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Treatment of the Acrylic Fibre Wastewater by Fenton Process

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ABSTRACT

The acrylic fibre is one of the major synthetic fibres commonly used in the mass production of clothing. However, the effluents of the acrylic fibre manufacturing contain amounts of organic and inorganic contaminants. Some of these pollutants are toxic and bio-refractory, which may cause serious environmental impacts if they are discharged directly into receiving waters without appropriate treatment. In this study, the Fenton process was used for the treatment of acrylic fibre wastewater. The operational parameters were evaluated according to the single factor experiments. The results showed that the H_2O_2 dosage, Fe^{2+} dosage, pH value and reaction time had an important role on the effect of treatment of the acrylic fibre wastewater. The most significant interactive influence of Fe^{2+} dosage and pH value was observed. Under the following reaction conditions, such as H_2O_2 dosage of 80 mmol/L, Fe^{2+} dosage of 25 mmol/L, initial pH value of 3 and a reaction time of 110 min, the COD removal efficiency of the acrylic fibre wastewater was 45.2%.

INTRODUCTION

Over the past decades, the acrylic fibre industry has rapidly progressed in many developing countries. The products of this technology are welcomed because of their high quality and versatility (Jiang et al. 2010). The acrylic fibre industry has continuously developed with the growth of the economy, accomplishing technological development for synthesizing and processing several highly valuable polymer materials, such as those used for electrical, electronic and biomedical devices. The acrylic fibre is one of the major synthetic fibres commonly used in the mass production of clothing. However, the effluents of the acrylic fibre manufacturing contain amounts of organic and inorganic contaminants (Li et al. 2012). Some of these pollutants are toxic and bio-refractory, which may cause serious environmental impacts if they are discharged directly into receiving waters without appropriate treatment. Advanced oxidation processes based on the generation of hydroxyl radicals are widely used in the treatment (Neyens & Haeyens 2003). Among the advanced oxidation processes, Fenton process is particularly attractive because of its simplicity and high removal efficiency of recalcitrant pollutants (Bosco et al. 2006). The initially biorefractory compounds can be mineralized or converted to more readily biodegradable intermediates, which can be removed by further biological treatment. Besides, since both ferrous and ferric ions are coagulants, the Fenton process can have the dual functions of oxidation and coagulation in the treatment process (Panizza & Cerisola 2009). In recent years, Fenton oxidation has been extensively studied for the treatment of a variety of toxic and biorefractory wastewaters, such as chemical manufacturing wastewater, pharmaceutical wastewater, textile dyeing wastewater, landfill leachate, and so on. In terms of acrylic fibre wastewater, due to the high organic load, toxicity and presence of biorefractory compounds (Dükkanct et al. 2010, Jagadevan et al. 2011, Rusevova et al. 2012).

The typical Fenton reagents include ferrous ions and hydrogen peroxide. In acidic conditions, strong oxidative ·OH is generated in the catalytic oxidation reaction (1) of H_2O_2 with Fe^{2+}. Apart from the main reaction, a series of chain reactions (2)-(5) involving Fe^{2+}, ferric ion, H_2O_2, superoxide, and ·OH are also possible. In these reactions, bio-refractory or toxic organic compounds can be oxidized to biodegradable small molecules or carbon dioxide and water (Zhao et al. 2012).

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

$$Fe^{2+} + \cdot OH \rightarrow Fe^{3+} + OH^- \quad ...(2)$$

$$H_2O_2 + \cdot OH \rightarrow HO_2^{\cdot} + H_2O \quad ...(3)$$

$$Fe^{2+} + HO_2^{\cdot} \rightarrow Fe^{3+} + HO_2^- \quad ...(4)$$

$$Fe^{3+} + HO_2^{\cdot} \rightarrow Fe^{2+} + O_2 + H^+ \quad ...(5)$$

According to the existing form of catalyst, the Fenton reaction can be divided into two types, such as classical homogeneous Fenton oxidation and novel heterogeneous Fenton oxidation (Tang & Huang 1996, Lucas & Peres 2009).

In this study, the Fenton process was used for the treatment of acrylic fibre wastewater. The effects of the operational parameters, such as H_2O_2 dosage, Fe^{2+} dosage, pH value and reaction time, have been discussed in detail.

MATERIALS AND METHODS

Materials: The acrylic fibre wastewater used in this test was collected from the manufacturing department of a fibre synthesis plant in Shaoxing, China. The characteristics of the acrylic fibre wastewater are listed in Table 1.

Experimental methods: The experiments were conducted in 500 mL batch reactors at room temperature. The 300 mL of acrylic fibre wastewater was added into the batch reactors. The pH value was adjusted with (1+1) H_2SO_4 solution. A weighed amount of $FeSO_4 \cdot 7H_2O$ was dissolved in the acrylic fibre wastewater. The tests were then initiated by the addition of H_2O_2 solution (30%, w/w). The reaction was terminated by adjusting the pH value in the solution to 10.0, which instantaneously consumed the remaining H_2O_2. The solution was left undisturbed for half an hour to settle out. The supernatant was withdrawn and filtered for analyses.

