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研究论文
ARTICLE

光电芬顿氧化反应降解染料罗丹明 B

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摘 要：以有机染料罗丹明 B 为目标污染物，利用光电芬顿方法对其进行降解研究。研究汇总考察了芬顿试剂（二价铁盐、双氧水）的用量及比例、初始 pH 值、电流强度等对罗丹明 B 降解效果的影响。实验结果表明：芬顿试剂 H_2O_2/Fe^{2+} 的最佳物质的量比为 1 : 1，Fe^{2+} 浓度降低或 H_2O_2 浓度升高，罗丹明 B 降解效果增强；罗丹明 B 在 pH＝3.0 的酸性体系中降解效果最好；电流强度为 0.005 A 时，罗丹明 B 降解效果好且耗能最低。在 H_2O_2/Fe^{2+} 物质的量比为 1 : 1、电流为 0.005 A、初始 pH＝3.0、H_2O_2 初始浓度为 1.0 mM 条件下，以 350 nm 紫外光照射的光电芬顿反应降解初始浓度为 0.1 mM 的罗丹明 B 溶液，处理 10 min 后罗丹明 B 去除效率可达 99% 以上。

关键词：光电芬顿；罗丹明 B；影响参数；降解效果

Dye Rhodamine B degradation by photo-Electric Fenton's oxidation

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Abstract: ***Background, aim, and scope*** The synthetic dyes is undesirable in water because the coloring agents are harmful to the aquatic environment. Rhodamine B (RhB) is an important representative of xanthene-based dyes. Human exposure to RhB can cause irritation in contact with skin, eyes and respiratory tract. Fenton reaction is proven to be an efficient oxidation process for organic pollutants in previous literatures, due to the fact that ferrous salt is widely available, non-toxic and relative cheap and that the end product is environmentally benign. The hydroxyl radical (·OH) is the active species to degrade target compounds in Fenton reaction. However, the rapid depletion and slow regeneration of ferrous ions throughout the reaction is one of the main drawbacks in classical Fenton process ($Fe^{2+}+H_2O_2 \rightarrow Fe^{3+}+OH^-+\cdot OH$). In the photo-Fenton and electro-Fenton process, the catalytic reaction is propagated by Fe(Ⅱ) regeneration to overcome the drawback in classical Fenton process. ***Materials***

