0$=28000mg/L,<br>pH:3,<br>TDS=4%,<br>CD=2.5A | dosage: 7.5, 15, 30, 45cc | the maximum of the COD removal was about 83±2%. | Alavi et al. 2019 |
| Metformin | by RSM model to compare the removal efficiency in different dosage | dosage: 100-400μg $L^{-1}$ | When the concentration of MET is constant, 300μg $L^{-1}$ achieves the maximum removal efficiency. | Dolatabadi and Ahmadzadeh 2019 |
| Cartap | $[Padan95SP]_0$=700 mg $L^{-1}$,<br>$V_{solution}$=250 mL,<br>$[Fe^{2+}]$=15 mM,<br>$[NaSO_4]$=0.05 M,<br>pH=3,<br>j=20 mA $cm^{-2}$ | initial concentration: 0.05M, 0.2M, 0.3M | 0.2M could realize rapid removal in a short time | Hoang and Holze 2020 |
| Malachite Green | initial MG concentration=200 mg/L;<br>pH=3;<br>current density=10 mA/cm2;<br>time=10 min | dosage:25, 50, 75, 100, 150, 200 mg/L | The removal efficiency reached the maximum when the dosage was 150mg/L. | Teymori et al. 2019 |
| Diazinon | pH=3,<br>voltage= 15V,<br>DZ concentration= 25 mg/L,<br>time=80 min,<br>$FeSO_4$ = 0.24 gr/L, NaCl= 0.1 g/L | initial concentration: 0, 10, 20, 30, 40, 50 mg/L | The removal efficiency reached the maximum at 30 mg/L. (88.6%) | Dargahi et al. 2021b |

**Table 1** (continued)

Effect of adding concentration (dosage)

| Organics/Pollutants | Reaction conditions | $H_2O_2$ conditions | Results | References |
|---|---|---|---|---|
| Quinoline | pH:3.0,<br>electrical conductivity: 15800 μs/cm,<br>voltage: 26.5 V<br>current density: 30.5 mA/cm$^2$ | initial concentration: 20-100 mmol/L | When the concentration of $H_2O_2$ was 71 mmol/L, the COD decline rate was the highest. | Zhang et al. 2021 |
| Phenol | initial phenol concentration: 200 mg/L,<br>pH:3,<br>$Fe^{2+}$=4 mg/L<br>current density: 12 mA/cm$^2$<br>time: 60 min | initial concentration:0-1000 mg/L | When $H_2O_2$ concentration was 600mg/L, phenol degradation rate was the highest, 89.5%, and COD removal rate was 59.8%. | Babuponnusami and Muthukumar 2012 |
| wastewater from liquid organic fertilizer plant | pH:3,<br>I=50 A/m$^2$ | dosage:5, 10, 25, 50mM | The removal efficiency at 25 mM was reached to 87% at 45 min (the maximum value) | Akyol et al. 2013 |
| Effect of feeding mode | | | | |
| Composting plant leachate | $H_2O_2$/COD: 0.6, other conditions are the same as above. | Feeding mode: 1-4 steps | Four steps feeding mode was the best on COD removal. | Alavi et al. 2019 |
| Landfill leachate | $H_2O_2$=0.17mol/L,<br>$Fe^{2+}$=0.014 mol/L,<br>pH:3,<br>I=2 A,<br>d=1.3 cm | feedings: 1, 2, 3, 4, continuous | When the feeding mode was continuous, the COD removal efficiency reached maximum was 79.4%. | Zhang et al. 2006 |
| Aniline | pH:3.2,<br>$Fe^{2+}$:1.07×10$^{-3}$ M,<br>aniline:0.01 M | Feeding mode:<br>①$H_2O_2$:58 mM<br>②$H_2O_2$:46.5 & 11.5 pm | The degradation rate of aniline does not change significantly with the feeding method. | Anotai et al. 2010 |

where $C_0$ and $C_t$ denote the concentration of the substance at the beginning of the reaction and after the reaction time $t$, respectively, and $K_1$ and $K_2$ denote the pseudo first-order and pseudo second-order reaction constants, respectively. The values of $K_1$ and $K_2$ can be determined from the analysis of the slopes of $\ln C_t$ versus t and $1/C_t$ versus $t$, respectively (Moussavi et al. 2011). From the experiments of Mansour et al. (2011), Teymori et al. (2019), it can be seen that the time required to reach the same $\ln(C_0/C_t)$ is increasing with the increase in the concentration of the reactants and also due to the above analyzed reasons, the values of $K_1$, $K_2$ are eventually decreasing.

On the other hand, for the mineralization current efficiency (MCE), an increase in the concentration of reactive substances leads to an increase in the final MCE value (Flores et al. 2016a, 2017; Guelfi et al. 2016). This is due to the increase in organic loading, where a larger organic load favors a larger amount of oxidized substances to react with the intermediates (Flores et al. 2016b), thus reducing the occurrence of associated parasitic reactions. Common parasitic reactions are (i) the oxidation of BDD (·OH) to $O_2$ (Equation (31)); (ii) the scavenging of ·OH by $Fe^{2+}$ with $H_2O_2$ (Equation (32)(33)) (Sirés and Brillas 2012; Sirés et al. 2014); (iii) the isolation of $S_2O_8^{2-}$ from $SO_4^{2-}$ with inhibition of emission of the reaction $H_2O$ (Equation (34)(36)); and (iv) the generation of other weaker oxidizing agents by reaction and at the anode (Equation (35)). The relevant reaction equations are given below:

$$2\mathrm{BDD}(\cdot\mathrm{OH}) \longrightarrow 2\mathrm{BDD} + \mathrm{O_2} + 2\mathrm{H^+} + 2\mathrm{e^-} \tag{31}$$

$$\mathrm{H_2O_2} + \cdot\mathrm{OH} \longrightarrow \cdot\mathrm{HO_2} + \mathrm{H_2O} \tag{32}$$

$$\mathrm{Fe^{2+}} + \cdot\mathrm{OH} \longrightarrow \cdot\mathrm{HO_2} + \mathrm{H_2O} \tag{33}$$

$$2\mathrm{SO_4^{2-}} \longrightarrow \mathrm{S_2O_8^{2-}} + 2\mathrm{e^-} \tag{34}$$

$$\mathrm{M} + \mathrm{H_2O} \longrightarrow \mathrm{M}(\cdot\mathrm{OH}) + \mathrm{H^+} + \mathrm{e^-} \tag{35}$$

$$3\mathrm{H_2O} \longrightarrow \mathrm{O_3} + 6\mathrm{H^+} + 6\mathrm{e^-} \tag{36}$$

However, Wang et al. (2010) in exploring the effect of concentration change of Acid Red 14 dye found that the MCE of the EF process increased to a maximum value when the initial AR14 concentration did not exceed a certain value, and thereafter, it gradually decreased over a longer electrolysis time. However, when the AR14 concentration exceeded a certain value, the MCE decreased to a minimum value and then increased with electrolysis time. This contradicts the results of some of the experiments, which they attribute to the ratio between the concentration of AR14 and the concentration of ·OH produced by the electro-Fenton reaction.

In addition to the degradation of substances, changes in the initial concentration of reactants can also have an impact on the reaction state and the synthesis of new substances in the conversion synthesis of related organic substances using the electro-Fenton process. For example, Galia et al. (2016), Lanzalaco et al. (2017) explored the effect of different PVP specific gravity on the synthesis of nanogels when polyvinylpyrrolidone (PVP) was synthesized using the electro-Fenton process and found that the degree of decay of the hydrodynamic radius ($R_h$) decreases as the specific gravity of the PVP is increased, which is attributed to the decrease in the average number of free radicals per chain due to a higher concentration of polymer chains and a fixed rate of generation of ·OH. Furthermore, in the experiments of Rekik et al. (2017) on the conversion of vanillic acid (VA) to protocatechuic acid (PCA), it was found by varying the concentration variable that: as the concentration of VA increased, the concentration of the converted PCA increased; in addition the energy consumption in the reaction decreased as the concentration of VA increased. This experimental phenomenon can be attributed to the reduction of ·OH loss in the secondary reaction (Equation (37)), which leads to an increase in process efficiency.

$$2 \cdot \mathrm{OH} \longrightarrow 1/2\mathrm{O_2} + \mathrm{H_2O} \tag{37}$$

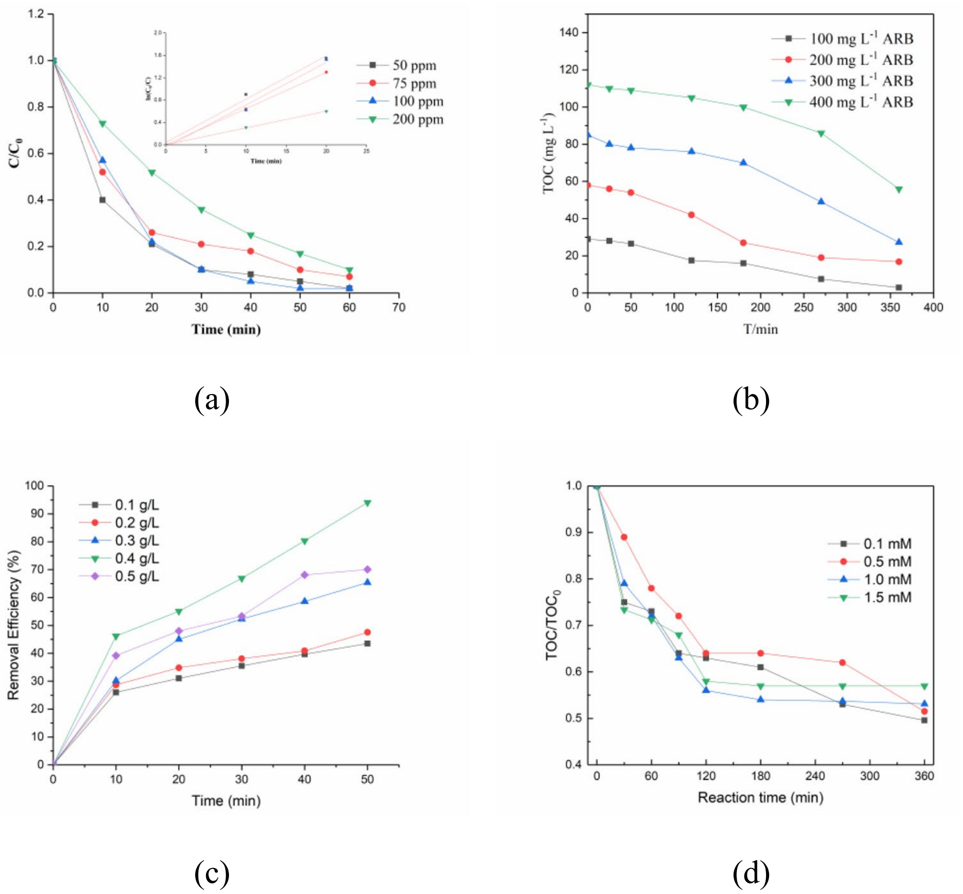
Figure 8 and Table 2 respectively show and list relevant literature studies on the effect of reactant concentration on the electro-Fenton process.

Table 2 Examples of effect of reactant concentration by Electro-Fenton reaction process

## Effect of reaction volume and electrode quality

The current study shows that varying the reaction volume and the electrode quality as two geometrical parameters in the electro-Fenton reaction will have an effect on the yield of $H_2O_2$ in the reaction.