Analytical methods: The value of pH was measured by using a digital pH meter according to APHA standard method. COD was measured by model MS-3 microwave digestion system.

Statistical analyses of data: All experiments were repeated in duplicate and the data of results were the mean and the standard deviation (SD). The value of the SD was calculated by Excel Software. All error estimates given in the text and error bars in figures are the standard deviation of the means (mean±SD). All statistical significance were noted at α=0.05 unless otherwise noted.

RESULTS AND DISCUSSION

Effect of H_2O_2 dosage: During the Fenton process, hydrogen peroxide is the most important parameter because of its source of ·OH and the main cost for scale up application. So, it plays a very important role during the reaction process. The dosage of H_2O_2 has an important effect on the removal efficiency of the acrylic fibre wastewater and decrease of treatment cost. The experiments were carried out under the Fe^{2+} dosage of 20.0 mmol/L, the initial pH value in solution of 4.0, the reaction time of 120 min and the dosage ranged from 30.0 to 180.0 mmol/L. The effects of H_2O_2 dosage on the COD removal efficiency of the acrylic fibre wastewater were shown in Fig. 1. From Fig. 1, when the H_2O_2 dosage is below 90.0 mmol/L, the COD removal efficiency increased sharply with the increasing of H_2O_2 dosage. When the H_2O_2 dosage is above the 90.0 mmol/L, the COD removal efficiency increased slowly with the increasing of H_2O_2 dosage.

Table 1: The characteristics of the acrylic fiber wastewater.

Parameter	pH	COD (mg/L)	BOD_5 (mg/L)	TN (mg/L)	NH_4^+-N (mg/L)
Value	6.4	1046	398	155	20.8

Effect of the Fe^{2+} dosage: Ferrous is one of the main affective factor in Fenton reaction. It can decompose H_2O_2 to generate ·OH. It is well known that higher hydrogen peroxide to substrate ratios result in more extensive substrate degradation, while higher concentrations of iron ions yielded faster rates (Lopez et al. 2004). The experiments were tested under the H_2O_2 dosage of 90.0 mmol/L, the initial pH value in solution of 4.0, the reaction time of 120 min and the Fe^{2+} dosage ranged from 5.0 to 30.0 mmol/L. The effects of Fe^{2+} dosage on the COD removal efficiency of the acrylic fibre wastewater are shown in Fig. 2. As seen from Fig. 2, the COD removal efficiency of the acrylic fibre wastewater increased when the Fe^{2+} dosage increased. In some literature, the same experiment results were reported (Wei et al. 2013). The experiment results indicated that more Fe^{2+} dosage did not mean more oxidation removal because the use of a much higher concentration of ferrous could lead to the self-scavenging effect of ·OH radical and induce the decrease in degradation efficiency of pollutants (Li et al. 2011).

Effect of the initial pH value in solution: The value of pH in solution is one of the important factors. The pH in solution has a decisive effect on the oxidation potential of ·OH because of the reciprocal relationship between the oxidation potential and the pH value. The tests about the effect of the initial pH value in solution should be investigated. The experiments were tested under the H_2O_2 dosage of 90.0 mmol/L, the Fe^{2+} dosage of 20.0 mmol/L, the reaction time of 120 min and the initial pH value in the solution ranged from 2.0 to 6.0. The effects of initial pH value in the solution on the COD removal efficiency of the acrylic fibre wastewater are shown in Fig. 3.

As shown from Fig. 3, the pH value in solution had an important effect on the COD removal efficiency of the acrylic fibre wastewater. When the initial pH value in solution was 4.0, the COD removal efficiency reached optimum. At extremely low pH value, oxidation removal decreased sharply, principally due to the formation of complex species $[Fe(H_2O)_6]^{2+}$, which reacted slower with peroxide when compared to that of $[Fe(OH)(H_2O)_5]^{2+}$ (Gallard et al. 1998). On the other hand, oxidation removal rapidly decreased with the increasing initial pH in solution above 4.0. The reasons for this inhibition might be explained not only by the decomposition of hydrogen peroxide, but also by the deactivation of a ferrous catalyst with the formation

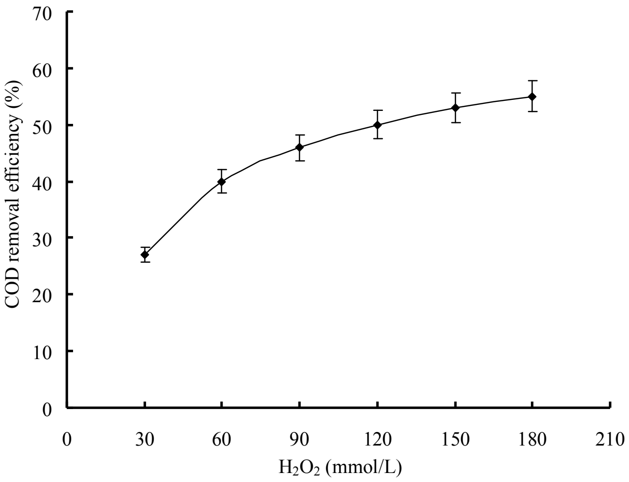

Fig.1: The effect of H_2O_2 dosage on COD removal efficiency.