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and methods In this study, a novel electric photo-Fenton process was employed, the Fenton's reagents of ferrous ions (Fe^{2+}) and H_2O_2 were used in the electrochemical cell to produce hydroxyl radicals, the iron steel worked as sacrificial anode for the generation of supplementary Fe^{2+}, and Fe^{2+} were also regenerated via the reduction of ferric ions by the cathode and UV light. The dye RhB was chosen as a target pollutant to evaluate the performance in aqueous solution. Three types of Fenton reactions (i.e. sole-Fenton, photo-Fenton and electro photo-Fenton) were studied and compared. The effect parameters such as pH, hydrogen peroxide concentration, ferrous ion dosage, and electric current were evaluated and optimized. ***Results*** Three types of Fenton processes (i.e. sole-Fenton, photo-Fenton and electro photo-Fenton) were conducted to identify the RhB degradation efficiency. The electro photo-Fenton shows best performance to RhB degradation. Meanwhile, in the electro photo-Fenton process, the effect of H_2O_2/Fe(Ⅱ) molar ratio was examined in the range from 1 : 10 (5 : 50) to 10 : 1, respectively. As the $[H_2O_2]_0$ fixed at 0.5 mM or 1.0 mM, the decay efficiency were both increased with the decrement of Fe (Ⅱ) and the optimum RhB decay was achieved at H_2O_2/Fe(Ⅱ) molar ratio = 1 : 1. Next, the influence of solution pH on RhB degradation was examined with the pH value in the range from 2.0 to 4.0. The highest RhB degradation efficacy was reached at an initial pH value around 3.0, and the degradation performances of above or below this pH value were worse. Finally, The influence of the electric current (I) on the RhB degradation was studied in the range from 0.002 A to 0.030 A at current-controlled conditions. A significant jump was observed when I increased from 0.002 A to 0.005 A. However, the decay rate leveled off as the electric current further increased. ***Discussion*** (a) The outstanding performance of electro photo-Fenton than other types of Fenton processes was achieved by an extra application of electrochemical method, where Fenton reagents of H_2O_2 and Fe^{2+} can both be re-generated. (b) The optimum RhB decay was achieved at H_2O_2/Fe(Ⅱ) molar ratio = 1, after which the decay performance levelled off. On the other hand, the increment of $[H_2O_2]_0$ always brought the performance improvement, while the decreament of Fe(Ⅱ) dosage improved the decay performance. (c) The increment of decay rate at higher current at the range of 0.002—0.005 A was likely attributed to the faster release of H_2O_2 on the cathode. ***Conclusions*** The electric photo-Fenton with a sacrificed anode showed the optimum performance for RhB degradation. The degradation curve can be defined as a two-stage reaction comprised of a rapid first stage and a retarded second stage. The effect of various parameters such as the Fenton reagent H_2O_2/ Fe(Ⅱ) molar ratio, initial solution pH value, and electric current was further investigated and optimized. The optimal conditions were H_2O_2 : Fe(Ⅱ) molar ratio = 1 : 1, pH = 3.0, and electric current = 0.005A. The experimental results showed that the RhB degradation efficiency under the optimal parameters condition can achieve more than 99% in 10 min. ***Recommendations and perspectives*** The photo-Electric Fenton's oxidation technology is a promising technology to removal synthetic dyes, and the generation of less ferric hydroxide sludge would reduce the workload for additional separation and disposal. It can be combined with other technologies to give full play to its advantages.

Key words: electric photo-Fenton; Rhodamine B; effect parameters; degradation efficiency

1 Introduction

Water pollution is already a global problem and causes some countries facing water shortage problem. One of water pollution sources is synthetic organic compounds. The application of synthetic organic compounds has major concern on the environmental and human health risk although they have employed as important role in the development of modern society, since they are nonbiodegradable, highly toxic and may be carcinogenic and mutagenic to organisms (Wilkinson et al, 2017). Synthetic dyes have been widely used in various fields such as textile, leather treatment, paper, food technology, pharmaceutical, agriculture research, photoelectrochemical cells, hair coloring (Shukla and Oturan, 2015). The water containing synthetic dyes is undesirable because

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the coloring agents are obvious and may be harmful to the aquatic environment. Dyes can reflect and absorb sunlight to the water that they can obstruct the population growth rate of bacteria and inhibit photosynthesis of aquatic plants and algae (Salleh et al, 2011).

Rhodamine B (RhB) is an important representative of xanthene-based dyes. It is extensively used as a colorant in various textile-processing industries and food stuffs, including dyeing silk, wool, jute, leather, cotton and so on. The LD_{50} for RhB in oral mouse is 887 $mg \cdot kg^{-1}$. Human exposure to RhB can cause irritation in contact with skin, eyes and respiratory tract (Palphramand et al, 2011). In California, USA, it is required that products containing RhB must contain a warning on its label.

Fenton reaction is proven to be an efficient oxidation process for organic pollutants in previous literatures, due to the fact that ferrous salt is widely available, non-toxic and relative cheap and that the end product is environmentally benign (Klavarioti et al, 2009). Besides, it is easy to operate and maintain as compared to other AOPs (advanced oxidation processes). The hydroxyl radical (·OH) is the active species to degrade target compounds, according to Eq. (1) (Epold et al, 2015).

