



Efficient removal of dyes using activated carbon fibers coupled with 8-hydroxyquinoline ferric as a reusable Fenton-like catalyst



Lijie Sun, Yuyuan Yao^{*}, Lie Wang, Yajun Mao, Zhenfu Huang, Daichuan Yao, Wangyang Lu, Wenxing Chen

National Engineering Lab of Textile Fiber Materials & Processing Technology (Zhejiang), Zhejiang Sci-Tech University, Hangzhou 310018, PR China

HIGHLIGHTS

- Developing a novel heterogeneous Fenton-like catalyst of QuFe@ACFs.
- Presenting a remarkable pH-tolerant performance at a wide pH range.
- Exhibiting the excellent sustained catalytic ability and reusable capability.

ARTICLE INFO

Article history:

Received 31 August 2013
Received in revised form 1 December 2013
Accepted 3 December 2013
Available online 9 December 2013

Keywords:

Enhanced activity
pH-tolerance
Activated carbon fibers
8-Hydroxyquinoline
Dyes

ABSTRACT

The development of a pH-tolerant Fenton-like catalyst is an active and challenging project in water treatment. In this study, activated carbon fibers (ACFs) supported 8-hydroxyquinoline ferric (QuFe) has been reported as a heterogeneous Fenton-like catalyst (QuFe@ACFs) for efficient removal of dyes. The catalyst possessed remarkable catalytic performance across a wide range of pH values (3–9). Moreover, it also presented excellent sustained catalytic ability and regeneration capability in the experiments, causing reducing secondary contamination. Importantly, with the introduction of ACFs, QuFe@ACFs exhibited a greatly enhanced catalytic activity in comparison with bare QuFe. The hydroxyl radicals ($\cdot\text{OH}$) are confirmed as the active species in QuFe@ACFs/ H_2O_2 system by the use of electron paramagnetic resonance (EPR) and fluorescence spectra. In addition, the central composite design (CCD) matrix and response surface methodology (RSM) were applied in designing the experiments for evaluating the interactive effects of temperature, pH, initial dye concentration and catalyst dosage. This study not only opens a novel avenue for designing pH-tolerant heterogeneous Fenton-like catalyst, but also provides a viable process for efficient elimination of dyes effluent.

© 2013 Elsevier B.V. All rights reserved.

1. Introduction

Fenton process has been considered as one of promising technologies among various advanced oxidation processes (AOPs) due to its simple operation and high catalytic efficiency, where hydroxyl radicals ($\cdot\text{OH}$) are usually main highly reactive oxidizing species generated to destroy recalcitrant organic contaminants completely [1–3]. However, pH is one of critical operating parameters in the traditional Fenton process because these Fenton systems can only work efficiently under highly acidic conditions (pH 2–3.5) [4,5], which means that strong acids have to be added into the wastewater to reach this pH requirement before the application of traditional Fenton reactions in most cases. Moreover, recycling of ferric iron back to ferrous iron, a key step in Fenton processes [6], will be inhibited owing to the formation of ferric

oxides or hydroxides precipitate at higher pH. What is worse, the separation of new pollutants including the superfluous iron ions [7] and stubborn sludge [8] from the reaction media at the end of the reaction increases the costs of the process. Hence, those non-ignorable drawbacks limit further application of the Fenton process in wastewater treatment engineering.

Therefore, the development of new approaches to enable the Fenton reaction to operate at a wider pH range is of great significance. Many researchers have made great efforts to develop heterogeneous Fenton systems using supported Fenton-like catalysts. Numerous support materials (such as nafion [9,10], clay [11,12], graphene [13], zeolite [14,15], and ion-exchange resin [16,17]) have been applied to immobilize Fe ions to prepare heterogeneous Fenton-like catalysts, which can efficiently prevent iron ions leaching and broaden the working pH range to a certain extent. However, these catalysts usually suffer from complex synthetic or applied processes, which will be a bottleneck for their large-scale practical applications [18,19].

^{*} Corresponding author. Tel.: +86 571 86843810; fax: +86 571 86843255.
E-mail address: yyy0571@126.com (Y. Yao).

Consequently, the selection of suitable catalyst support is vital because appropriate supports will bring excellent catalytic performances and practicabilities. In comparison with the aforementioned supports, activated carbon fibers (ACFs) may be a better candidate to construct excellent heterogeneous Fenton catalysts for its several remarkable characteristics: (i) with uniform microporous structure and high adsorption ability, ACFs can enrich the organic substances around the catalysts and promote removal efficiency; (ii) as the felt or cloth form, ACFs are facile to handling into diverse shapes to suit different application; (iii) with the versatility of its texture and surface chemistry, ACFs allow for a wide range of strategies to firmly attach catalytic active species and thus to increase catalyst stability and its possible reuse in the process. Based on above considerations, we have reported that Fe@ACFs could be used as an effective heterogeneous Fenton catalyst for the removal of organic dyes [20]. Although Fe@ACFs achieved some preliminary effects in improvement of catalytic activity, developing a pH-tolerant microenvironment for the heterogeneous Fenton reaction is still a major challenge.

8-Hydroxyquinoline (Qu) has been widely used in dyes and drug manufacturing as an important intermediate with low hazard [21], as well as having attracted much of researchers' attention in light-emitting material field owing to its chelating ability towards various metal ions and high luminescence efficiency [22,23]. However, to the best of our knowledge, the researches on 8-hydroxyquinoline chelating iron ions in water disposal area are almost blank. To explore its potential application, 8-hydroxyquinoline ferric (QuFe) as appropriate and efficient catalytic active sites that were supported onto ACFs (QuFe@ACFs) to eliminate organic dyes based on the following considerations: (i) The tripodal ligand possessing three bidentate 8-hydroxyquinoline subunits is a very strong chelator for ferric ions over a large range of pH from acidic to basic medium [24]; (ii) as a result of π - π dispersive interaction of ACFs for aromatic structure [25,26], QuFe can be anchored onto ACFs firmly; (iii) π - π conjugate interaction exists in benzene rings [27], which may become a convenient and possible pathway for the transfer of the free electrons between ACFs and QuFe.