Xia et al. (2015) investigated the ability of polyacrylonitrile-based carbon fiber brush (PAN-CFB) cathodes to produce $H_2O_2$ in different volumes of $Na_2SO_4$ electrolyte. The experimental results showed that the electro-generated yield of $H_2O_2$ tended to increase and then decrease when the reaction volume was gradually decreased. However, their theoretical speculations showed that the yield of $H_2O_2$ should increase with decreasing reaction volume. They analyzed that the reason for this experimental deviation is that the reaction solution in a very small volume is not conducive to the accumulation of the yield of $H_2O_2$ and produces a larger decomposition of $H_2O_2$, which is reflected in the action of the anode on $H_2O_2$.



(a) (b)

(c) (d)

**Fig. 8** Experimental research on the influence of reactant concentration (presented from (**a**) Annabi et al. 2016; (**b**) Wang et al. 2010; (**c**) Gomathi et al. 2018; (**d**) Lan et al. 2016)

Chen et al. (2019), on the other hand, investigated the effect of different reactor volumes and electrode qualities on the yield of $H_2O_2$. They found that increasing the electrode quality of the cathode increases the yield of $H_2O_2$ while increasing the reactor volume decreases the yield of $H_2O_2$ but the current efficiency increases. For the electrode quality, increasing the electrode quality increases the sites where $O_2$ reduction occurs, which in turn leads to an increase in the yield of $H_2O_2$. And increasing the reactor volume, although the concentration of $H_2O_2$ will decrease, the consumption of $H_2O_2$ will be weakened at low concentration of $H_2O_2$, and at the same time, the electron utilization efficiency is increased and the current efficiency is increased.

## Effect of reaction time

As one of the influential parameters of the electro-Fenton process, the reaction time not only reflects the trend of the relevant parameters over time during the degradation or oxidation of pollutants or organics, but also provides an assessment and reference for finding the optimal degradation efficiency of pollutants or organics with the optimal process cost.

Flores et al. (2017) in the treatment of 4-hydroxyphenylacetic acid in olive oil mill wastewater by the electro-Fenton process under certain conditions, concluded that the TOC content decreased at a fast rate during the first 120 minutes, and then slowly with increasing time after 120 minutes. Mansour et al. (2015) in the treatment of sulphadiazine by using the electro-Fenton process under certain conditions, found that: electrolysis at mineralization was only 2.1% and 18.1% at 30 and 60 min and the oxidation level was very low, their experiments indicated that intermediates were produced during the degradation process. Typically, the rate of degradation or mineralization of a contaminant increases at a certain rate over time for a short period of time; after a certain degradation or mineralization value has been reached, the rate of mineralization or degradation increases slowly over a longer period of time. This is usually attributed to the formation of degradation by-products (e.g., short-chain carboxylic acids) with reduced reaction kinetics with hydroxyl radicals (Ouarda et al. 2020). On the one hand, an increase in electrolysis time is directly related to an increase in degradation efficiency due to certain reaction conditions. By increasing the electrolysis time, the production of ·OH is also enhanced and the increase in the amount of ·OH leads to a higher efficiency in the evaluation process. However, with

**Table 2** Examples of effect of reactant concentration by Electro-Fenton reaction process

| Organics/Pollutants | Reaction conditions | Reactant concentration | Results | References |
|---|---|---|---|---|
| azo dye Acid Red 14 | solution:500 mL,<br>$Na_2SO_4$:0.05 mol $L^{-1}$,<br>pH:3.0,<br>I:0.36 A | AR14 concentration:100, 200, 300, 400 mg $L^{-1}$ | TOC removal rate decreases with the increase of dye concentration. At the same time, when the initial AR14 concentration is not in excess of 200 mg $L^{-1}$, the MCE of the EF process increases to a maximum, after which it falls progressively over longer periods of electrolysis. However, when the AR14 concentration exceeds 200 mg $L^{-1}$, the MCE falls to a minimum value and then increases over time with electrolysis. | Wang et al. 2010 |
| Dimethylacetamide (DMAC) | initial pH:3,<br>current density:50 mA $cm^{-2}$,<br>AuPd/CNTs:1 g/$L^{-1}$(0.5 wt% Au, 0.5 wt% Pd), | DMAC concentration:20, 100, 250, 500 mg $L^{-1}$ | The obtained kinetic constants decreased with increasing initial DMAC concentration. | Sun et al. 2015 |
| glyphosate | pH:3.0,<br>current density:0.36 A,<br>$Fe^{2+}$:1 mM,<br>t:360 min | glyphosate concentration:0.1 mM, 0.5 mM, 1.0 mM, 1.5 mM | The TOC removal was 50.4, 48.5, 46.9, and 43.0% at initial glyphosate concentration of 0.1, 0.5, 1.0 and 1.5 mM, respectively | Lan et al. 2016 |
| enoxacin | $Fe^{2+}$:0.2 mmol $L^{-1}$,<br>V:250 mL,<br>$Na_2SO_4$:50 mmol $L^{-1}$,<br>I:200 mA,<br>t:60 min | initial ENO concentration:50, 75, 100, 200 ppm | The degradation rate decreased with the increase of concentration, and the final degradation rate was similar after 60 min. | Annabi et al. 2016 |
| Reactive Yellow 160 azo dye | $Na_2SO_4$:0.05 mol $dm^{-3}$,<br>pH:3.0,<br>current density:100 mA $cm^{-2}$,<br>T:25°C,<br>$Fe^{2+}$:0.50 mmol $dm^{-3}$ | RY160:0.167 mmol $dm^{-3}$, 0.334 mmol $dm^{-3}$ | With the increase of dye concentration, the decolorization time of solution became longer. | Bedolla-Guzman et al. 2016 |
| trans-ferulic acid | $Fe^{2+}$:0.50 mM,<br>current density:33.3 mA $cm^{-2}$,<br>t:360 min,<br>$Na_2SO_4$:0.05 M<br>pH:3.0,<br>T:35°C | trans-ferulic acid concentration:0.167-1.668 mM | The kinetic constant decreases with the increase of concentration, and the TOC removal, mineralization current efficiency increased with concentration. | Flores et al. 2016a |
| poly(vinylpyrrolidone) | $Na_2SO_4$:0.05 mol $dm^{-3}$,<br>$FeSO_4$:1.0 mmol $dm^{-3}$,<br>pH:2.8,<br>current density:33.3mA $cm^{-2}$,<br>T:20°C | PVP concentration: 0.25-0.75 wt.% | The attenuation of hydrodynamic radius decreases with the increase of concentration. | Lanzalaco et al. 2017 |

**Table 2** (continued)

| Organics/Pollutants | Reaction conditions | Reactant concentration | Results | References |
|---|---|---|---|---|
| Propoxur (PX) | $Na_2SO_4$:0.05 M,<br>pH:3.0,<br>T:25°C,<br>current density:100 mA $cm^{-2}$,<br>$Fe^{2+}$:0.50 mM | PX concentration:0.19 mM-0.76 mM (100 mL) | A gradually slower decay of the normalized concentration as the initial amount of PX was raised. | Guelfi et al. 2016 |
| 4-Hydroxyphenylacetic acid | $Na_2SO_4$:0.05 M,<br>$Fe^{2+}$:0.50 mM,<br>current density:33.3 mA $cm^{-2}$,<br>pH:3.0,<br>t:360 min | 4-Hydroxyphenylacetic acid concentration:0.21 mM, 0.51 mM, 1.03 mM, 2.06 mM | At 360 min, TOC was reduced by 72.1%, 82.3%, 90.1% for 0.21, 0.51, 1.03 and 2.06 mM, respectively. MCE increased with the increase of concentration. | Flores et al. 2017 |
| scarlet red dye | pH:3,<br>$H_2O_2$:0.5 g/L,<br>$Fe^{2+}$:0.025 g/L,<br>CD:0.4 mA $cm^{-2}$,<br>t:50 min | scarlet red dye concentration:0.1, 0.2, 0.3, 0.4, 0.5 g/L | The optimum dye concentration was found to be 0.4 g/L for which maximum degradation was obtained. | Gomathi et al. 2018 |
| tyrosol | $Fe^{2+}$:0.50 mM,<br>KCl:1 mM,<br>$Na_2SO_4$:12 mM,<br>$NaNO_3$:0.01 mM,<br>current density:10 mA $cm^{-2}$,<br>pH:3.0 | $[Tyrosol]_0$:0.723, 1.047, 2.096, 5.239 mM | TOC removal rate decreases with the increase of substance concentration, MCE increases with the increase of concentration, and energy consumption decreases with the increase of concentration. | Flores et al. 2018 |
| Metribuzin (MTZ) | $Na_2SO_4$:0.05 M,<br>$Fe^{2+}$:0.5 mM,<br>pH:3.0,<br>T:25°C ,<br>current density:100.0 mA $cm^{-2}$ | $[MTZ]_0$:0.262, 0.523, 1.046 mM | The degradation was decelerated at higher herbicide content, reaching complete removal at increasing times of 90, 105 and 180 min for 0.262, 0.523 and 1.046 mM, respectively. | Guelfi et al. 2018 |
| Metformin (MET) | t:10 min,<br>pH:3,<br>current density:6 mA $cm^{-2}$,<br>$H_2O_2$ dosage:250 μL $L^{-1}$ | initial MET concentration:10-50 mg $L^{-1}$ | When the concentration of MET was 10 mg L-1, the maximum removal rate was obtained. | Dolatabadi and Ahmadzadeh 2019 |
| vanillic acid (VA) | $[Fe(NO_3)_3]$:0.5 mmol $L^{-1}$,<br>$NaNO_3$:0.05 mol $L^{-1}$,<br>T:20°C,<br>pH:3,<br>Vs:130 mL,<br>I:20 mA | $[VA]_0$:6.5, 10, 20, 30 mmol $L^{-1}$, | Protocatechuic acid (PCA) production increased with the increase of VA concentration, the energy consumption decreases by increasing the initial VA concentration. | Rekik et al. 2017 |
| Malachite Green (MG) | CD:10 mA/$cm^2$,<br>$C_{H2O2}$:50 mg/L,<br>pH:3,<br>t:30 min | MG concentration:200, 600, 1000, 1500, 2000, 3000 mg/L | The degradation rate decreased with the increase of concentration. | Teymori et al. 2019 |

**Table 2** (continued)

| Organics/Pollutants | Reaction conditions | Reactant concentration | Results | References |
|---|---|---|---|---|
| Amoxicillin (AMX) | I:600 mA,<br>[$Fe^{2+}$]:2 mmol $L^{-1}$,<br>[$Na_2SO_4$]:50 mmol $L^{-1}$,<br>pH:3,<br>T:25°C,<br>ω:360 r $min^{-1}$ | [AMX]:0.082 mmol $L^{-1}$, 0.164 mmol $L^{-1}$ | The degradation rate and mineralization rate decreased with the increase of initial concentration. | Kadji et al. 2020 |
| phenol | $Fe^{2+}$:4 mg/L,<br>current density:12 mA/$cm^2$,<br>pH:3,<br>$H_2O_2$ concentration:500 mg/L,<br>reaction time:60 min | initial phenol concentration:100 mg/L-400 mg/L | With the increase of concentration, the reaction kinetic constant ($k_{ap}$) decreased from 0.04 to 0.0286 $min^{-1}$. | Babuponnusami and Muthukumar 2012 |
| Sulfamethazine (SMT) | [$Fe^{2+}$]:0.1 mM,<br>I:300 mA,<br>pH:3,<br>T:18°C,<br>[$Na_2SO_4$]:50 mM,<br>V:1 L,<br>t:60 min | $[SMT]_0$:0.09 mM, 0.20 mM, 0.36 mM, 0.71 mM | The degradation efficiency of SMT was inversely proportional to the initial concentration. As the initial concentration increased from 0.09 to 0.71 mM, the degradation efficiency of SMT after 60 min reaction time decreased from 100 to 75.8%. | Mansour et al. 2011 |
| poly(vinylpyrrolidone) (PVP) | j:33.3 mA $cm^{-2}$,<br>time:3600 s,<br>$Na_2SO_4$:50 mM,<br>$FeSO_4$:1 Mm,<br>pH:2.8 | [PVP](wt %):0.25, 0.50 | With the increase of the specific gravity of PVP and the mineralization efficiency, the ratio of extracting 1 H atom per monomer unit and 1 H atom per polymer chain decreased. | Galia et al. 2016 |

the increase of electrolysis time, the content of target pollutants gradually decreased, which made the collision chance between ·OH and target pollutants decreased, and eventually led to a slow increase in the removal rate; on the other hand, since the target pollutants produce by-product intermediates during the degradation process, which are difficult to be mineralized, it takes a longer time for their degradation, which leads to an increase in the energy consumption of the process, and thus an increase in the operating cost.