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + \cdot OH \qquad (1)$$

However, the rapid depletion and slow regeneration of ferrous ions throughout the reaction is one of the main drawbacks in classical Fenton process (Rahim Pouran et al, 2015). The alternative to the Fenton's reagent is the photo-Fenton and electro-Fenton process, in which the catalytic reaction is propagated by Fe(Ⅱ) regeneration via equation (2) and (3), respectively (Rahim Pouran et al, 2015).

$$Fe^{\mathrm{III}}(OH)^{2+} + h\nu \rightarrow Fe^{2+} + \cdot OH \qquad (2)$$

$$Fe^{3+} + e^- \rightarrow Fe^{2+}(\text{Cathode}) \qquad (3)$$

In this study, a novel electric photo-Fenton process was employed, in which Fenton's reagents of ferrous ions (Fe^{2+}) and H_2O_2 were used in the electrochemical cell to produce hydroxyl radicals, the iron steel worked as sacrificial anode for the generation of supplementary Fe^{2+}, and Fe^{2+} were also regenerated via the reduction of ferric ions by the cathode and UV light. The dye RhB was chosen as a target pollutant to evaluate the performance in aqueous solution. Three types of Fenton reactions (i.e. sole-Fenton, photo-Fenton and electro photo-Fenton) were studied and compared. The effect parameters such as pH, hydrogen peroxide concentration, ferrous ion dosage, and electric current were evaluated and optimized.

2 Experiment

The dye RhB (Rhodamine B, $C_{28}H_{31}N_2O_3Cl$, N-[9-(ortho-carboxyphenyl)-6-(diethyl-amino)-3H-xanthen-3-yli-dene] diethyl ammonium chloride) was purchased from Sigma-Aldrich Inc (USA). Fenton reagents, including hydrogen peroxide (35%) and Iron(Ⅱ) sulfate heptahydrate (99.0%) were of analytic reagent grade and obtained from Sigma-Aldrich Inc. (USA). Sulfuric acid was employed to adjust pH values. Sodium thiosulfate was used to quench the Fenton reaction. Sodium sulfate anhydrous at the concentration of 0.05 M ($mol \cdot L^{-1}$) was employed as background electrolyte for the electric photo-Fenton process. The deionized and distilled water with an 18.2 MΩ resistivity was generated by a Bamstead NANO pure water treatment system (USA).

The electric photo-Fenton experiments were conducted in an electrochemical cell, composing of a single-compartment quartz beaker, a steel sheet with total surface area of 12.5 cm^2 as sacrificial anode, and a platinum sheet as the cathode. An Agilent E3641A DC potentionstat-galvanostat power supply was employed to provide the constant current. Two 8 W fluorescent lamps with emitting wavelength at 350 nm were placed on the top of the cell to supply UV light. Experiments were carried out in 100 mL 0.1 mM RhB aqueous solution containing 0.05 M Na_2SO_4 as background electrolyte with pH adjustment. The reaction was initialized by pipetting a suitable amount of H_2O_2 and Fe^{2+} into the reactor and turning on the UV lamps simultaneously when a specified constant electrical current was supplied. For photo-Fenton process, the anode, electrolyte and current were disposed. For sole-Fenton process, the UV lamps were

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disposed also.

The solution was mixed by a magnetic stirrer to maintain a complete homogeneity solution throughout the reaction. At determined time intervals, the defined portion of sample was taken from the reactor. The portion sample was mixed immediately with specific amount of sodium thiosulfate to quench the reaction. After that, the solution was then analyzed by a UV-VIS spectrophotometer at the wavelength of 553 nm to quantify the remaining dye. All the experiments were carried out at (23 ± 1)℃. Parts of test were examined in triplicate. The relative standard deviations of the determination were less than 10%.

