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Treatment of Dye C.I. Reactive Red 15 in Aqueous Solution Using the Activated Carbon Supported Zero Valent Iron

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ABSTRACT

In recent years, the use of zero valent iron for the treatment of toxic chemicals in water has received wide attention. The zero valent iron particles with large surface area per unit mass have been found to be a highly efficient reducing agent capable of remediate contaminated land, surface and groundwater. In this paper, the treatment of dye C.I. Reactive Red 15 in aqueous solution by using the activated carbon supported zero valent iron was tested. The effect of parameters such as pH of solution, the reaction time, the initial concentration of the dye in aqueous solution and the activated carbon supported zero valent iron dosage on the treatment was also investigated. The results showed that all the parameters have a significant effect on the degradation rate of the dye. The treatment method of the activated carbon supported zero valent iron was also proved to be a universal and efficient reductant for rapid degradation of the dye C.I. Reactive Red 15 in aqueous solution.

INTRODUCTION

Dyes are widely used in the textile and dyestuff industries. Effluents from these industrial facilities are typically of high organic contents and colour strength. Thus, textile wastewater must be treated before discharge in order to minimize the threat to the environment (Mielczarski et al. 2005, Ren et al. 2006). The strong colour in wastewater can decrease the transparency of water and influence photosynthetic activity (Choe et al. 2001). They would also hinder the activities of submerged organisms. Moreover, the dye wastewater is recalcitrant, non-biodegradable and persistent (Gao et al. 2010). Various treatment methods, such as coagulation and flocculation, adsorption and filtration have been investigated to remove dye from wastewater (Gomes et al. 2007, Fanchiang & Tseng 2009, Resales et al. 2009). These high cost processes do not destroy dye molecules, but only transfer them from one phase to another. The most extensively used ways to treat dye wastewater is biological (Arsan et al. 2008, Pu et al. 2010). This method is a relatively inexpensive way to remove dyes (Guimarães et al. 2012). However, the disadvantage of this method is related to its long treatment period with a rather limited success as they cannot achieve destructive degradation due to the fact that dye wastewaters are intentionally designed to resist biological, photolytic and chemical degradation (Robinson et al. 2001, Chang & Cheng 2006).

In recent years, the use of zero valent iron for the treatment of toxic chemicals in water has received wide attention (Soon & Hameed 2011). The zero valent iron particles with large surface area per unit mass have been found to be highly efficient reducing agent capable of remediating contaminated land, surface and groundwater (Hu et al. 2005, Chen et al. 2012). The advantages of the technology are low cost, low toxicity, small footprint and easy to get (Oh et al. 2004). So, this technology can satisfy the basic requirements for industrial application. Zero valent iron is a strong reducing agent, cheap and easy to produce (Bayer & Finkel 2005). It has also already been proven effective in reducing chlorinated solvents, including chlorinated organics, nitroaromatic compounds, pesticides, nitrate, chlorinated organic compounds and metal ions (Devlin et al. 1998, Kanel et al. 2005, Alkan et al. 2008).

In this paper, the treatment of dye C.I. Reactive Red 15 in aqueous solution by using the activated carbon supported zero valent iron was tested. The effect of several parameters such as pH of solution, the reaction time, the initial concentration of dye C.I. Reactive Red 15 in aqueous solution and the activated carbon supported zero valent iron dosage are discussed in detail.

MATERIALS AND METHODS

Materials

The dye of C.I. Reactive Red 15 was chosen as an object in this experiment. It was purchased from Shanghai Chemical Co. Ltd. in China. Its molecular formula is $C_{25}H_{14}ClN_7Na_4O_{13}S_4$. Its chemical structure is shown in Fig. 1.

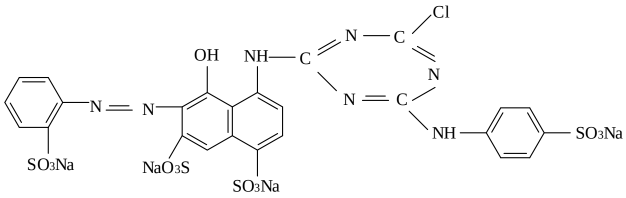

Fig. 1: The chemical structure of the C.I. Reactive Red 15.

The activated carbon supported zero valent iron was prepared using conventional liquid-phase method by the reduction of ferric iron by borohydride. Appropriate weight ratios of the activated carbon and zero valent iron were 1:1. Ferric ion was reduced to elemental iron according to the following reaction:

$$2Fe^{3+}_{(aq)} + 6BH^{-}_{4(aq)} + 18H_2O \rightarrow 2Fe^{0}_{(s)} + 6B(OH)_{3(aq)} + 21H_{2(g)} \quad ...(1)$$

The other chemicals used were of analytical grade.

Experimental Methods

The experiments were carried out by mixing 100 mL of dye solution and the activated carbon supported zero valent iron particles in 250 mL Erlenmeyer flasks. The flasks were shaken at 120 rpm and a constant temperature of 298 K. The value of pH in solution was adjusted with (1+1) HCl or 10% NaOH solutions. When the test was finished, the sample was centrifuged for 10 min at the 2000 rpm and then measured. All the tests were preformed twice, and the average values were used.