In the electro-Fenton reaction, the mineralization current efficiency (MCE) usually exhibits a rapid increase at some time after the onset of degradation, after reaching a maximum value, followed by a gradual decrease and slow leveling off with time (Guelfi et al. 2016; Flores et al. 2016b). There are two main reasons for this phenomenon: (i) a decrease in organic matter content as degradation proceeds; and (ii) the formation of more difficult-to-degrade products (Sirés et al. 2014; Panizza and Cerisola 2009).

$$\mathrm{MCE} = \frac{n\,F\,V\;\;\Delta(TOC)_{exp}}{4.32\times10^{7}\;m\,I\,t}\times100\% \tag{38}$$

where $F$ is Faraday's constant (96487 C $\mathrm{mol}^{-1}$), V is the volume of solution (L), $\Delta(TOC)_{exp}$ is the experimental TOC reduction (mg $\mathrm{L}^{-1}$), $4.32\times10^{7}$ is the conversion factor (3600 s $\mathrm{h}^{-1}$ × 12000 mg C $\mathrm{mol}^{-2}$), $m$ is the number of carbon atoms contained in the target pollutant or organic matter, and $n$ is the number of electrons from the residual reaction during the mineralization process (which needs to be calculated from the reaction equation).

It is easy to see from the equation that when the conditions of the reaction are certain (i.e., $n$, $F$, $V$, $m$, and $I$ are certain), the TOC reduction in the experiment increases in the pre-reaction as the time increases, which results in the rate of change of the numerator in the equation being greater than the rate of change of the denominator, which leads to an increase in the mineralization current efficiency. When the maximum value is passed, the rate of change of the denominator is greater than the rate of change of the numerator as time increases because the reduction of TOC tends to level off, so the mineralization current efficiency starts to decrease, and when it goes down to a certain value, the $\Delta(TOC)_{exp}/t$ ratio tends to a constant value, so the mineralization current efficiency eventually levels off.

In the absence of $Fe^{2+}$, the production of $H_2O_2$ may be produced faster for a short period of time, followed by a stabilization of the yield over a longer period of time. This phenomenon may be due to the increasing decomposition rate of $H_2O_2$, and when the $H_2O_2$ production is in equilibrium with the decomposition rate, the yield of $H_2O_2$ in solution tends to stabilize (Mounia and Djilali 2012). In the traditional electrochemical reaction system, the $H_2O_2$ content shows a tendency of increasing and then decreasing. The main reason may be that as the reaction proceeds, a large amount of $Fe(OH)_3$ precipitates in the solution, which hinders the transfer of electrolyte in the solution. In contrast, for the electro-Fenton reaction using a composite cathode, the content of $H_2O_2$ is continuously increasing with time (Li et al. 2019a).

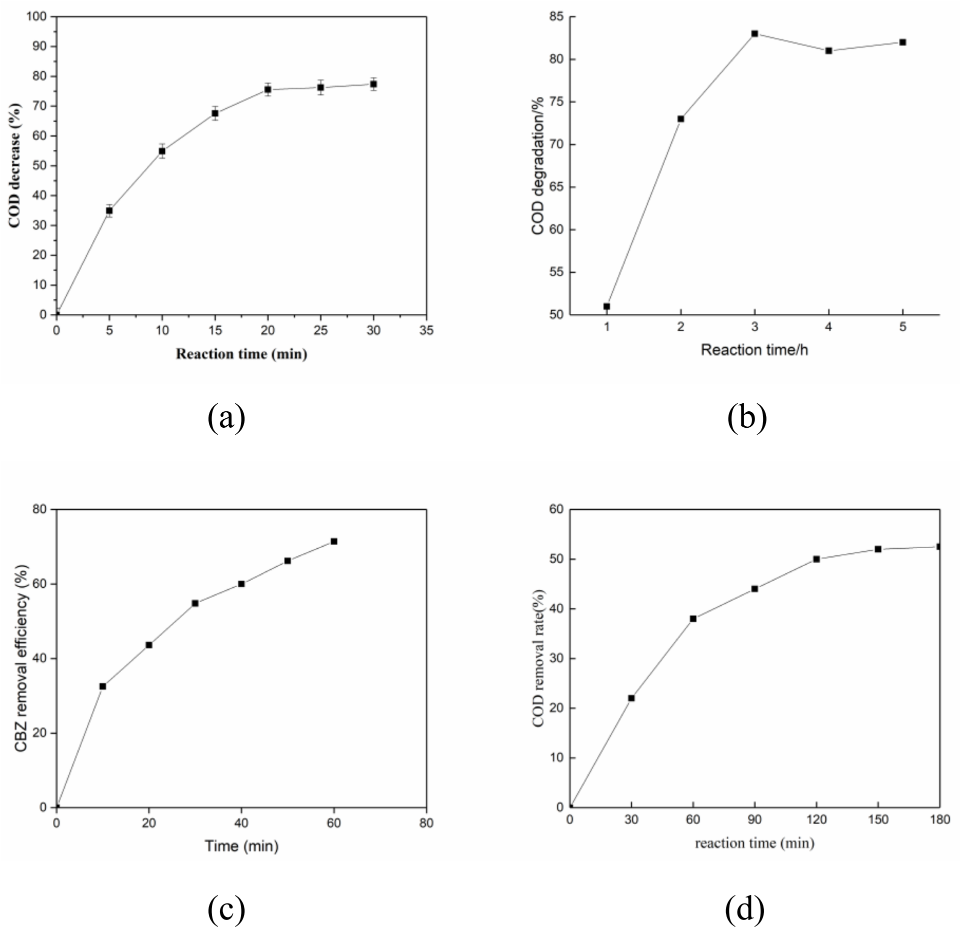
In summary, too short reaction time will make the process ineffective treatment; too long a reaction time will increase the operating cost of the process. As an important parameter, the reaction time is of great significance for the electro-Fenton process reaction. Figure 9 and Table 3 respectively show and list relevant literature studies on the effect of reaction time on the electro-Fenton process.

Table 3 Examples of effect of reaction time by Electro-Fenton reaction process

## Effect of voltage (potential)

Voltage (potential), as an influential parameter in the electro-Fenton reaction, bears a strong resemblance to current. Related studies also illustrate that varying the voltage (potential) can also have an effect on Fenton reagents and the degradation of organic pollutants.

First of all, in terms of the production of $H_2O_2$, researchers such as Li et al. (2019b), Diouf et al. (2018) and Zheng et al. (2021) have investigated the effect of different voltages (potentials) on the production of $H_2O_2$. Their experimental studies all show the same trend, i.e., the maximum yield of $H_2O_2$ is reached at a certain potential or voltage, while the yield of $H_2O_2$ decreases beyond that potential or voltage. The applied voltage will directly affect the production of $H_2O_2$ (Haider et al. 2019) , on the one hand, according to Faraday's law, the current density increases with the increase of voltage, and this leads to an increase in the production rate of $H_2O_2$. On the other hand, when the voltage is too high, the cathode produces some side reactions, such as hydrogen precipitation reaction, $H_2O_2$ decomposition, or $O_2$ undergoes four-electron reduction to $H_2O$ (Li et al. 2018; Xu et al. 2016; Özcan et al. 2008; Kuang et al. 2008), which prevents the generation of $H_2O_2$, thus resulting in a decrease in the yield of $H_2O_2$. Secondly, in terms of $Fe^{2+}/Fe^{3+}$, the experiments of Kim et al. (2018) showed that the concentration of $Fe^{2+}$ increased with increasing potential, which indicated that the rate of conversion of $Fe^{3+}$ to $Fe^{2+}$ was accelerated. Li et al. (2019a, b, c) obtained the same experimental phenomenon, but they additionally concluded that the increasing voltage resulted in a decrease in the concentration of the total ferric ions in the solution; they concluded that the localized alkalinization of the cathode affected the pH near the cathode with the increase of the voltage (Petrucci et al. 2016), which led to the sedimentation of iron ions and decay of the rate of reduction of iron ions, resulting in a gradual decrease in the concentration of total iron ions.



(a) (b)

(c) (d)

**Fig. 9** Experimental research on the influence of reaction time (presented from (**a**) Zhang et al. 2021; (**b**) Wang et al. 2020a; (**c**) Wang et al. 2017; (**d**) Li et al. 2019c)

Finally, voltage can also have an effect on the degradation efficiency of organic pollutants. Malakootian and Moridi (2017) investigated the effect of applied voltage on the degradation of Acid Red 18 dye. They found a significant increase in the dye removal efficiency by increasing the voltage from 10 V to 30 V and a decrease in the dye removal efficiency when the voltage was further increased to 40 V. George et al. (2013) studied the effect of applied voltage on the oxidation of Salicylic Acid and also came up with the same experimental pattern. Since voltage affects Fenton's reagent, which is a key factor in the degradation of organic pollutants, changing the voltage also directly affects the degradation efficiency of the pollutant. Ren et al. (2016) also found in the degradation effect of tartrazine that the degradation efficiency of the pollutant is close to 100% when the applied voltage is of a certain value and that the degradation efficiency of the pollutant reached a steady state when the applied voltage was higher than this voltage value. Cruz del Álamo et al. (2020) also showed that an increase in voltage led to a decrease in the efficiency of the mineralization current, which was also attributed to the generation of parasitic reactions. However, Lim et al. (2017) in the electro-Fenton treatment of palm oil mill wastewater found that the trend of COD removal efficiency was opposite to the trend of increasing applied voltage, with the highest COD removal efficiency being reached at the lowest voltage. They analyzed that the decrease in COD removal efficiency could be due to the oxidation of $Fe^{2+}$ to $Fe^{3+}$ at an oxidation potential of 0.771 V and the decomposition of $H_2O_2$ to $H_2O$ in an acidic solution at a reduction potential of 1.77 V, which would lead to a decrease in the concentration of the components that make up the Fenton reagent, which would lead to a decrease in the concentration of COD.