3 Results and discussion

3.1 Comparison of different Fenton processes

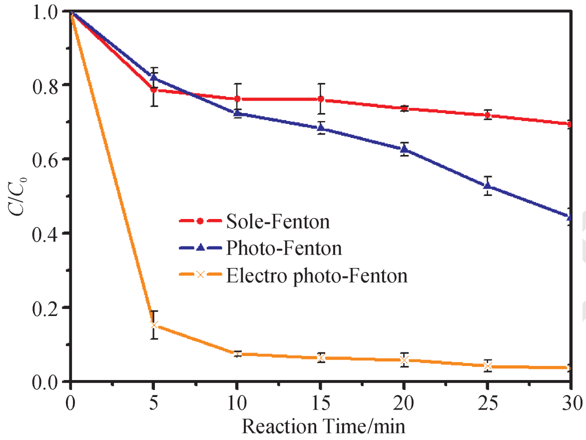
Three types of Fenton processes (i.e. sole-Fenton, photo-Fenton and electro photo-Fenton) were conducted to identify the RhB degradation efficiency. As shown in Fig.1, the sole-Fenton process showed about 30% of RhB decay in 30 min, while 55% of RhB decay was observed in photo-Fenton process. Among the three processes, the electro photo-Fenton was the optimum one with 95% RhB decay. The best performance of electro photo-Fenton should be ascribed to its different Fenton reactions. Apparently, the sole-Fenton process in the dark involved the generation of hydroxyl radicals as stated in Eq. (1). When in the photo-Fenton process, the utilization of UVA ($\lambda = 350$ nm) contributed a positive effect of photo-reduction of ferric ions to ferrous ions and hydroxyl radicals (·OH), via Eq. (2). Besides, the direct photolysis of Fenton reagent H_2O_2 also contributes to the production of ·OH radicals according to Eq. (4).

$$H_2O_2 + h\nu \rightarrow 2 \cdot OH \ (\lambda < 400 \text{ nm}) \quad (4)$$

As a result, the photo-Fenton process showed a faster RhB decay than sole-Fenton. The outstanding performance of electro photo-Fenton was achieved by an extra application of electrochemical method, where both Fenton reagents of H_2O_2 and Fe^{2+} can be yielded. Theoretically, H_2O_2 can be electro-generated via the reduction of dissolved oxygen at the cathode (Eq. (5)) (Özcan et al, 2008), while Fe^{2+} ions can be electrically generated on a sacrificial anode through iron oxidation (Eq. (6)) and electro-regenerated on the cathode (Eq. (3)).

$$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2(\text{Cathode}) \quad (5)$$

$$Fe \rightarrow Fe^{2+} + 2e^-(\text{Anode}) \quad (6)$$



Experimental conditions: $[RhB]_0 = 0.1$ mM, $[H_2O_2]_0 = 0.5$ mM, $[Fe(\text{II})]_0 = 0.1$ mM, pH = 3.0, and $I = 0.010$A.

Fig.1 RhB degradation at different Fenton processes

3.2 Effect of Fenton reagents

In the electro photo-Fenton process, the effect of $H_2O_2/Fe(\text{II})$ molar ratio was determined in the range from 1 : 10 (5 : 50) to 10 : 1 with $[H_2O_2]_0$ fixed at 0.5 mM and 1.0 mM, respectively. The results were shown in Fig.2a and Fig.2b. The RhB showed a two-stage decay with a rapid degradation in the first several minutes, and then a retarded slow second reaction. The rapid first-stage was caused by the abundant amount of Fenton reagents, contributing to the fast yield of hydroxyl radical (Eq. (1)). The slow second-stage resulted from its lower concentration of hydroxyl radicals because of the fast depleting of Fenton reagents Fe(II) and H_2O_2, and the slow regeneration rate of Fe(II) and H_2O_2 (Qiang et al, 2003). Besides, after the first stage, the RhB molecule is almost completely degraded, the byproducts should be the competitor for radicals and retard the RhB decay in the second stage (Chen and Chu, 2014).