Analytical Methods

The value of pH was measured with a pH probe. The concentration of dye C.I. Reactive Red 15 was measured by a UV-1600 spectrophotometer at 510 nm.

The removal rate of dye C.I. Reactive Red 15 was calculated as follows:

$$Q = \frac{C_0 - C_t}{C_0} \times 100\% \quad ...(2)$$

Where, C_0 and C_t (mg/L) are the initial and equilibrium concentrations of the dye in solution respectively. Q is the degradation rate of the dye.

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 standard deviation of means (mean ± SD). All statistical significance was noted at α=0.05 unless otherwise noted.

RESULTS AND DISCUSSION

Effect of Reaction Time on the Degradation Rate

Because the reaction time of degradation rate could help to understand the treatment of dye, the experiments were conducted. The reaction time was 10, 20, 30, 50, 80 and 120 min. The experiments were carried out by mixing 100 mL of 100 mg/L of dye solution and 4 g of the activated carbon supported zero valent iron particles in 250 mL Erlenmeyer flasks. The value of pH in solution was 2. The flasks were shaken at 120 rpm at a constant temperature of 298 K. The results are presented in Fig. 2.

As shown in Fig. 2, it can be concluded that reaction time is an important factor affecting the degradation process of the dye in aqueous solution by the activated carbon supported zero valent iron. At first stage, the degradation increased quickly. The removal efficiency reached 94.86% at 20 min of reaction time. The degradation rate of the dye increased slowly above 20 min of reaction time.

Effect of Dye Concentration on the Degradation Rate

The initial concentration of dye is an important factor for practical application. Thus, it is necessary to study the effect of initial dye concentration on its treatment. The initial dye concentration ranged from 100 mg/L to 500 mg/L. The experiments were done by mixing 100 mL dye solutions and 4 g of the activated carbon supported zero valent iron particles in 250 mL Erlenmeyer flasks. The value of pH in solution is 2. The flasks were shaken at 120 rpm at a constant temperature of 298 K. The reaction time is 120 min. The results are shown in Fig. 3.

As shown in Fig. 3, it can be concluded that the degradation rate of the dye decreased with the increasing of initial concentration.

Effect of pH in Solution on the Degradation Rate

The value of pH in solution is an important parameter affecting dye degradation. The actual dye containing wastewater has a wide range of pH values, so it is necessary to investigate the influence of pH on degradation process. In this experiment, the value of pH in solution was adjusted with (1+1) HCl or 10% NaOH solutions. The value of pH in solution was kept at 2, 4, 6, 8 and 10. The experiments were carried out by mixing 100 mL of 100 mg/L of dye solutions and 4 g of the activated carbon supported zero valent iron particles in 250 mL Erlenmeyer flasks. The flasks were shaken at 120 rpm at a constant temperature of 298 K. The reaction time is 120 min. The results are shown in Fig. 4.

From Fig. 4, it can be seen that the value of pH in solution has an important effect on the degradation rate of the

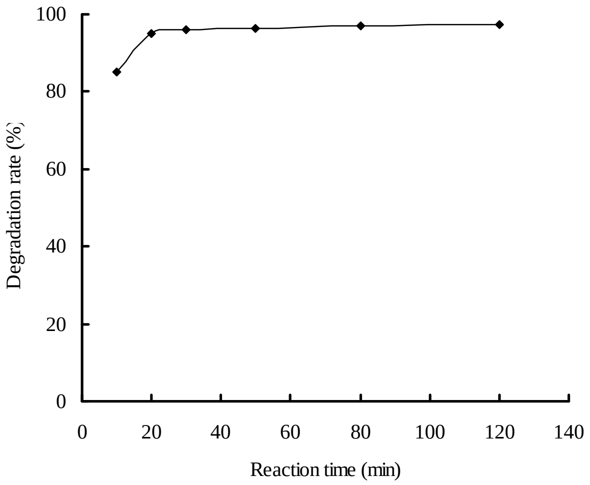

Fig. 2: Effect of reaction time on the degradation rate.

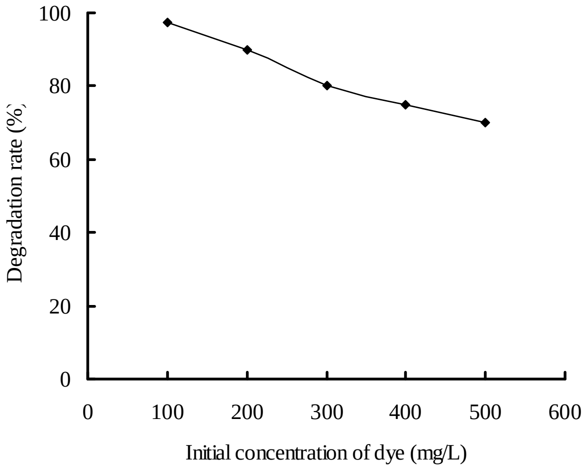

Fig. 3: Effect of dye concentration on the degradation rate.