In addition, Li et al. (2011) also removed phenol wastewater by using a three-dimensional electro-Fenton system, and they found that when the voltage was elevated, the degradation of organic pollutants was accelerated, the level of polarization of the particle electrodes increased, and the number of working electrodes increased. They analyzed that the increase in voltage makes the main potential difference between the electrode and the conductive particles and electrolyte increase improves the impact of the electrochemical oxidation reaction, accelerates the direct and indirect oxidation rate, and makes the system more effective in treatment. Figures 10 and Table 4 respectively show and list relevant

**Table 3** Examples of effect of reaction time by Electro-Fenton reaction process

| organics/pollutants | reaction condition | reaction time | Results | References |
|---|---|---|---|---|
| N,N Methylenebismorpholine | potential:-500 mV/SCE(1.4 mA/cm$^2$),<br>$Na_2SO_4$:0.5 M,<br>pH:3 | 5 h | A rapid increase $H_2O_2$ production for times less than approximately 2h, then the production of hydrogen peroxide stabilizes, after two hours of electrolysis, production and decomposition occur simultaneously. The decomposition became higher, and the output increased slightly. | Mounia and Djilali 2012 |
| sulfamethazine (SMT) | pH:3,<br>T:18°C,<br>I:200 mA<br>$Na_2SO_4$:0.05 M,<br>$FeSO_4{\cdot}7H_2O$:0.1 M<br>SMT:0.36 M(100 mg L$^{-1}$) | 1 h | After 1 h, SMT degraded rapidly until it was eliminated, but the level of mineralization and oxidation was low. | Mansour et al. 2014 |
| sulfamethazine (SMT) | $[Fe^{2+}]_0$=0.5 M,<br>$Na_2SO_4$=50 mM,<br>pH=3,<br>T=18°C,<br>I=500 mA,<br>V=1 L,<br>SMT=0.2 mM | 1 h | SMT was eliminated by 100% after 30 min of electro Fenton pretreatment, but the salinity was still limited, which was 2.1 and 18.1% after 30 minutes and 60 minutes of electrolysis, respectively. | Mansour et al. 2015 |
| Reactive Yellow 160 azo dye | pH:3.0,<br>T=25°C,<br>$Na_2SO_4$:0.05 mol dm$^{-3}$,<br>I:100 mA cm$^{-3}$,<br>RY160:0.167 mmol dm$^{-3}$ | 6 h | The mineralization current efficiency decreased rapidly with time, then decreased slowly and tended to be stable. | Bedolla-Guzman et al. 2016 |
| Landfill Leachate's Organic | electrode distance:5 cm,<br>initial pH:3.5,<br>electrode material: Fe-Fe,<br>$H_2O_2$ concentration:15 mg/L | 3 h | The maximum removal efficiency of COD (85%), phosphate (78.6%), $NH_3$-N(30.9%), and turbidity (56.2%) occurred in the first 30 min of reaction time, and later it gradually slowed down. | Aval et al. 2017 |
| carbamazepine | CBZ:50 mg L$^{-1}$,<br>FeSO4:4.56 g L$^{-1}$,<br>pH:3.0,<br>current density:5.0 mA cm$^{-2}$,<br>gas diffusion cathode (GDC) with the $Na_2SO_4$ to carbon black ratio of 150% (w/w) | 60 min | The degradation efficiency increased with time, reached 71% of the maximum degradation efficiency of CBZ at 60 min. | Wang et al. 2017 |
| -COOH and succinimide groups | $Na_2SO_4$:0.05 mol dm$^{-3}$,<br>pH:2.8,<br>T:20°C,<br>current density:33.3 mA cm$^{-3}$,<br>$FeSO_4$:1.0 mmol dm$^{-3}$ | 180 min | the $H_2O_2$ concentration showed a continuous increase with time, reaching 27.0 mmol dm$^{-3}$ at 180 min. | Lanzalaco et al. 2017 |

**Table 3** (continued)

| organics/pollutants | reaction condition | reaction time | Results | References |
|---|---|---|---|---|
| insecticide propoxur | $Na_2SO_4$:0.05 M,<br>pH:3.0,<br>T:25°C,<br>current density:100 mA $cm^{-3}$,<br>$Fe^{2+}$:0.50 mM | 600 min | At the beginning of electrolysis, the maximum current efficiency was reached, then it decreased rapidly, and then became stable. | Guelfi et al. 2016 |
| trans-cinnamic acid | trans-cinnamic acid:0.926 mM(100 mL),<br>$Na_2SO_4$:0.05 M,<br>pH:3.0,<br>T:25°C,<br>current density:33.3 mA $cm^{-3}$ | 360 min | The MCE value rose from an initial 14 to 28 % at 120 min, further decaying to 13 %. | Flores et al. 2016b |
| 4-hydroxyphenylacetic acid | $Na_2SO_4$:0.05 M,<br>pH:3.0,<br>T:35°C,<br>current density:16.7 mA $cm^{-2}$,<br>$Fe^{2+}$:0.5 mM | 360 min | With the increase of time, TOC gradually decreased and finally decreased by 86%. | Flores et al. 2017 |
| acetaminophen | ACE:1 mM(500 mL),<br>pH:3.0,<br>$Na_2SO_4$:50 mM,<br>$FeSO_4{\cdot}7H_2O$:0.2 mM,<br>I:500 mA | 480 min | The toxicity of mixtures decreased gradually as a function of treatment time, 18.7%; 18.1% and 0%, respectively, from 210 to 480 min. | Le et al. 2017 |
| tebuthiuron | $Fe^{2+}$:0.50 mM,<br>pH:3.0,<br>$Na_2SO_4$:0.05 M,<br>T:35°C,<br>TOC:20 mg $L^{-1}$,<br>current density:50 mA $cm^{-2}$ | 360 min | The content of tebuthiuron decreased with time, and the degradation was completed within 300 min. | Gozzi et al. 2017 |
| lignin wastewater | Fe&$Fe_2O_3$/ACF,<br>Voltage:14 V,<br>I:0.7 A,<br>pH:2.0 | 30 min | The $H_2O_2$ continued to grow with the increase of reaction time under different operating conditions. | Li et al. 2019a |
| Aluminum Milling Wastewater | $Fe^{2+}$:0.5 mmol/L,<br>$Na_2SO_4$:10 g/L,<br>plate spacing:5 cm,<br>aeration volume:125 L/h,<br>milling wastewater:4 L | 180 min | The removal rate of COD increased with the increase of time, but its rate of increase decreased gradually after 120 min. | Li et al. 2019c |
| metformin | pH:3,<br>MET concentration:30 mg $L^{-1}$,<br>current density:5 mA $cm^{-2}$,<br>$H_2O_2$ dosage:200 μL $L^{-1}$ | 12 min | The degradation rate increased with time, reached 80% at 12 min. | Dolatabadi and Ahmadzadeh 2019 |
| Yellow 2G dye | pH:6.6,<br>current density:12.5 mA $cm^{-2}$,<br>NaCl:1 g $L^{-1}$, | 25 min | The color removal rate increases with time and reaches the maximum color removal (≈ 90%) after 25 min. | Benhadji and Ahmed 2020 |

**Table 3** (continued)

| organics/pollutants | reaction condition | reaction time | Results | References |
|---|---|---|---|---|
| Biotreated Coking Wastewater | voltage:10 V,<br>pH:3,<br>plate spacing:1 cm,<br>reaction capacity:400 mL | 5 h | The COD removal rate does not change significantly when the reaction time is extended from 3 to 5 h, demonstrating a basically stable value of approximately 83%. | Wang et al. 2020a |
| Quinoline | pH:3.0,<br>$H_2O_2$ dosage:71 mmol/L,<br>current density:30.5 mA/cm$^2$,<br>voltage:26.5 V,<br>electrical conductivity:15800 μs/cm | 30 min | The rapid COD reduction during the first 20 min was attributed to oxidation of 8eshes8ed, the degradation rate increased slowly after 20 min. | Zhang et al. 2021 |
| drinking water treatment sludge (DWTS) | j:14 mA cm$^{-2}$,<br>t:60 min,<br>$m_{DWTS}$:2.0 g,<br>pH:4.0 | 180 min | Decolorization and COD degradation efficiency increased with increasing time, instantaneous current efficiency (ICE) | Nguyen et al. 2021 |

literature studies on the influence of voltage (potential) on the electro-Fenton process.

Table 4 Examples of effect of voltage (potential) by Electro-Fenton reaction process

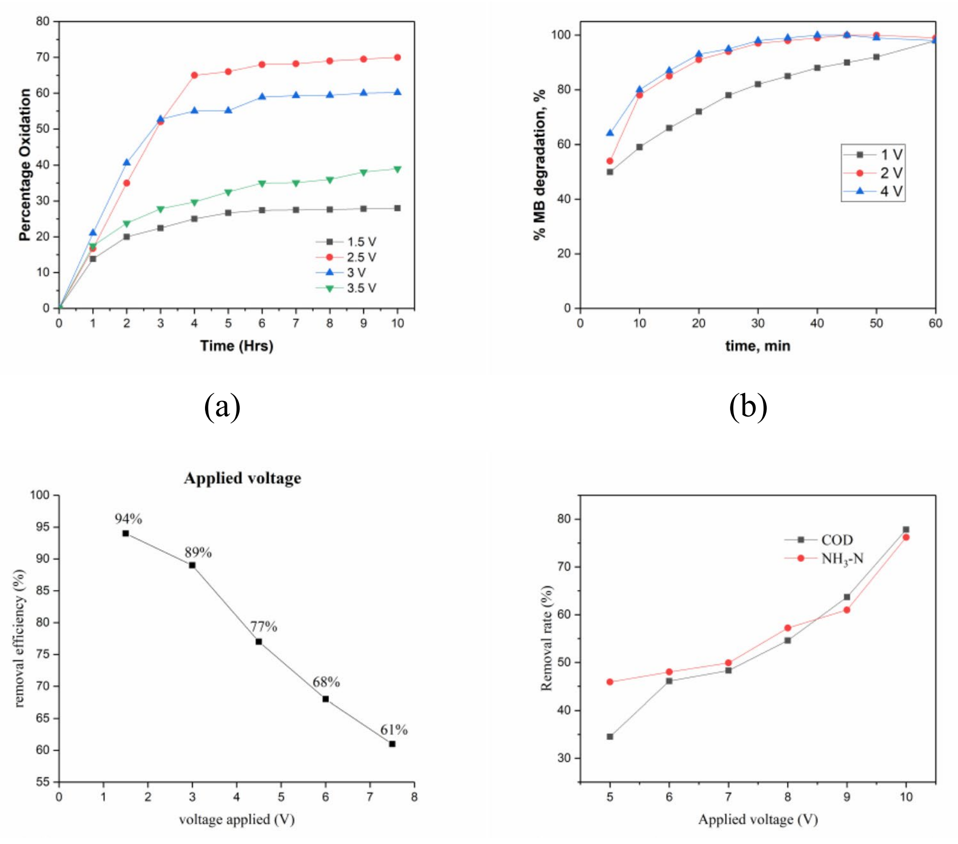
## Combination technology

### Combined with ultrasound technology

Studies have shown that ultrasound technology has demonstrated excellent capabilities in numerous applications, such as in chemical processing (Kiss et al. 2018), chemical synthesis (Sancheti and Gogate 2017), cleaning technology (Tiwari 2015), water treatment (Goncharuk et al. 2008) and others. The application of ultrasound at frequencies higher than 20 kHz leads to the growth of cavitation bubbles that become stable after a high number of cycles; each bubble can be seen as a hot spot and collapses, thus generating energy to increase the temperature (up to 5000 K) and the pressure (up to 500 atm), while the bubbles cool down at a rate of up to 109 K/s (Nazari et al. 2018).

The ultrasound process in the degradation of pollutants in the mechanism embodied in the physical decomposition of the water acoustic cavitation generated cavities or micro bubbles (micro jets and shock waves) and ultrasound through the propagation of the liquid medium thereby causing this physical effect (Hasani et al. 2020; Wu et al. 2013). In the event of surface instability, the cavity collapse can be very asymmetric and generate high velocity liquid jets (microjets), thus increasing the mass transfer rate (Liang et al. 2007; Li et al. 2013a; Pokhrel et al. 2016). Ultrasound decomposition then occurs through three reaction zones, namely the gas phase region inside the bubble (pyrolysis reactions), the interfacial region (reactions occurring in the pressure/temperature gradient in the aqueous phase) and the native solution (Riesz and Kondo 1992). Although ultrasound technology has been shown to be effective in treating pollutants, there are many parameters related to cavitation and bubble breakup that affect the process of performing ultrasound, such as pressure amplitude, acoustic frequency, type of signal, and other factors such as liquid temperature, which affect the effectiveness of ultrasound in treating pollutants in the water column (Wood et al. 2017).

