

# Effective removal of phenol by using activated carbon supported iron prepared under microwave irradiation as a reusable heterogeneous Fenton-like catalyst



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## ABSTRACT

In this paper, an iron-based catalyst supported on activated carbon was prepared in a microwave furnace as a heterogeneous Fenton-like catalyst for phenol degradation. The performance of Fe/AC in the process of catalytic wet peroxide oxidation was evaluated. The adsorption of the catalyst and its support in the presence of hydrogen peroxide and the contribution of a homogeneous reaction in this system was reassessed. We found the iron-based catalyst supported on activated carbon showed a high rate of activity in the process of phenol removal. Phenol and TOC removal were 91% and 48% respectively, in optimal conditions. In the case of the Fe/AC/H<sub>2</sub>O<sub>2</sub> system in the conditions we investigated, the catalyst adsorption capacity decreased due to the introduction of oxygen-containing carbon surface groups in the presence of hydrogen peroxide. The relatively high stability of this catalyst was observed. Only a 10% decrease of catalytic activity was reached after the fourth cycling run. The iron leaching which led to the homogeneous reaction can be ignored because only 1.6% of phenol removal was achieved in the homogeneous condition.

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## 1. Introduction

Fenton reaction as an advanced oxidation process (AOP) is considered a promising technology for treating wastewaters containing organic compounds [1,2]. The highly active intermediate species produced in this process can decompose the organic contaminants into harmless compounds like water and CO<sub>2</sub>, and thus remove pollutants from water [3]. The most important disadvantage of classic Fenton reagents is the high concentration of iron (50–80 ppm) in solution which produces superfluous iron ions and stubborn sludge after reaction. Additional separation processes are needed to remove the new pollutions from the wastewater [4,5].

In order to overcome these problems, a heterogeneous Fenton system which loads iron on a solid support has been developed and is widely researched [2,4,6,7]. Numerous support materials such as zeolite [8,9], ion-exchange resin [10,11], clay [12], nafion [13] and

carbon materials [14–16] have been applied to fix iron ions to prepare heterogeneous Fenton-like reagents. These heterogeneous Fenton systems could offer some advantages over the classic homogeneous Fenton reagents, such as no sludge formation, the possibility of recycling the iron promoter and operation at near neutral pH. Compared with the aforementioned supports, activated carbon may be a better candidate, as it can offer the most desirable properties required for catalytic supports such as extended surface area, well-developed porosity and inexpensive and variable surface chemistry characters [17].

In addition, unlike the other carbon materials (activated carbon fibers and multi-walled carbon nanotube) with uniform pore size, the pore size of activated carbon is dispersed, making it more suitable for wastewater because the contaminants in wastewater vary. Different contaminants with various molecular dimensions can be absorbed by different pore sizes in activated carbon. Besides the advantages mentioned above, it is important that the activated carbon itself shows some catalytic effect on the decomposition of hydrogen peroxide to hydroxyl radicals under neutral conditions [18,19]. Considering the unique properties of activated carbon mentioned above, research on Fenton-like systems which support

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activated carbon has been widely studied and discussed [14,20–23], and some synergistic effects have been observed among the activated carbon, iron ions and oxidants [24–26].

When reviewing the recent literature, it became apparent that the present study on this complicated system has room for improvement. First, based on the assumption that the adsorption capacity of carbon is similar whether an oxidant is present or not, the adsorption data of activated carbon is derived in the absence of an oxidant. However in practice, the adsorption capacity of carbon would change in the presence of oxidant, which may modify the porosity and surface chemistry [27,28]. Second, a parallel experiment was conducted to evaluate the contribution of the homogeneous process (iron-leaching). The iron concentration in this experiment did not correspond to the actual concentration because the iron concentration in solution increased with the reaction time [29]. Third, in most cases, the conventional electric calcination method for the preparation of a supported-catalyst is adopted, unavoidable temperature gradients occur, and this gradient of temperature would adversely affect the catalyst distribution on the support surface. However, microwave heating, which uses electromagnetic energy, has some unique characteristics to help avoid the existence of temperature gradient because microwave can penetrate materials, and heat can be generated throughout the volume of the materials, in addition, the transfer of energy in microwave furnace does not rely on diffusion heat from the surfaces [23].

In this work, iron ions supported on activated carbon calcined in a microwave furnace were prepared and used as a heterogeneous Fenton catalyst. An improved experimental method was applied to evaluate the adsorption and catalysis of this system in the process of phenol degradation.

## 2. Materials and methods

### 2.1. Materials

Activated carbon with a particle size between 60 and 80 mesh was supplied by Laiyang Fine Chemical Factory, China. Phenol, iron nitrate nonohydrate ( $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ ), hydrochloric acid, sulfuric acid (98%), sodium hydroxide, sodium sulfide and hydrogen peroxide (30.0w/v) were purchased from Sinopharm Chemical Reagent Co Ltd, China. All chemicals mentioned in this work are of analytical grade.