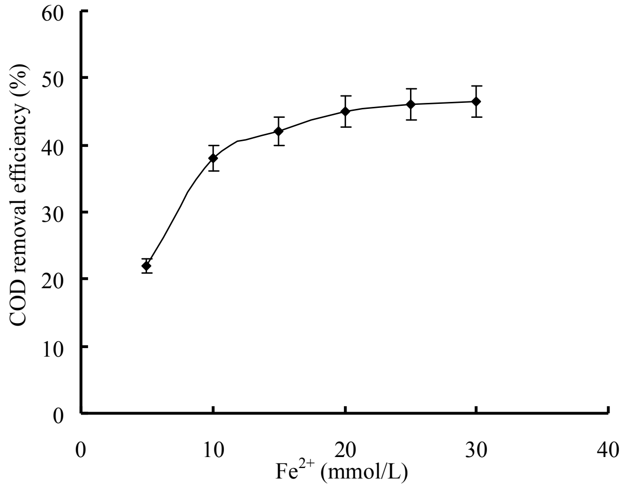

Fig. 2: The effect of Fe^{2+} dosage on the COD removal efficiency.

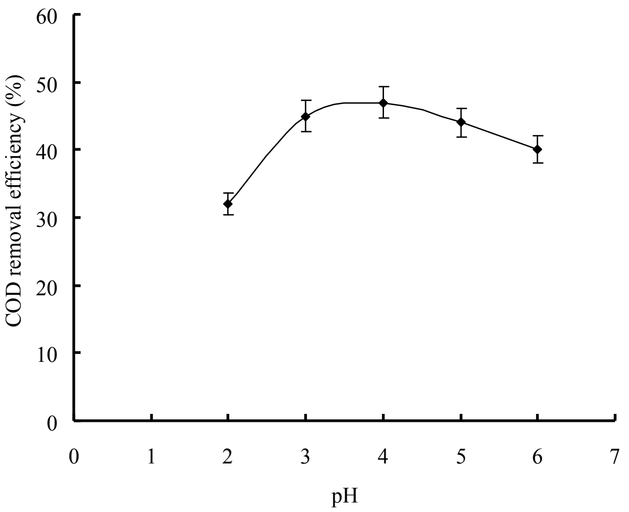

Fig. 3: The effect of pH value in solution on the COD removal efficiency.

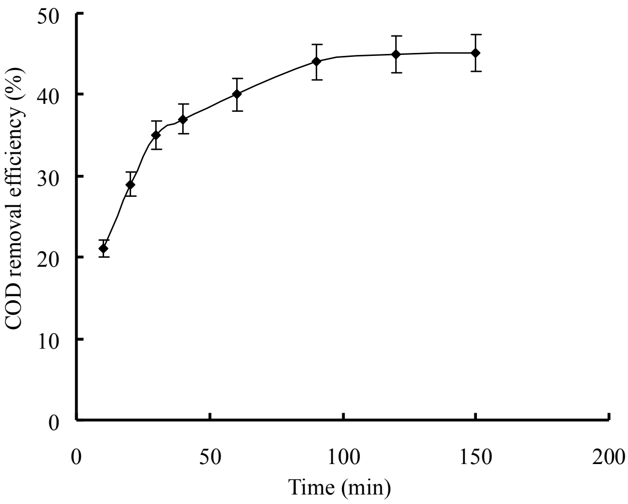

Fig. 4: The effect of reaction time on the COD removal efficiency.

of ferric hydroxide complexes leading to a reduction of ·OH radical.

Effect of the reaction time: Reaction time is a key parameter in Fenton process, because not only treatment performance, but also reactor volume is associated with it. The experiments were tested under the following conditions, such as the H_2O_2 dosage of 90.0 mmol/L, the Fe^{2+} dosage of 20.0 mmol/L and the initial pH value in solution of 4.0. The reaction time ranged from 5 to 150 min. The effects of initial pH value in solution on the COD removal efficiency of the acrylic fibre wastewater are shown in Fig. 4.

As seen from Fig. 4, the reaction time had an important effect on the COD removal efficiency of the acrylic fibre wastewater during the Fenton processes. The COD removal efficiency of the acrylic fibre wastewater increased rapidly in the first 30 min. Then, the COD removal efficiency of the acrylic fibre wastewater increased slightly. No further COD removal efficiency of the acrylic fibre wastewater increase was observed with further prolonged reaction time.

CONCLUSION

In this work, the Fenton process was used for the treatment of acrylic fibre wastewater. The operational parameters were evaluated according to the single factor experiments. The results showed that the H_2O_2 dosage, Fe^{2+} dosage, pH value and reaction time had an important role on the effect of treatment of the acrylic fibre wastewater. The most significant interactive influence of Fe^{2+} dosage and pH value was observed. Under the following reaction conditions, such as H_2O_2 dosage of 80 mmol/L, Fe^{2+} dosage of 25 mmol/L, initial pH value of 3 and a reaction time of 110 min, the COD removal efficiency of the acrylic fibre wastewater was 45.2%.

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