Ultrasound-electro-Fenton technology is a hybrid oxidation technology that combines ultrasound and electro-Fenton technology. In a system that combines electro-Fenton and ultrasound, the physical degradation of pollutants still occurs through the cavities or microbubbles generated by acoustic cavitation in the water, which are mixed and cleaned at high speeds on the surface of the electrodes through the solvation inhibition layer, leading to the enhanced transfer between the electrodes and the solution (Babuponnusami



(a) (b)

(c) (d)

**Fig. 10** Experimental research on the influence of voltage (potential) (presented from (**a**) George et al. 2013; (**b**) Do et al. 2017; (**c**) Lim et al. 2017; (**d**) Wang et al. 2020b)

and Muthukumar 2012). The chemical effect, on the other hand, is due to the dramatic collapse of the microbubbles, which or cavities concentrate the ultrasound energy into the microreactor and reach the desired conditions in a short time (<1ns), where the microbubbles produce oxidized substances through homolytic cleavage of molecules (Trabelsi et al. 1996), and these oxidizing radicals enable pyrolysis of the organic matter and an increase in the rate of the reaction (Cai et al. 2014; Cai et al. 2016). In this case, the cavities are involved in the generation of hydroxyl radicals and free hydrogen, while the hydrogen radicals react with oxygen to generate hydroperoxide radicals (Li et al. 2013b). The relevant reaction equations are as follows (Equation (39)(40)):

$$H_2O + ))) \longrightarrow \cdot HO + H\cdot \tag{39}$$

$$H\cdot + O_2 \longrightarrow HO_2\cdot \tag{40}$$

In summary, ultrasound-electro-Fenton technology is a combination of physical and chemical mechanisms for effective pollutant degradation. This combination of technologies improves the cleanliness of the electrode surface, reduces process time, lowers energy consumption and improves environmental suitability and compatibility, and ultimately improves the pollutant removal efficiency through the resulting chemical and physical reactions (Hasani et al. 2020; Ranjit et al. 2008). Studies have also shown that ultrasound-electro-Fenton technology is more effective in the treatment of contaminants than electro-Fenton treatment alone. Hasani et al. (2020) utilized ultrasound-electro-Fenton for the removal of cefixime antibiotics from aqueous solutions and found that the maximum removal efficiency of the process under optimal conditions could reach 97.5%. Whereas, also under optimal conditions, the removal efficiency of the electro-Fenton process and ultrasound only reached 81.7% and 9%, respectively; Mahmoudi et al. (2021) compared the removal efficiencies of the ultrasound-electro-Fenton and electro-Fenton processes for Acid Black 172 (AB172) and Dispersed Blue 56 (DB56), and found that the maximal removal efficiency of the electro-Fenton was 82-88%, whereas the ultrasound-electro-Fenton could reach 89.5-91%; Babuponnusami and Muthukumar (2012) compared the effect of electro-Fenton and ultrasound-electro-Fenton on the degradation performance of phenol, and the results showed that the ultrasound-electro-Fenton method could achieve complete phenol removal in a shorter period of time compared to the electro-Fenton process.

**Table 4** Examples of effect of voltage (potential) by Electro-Fenton reaction process

| organics/pollutants | reaction conditions | voltage conditions | results | references |
|---|---|---|---|---|
| Landfill Leachate's Organic | time:30 min,<br>electrodes distance:5 cm,<br>initial pH:3.5,<br>$H_2O_2$ concentration:15 mg/L | voltage:5, 10, 15, 20, 30, 40 V | By increasing the voltage from 10 V to 30 V, the removal efficiencies of COD, $NH_3$-N, phosphate, and turbidity were obtained as 85.3%, 30.9%, 78.2%, and 56.2%, respectively. By increasing the voltage from 30 V to 40 V, significant changes were not observed in the removal of the organic material. | Aval et al. 2017 |
| methylene blue | pH:3,<br>$Na_2SO_4$:10 mmol $L^{-1}$,<br>time:20 min,<br>$Fe^{3+}$:0.2 mmol $L^{-1}$,<br>methylene blue =20 mg $L^{-1}$ | voltage condition:3, 5, 7, 9 V | ①As the voltage increased, the yield of $H_2O_2$ increased and then reached a maximum when the voltage was 7 V.<br>②When 9eshes9ed9ed for 20 min at voltages of 3, 5, 7 and 9 V, the total iron ions decreased by 0.91%, 8.55%, 12.43%, and 19.33%, respectively.<br>③After 20 min of reaction, the removal rates of methylene blue at voltages 3, 5, 7 and 9 V were 86.55%, 96.16%, 97.80% and 90.33%, respectively. | Li et al. 2019b |
| Aluminum Milling Wastewater | pH:3,<br>$Fe^{2+}$:0.5 mmol $L^{-1}$,<br>electrodes distance:5 cm,<br>$Na_2SO_4$:10 g/L,<br>aeration:125 L/h,<br>reaction time:120 min, | voltage:20, 25, 30, 35, 40, 45 V | When the voltage was 40 V, the maximum removal rate of COD is achieved, with the value greater than 50%. | Li et al. 2019c |
| Biotreated Coking Wastewater | reaction time:5 h,<br>plate spacing:1 cm,<br>V:400 mL,<br>Initial pH:3 | voltage:4-10 V | When the applied voltage is 10 V, the treatment effect of the cathode EF oxidation system on the BTCW is the best, and the removal rate reaches 84.79%. | Wang et al. 2020a |
| crystal violet | V:250 mL,<br>$Na_2SO_4$:3 g $L^{-1}$,<br>time:90 min, | voltage:-200— -1000 mV | The amount of generated $H_2O_2$ increased with the cathodic potential until an optimal voltage specific to each electrode. These optimum voltages were -600 mV for Gr-Bat electrode and -700 mV for VC electrode. | Diouf et al. 2018 |
| Diazinon (DZ) | $H_2O_2$ concentration: 30 mg/L,<br>pH:3,<br>electrolysis time:50 min,<br>$FeSO_4$:0.24 gr/L,<br>NaCl:0.1 g/L,<br>DZ concentration:50 mg/L | voltage:5-15 V | Development of the percentage removal by increasing the applied voltage is detectable, so that it 10eshes10ed from 47.76 to 69.55% by changing the voltage from 5 to 15 V. | Dargahi et al. 2021b |

**Table 4** (continued)

| organics/pollutants | reaction conditions | voltage conditions | results | references |
|---|---|---|---|---|
| quinoline | pH:3.0,<br>conductivity:15,800 μs/cm,<br>j:30.5 mA/cm$^2$,<br>$H_2O_2$ concentration:71 mmol/L,<br>NaCl:3 g/L | voltage:12.5-37.5 V | The COD decrease gradually rose from 22.97% to 66.64% as the applied voltage increased from 12.9 V to 26.5 V. The COD decrease was greatest with the voltage 26.5 V | Zhang et al. 2021 |
| real dyeing wastewater | pH:4.0,<br>drinking water treatment sludge (DWTS):1.0 g,<br>electrodes distance:3.0 cm,<br>reaction time:60 min,<br>T:25°C, | voltage:5-25 V | The current density increased with the increase of applied voltage, the best values of decolorization and COD degradation efficiencies at 20.0 V were achieved at 96.8% and 86.8%, respectively. | Nguyen et al. 2021 |
| Phenol Wastewater | pH:3,<br>$Fe^{2+}$:0.6 mmol/L,<br>phenol concentration:300 mg/L | voltage:6, 9, 12, 15, 18, 21 V | When the electrolysis voltage was less than 15V, the phenol removal efficiency increased significantly, while the electrolysis voltage was more than 15 V, removal efficiency began decreased. | Li et al. 2011 |
| Organic Compounds in Actual Wastewater | pH:3<br>$Fe^{3+}$ concentration:20 mg/L,<br>reaction time:2 h | potential:0.5, 0.7, 1, 1.2, 1.4 V | The content of $Fe^{2+}$ in the solution increased with increasing potential. | Kim et al. 2018 |
| methylene blue (MB) | pH:5,<br>catalyst loading:1.8 g/L,<br>air flow rate:20 mL/min | voltage:1, 2, 3 V | The MB degradation rate and TOC removal rate increase with increasing voltage, but there is no significant improvement in the later stage. The mineralization current efficiency decreases with increasing voltage. | Cruz del Álamo et al. 2020 |
| microalgae | pH:7,<br>$Na_2SO_4$:0.05 M,<br>reaction time:60 min | voltage:-0.8, -1.0, -1.2 V | Obtain the maximum production of $H_2O_2$ when the voltage was -1.0 V. | Zheng et al. 2021 |
| chlorbromuron | pH:2,<br>chlorbromuron concentration:1.19×10$^{-4}$ M,<br>$FeSO_4·7H_2O$:1 mM,<br>$Na_2SO_4$:0.5 M | voltage:1.0, 2.0, 3.0, 4.0 V | At the voltage of 3.0 V, the maximum TOC removal rate was achieved. | Martínez and Bahena 2009 |
| tartrazine | pH:3,<br>flow rate:40 mL/min,<br>tartrazine:100 mg/L,<br>$Fe^{2+}$:0.4 mmol/L | voltage:2.5, 3, 3.5, 4.5, 5 V | When the voltage is higher than 2.5 V, the degradation rate of tartrazine can reach 100%, and the TOC degradation rate increases with the increase of voltage. | Ren et al. 2016 |

**Table 4** (continued)

| organics/pollutants | reaction conditions | voltage conditions | results | references |
|---|---|---|---|---|
| Palm oil mill effluent (POME) | $H_2O_2$:0.05 M(75 mL),<br>$Na_2SO_4$:0.25 M(75 mL),<br>time:4 h,<br>$FeSO_4$:0.005 M(50 mL),<br>POME:50 mL | voltage:1.5, 4.5, 6.0, 7.5 V | When the voltage was 1.5 V, the COD removal rate was 94%, and thereafter, as the voltage increased, the COD removal rate decreased. | Lim et al. 2017 |
| high-ammonia-nitrogen landfill leachate nanofiltration concentrate | pH:3,<br>electrode distance:1 cm,<br>reaction time:300 min | voltage:5, 6, 7, 8, 9, 10 V | The removal rates of COD and $NH_3$-N increased with the increase of voltage. | Wang et al. 2020b |
| Pentachlorophenol (PCP) | pH:2.5,<br>$Na_2SO_4$:0.14 mol/L,<br>time:120 min,<br>PCP solution:16 mg/L(200 mL),<br>electrolyte: $Na_2SO_4$ | voltage:3, 5, 7.5 V | At the voltage of 7.5 V for 30-60 min of reaction, the maximum reduction in PCP was achieved. | Yuan and Lu 2005 |
| Acid Red 18 | pH:3,<br>dye:100 mg/L,<br>$H_2O_2$:10 ml/L,<br>NaCl:100 mg/L,<br>electrodes distance:2 cm | voltage:10, 20, 30, 40 V | The dye degradation rate increased from 10 V to 30 V, and further increased to 40 V, resulting in a decrease in the dye degradation rate. | Malakootian and Moridi 2017 |
| methylene blue (MB) | catalyst:0.12 g/L,<br>pH:3,<br>MB:10 mg/L,<br>$Na_2SO_4$:0.05 M | voltage:1, 2, 4 V | As the voltage increased, the MB removal efficiency increased. | Do et al. 2017 |
| Salicylic Acid (SA) | pH:2.5,<br>flow rate:10 mL/min,<br>electrodes distance:3 cm,<br>$Fe^{2+}$ concentration:5 mg/L | voltage:1.5, 2.5, 3, 3.5 V | The maximum oxidation rate is obtained at the voltage of 2.5 V. | George et al. 2013 |
| Refinery Wastewater | pH:3,<br>NaCl:3 g,<br>reaction time:60 min,<br>Fe particles:4 g,<br>gas flow rate:1.5 L/min,<br>volume:300 mL | voltage:8, 10, 12, 15 V | When the voltage exceeded 12 V, the reduction in COD decreased. | Yan et al. 2016 |

With the in-depth study of ultrasound-electro-Fenton, the reaction system using three-dimensional electro-Fenton in ultrasound-electro-Fenton has begun to emerge, but there are still very few techniques on ultrasound-electro-Fenton that include three-dimensional electro-Fenton. Currently, Dargahi et al. (2021a) evaluated the degradation of the herbicide 2,4-dichlorophenoxyacetic acid (2,4-D) by three-dimensional ultrasound-electro-Fenton using PAC/$Fe_3O_4$ as a particle electrode, and explored the relevant influencing factor variables in the degradation process. The experimental results showed that the removal of 2,4-D, COD and TOC was 96.2%, 92.31% and 86.5%, respectively, under the optimal reaction conditions. In addition, a significant reduction in the biotoxicity of the outlet effluent from the 3D/SEF process was detected. Finally, the 2,4-D herbicide was completely degraded by hydroxyl radicals generated by the electrocatalytic process and converted to $CO_2$ and $H_2O$ products. In summary, as an enhanced technology of electro-Fenton, ultrasound-electro-Fenton showed better results in degrading pollutants compared to the single electro-Fenton system. Table 5 lists the relevant literature research on the combination of electro-Fenton process and ultrasound technology.