Fig.3 summarized the alternation trends of remaining RhB as $H_2O_2/Fe(\text{II})$ molar ratio varied. It can be found that as the $[H_2O_2]_0$ fixed at 0.5 mM or 1.0 mM, the decay efficiency were both increased with the decrement of Fe(II). The optimum RhB decay

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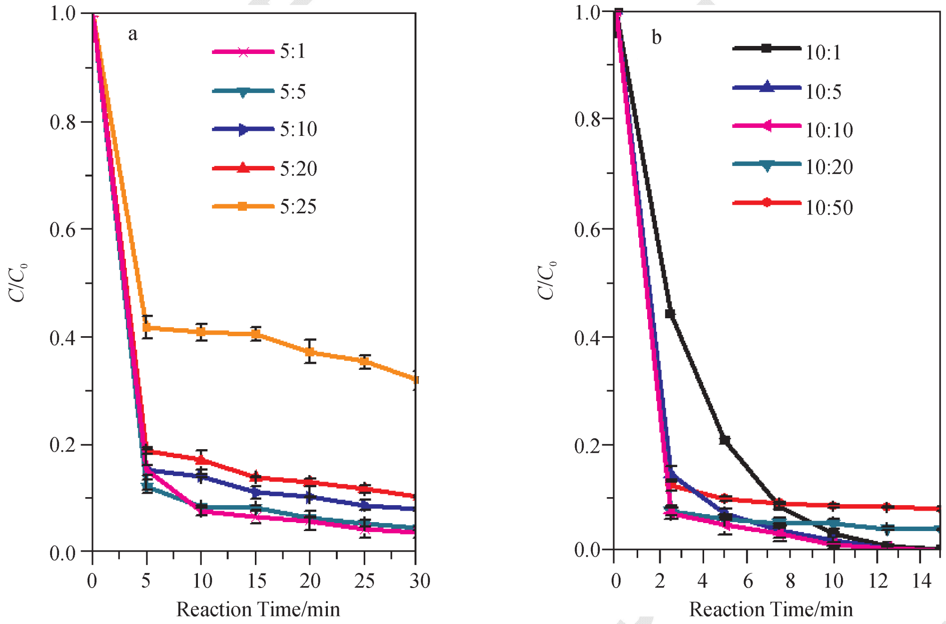
was achieved at H_2O_2/Fe(Ⅱ) molar ratio = 1, after which the decay performance levelled off. On the other hand, the increment of $[H_2O_2]_0$ always brought the performance improvement as shown in Fig.3. In Masomboon and coworker's study (Masomboon et al, 2010), both the increment of Fe(Ⅱ) dosage and $[H_2O_2]_0$ benefitted the performance of electro photo-Fenton process. Differently, in this study, the decreament of Fe(Ⅱ) dosage improved the decay performance. This property is a great advantage for the practical application of Fenton process, because the generation of less ferric hydroxide sludge would reduce the workload for additional separation and disposal (Chou et al, 1999).



Experimental conditions: $[RhB]_0$=0.1 mM, pH=3.0, and I=0.010A.

Fig.2 Effect of molar ratios of H_2O_2/Fe(Ⅱ) at (a) $[H_2O_2]_0$=0.5 mM, and (b) $[H_2O_2]_0$=1.0 mM

