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Personal Review:

Progress and prospect in electro-Fenton process for wastewater treatment*

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Abstract: As a novel advanced oxidation process (AOP), electro-Fenton process is powerful for degrading most organic compounds including toxic and non-biodegradable ones, and so has attracted great attention. This paper reviews this process in detail including the mechanism, electrolytic bath, electrode materials, aerations and operation parameters. The application of electro-Fenton method in wastewater treatment is evaluated and summarized. Future work in this field is suggested, and three main directions of new electrode exploitation, development of assisted technologies and mechanistic study should be strengthened.

Key words: Electro-Fenton, Hydroxyl radicals, Advanced oxidation process (AOP), Refractory wastewater
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INTRODUCTION

Due to the increasing species and amount of toxic and bio-refractory organic pollutants in wastewater, great attention has been paid to one of the advanced oxidation processes (AOPs), i.e. the electro-Fenton process. The electro-Fenton process was carried out with a conventional anode and a graphite-based cathode, and on the cathode the H_2O_2 is produced electrochemically via oxygen reduction; then addition of Fe^{2+} into the system generates analogously the OH· radicals in the classical Fenton's reaction between Fe^{2+} and H_2O_2 (Brillas and Casado, 2002). Compared to the conventional Fenton process, the electro-Fenton process has the advantage of allowing better control of the process and avoiding the storing and transport of the H_2O_2. Moreover, electricity as a clean energy source is used in the process, so the overall process does not create secondary pollutants.

Considerable study of the application of electro-Fenton process in wastewater treatment has been undertaken to date. The process efficiency has been experimentally confirmed by different authors for the treatment of media containing phenol (Fockedey and Lierde, 2002), aniline (Brillas and Casado, 2002), azo dyes (Guivarch *et al*., 2003b; Lahkimi *et al*., 2007), acetic acid (Casado *et al*., 2005), and 2,4-D (Brillas *et al*., 2000). It has also been shown that the process efficiency is improved by illumination with UV or visible light during electrolysis (Casado *et al*., 2005). The combined method of electro-Fenton process and UV has been called photoelectro-Fenton process. Photoelectro-Fenton process was first proposed and applied to degrade 4-chlorophenol by Brillas *et al*.(1998a). With the irradiation of UV light, a higher regeneration rate of Fe^{2+} can be obtained, so that the catalytic effect of Fe^{2+} can be enhanced.

However, there are few detailed general overviews on the electro-Fenton process. The aim of this paper is to review the progress of the electro-Fenton process reported during recent years. On the base of

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this review, further work in this field is also tentatively suggested.

MECHANISM

In the electro-Fenton process, H_2O_2 is formed by the reduction of the dissolved oxygen on the cathode surface (Eq.(1)). H_2O_2 can then react with the externally added Fe^{2+} to produce $OH\cdot$ (Eq.(2)), and this reaction is Fenton's reaction.

$$O_2+2H^++2e^-\rightarrow H_2O_2, \tag{1}$$

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

$$Fe^{3+}+e^-\rightarrow Fe^{2+}. \tag{3}$$

$OH\cdot$ has high oxidation potential that is only lower than fluorine, and must react with most organic pollutants completely and rapidly. Moreover, the regeneration of Fe^{2+} from the reduction of Fe^{3+} at the cathode (Eq.(3)) is crucial for the electro-Fenton process (Oturan and Pinson, 1995; Brillas *et al.*, 2000). The sum of reactions in Eqs.(1)~(3) gives the global reaction that takes place in the cathodic compartment (Eq.(4)). In the anodic compartment, the reaction that takes place is simply the oxidation of water (Eq.(5)). The overall reaction ((Eq.(6)) taking place in the electrolysis cell and leading to the production of hydroxyl radicals emphasizes the catalytic character of elector-Fenton process. Thus, the only reagents necessary for the production of $OH\cdot$ are oxygen and energy from the electrical generator (Oturan, 2000).