Table 5 Research on the combination of Electro-Fenton and ultrasonic technology

## Combined with anodic oxidation technology

As a type of electrochemical advanced oxidation processes (EAOP), anodic oxidation (AO) is probably the simplest and most effective alternative for the direct or indirect generation of reactive substances at the anode (Hu et al. 2020). In the anodic oxidation process, high current densities are used for the anodic oxidation of pollutants by applying high current densities to the anode and generating oxidizing physisorbed hydroxyl radicals, M(·OH), which are intermediates of water discharged to $O_2$ at the anode surface by reaction (Equation (41)) (Panizza and Cerisola 2009; Marselli et al. 2003; Martínez-Huitle and Ferro 2006; Brillas and Martínez-Huitle 2015):

$$M + H_2O \longrightarrow M(\cdot OH) + H^+ + e^- \tag{41}$$

In terms of materials used for anodizing, the use of inactive anodes such as $Ti_4O_7$ or boron doped diamond (BDD) instead of active anodes such as mixed metal oxides (MMOs) and dimensionally stabilized anodes (DSAs) allows for a better degradation of organic matter and improved mineralization of the solution (Ouarda et al. 2020). Many works have demonstrated that some inactive anodes, such as BDD, $SnO_2$ and $PbO_2$, are ideal anodes for mineralization of organic pollutants into $CO_2$ and aqueous end products (Sirés et al. 2014). And as an effective technology for treating organic pollutants, anodic oxidation can be realized in combination with physical, chemical or biological processes (Becerril-Estrada et al. 2020; Chu et al. 2012; Ouarda et al. 2018) to obtain higher removal efficiency of pollutants. The electro-Fenton process, as a chemical process, can be combined with anodic oxidation to enhance the removal efficiency of pollutants. Ouarda et al. (2020) compared the removal of TOC from acetaminophen synthetic solutions by EF, AO-$H_2O_2$ and EF-AO techniques, respectively. It was found that after 8 h of reaction, the removal of TOC from the solution using a single EF process was 76 ± 5%, the removal efficiency achieved by the AO-$H_2O_2$ method was 81.0 ± 3.4%, while the combined EF-AO technique achieved a removal efficiency of 87 ± 2.7%. They concluded that the ability of the EF-AO process to achieve higher TOC removal rates can be explained by the generation of hydroxyl radicals on the anode surface and in the native solution of the Fenton reaction. However, different experimental results were obtained by Vasconcelos et al. (2016), whom, when comparing the removal of the dye Reactive Black 5 (RB-5) through the use of the AO, EF, and AO-EF processes, respectively, found that the removal efficiency of the dye color and TOC was higher than that of the combined AO-EF process when using either the AO or EF process alone. Through their analysis, they concluded that RB-5 was oxidized by heterogeneous BDD (·OH) on the BDD surface and also by homogeneous ·OH radicals formed by the Fenton reaction that electrically generates hydrogen peroxide. However, it is also possible that the RB-5 dye is reduced at the cathode surface, ultimately leading to a decrease in color removal efficiency.

However, in general, anodic oxidation combined with electro-Fenton are efficient and significant in terms of degradation on organic pollutants, and repreparation of substances. For example, Olvera-Vargas et al. (2018) proposed an integrated dynamic cross flow electro-Fenton (DCF-EF) during which the anodic oxidation of acetaminophen was performed using a $Ti_4O_7$ anode. The experimental results showed that under optimal reaction conditions, complete degradation of the drug and 44% mineralization efficiency could be achieved, and that $Ti_4O_7$ rods as anode contributed significantly to the mineralization of acetaminophen. Labiadh et al. (2016) treated the concentrate obtained from sanitary waste leachate during reverse osmosis by such a combined process, where boron-doped diamond was used as an anode electrode and pretreated, and finally electro-Fenton treatment was carried out, and the experiments showed that this combined process could be effective for the treatment of organic loads and nitrogenous substances in the reverse osmosis concentrate. In this combined process, anodic oxidation dominates, and the anodic oxidation process removes nitrogen-containing compounds more effectively than electro-Fenton, and the combined process also enhances the biodegradability index of the effluent. Ou et al. (2019) treated

**Table 5** Research on the combination of Electro-Fenton and ultrasound technology

| Organics/Pollutants | Electrodes | Reaction conditions | Results | References |
|---|---|---|---|---|
| phenol | Cathode/Anode: Stainless steel | Initial phenol concentration: 200 mg/L,<br>$Fe^{2+}$: 4 mg/L,<br>current density:12 $mA/cm^2$,<br>$H_2O_2$ concentration: 500 mg/L,<br>pH:3,<br>ultrasonic frequency: 34kHz | Complete degradation of phenol in 40 minutes. | Babuponnusami and Muthukumar 2012 |
| 2,4-Dichlorophenoxyacetic acid herbicide | Cathode:SS316<br>Anode:SS316/β-$PbO_2$,<br>Particle electrode: (PAC)/$Fe_3O_4$ | 2,4-D concentration: 50 mβL,<br>pH:3,<br>$FeSO_4$: 0.08 mg/250 mL,<br>electrolysis time:60 min,<br>$H_2O_2$ concentration:0.2 mL/L,<br>current density:5 $mA/cm^2$,<br>catalyst dose:5 g/L,<br>$Na_2SO_4$ concentration: 0.3g/250 mL,<br>ultrasonic frequency: 37kHz | Removal efficiencies of the 2,4-D, COD and TOC were 96.2%, 92.31% and 86.5%, respectively. | Dargahi et al. 2021a |
| diazinon | Cathode:SS316<br>Anode:SS316/β-$PbO_2$, | Diazinon initial concentration: 45.6 mg/L,<br>voltage:13.8 V,<br>pH=5.3,<br>$H_2O_2$:30 mg/L,<br>$FeSO_4$: 0.24 gr/L,<br>NaCl:0.1 g/L,<br>ultrasonic frequency: 37kHz | the DZ degradation, COD, and TOC removal efficiencies of 92.2%, 73.2%, and 67.4%, respectively. | Dargahi et al. 2021b |
| Cefixime antibiotic | Cathode/Anode: Fe, | pH:3.07,<br>$H_2O_2$ concentration:0.85 ml/L,<br>Voltage:15 V,<br>Initial antibiotic concentration:10.4 mg/L,<br>electrolysis time:81.5 min,<br>ultrasonic frequency: 37kHz | Under the optimum conditions, the maximum efficiency of removing cefixime antibiotic from the aqueous solution by Sono-electro-Fenton was 97.5%. | Hasani et al. 2020 |
| Acid Black<br>172 and Disperse Blue 56 | Cathode/Anode: Fe | [dye]=200 mg/L,<br>pH=3,<br>I=2 $mA/cm^2$,<br>time=30 min for DB56 and 50 min AB172<br>$H_2O_2/C_0$ of 0.33for AB172 and 0.78 for DB56,<br>Ultrasonic power:11 W | The degradation efficiency of AB172 and DB56 was 89.5% and 91% respectively, and the COD removal efficiency was 72.4% and 81.2% respectively. | Mahmoudi et al. 2021 |
| 4-Chlorophenol | Cathode/Anode: Ti-based mixed metal oxide 14eshes (Ti/MMO,<br>$IrO_2/Ta_2O_5$ coating on titanium mesh type) | $Fe^{2+}$:80 mg $L^{-1}$,<br>Pd/$Al_2O_3$:10 mg $L^{-1}$,<br>pH:3,<br>$Na_2SO_4$:10 mM,<br>current density:16.94 mA $cm^{-2}$,<br>time:180 min<br>ultrasonic frequency: 20kHz | The maximum removal rate of 4-chlorophenol (4-CP) was>99.9%. | Nazari et al. 2018 |
| Reactive Black 5 (RB 5) | Cathode/Anode: cast iron plates | pH:3,<br>I:0.25 A,<br>$H_2O_2$:800 mg/L,<br>electrode distance:2.5 cm,<br>ultrasonic frequency: 35kHz,<br>time:15 min | The removal efficiency of color and COD could reach more than 90%, and the first-order kinetic constants were 0.9828 and 0.8148 respectively. | Şahinkaya 2013 |

**Table 5** (continued)

| Organics/Pollutants | Electrodes | Reaction conditions | Results | References |
|---|---|---|---|---|
| 2,4-dichlorophenoxyacetic acid | Cathode/Anode: nickel foil (0.125 mm thin) | 2,4-D concentration:1.2 mM,<br>$H_2O_2$:3.0 mM,<br>$Fe^{2+}$:3.0 mM,<br>pH:3,<br>current density: Not more than 100mA.<br>Ultrasonic frequency: 20kHz, | Almost 50% oxidation of 2,4-D solution was completed in 60 seconds. | Yasman et al. 2004 |
| 2,4-dichlorophenoxyacetic acid(2,4-D), 4,6-dinitro-o-cresol (DNOC) and azobenzene (AB) | Cathode: carbon-felt,<br>Anode: Pt mesh | 2,4-D:1 mM,<br>DNOC:0.5mM,<br>AB:0.025 mM,<br>$Na_2SO_4$:0.05 M,<br>$Fe^{3+}$:0.1 mM,<br>pH:3.0,<br>I:200 mA,<br>Ultrasonic power:20 W<br>ultrasonic frequency: 28kHz, | 2,4-D and DNOC were degraded within 90 minutes, and AB was degraded within 10 minutes. | Oturan et al. 2008 |

polyaniline (PANI) wastewater and re-preparation of PANI by this process. The experimental results showed that the COD removal in PANI wastewater could reach 89% under certain reaction conditions. Moreover, the yield of PANI-T can reach 92.1% using treated PANI wastewater, which indicates that it is efficient and environmentally friendly to treat PANI wastewater and re-prepare PANI using this process.

Currently, boron-doped diamond (BDD) has a higher oxidizing capacity than other common anodes in AO (Hamza et al. 2009; Ciríaco et al. 2009; Rodrigo et al. 2010; Cavalcanti et al. 2013). This is due to the fact that the BDD anode $O_2$ overpotential is larger and the interaction between the surface and ·OH is very low, thus favoring the reaction of BDD (·OH) with organics, which allows the BDD anode to achieve the best performance in destroying organics (Rocha et al. 2011), which makes this type of electrode the electrode of choice for anodic oxidation. Table 6 lists the relevant literature research on the combination of electro-Fenton process and anodic oxidation technology.