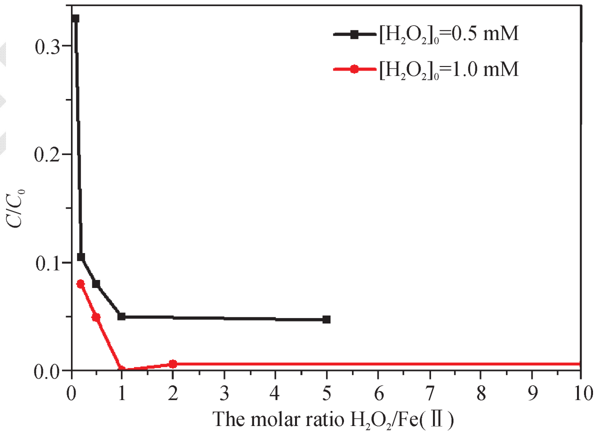


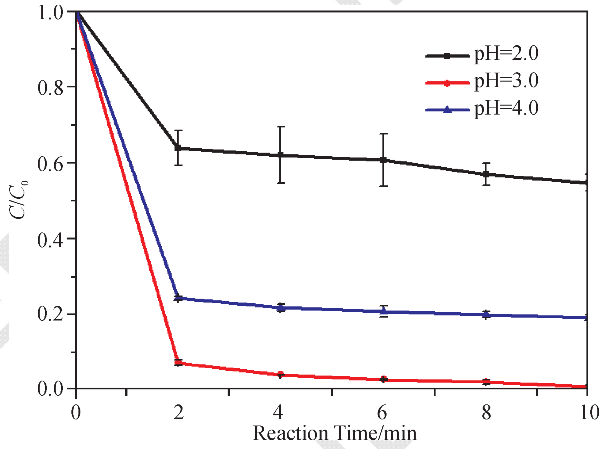
Fig.3 The remaining RhB at 15 min reaction as a function of H_2O_2/Fe(Ⅱ) molar ratio

3.3 Effect of pH level

The influence of solution pH on RhB degradation was examined with the pH value in the range from 2.0 to 4.0. The results were depicted in Fig.4. The highest RhB degradation efficacy was reached at an initial pH value around 3.0. The degradation performances of above or below this pH value were worse. In an aqueous solution, the generated Fe(Ⅲ) underwent spontaneous-hydrolysis with water to form four species Fe(Ⅲ)-hydroxo complexes of $Fe^{Ⅲ}OH^{2+}$, $Fe^{Ⅲ}(OH)_2^{+}$, $Fe^{Ⅲ}_2(OH)_2^{4+}$, and $Fe^{Ⅲ}(OH)_3^{0}$ (Martin et al, 1998). At the investigated pH level of 3.0, the predominant specie is the monohydroxy complex, $Fe^{Ⅲ}OH^{2+}$, which is the most photosensitive species of the four (Flynn, 1984). Such a complex is capable of producing hydroxyl radicals directly through photo-sensitization reaction, as well as the re-generation of ferrous reagent (Eq. (7)).

$$\mathrm{Fe^{III}OH^{2+}} + h\nu \rightarrow \mathrm{Fe^{2+}} + \cdot\mathrm{OH} \qquad (7)$$

At pH 2.0 and 4.0, the photolysis of $\mathrm{Fe^{III}OH^{2+}}$ in Eq. (7) was restrained because the lower amount of $\mathrm{Fe^{III}OH^{2+}}$. Besides, the precipitant of $\mathrm{Fe^{III}(OH)_3^0}$ is formed at pH 4.0 (Ensing et al, 2003), which is known as an adsorbent for organic pollutant (Peng et al, 2006). The RhB could be adsorbed on the $\mathrm{Fe^{III}(OH)_3^0}$ and precipited from the aqueous solution. Therefore, the RhB removal efficacy at pH 4.0 is much better than 2.0.



Experimental conditions: $[\mathrm{RhB}]_0 = 0.1$ mM, $[\mathrm{H_2O_2}]_0 = 1.0$ mM, $[\mathrm{Fe(II)}]_0 = 1.0$ mM, and $I = 0.010$A.