$$O_2+3H^++3e^-\rightarrow H_2O+OH\cdot, \tag{4}$$

$$H_2O\rightarrow\frac{1}{2}O_2+2H^++2e^-, \tag{5}$$

$$\frac{1}{2}O_2+H_2O\rightarrow 2OH\cdot. \tag{6}$$

In fact, the regeneration of Fe^{2+} can also be achieved by the oxidation of an organic, by the reaction with H_2O_2 or by the reaction with hydroperoxyl radical as described below (Brillas *et al.*, 2000):

$$Fe^{3+}+R\cdot\rightarrow Fe^{2+}+R^+, \tag{7}$$

$$Fe^{3+}+H_2O_2\leftrightarrow[Fe\text{-}O_2H]^{2+}+H^+\leftrightarrow Fe^{2+}+HO_2\cdot+H^+, \tag{8}$$

$$Fe^{3+}+HO_2\cdot\rightarrow Fe^{2+}+H^++O_2. \tag{9}$$

The typical mechanism of electro-Fenton process is illustrated in Fig.1.

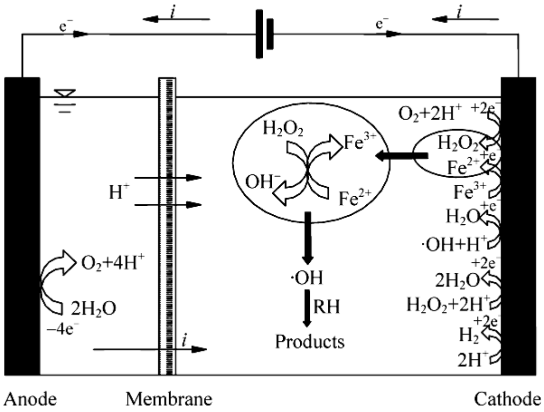

Fig.1 Illustration of the reaction mechanism of electro-Fenton process (Qiang, 2002)

AFFECTING FACTORS

Electrolytic bath

The integrated conventional electrode system (ICES) is commonly employed as illustrated in Fig.2a. Also the anode and cathode compartments can be separated by a membrane (Fig.2b), or a glass frit (Guivarch *et al.*, 2003a; Kaichouh *et al.*, 2004). This electrolytic bath is called separated conventional electrode system (SCES). The separation is beneficial to the anode and cathode reaction individually, and these two different reactions do not disturb each other. Moreover, it can prevent the anodic electrode from being polluted.

The concept of three-dimensional electrodes developed in the late 1960s, and is a novel electrochemical reactor (Xiong *et al.*, 1998). The three-dimensional electrode system (TDES) is also called a particle electrode system or a bed electrode system. In this kind of electrode system, granular or other drossy working electrode materials are filled in the region between the electrodes of conventional two-dimensional electrodes. The surface of working electrode materials is charged, and the electrochemical reaction takes place on the charged surface. The particle electrode is like a flow bed, with a large area-volume ratio, and the main mechanism of TDES is micro-electrolysis. Owing to the short distance between the working granules, high efficiency of mass transfer can be achieved in TDES, particularly when the conductivity of the raw water is low. Therefore, a major disadvantage of the limitation of specific surface area of broad electrode in the conventional electrode system can be overcome.