Table 6 Research on the combination of Electro-Fenton and anodic oxidation technology

## Combined with electrocoagulation technology

Electrocoagulation (EC) is one of the most widely used electrochemical methods in wastewater treatment (Khaled et al. 2015), a well-established wastewater treatment method that has been widely explored for treating a wide range of wastewater pollutants due to its ease of operation, versatility, environmental friendliness and low footprint (Das et al. 2022). In terms of the mechanism of treating pollutants in wastewater, electrocoagulation involves the production of coagulants by electrochemical oxidation in situ at a sacrificial anode. By releasing metal ions and further converting them to hydroxides, these hydroxides can neutralize charges or act as scavenging flocs with large surface areas to promote aggregation or precipitation into sludge and adsorption of dissolved pollutants (Ghernaout 2013; Ghernaout and Ghernaout 2012). During electrocoagulation, pollutants can be removed by adsorption, precipitation, electrostatic attraction, anodic oxidation, and cathodic reduction (Thiam et al. 2014); however, the process of removal that occurs can be affected by the type and concentration of the electrolyte medium in solution (Govindan et al. 2014a, 2015b, 2020a). As for the choice of anode materials, Fe and Al are the traditional anode soluble metals used in electrocoagulation process due to their low cost and high valence (Ilhan et al. 2008; Chafi et al. 2011).

When the anode electrode is Fe, Fe is oxidized to form $Fe^{2+}$, while at the same time $H_2O$ at the cathode undergoes reduction and produces $OH^-$ , and the combination of $Fe^{2+}$ and $OH^-$ produces insoluble $Fe(OH)_n$ (n=2,3). These $Fe(OH)_n$ will act as a coagulant to the suspended solids, producing dense flocs and precipitation. In addition, the production of $H_2$ at

**Table 6** Research on the combination of Electro-Fenton and anodic oxidation technology

| Organics/Pollutants | Electrodes | Reaction conditions | Results | References |
|---|---|---|---|---|
| Reactive Black 5 dye (RB-5) | Cathode: Reticulated Vitreous Carbon (RVC)<br>Anode: Boron-Doped Diamond (BDD) | initial RB-5 concentration: $2\times10^{-5}$ mol $dm^{-3}$,<br>$Na_2SO_4$:0.5 mol $dm^{-3}$,<br>liquid flow rate:100 $dm^3$ $h^{-1}$,<br>pH:3 (cathode chamber),<br>$Fe^{2+}$:$1.0\times10^{-4}$ mol $dm^{-3}$,<br>T:23°C,<br>reaction time:90 min,<br>j:41.1 mA $cm^{-2}$ | The process using AO–EF combined process resulted in reasonable TOC (82%) and colour removal (>98%) after 90 min of electrolysis, but the EC values (745 kWh $kg^{-1}$) were slightly higher. | Vasconcelos et al. 2016 |
| Acetaminophen (Paracetamol (PCTM)) | Cathode: carbonaceous electrocatalytic membrane<br>Anode: $Ti/Ti_4O_7$ | liquid flow rate:3 L $min^{-1}$,<br>PCTM:0.1 mM,<br>$O_2$ pressure:0.2 bar,<br>I:100 mA,<br>$Na_2SO_4$:50 mM,<br>pH:3,<br>T:18°C,<br>$Fe^{2+}$:0.2 mM,<br>V:1.5 L | Under the optimal conditions, the EF system achieved complete degradation of the drug and simultaneously achieved 44% mineralization, while using $Ti_4O_7$ anode could achieve up to 20% mineralization of p-benzoquinone (p-BQ), a by-product of paracetamol. | Olvera-Vargas et al. 2018 |
| acetaminophen (ACT) | AO:<br>Anode:$Ti_4O_7$<br>EF:<br>Cathode: carbon felt (CF),<br>Anode:Ti/Pt | $Fe^{2+}$:0.2 mM,<br>$Na_2SO_4$:50 mM,<br>air aeration:1 L $min^{-1}$,<br>ACT:10 mg $L^{-1}$,<br>V:250 mL,<br>pH:3,<br>time:8 h,<br>I:250 mA | After 8 h of reaction, the maximum TOC content removed by the EF/AO process was about 87%. | Ouarda et al. 2020 |
| concentrate from reverse osmosis of sanitary landfill leachate | AO:<br>Anode: BDD<br>EF:<br>Cathode: carbon felt<br>Anode: BDD | AO pretreatment condition:<br>pH:8.8,<br>I:4.9 A,<br>flow rate:500 L/h,<br>time:8 h<br>EF condition:<br>air aeration:1 L/min (10 min),<br>I:0.3 A,<br>pH:3,<br>time: 8 h,<br>$Fe^{3+}$:16 mg /L | After AO pretreatment, the removal rates of COD, DOC, $NH_4^+$, and total nitrogen were 48%, 42%, 50%, and 35%, respectively; subsequently, using the most favorable EF experimental conditions, the removal rates of COD, DOC, $NH_4^+$, and total nitrogen were 60%, 53%, 33%, and 22%, respectively. | Labiadh et al. 2016 |
| 2-Nitrophenol | AO: BDD thin film<br>EF:<br>Cathode: carbon felt (CF)<br>Anode: BDD thin film | air aeration: 1 L $min^{-1}$ (10 min),<br>pH:3,<br>$Na_2SO_4$:0.05 M,<br>I:300 mA,<br>V:250 mL,<br>2-NP: 367 mg $L^{-1}$ | In the AO-EF coupling process, when the processing time was 2 hours, the TOC removal rate was 67%. | Oturan et al. 2011 |

**Table 6** (continued)

| Organics/Pollutants | Electrodes | Reaction conditions | Results | References |
|---|---|---|---|---|
| perfluorooctanoic acid (PFOA) | Cathode:Fe/10MnC (EF)<br>Anode:BDD (AO) | T:25°C,<br>V:100 mL,<br>pH:3,<br>electrolyte: $Na_2SO_4$,<br>$N_2$ flow rate:100 mL $min^{-1}$,<br>$FeSO_4 \cdot 7H_2O$:0.0278 g,<br>reaction time:270 min | When the cathode was Fe/10MnC, 97% of PFOA and 93% of TOC were removed after 270 minof reaction. | Wang et al. 2019 |
| 2,4-dichlorophenoxyacetic acid (2,4-D) | Cathode:CB-GF (EF)<br>Anode: BDD films on conducting p-Si substrate | V:100 mL,<br>2,4-D:20 mg $L^{-1}$,<br>pH:5,<br>AO:<br>j:5 mA $cm^{-2}$ (15 mA),<br>$Na_2SO_4$:0.1 M,<br>EF:<br>$Fe^{2+}$:1 mM,<br>time:2 h | 2,4-D could be completely removed within 2 h, and the removal rate of TOC could reach 69%. | Cai et al. 2019 |
| polyaniline (PANI) wastewater | Cathode: graphite felt<br>Anode: $IrO_2$/Ti | V:360 mL,<br>PANI:0.3 M,<br>T:25°C,<br>electrodes distance:2 cm,<br>pH:3.0,<br>$O_2$ flow rate:160 mL min-1 (30 min),<br>$Fe^{2+}$:0.2 mM,<br>j:14 mA $cm^{-2}$,<br>time:360 min | The removal rate of COD in wastewater could reach 89%. | Ou et al. 2019 |
| 4-nitrophenol | Cathode 1: gas diffusion electrode (GDE),<br>Cathode 2: graphite, (EF)<br>Anode: Ti/$SnO_2$–$Sb_2O_5$–$IrO_2$ (AO) | pH:3.0,<br>$Na_2SO_4$:0.1 mol $L^{-1}$,<br>4-nitrophenol:200 mg $L^{-1}$,<br>stirring:500 rpm,<br>electrodes distance:15 mm,<br>air flow rate:25 mL $s^{-1}$,<br>$Fe^{2+}$:0.1 Mm,<br>time:600 min | After 600 min treatment, 74.5% of the original TOC was removed by the dual-cathode oxidation system. | Chu et al. 2012 |

the cathode leads to flotation of particles at the surface, and the production of insoluble particles of different densities and sizes during the re-precipitation process can be eliminated by filtration (Hosny 1996; Chen et al. 2002; Kyzas and Matis 2016). Two production mechanisms have been proposed for the production of iron hydroxide material (Mollah et al. 2001):

Mechanism 1

Anode.

$$4Fe \longrightarrow 4Fe^{2+} + 8e^{-} \tag{42}$$

$$4Fe^{2+} + 10H_2O + O_2 \longrightarrow 4Fe(OH)_3 + 8H^{+} \tag{43}$$

Cathode.

$$8H^{+} + 8e^{-} \longrightarrow 4H_2 \tag{44}$$

Overall.

$$4Fe + 10H_2O + O_2 \longrightarrow 4Fe(OH)_3 + 4H_2 \tag{45}$$

Mechanism 2

Anode.

$$Fe \longrightarrow Fe^{2+} + 2e^{-} \tag{46}$$

$$Fe^{2+} + 2OH^{-} + O_2 \longrightarrow 4Fe(OH)_2 \tag{47}$$

Cathode.

$$2H_2O + 2e^{-} \longrightarrow H_2 + 2OH^{-} \tag{48}$$

Overall.

$$Fe + 2H_2O \longrightarrow 4Fe(OH)_2 + H_2 \tag{49}$$

When the material is Al, Al is first oxidized in solution to produce $Al^{3+}$, which is subsequently converted to $Al(OH)_3$ and then polymerized to $Al_n(OH)_{3n}$ at a suitable pH value. The relevant reaction equation is (Chen 2004):

$$Al \longrightarrow Al^{3+} + 3e^{-} \tag{50}$$

$$Al^{3+} + 3OH^{-} \longrightarrow Al(OH)_3/Al^{3+} + 3H_2O \longrightarrow Al(OH)_3 + 3H^{+} \tag{51}$$

$$nAl(OH)_3 \longrightarrow Al_n(OH)_{3n} \tag{52}$$

Although electrocoagulation is very effective for the treatment of pollutants in wastewater, on the one hand, electrocoagulation constantly consumes the electrode plates as they pass through the electrolytic reaction, and therefore needs to be replaced frequently; secondly, oxidized films and precipitates are formed on the surface of the plates, which leads to a decrease in the efficiency of electrocoagulation; and thirdly, the colloids produced in the process of electrocoagulation are sometimes re-dissolved. In order to solve the above problems, the combination of electrocoagulation with another process is a promising method to improve the removal efficiency of water pollutants. For example, free radical-assisted electrocoagulation is a new combination that shows higher performance (Al-Qodah et al. 2018). While electro-Fenton, an advanced oxidation technology capable of generating reactive free radicals, is used in combination with the electrocoagulation process, usually with Fe electrodes as sacrificial anodes and $H_2O_2$ externally added to degrade the pollutants. When electrocoagulation occurs and $Fe^{2+}$ is electrochemically generated, $Fe^{2+}$ activates $H_2O_2$ and generates reactive free radicals that accelerate the degradation of the contaminants (Govindan et al. 2014b, 2015a). Figure 11 shows a diagram of the $H_2O_2$ assisted EC process and the mechanism of pollutant degradation by the Fe electrode.