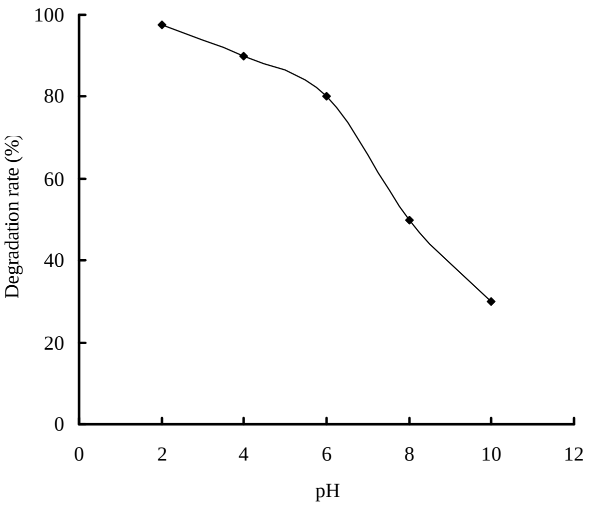

Fig. 4: Effect of pH in solution on the degradation rate.

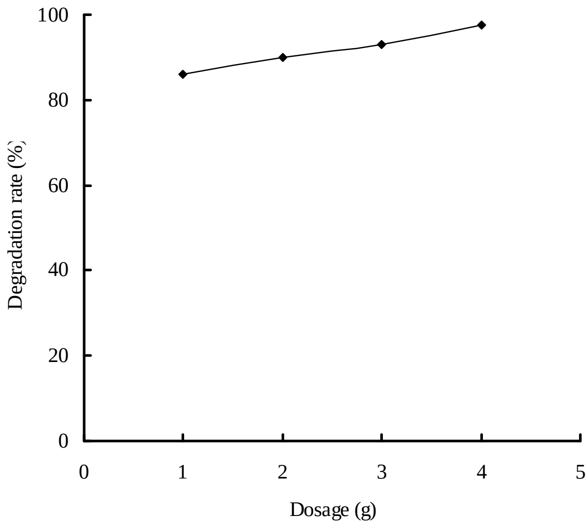

Fig. 5: Effect of the activated carbon supported zero valent iron dosage on the degradation rate.

dye. The degradation rate of the dye decreased with increasing the value of pH in solution. Apparently, the degradation process for the dye was acid driven. The main reason might be the strong reductive reaction and the oxidation of the activated carbon supported zero valent iron. When the effective collision between dye molecular and zero valent iron particles happened, the zero valent iron was combined with H^+ and turned into the transitional product. The iron corrosion in the presence of oxygen can generate hydrogen peroxide, because the reaction process was not protected by nitrogen gas. The hydrogen peroxide could react with the Fe^{2+} and produce strong oxidants such as hydroxyl radical and ferryl. Fe^{2+} ions could easily form ferrous hydroxide precipitates on the surface of the activated carbon particles with pH increasing, covering the reactive sites and holding back the reaction process. This explained why degradation rate was low for dye in high value of pH in solution.

Effect of the Activated Carbon Supported Zero Valent Iron Dosage on the Degradation Rate

In order to investigate the role of the activated carbon supported zero valent iron dosage on the degradation rate of dye C.I. Reactive Red 15, a series of experiments were conducted. The activated carbon supported zero valent iron dosage ranged from 1 g to 4 g. The experiments were performed by mixing 100 mL of 100 mg/L of dye solutions and the activated carbon supported zero valent iron particles in 250 mL Erlenmeyer flasks. The value of pH in solution is 2.

The flasks were shaken at 120 rpm at a constant temperature of 298 K. The reaction time was 120 min. The results are shown in Fig. 5.

From Fig. 5, it can be seen that the dosage of the activated carbon supported zero valent iron has an important role on the degradation rate. The degradation rate of the dye in aqueous solution increased with the increasing of the activated carbon supported zero valent iron. The increased total surface area and the availability of more activated carbon supported zero valent iron sites may be the reason for the rise in degradation rate with increasing dosage.

CONCLUSIONS

The treatment of the dye C.I. Reactive Red 15 in aqueous solution by using the activated carbon supported zero valent iron was tested. The effect of the parameters like pH in solution, the reaction time, the initial concentration of the dye in aqueous solution and the activated carbon supported zero valent iron dosage was investigated. The results showed that the operational parameters had an important effect on the degradation of the dye in aqueous solution. The degradation rate of the dye C.I. Reactive Red 15 in aqueous solution reached 97.5% at pH 2.0, the reaction time of 120 min, the initial dye concentration of 100mg/L, the dosage of 4g, 120 rpm and temperature of 298 K. The activated carbon supported zero valent iron can be applied into the degradation of dye C.I. Reactive Red 15 in aqueous solution.

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