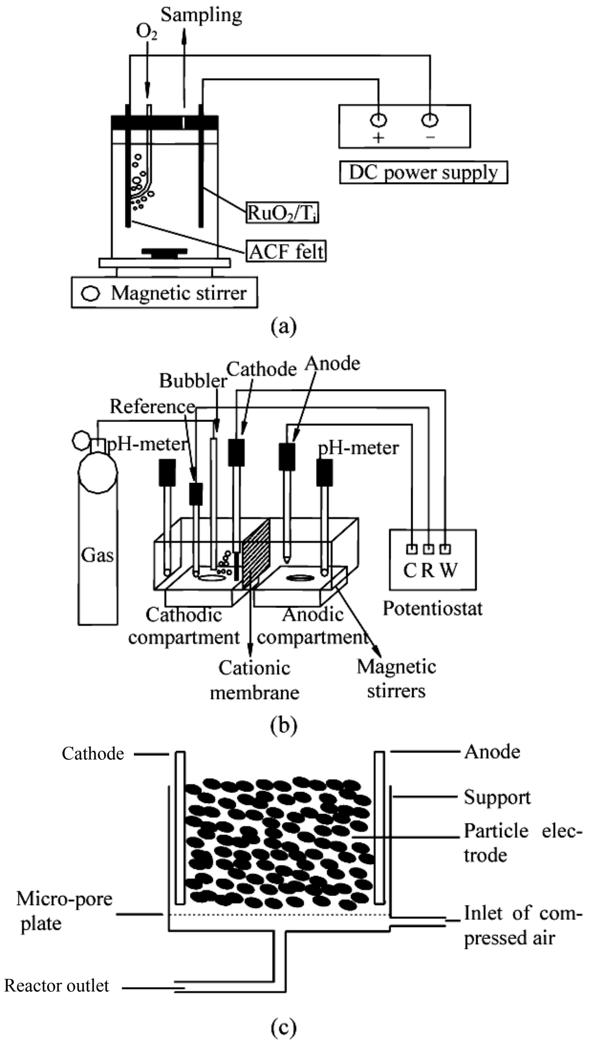

Fig.2 Scheme of electrolytic bath
(a) ICES (Wang *et al.*, 2005); (b) SCES (Pozzo *et al.*, 2005a); (c) TDES (He *et al.*, 2002)

Electrode materials

Pt anode has been successfully used to degrade herbicides and pesticides owing to its stability and excellent transmission of electrons (Brillas and Casado, 2002; Brillas *et al.*, 2000; 1998b; Oturan, 2000). However, Pt anode is hardly used in practical wastewater treatment because of its high cost. Therefore, Fe anode is widely used, especially in the practical wastewater treatment. Pt and Fe are the main anode materials in electro-Fenton process. Besides, new materials, for instance, boron-doped diamond (BDD) electrodes have emerged (Brillas *et al.*, 2004; Flox *et al.*, 2006). However, BDD electrode is more expensive than Pt.

Cathode is the working electrode in the electro-Fenton system, so the cathode efficiency is critical for the system. So far, various cathode materials have been used laboratory-scale or even pilot-scale. The graphite electrode is widely applied as the working electrode (cathode), and Pt as the anode to treat wastewater containing biosphenol A (BPA) (Gözmen *et al.*, 2003), pentachlorophenol (PCP) (Oturan *et al.*, 2001), p-nitrophenol (PNP) (Oturan *et al.*, 2000), organophosphorus pollutants (Guivarch *et al.*, 2003a), with the removal percentage of all these pollutants being over 80%. Activated carbon fiber (ACF) is a new material with strong ability of adsorption. Its feasibility in the electro-Fenton process has been identified by Wang *et al.*(2005). They used ACF as cathode to degrade an azo dye and Acid Red 14. After 360 min of electrolysis and under the operation conditions of 0.36 A current, 1 mmol/L Fe^{2+} at pH 3, 70% TOC (total organic carbon) was removed and complete decolorization was achieved. They also compared graphite and ACF cathodes under the same condition, with the results showing that more H_2O_2 was electro-generated from oxygen reduction due to the large surface area of ACF. Carbon-polytetraflouoroethylene (Carbon-PTFE) was used as a typical composite cathode in the electro-Fenton process by Brillas and co-workers to destroy many organics, for instance, 2,4-D (Brillas *et al.*, 2000), aniline (Brillas and Casado, 2002; Brillas *et al.*, 1998b). Reticulated Vitreous Carbon (RVC) (Fockedey and Lierde, 2002) and Hg pool (Ventura *et al.*, 2002) were also used as cathode in some studies, however, Hg is fatal for the environment.

Aeration

In the electro-Fenton process, H_2O_2 is generated from oxygen reduction. The efficiency of aeration is not only important for the generation of H_2O_2, but for the running cost. There are generally two approaches to improve the production of H_2O_2. One is to select a cathode with large surface area, and the other is to enhance the aeration efficiency. Oxygen is introduced into the system mainly by aeration. The simple mode of aeration is shown in Fig.2a. It is easy to operate and maintain, but has the limitation of mass transfer of oxygen.