### 2.2. Preparation of Fe/AC catalyst

Incipient wet impregnation in aqueous solution was used in this study to prepare activated carbon supported iron catalyst with  $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  as the catalyst precursor. The theoretical ratio of iron to activated carbon is 10% by weight. About 90 g of activated carbon sieved to 40–60 mesh was impregnated with 0.89 mol/L  $\text{Fe}(\text{NO}_3)_3$  solution after impregnation, the slurry was stirred for 24 h, and then dried in air at 105 °C to reach a constant weight, and finally treated for 2 h at 400 °C under He flow in a microwave furnace for the decomposition of iron precursor.

### 2.3. Catalyst characterization

The catalysts were characterized by physical adsorption of  $\text{N}_2$  at  $-196^\circ\text{C}$  in a micromeritics ASAP 2020 apparatus. Before  $\text{N}_2$  adsorption, the samples were degassed at 250 °C for 12 h. The surface area was calculated from  $\text{N}_2$  adsorption isotherms using the BET method. The micropore volumes of the catalysts were calculated using the Dubinin–Radushkevich (DR) method. Fourier Transform Infrared (FTIR) spectra were acquired using a Bruker TENSOR27 FTIR spectrophotometer in the 4000–400  $\text{cm}^{-1}$  spectral

range to characterize the functional groups on the surface of the samples. Iron concentration of the catalyst was determined by inductively coupled plasma–optical emission spectroscopy (ICP–OES).

The chemical characteristics of catalysts were characterized by temperature programmed desorption (TPD). The amounts of CO and  $\text{CO}_2$  that evolved after heat treatment in He flow (60  $\text{cm}^3/\text{min}$ ) to 1000 °C were analyzed by using a Prisma mass spectrometer (Thermocube, Balzers) and the oxygen content was calculated from the CO and  $\text{CO}_2$  released in the TPD experiments.

### 2.4. Catalyst experiments procedure

The catalytic tests were performed in a 1000 ml slurry batch reactor with a rotation speed of 100 rpm. 500 ml of phenol aqueous solution and 0.1 g catalyst (Iron concentration: 8.65%) were used in all experiments at 30 °C. The catalysts were added into the phenol solution with the desired initial concentrations after adjusting the solution to a pH of 3 with sulfuric acid (0.1 mol/L) and sodium hydroxide (0.1 mol/L) solution. The reaction was triggered by adding different concentrations of  $\text{H}_2\text{O}_2$  to the solution. After the reaction time, sodium sulfide was added in a stoichiometric ratio of 1.2:1 ( $\text{Na}_2\text{SO}_3 : \text{H}_2\text{O}_2$ ) to stop the reaction.

Phenol adsorption tests were carried out in the absence of  $\text{H}_2\text{O}_2$ , with the other conditions identical to the reaction tests. Before the adsorption tests, the catalysts were soaked in  $\text{H}_2\text{O}_2$  solution (0.25 g/L) for 1 h at room temperature to discriminate the influence of oxidants on activated carbon support. The time of zero adsorption was coincident with the addition of Fe/AC.

In order to evaluate the homogenous effect of any leached iron contributing to the process, an additional homogenous test was carried out with the iron concentration the same as that found in the solution at a different time. To evaluate reusability, the catalyst was recovered by filtering the solution after the cycle reaction and reused under the same operating conditions.

### 2.5. Analytical techniques

The iron concentration in solution was measured by analyzing the samples at different degradation times using a UV/Vis spectrophotometer (model 722) at 510 nm by means of the o-phenantroline method. Phenol concentration was determined following the ASTM standard 4-aminoantipyrine colorimetric

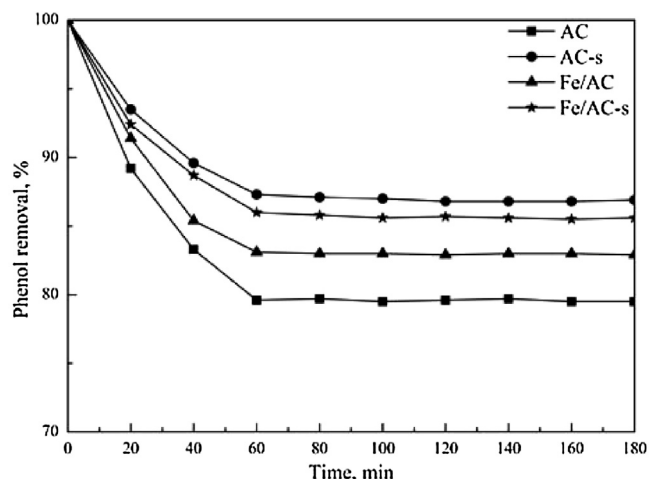


Fig. 1. Phenol adsorption performance for different samples.

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