The advantages of the electro-Fenton process for the removal of organic matter became apparent when it was combined with the electrocoagulation process. Zhao et al. (2012) tested the removal efficiency of plugboard wastewater under a single electrocoagulation process versus a combined electrocoagulation-electro-Fenton process, and it was found that, with a single electro-coagulation process, only nearly 30% of the COD could be removed, but with the addition of $H_2O_2$, the removal of COD in the wastewater was dramatically increased to 76%. This is due to the fact that the in situ generation of $Fe^{2+}$ can react with $H_2O_2$ to produce ·OH after the addition of $H_2O_2$ to the solution, resulting in an increase in COD removal. Anfruns-Estrada et al. (2017), on the other hand, compared the ability of two electrochemical techniques, electrocoagulation and electro-Fenton, to disinfect both primary and secondary effluents from municipal wastewater treatment plants. It was shown that the EF process at pH = 3.0 achieved substantial or even complete microbial inactivation within 30 min compared to single EC, but was inferior to the EC process in TOC removal, whereas in continuous combined EC/EF treatment, the bacterial membranes were weakened due to interactions with $Fe(OH)_n$ floc and oxidants, and subsequently further subjected to rapid oxidant inactivation, higher TOC removal than the single EC and EF processes, achieving more effective effluent treatment of the effluent.

Related studies have also shown that the combined process of EC and EF not only has a higher removal efficiency in treating organic pollutants compared to a single EC process, but this combined process is also more powerful than EC combined with other processes. Afanga et al. (2020) tested a hybrid electrochemical process that included electrocoagulation (EC) alone and combined with electro-Fenton (EF), anodic oxidation (AO), and peroxic coagulation (PC) for the treatment of textile industry wastewater using an intermittent reactor; the experimental results showed that the treatment effectiveness of textile industry wastewater decreased in the order of EC-EF>EC-AO>EC-PC>EC. The

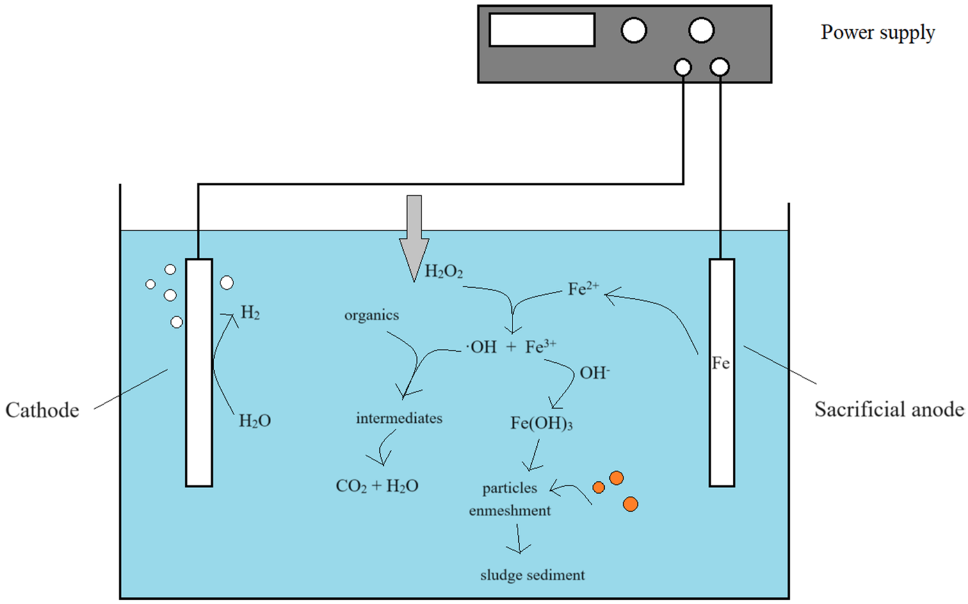


**Fig. 11** The mechanism of hydrogen peroxide assisted EC with Fe electrodes (EC-EF process)

study also showed that the energy consumption using the integrated process was lower than that of EC alone; where the energy consumption required for TOC removal was 3 kWh·kg$^{-1}$ when the EC process was used alone, whereas when EC was combined with EF, the energy consumption decreased to 0.45 kWh·kg$^{-1}$. Similarly, Zazou et al. (2019) treated a textile industrial wastewater containing reactive dyestuff mixtures by electrocoagulation combined with different electrochemical advanced oxidation processes (PC, AO and EF) to treat the textile industrial wastewater containing a mixture of reactive dyes; the experimental results showed that among the tested electrochemical advanced oxidation processes, the sequential EC-EF treatment was preferred under optimal reaction conditions and was very effective because it provided electrically generated hydroxyl radicals ·OH. By using the combined EC-EF process, 97%, 100%, and 100% of the TOC, turbidity, and color removal rates. In contrast, TOC removal was only 92% and 68% for the EC-AO and EC-PC processes, respectively. In terms of energy consumption, the single EC process required 3 kWh to remove 1 kg of TOC while treating wastewater under optimal conditions, while the EC-EF process required only 0.1 kWh.

It is concluded that EC-EF as a combined process is not only more significant in treating organic pollutants compared to a single EC process, requires less energy than a single EC process, but is also more effective than the combination of EC with other processes. In addition, studies related to the treatment of other pollutants using the EC-EF process in addition to the above studies are also listed in Table 7.

Table 7 Research on the combination of Electro-Fenton and electrocoagulation technology

## Conclusion

This paper lists and analyzes the influencing factors and combination technologies related to the electro-Fenton process; among the influencing factors analyzed above, the addition of $H_2O_2$ can improve the removal efficiency of the electro-Fenton process, but the excessive addition will have a scavenging effect on the ·OH, and in addition, it will also increase the pH of the solution and the operating cost of the reaction. In terms of feeding mode, the removal efficiency of multiple additions is better than a single addition. An increase in reactant concentration decreases the reaction constant, which in turn has an effect on the degradation efficiency of the pollutant. The two geometric parameters, reaction volume and electrode quality, are less studied, but from the only studies, increasing the electrode quality favors the reaction, while increasing the reaction volume hinders the generation of $H_2O_2$. The reaction time not only reflects the trend of the relevant parameters with time during the degradation or oxidation of pollutants or organics, but also provides an assessment and reference for finding the optimal degradation efficiency and optimal process cost of pollutants or organics. Finally, there is a similarity between the effects of voltage (potential) and current, and excessive voltage (potential) can also produce the occurrence of side reactions such as hydrogen precipitation reaction in solution and $H_2O_2$

**Table 7** Research on the combination of Electro-Fenton and electrocoagulation technology

| Organics/Pollutants | Electrodes | Reaction condition | Results | References |
|---|---|---|---|---|
| plugboard wastewater | Cathode: stainless steel<br>Anode:$Ti_{0.7}Ru_{0.3}O_2$ | pH:3,<br>current density:104.2 A $m^{-2}$,<br>$H_2O_2$ dosage:600 mg $L^{-1}$,<br>reaction solution:1 L,<br>electrocoagulation process,<br>reaction time:30 min | After 30 min of reaction, the COD content in the wastewater decreased from 232 mg $L^{-1}$ to 50 mg $L^{-1}$. | Zhao et al. 2012 |
| bronopol | Cathode: Fe,<br>Anode: Fe (EC),<br>Cathode: carbon-PTFE air-diffusion<br>Anode: BDD thin film (EF) | Electrocoagulation process:<br>pH:2,<br>I:200 mA,<br>electrode distance: 1 cm,<br>reaction time: 60 min,<br>bronopol concentration: 2.78 mM,<br>solution;150 mL<br>Electro-fenton process:<br>pH:3,<br>solution:100 mL,<br>I:100 mA,<br>aeration rate:1 L/min | After 420 min of combined process reaction, 85% TOC removal was achieved. | Bocos et al. 2016 |
| microbiota from urban wastewater | Cathode: Fe,<br>Anode: Fe (EC),<br>Cathode:, carbon-PTFE air-diffusion<br>Anode: BDD (EF) | Electrocoagulation process:<br>solution:200 mL,<br>j:200 A $m^{-2}$,<br>T:25°C,<br>reaction time:30 min,<br>Electro-fenton process:<br>initial pH:7,<br>j:333 A $m^{-2}$,<br>$Na_2SO_4$:0.05 M,<br>stirring:800 rpm,<br>reaction time:30 min | After 30 min of EC treatment, all the active microbiota (except the heterotrophic bacteria) were inactivated within 30 min of EF, including the spores. | Anfruns-Estrada et al. 2017 |
| textile industry wastewater | Cathode: iron plate,<br>Anode: iron plate (EC),<br>Cathode: tridimensional carbon felt;<br>Anode: BDD deposited on a niobium substrate (EF) | solution: 400 mL,<br>j:10 mA $cm^{-2}$<br>EC process:<br>pH:8.75,<br>electrode distance:1.5 cm<br>EF process:<br>pH:3,<br>stirring: 200 rpm,<br>aeration time: 10 min | The mineralization rate of organic matter in wastewater can reach 97%, the removal rate of total suspended solids could reach 98%. | Afanga et al. 2020 |

**Table 7** (continued)

| Organics/Pollutants | Electrodes | Reaction condition | Results | References |
|---|---|---|---|---|
| textile industry wastewater | Cathode: iron plate,<br>Anode: iron plate (EC);<br>Cathode: carbon felt (CF),<br>Anode: BDD thin film (deposited on a niobium substrate) (EF) | EC process:<br>electrode distance:1.5 cm,<br>solution:400 mL,<br>j:20 mA $cm^{-2}$,<br>pH:8.75,<br>time:60 min<br>EF process:<br>aeration time: 10 min,<br>pH:3,<br>j:5 mA $cm^{-2}$,<br>$Fe^{2+}$ concentration: 0.2 mM,<br>time:60 min | Through the EC process, the removal rates of TOC and COD in wastewater were 44% and 62%, respectively; subsequently, using the EF process, a 94% TOC removal rate was obtained under this reaction condition. | Zazou et al. 2019 |
| domestic wastewater | Cathode: iron,<br>Anode: vitreous carbon | reaction time:60 min,<br>recycling flow rate:0.4 L $min^{-1}$,<br>j:0.34 A $dm^{-2}$ | Under this reaction condition, the removal rates of turbidity, TSS, and COD in domestic wastewater were 90.40%, 70.8%, and 50.07%, respectively. | Daghrir and Drogui 2012 |
| copper-ethylenediaminetetraacetic acid (Cu-EDTA) | Cathode: stainless steel,<br>Anode: iron plate | Cu-EDTA: 1.0 mM,<br>$Na_2SO_4$:0.05 M,<br>pH:3.0,<br>$H_2O_2$ concentration: 49.4 mM,<br>j: 72.92 $A/m^2$,<br>$NaClO_4$:10 mM,<br>time:30 min | The removal efficiency of Cu-EDTA could reach over 90%. | Guan et al. 2018 |
| Food industry wastewater | Cathode: Fe,<br>Anode: Fe | EF process:<br>j:5 $mA/cm^2$,<br>pH:2,<br>time:120 min<br>EC process:<br>pH:7.5±0.3,<br>j:15 $mA/cm^2$,<br>time:180 min | This tandem sequential treatment processes resulted in 58.7 % TOC, 93.9 % total phosphate, 82.8 % TSS and 74.4 % turbidity reduction at 120 min EF (5mA/$cm^2$) and 180 min EC (15mA/$cm^2$) by applying optimum operation conditions. | Dindaş et al. 2018 |
| oxyfluorfen, lindane | Electrode: BDD (EF), | j:254.67 $A/m^2$, | After electrocoagulation, EF could remove more than 94% of the pollutants; in terms of energy consumption, the reduced value of oxyflufen (1-log removal) is as low as 14.51 kWh $m^{-3}$, and the reduced value of lindane was as low as 49.7 kWh $m^{-3}$. | Raschitor et al. 2019 |

decomposition. With the synergistic effect of electro-Fenton with ultrasound, anodic oxidation, and electrocoagulation technology, the efficiency of pollutant degradation is better than the efficiency of single electro-Fenton process.

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