Brillas *et al.*(1996) also first introduced a gas diffusion electrode (GDE) into electro-Fenton process. High-usage of oxygen was obtained through this kind of cathode. As shown in Fig.3, the GDE is always composed of a carbon-PTFE layer in contact with a metal mesh as current collector. The gas flow passed over the inner face of the cathode through a tube inside the holder, the top of which was connected to a drechsel to maintain the right pressure in the Plexiglas holder (Pozzo *et al.*, 2005a). The compressed oxygen or air flow was fed to the carbon-PTFE cathode to electrogenerate H_2O_2 via Eq.(1) by which organics were decomposed (Shen *et al.*, 2005). In GDE, oxygen is not aerated from the aqueous solution to the cathode surface, but escapes from the cathode itself. Oxygen reaches the cathode surface much more easily. No bubbles are observed on the cathode surface if operated properly. Obviously, it can be used to treat wastewater containing volatile organic compounds. Distinctly, the GDE was found to have higher selectivity for H_2O_2 production without any significant loss of the iron regeneration. The GDE was also proved effective even using air as the oxygen source with a significant decrease of operating cost (Pozzo *et al.*, 2005a).

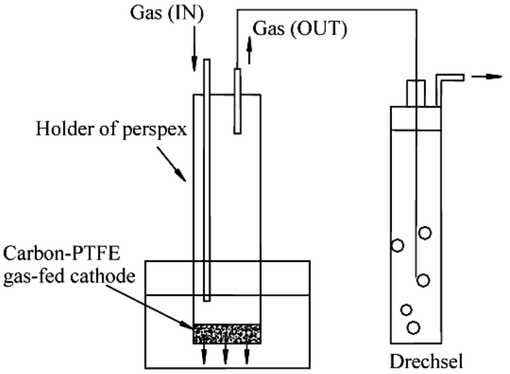

Fig.3 Gas diffusion electrode (GDE) (Pozzo *et al.*, 2005a)

So far, there is no special discussion about the difference of the several types of electrolytic baths and electrodes. However, the GDE has shown obvious advantages, and the composite electrodes with high efficiency and low cost are promising.

Operation parameters

It has been proved that the Fenton's reaction is a chain reaction. Various contaminants were studied in the laboratory to determine the optimal conditions. Practical applications of electro-Fenton process to degrade contaminants have also been examined in pilot-scale and continuous treatment systems.

1. pH

The pH is one of the most important factors for the electro-Fenton process. It has been confirmed that the optimal value of pH is 2~4. On one hand, when the pH increases, the iron ions especially the Fe^{3+} precipitate. Therefore, the amount of catalyst of Fenton's reaction decreases. On the other hand, when $pH<2$, H_2O_2 cannot be discomposed to OH· by Fe^{2+}. In this case, H_2O_2 turns into $H_3O_2^+$ by capturing one proton. $H_3O_2^+$ is electrophilic, leading to the decreasing rate of reaction between H_2O_2 and Fe^{2+} (Han and Xia, 2004).

In fact, the optimal pH indicates a disadvantage of electro-Fenton process because the pH of most waters is not within the optimal range. There are two ways to decrease the pH of wastewater. One is to add acid, and the other is to mix the target wastewater with some acidic wastewater. Some researchers investigated the wastewater treatment in neutral pH and the organics can also be removed successfully. But, in that case, the wastewater is depolluted mainly by coagulation rather than by degradation of OH· (Chen *et al.*, 2002).

2. Applied voltage and current

These two operation parameters are related directly to the cost of electro-Fenton process. With the increase of the voltage or current applied, the degradation of substrate increases at the beginning, and then tends to be stable and even worse because of the side reactions and polarization. In the electro-Fenton process, the current density is always low. This is the other disadvantage of the electro-Fenton process, because higher current density means larger reaction rate. Some researchers suggested that the current density in the electro-Fenton process should be no larger than 10 A/m^2, while others thought that the upper limit value should be 6.4 A/m^2 (Zhang *et al.*, 2004).