Fig.4　Effect of initial solution pH

3.4　Effect of electric current

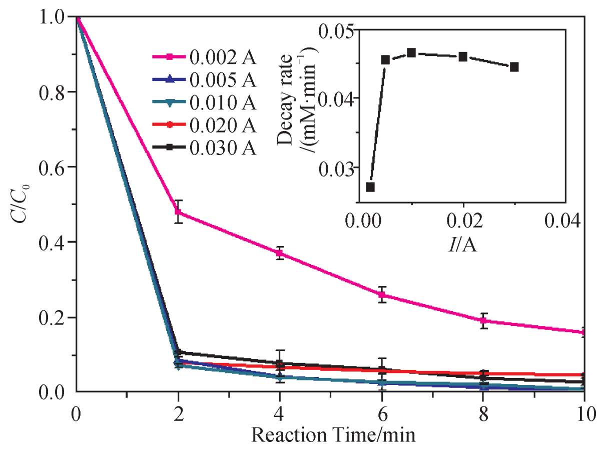
The influence of the electric current (I) on the RhB degradation was studied in the range from 0.002 A to 0.030 A at current-controlled conditions. Fig.5 showed that the electric current presented a certain effect on the RhB decay. The insert of Fig.5 depicted the variation of the decay rate in the first two minutes with I changed, where a significant jump was observed when I increased from 0.002 A to 0.005 A. However, the decay rate leveled off as the electric current further increased. The increment of decay rate at higher current (from 0.002 A to 0.005 A) was likely attributed to the faster release of $\mathrm{H_2O_2}$ on the cathode (see Eq. (4)). When the current further increased, excess Fe(Ⅱ) produced nearby the anode via Eq. (6) and the cathode through Eq. (3), which may become ahydroxyl radical scavenger as shown in Eq. (8) (Buxton et al, 1988).

$$\mathrm{Fe^{2+}} + \mathrm{OH}\cdot \rightarrow \mathrm{Fe^{3+}} + \mathrm{OH^-} \qquad (8)$$

The energy consumption for the photoelectro-Fenton process was also examined. The energy consumpiton (E, $\mathrm{Wh \cdot m^{-3}}$) was calculated via the following Eq. (9),

$$E = (U \times I \times t)/V \qquad (9)$$

where, U is the voltage measured during the reaction (volt), I is the electric current (A), t is the electrolysis time (h), and V is the volume of reaction solution ($\mathrm{m^3}$). The energy consumptions for 90% RhB removal at different electric current were listed in Tab.1. It deserved to note that both the lowest energy consumption and the shortest reaction time were achieved at $I = 0.005$ A. Therefore, the electric current of 0.005 A is the optimal condition in consideration of the RhB decay efficiency, hydraulic retention time and energy consumption.



The insert shows the variation of the decay rate in the first two minutes. Experimental conditions: $[\mathrm{RhB}]_0 = 0.1$ mM, $[\mathrm{H_2O_2}]_0 = 1.0$ mM, $[\mathrm{Fe(II)}]_0 = 1.0$ mM, and pH = 3.0.

Fig.5　Effect of electric current

Tab.1　The energy consumption for 90% of 0.1 mM RhB degradation at different electric current

Electric current /A	Hydraulic retention time/min	Energy consumption /($\mathrm{Wh \cdot m^{-3}}$)
0.002	7.3	1.3
0.005	1.9	1.1
0.010	1.9	2.5
0.020	1.9	7.0
0.030	2.5	15.0

4　Conclusions

The degradation of RhB was studied by using various Fenton reactions (i.e. sole-Fenton, photo-

Fenton and electric photo-Fenton). The electric photo-Fenton with a sacrificed anode showed the optimum performance for RhB degradation. The degradation curve can be defined as a two-stage reaction comprised of a rapid first stage and a retarded second stage. The effect of various parameters such as the Fenton reagent $H_2O_2/Fe(Ⅱ)$ molar ratio, initial solution pH value, and electric current was further investigated and optimized. The optimal molar ratio of $H_2O_2/Fe(Ⅱ)$ was 1 : 1 where the higher $[H_2O_2]_0$ could benefit the RhB decay. The optimal pH for RhB degradation was determined at pH 3.0 in the solution. Moreover, the electric current had a great effect on the RhB degradation process. In consideration of both the RhB decay rate and energy consumption, the optimal electric current is examined as 0.005 A in our electric photo-Fenton process.

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