Herein, QuFe ligands supported on ACFs were employed for the first time as a novel heterogeneous Fenton-like catalyst. The catalytic properties of QuFe@ACFs were evaluated in terms of the removal rates of dyes in the presence of H_2O_2 . The effects of the main variables (pH, initial dye concentration, temperature and catalyst dosage) were investigated by using response surface methodology (RSM), which is an effective instrument [28–30] for optimizing the process when a combination of a few independent variables and their interactions affects requests and responses. Electron paramagnetic resonance (EPR) and fluorescence spectra were performed to detect active species in QuFe@ACFs/ H_2O_2 system. It is anticipated that this investigation will develop the potential of 8-hydroxyquinoline in the environmental remediation engineering.

2. Experiment

2.1. Materials and reagents

ACFs (specific area: 1300 m^2/g) were provided by Jiangsu Sutong Carbon Fiber Co., Ltd. (Jiangsu, China). Ferric chloride hexahydrate ($FeCl_3 \cdot 6H_2O$) and 30% (w/w) hydrogen peroxide (H_2O_2) were purchased from Sinopharm Chemical Reagent Co., Ltd. 8-Hydroxyquinoline and coumarin were from Aladdin. The spin trapping reagent 5,5-dimethyl-pyrroline-oxide (DMPO) was obtained from Tokyo Chemical Industry Co., Ltd. All dyes, including Reactive Brilliant Red X-3B (RR X-3B), Reactive Brilliant Red K-2BP (RR K-2BP), Reactive Brilliant Red M-3BE (RR M-3BE), Reactive Black

KN-B (RB KN-B), Neutral Orange RL (NO RL), Basic Brilliant Green (BG), Acid Orange 7 (AO 7), and Acid Red 1 (AR 1), Rhodamine B (Rh B) are commercial compounds. The molecular structures of these dyes are shown in Fig. S1 (Supplementary material). All chemicals were used as received without further purification. Double distilled water was used throughout this study.

2.2. Preparation of heterogeneous Fenton-like catalyst

1.350 g $FeCl_3 \cdot 6H_2O$ and 2.177 g 8-hydroxyquinoline were dissolved in 50 ml and 250 ml anhydrous ethanol, respectively, and the $FeCl_3 \cdot 6H_2O$ solution was dropwise added into the 8-hydroxyquinoline solution with continuous stirring to obtain mother solution.

2.2.1. Preparation of the QuFe

Adding ammonia into the mother solution and adjusting pH to 6–7 (measured using Mettler-Toledo pH meter, China). Then, the reaction mixture was refluxed for 4 h, and the resulting precipitate was filtrated, washed with ethanol and distilled water for at least three times respectively, dried in vacuum at room temperature to obtain 8-hydroxyquinoline ferric (QuFe).

2.2.2. Preparation of QuFe@ACFs

ACFs (5 g) were immersed in the mother solution controlling pH (3–4) at room temperature for 24 h. Afterwards, the treated ACFs were taken out from the solution, rinsed repeatedly with ethanol and distilled water at least three times to remove free QuFe and other residues, dried in vacuum at room temperature to obtain QuFe@ACFs. The QuFe@ACFs was characterized by field emission scanning electron microscopy (FESEM) and energy dispersive spectrometer (EDS) (Figs. S2 and S3). In addition, calculated according to the ferric content in QuFe@ACFs, the content of QuFe in QuFe@ACFs was 122.6 $\mu mol/g$, as detected by atomic absorption spectrometer (Hitachi 170-70 atomic absorption spectrometer).

2.3. Experimental procedures and analysis

Experimental procedures and analysis experiments on the catalytic oxidation of dyes were carried out in a 40 mL glass bottle, with the reaction temperature constant at 50 °C (or other temperatures) using a constant temperature shaker water bath (DSHZ-300A, Taicang, Jiangsu). In all experiments, a reaction volume of 25 mL was used. The reaction mixture was composed of the dye (50 μM), QuFe@ACFs (6 g/L) or ACFs (6 g/L), and H_2O_2 (40 mM), followed by pH adjustment with a diluted aqueous solution of NaOH or $HClO_4$. The UV-vis absorption and fluorescence spectra were recorded on a Hitachi U-3010 UV-visible spectrophotometer and a PE-LS-45 fluorescence spectrophotometer, respectively. EPR spectra of radicals trapped by DMPO were examined by using a Bruker A300 spectrometer at ambient temperature. The settings for the EPR spectrometer were as follows: center field, 3520 G; sweep width, 100 G; microwave frequency, 9.77 GHz; modulation frequency, 100 kHz; power, 12.72 mW.

2.4. Experimental response surface methodology (RSM)

RSM is an efficient technique for the optimization of a multivariable system. In this study, optimal removal of RR X-3B was obtained by RSM using Design Expert 8.0. The four independent parameters chosen in this study were pH, temperature, catalyst dosage, initial dye concentration, with the removal rate of RR X-3B as response. The ranges and levels of independent variables are given in Table 1. Central composite design (CCD), which is the most frequently used form of RSM, was employed to evaluate the influence of the four independent variables in 30 sets of exper-

Download English Version:

<https://daneshyari.com/en/article/147720>

Download Persian Version:

<https://daneshyari.com/article/147720>

[Daneshyari.com](https://daneshyari.com)