3. Distance between electrodes

The decrease of the distance between the electrodes leads to a decrease of the ohmic drop through the electrolyte and then an equivalent decrease of the cell voltage and energy consumption (Fockedey and

Lierde, 2002). It can be concluded that the closer the electrodes are, the better the performance is. However, it is necessary to keep appropriate distance between the electrodes for installation and avoidance of short circuit between anode and cathode.

4. Ratio of Fe^{2+} and H_2O_2

During the degradation of phosphorus compounds by electro-Fenton process, Pozzo *et al.*(2005b) considered the iron dosage as the crucial step whose optimal concentration was found to be in the range of 50~150 mg/L. That is, although Fe^{2+} can react with H_2O_2 to generate OH·, Fe^{2+} and H_2O_2 cannot be excessive unilaterally because of the occurrence of undesired side reactions as follows:

$$Fe^{2+}+OH\cdot\rightarrow Fe^{3+}+OH^-, \qquad (10)$$

$$H_2O_2+OH\cdot\rightarrow HO_2\cdot+H_2O. \qquad (11)$$

In Eq.(11), the HO_2 is also an oxidant, but has an oxidation potential much less than that of OH·. The relationship between the Fe^{2+} and H_2O_2 can be expressed as described below:

$$\text{Scavenger rate }[Fe^{2+}]=\text{Scavenger rate }[H_2O_2], \qquad (12)$$

that is,

$$k_{\text{ferrous ion}}[Fe^{2+}][OH\cdot]=k_{\text{hydrogen peroxide}}[H_2O_2][OH\cdot]. \qquad (13)$$

Therefore, the optimal ratio between Fe^{2+} and H_2O_2 can be obtained as Eq.(14):

$$R=[Fe^{2+}]/[H_2O_2]=k_{\text{hydrogen peroxide}}/k_{\text{ferrous ion}}=3.3\times10^9/(3.0\times10^8)=11. \qquad (14)$$

However, in the actual operation, the optimal ratio between Fe^{2+} and H_2O_2 is much lower than the theoretical value. For instance, in Pozzo *et al.*(2005b)'s study, they compared several different mole ratios of R. The results are shown in Fig.4. The oxidation of hypophosphite reaches maximum for a mole ratio of about 2, but has no significant improvement with further increase in the iron dosage. The reason for the difference between the actual and calculated value could be understood from the fact that several side reactions contributed to the Fe^{2+} consumption. Pozzo *et al.*(2005b) suggested that the crucial step appeared to be the iron dosage if H_2O_2 is continuously provided by the cathodic reduction of oxygen, and that the optimal concentration of iron dosage was found to be in the range of 50~150 mg/L. However, when using Fe^{3+} as the catalytic agent, the dosage of Fe^{3+} showed to be much lower than that of Fe^{2+}, for instance, in Bellakhal *et al.*(2006)'s study with a carbon-felt cathode, the maximum degradations could be obtained in the electro-Fenton process using only 0.5 mmol/L (28 mg/L) Fe^{3+}.

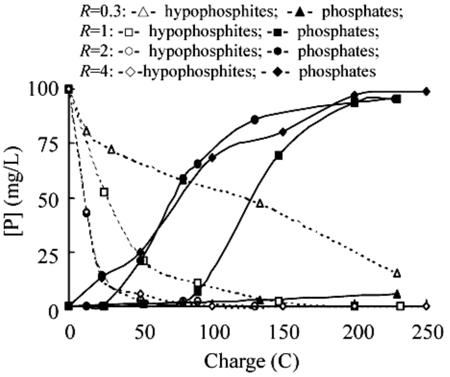

Fig.4 Influence of mole ratio *R* on phosphorus oxidation at $[H_2O_2]_{in}$=50 mg/L (Pozzo *et al.*, 2005b)

5. Supporting electrolyte

Sodium sulfate is generally used as the supporting electrolyte. Such electrolyte improves the solution conductivity, and accelerates the electron transfer, thus benefits the electro-Fenton reaction. Therefore, supporting electrolyte is necessary, especially in the solution without enough conductivity.

The concentration of the supporting electrolyte also affects the conductivity of the solution. Yuan *et al.*(2004) compared several concentration levels of NaCl electrolyte in the treatment of nitrophenol wastewater. It was observed that no notable increase of COD removal was obtained when the NaCl concentration exceeded 3.6 g/L. In the wastewater treatment, it is not an efficient way to increase the treatment efficiency by adding excessive supporting electrolyte which only leads to extra cost.

APPLICATION

The electro-Fenton process is widely used to treat non-biodegradable or refractory organic compounds with moderate energy costs (Brillas and Casado, 2002), such as dye, herbicide, pesticide, and landfill leachate.

Herbicide and pesticide

Chlorophenoxy herbicides and pesticides are used worldwide on a large scale for weeds and pests control. These compounds resist biodegradation and have potential toxicity towards humans and animals. Their widespread use for agricultural and non-agricultural purposes results in rinsate, waste product and contaminated soil continuously in hundreds of thousands of application sites. An increasing accumulation of these products in natural waters has been detected, with contamination levels up to 10 μg/L in US and European ground waters. Thus, there is a growing interest in AOPs technology for their degradation (Brillas *et al.*, 2004). The electro-Fenton process had been proved to be powerful enough to degrade Chlorophenoxy herbicides (Boye *et al.*, 2002; Brillas *et al.*, 2000; 2004). However, many studies found that the electro-Fenton process with Pt anode is not potent enough to mineralize chlorophenoxy herbicides and pesticides due to the formation of stable complexes of Fe^{3+} with generated carboxytic acids. Therefore, BDD anode was developed and applied. This new anode material possesses technologically important characteristics such as an inert surface with low adsorption properties, remarkable corrosion stability and an extremely wide potential window in aqueous medium. BDD anode has much higher O_2 overvoltage than that of conventional anodes such as PbO_2, doped SnO_2, IrO_2 and Pt, then producing larger amount of adsorbed OH· on the anode's surface by reaction Eq.(15) (Brillas *et al.*, 2004). Brillas' group compared several methods to mineralize four chlorophenoxy herbicides: 4-CPA, MCPA, 2,4-D and 2,4,5-T. The removal of TOC of one of the chlorophenoxy herbicides is shown in Fig.5. Analogous treatment with a Pt anode does not allow complete depollution, confirming the higher oxidizing power of BDD anode in both methods. The combination of BDD anode and GDE cathode is much more powerful to mineralize completely the chlorophenoxy herbicide and pesticide.

$$H_2O \rightarrow OH_{ads}\cdot + H^+ + e^-. \quad (15)$$

Other kinds of herbicide and pesticide can be also degraded successfully by the electro-Fenton process. Edelahi *et al.*(2003) degraded diuron by the electro-Fenton process and diuron can be degraded in less than 10 min, and with as high as 93% COD removal. This process is also efficient for imazapyr degradation, and the COD removal was found to be higher than 95% (Kaichouh *et al.*, 2004). In Guivarch *et al.*(2003a)'s work, the wastewater containing organophosphorus pesticides could be mineralized over 80% by electro-Fenton process. During this degradation, the OH· contributed to breaking down the aromatic ring and removing the toxicity of the pesticide solution. Although the ozone oxidation technology is thought to be a common process for degrading non-biodegradable organic compounds in drinking water treatment plant, some pesticides such as atrazine, were refractory to ozone, while they can be degraded efficiently by electro-Fenton process (Ventura *et al.*, 2002).

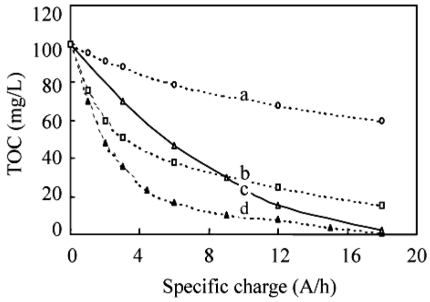

Fig.5 TOC abatement with specific charge for the degradation

Hundred ml of 230 mg/L 2,4-D solutions of pH 3.0 at 300 mA and at 35 °C. Methods: (a, ○) anodic oxidation with a 10 cm^2 Pt anode and a 3.1 cm^2 O_2-diffusion cathode; (b, □) electro-Fenton with a 10 cm^2 Pt anode, a 3.1 cm^2 O_2-diffusion cathode and 1 mmol/L Fe^{2+} in the solution; (c, Δ) anodic oxidation with a 3 cm^2 BDD anode and a 3 cm^2 graphite (without H_2O_2 production); (d, ▲) electro-Fenton with a 3 cm^2 BDD anode, a 3.1 cm^2 O_2-diffusion cathode and 1 mmol/L Fe^{2+} in the solution (Brillas *et al.*, 2004)

Dyes

Synthetic dyes are used extensively by several industries dyeing textile, paper and plastic matter. The textile industry uses about 10000 different dyes. The worldwide annual production of dyestuffs is over 7×10^5 t. It is estimated that 10%~15% of the dye is discharged into the effluent during dyeing processes. Furthermore, some azo dyes, their precursors and a number of their reaction products are carcinogenic. Therefore, the treatment of the effluent containing these compounds is important for the protection of natural waters. However, it is very difficult to successfully treat such refractory and non-biodegradable wastewater such as the azo, nitro or sulpho groups using traditional biological processes. The electro-Fenton process has been identified to be powerful for decreasing their strong color and toxicological effects. This process is efficient for azo dye degradation, achieving an efficient removal (over 80%) of COD (Guivarch *et al.*, 2003b). Wang *et al.*(2005) reported the mineralization of an azo dye Acid Red 14 by the electro-Fenton process, and 70% TOC removal as well as complete decolorization was achieved after 360 min of electrolysis under the operation conditions of 0.36 A current, 1 mmol/L Fe^{2+} at pH 3. Complete mineralization of acidic aqueous solutions containing concentrations of the dye indigo carmine up to 0.9 g/L had been proved to be feasible when using the electro-Fenton process with a BDD anode and 1.0 mmol/L Fe^{2+} as catalyst (Flox *et al.*, 2006). Similar successful applications were observed in the degradation of Reactive Red 120 azo dye and Reactive Black 5 azo dye (Kusvuran *et al.*, 2005).

Additionally, many other industry reagents can be degraded efficiently by the electro-Fenton process, such reagents are phenol (Fockedey and Lierde, 2002), pentachlorophenol (PCP) (Oturan *et al.*, 2001), aniline (Brillas and Casado, 2002), etc.

PROSPECT

At present, the electro-Fenton process is mainly studied in the laboratory, and there are many efforts to make it available in wastewater treatment project.

The form of electrolytic bath and electrode with low cost and high catalysis efficiency is of great importance in the development of electro-Fenton process. Although the conventional anode is Pt, it is identified to be less efficient than other anodes such as PbO_2, doped PbO_2, doped SnO_2, and IrO_2 (Boye *et al.*, 2002). Therefore, it is important to develop new electrode materials and new manufacture process. Besides, Zhang *et al.*(2004) suggested the research of the electro-Fenton process should be diverted healthfully along with the exploitation of highly active cathode materials.

Owing to its oxidation characters, the electro-Fenton process can be used as the pretreatment unit for the toxicity reduction of refractory waste. It is though that the oxidizing efficiency of H_2O_2 can be significantly enhanced by a combination with UV radiation (Brillas *et al.*, 1998a). The degradation process can be accelerated by the photolysis of Fe^{3+} complexes and by the enhancement of Fe^{2+} regeneration from Fe^{3+} species via photo-Fenton reaction. Thus, further tasks of electro-Fenton process should be conducted intensively on the combination of electro-Fenton process with other technologies.

Also, the mechanism of Fenton reaction is required to be identified in more depth, and the mathematical model of quantitative yield of H_2O_2 should be established in order to apply this process practically and predictably. The reactions in the electro-Fenton process are very complicated, so one aim of further study is to elucidate the mechanisms governing the process, and then to avoid most of the undesired side reactions consuming the key reagent in the degradation, OH·.

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