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Study on Degradation of Benzoic Acid Wastewater by Oxidation Processes of Heterogeneous Fenton

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Abstract

This study used self-made heterogeneous catalyst to investigate the degradation of benzoic acid wastewater, and the optimum conditions were obtained: Adjust the initial pH of the solution to 3, add an appropriate amount of catalyst to 1 g/L, adsorb for 210 min under magnetic stirring, then add 0.5 times the calculated theoretical amount of hydrogen peroxide (ie. 0.5Qth = 6.15 mmol/L). Finally TOC removal rate was reached of 76.76% after 90 minutes of reaction at room temperature.

Keywords

Heterogeneous; Fenton; Benzoic Acid.

1. Introduction

The traditional homogeneous Fenton method has higher requirements for pH in practical application. Adjusting the pH of wastewater will increase the cost of treatment. In addition, the wastewater was treated by this method contains iron ions. To avoid re-contamination of the environment, wastewater must be reprocessed, which is an economic cost unacceptable to sewage treatment plants [1,2]. After scholars' research on Fenton for many years, people have paid more and more attention to the research of heterogeneous systems.

In this study, a 100 mg/L benzoic acid wastewater was prepared for the wastewater from the benzoic acid production process of Tianjin Dongda Chemical Group to simulate the wastewater after the three-stage extraction treatment. In this experiment, the degradation of benzoic acid simulated wastewater was studied using a self-made catalyst. The effects of the length of adsorption time, the moment of hydrogen peroxide addition, the length of the reaction time, the acidity and alkalinity, the amount of hydrogen peroxide added, the amount of catalyst added, and the initial concentration of benzoic acid in the solution on the removal efficiency of degradation. It provides valuable basic data for the development and design of a new process for the extraction of waste water from production heterogeneous Fenton oxidation treatment.

2. Materials and methods

2.1 Materials

Benzoic acid, hydrogen peroxide(30 wt%), sodium hydroxide, sulfuric acid, phosphoric acid, sodium persulfate, hydrochloric acid, hydroxylamine hydrochloride, ammonium acetate, glacial acetic acid, o-phenanthroline, and sodium acetate was purchased from Tianjin. Jiangtian Chemical Technology Co., Ltd. All of the chemicals used in this study were analytical grade.

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2.2 Experimental methods

Take 500mL of benzoic acid simulated wastewater in the reactor, stir the reactor on a constant temperature magnetic stirrer, add a certain amount of catalyst, and stir overnight. After reaching saturation, adjust the solution to the desired pH with dilute H_2SO_4 or NaOH solution. Add a certain amount of H_2O_2 (30 wt%) to start the reaction. Sampling at a prescribed interval, suction filtration with a 0.22 μm filter membrane immediately, adjusting the pH of the filtrate to about 10 to stop the reaction, and taking the test solution for various index analyses.

2.3 Analysis method

The TOC were measured by the GE Sievers InnovOx laboratory total organic carbon (TOC) analyzer. PH were determined by Hanna instruments pH211 precision pH meter.

3. Study on the degradation of benzoic acid by Fe / AC

3.1 Determination of adsorption time and H2O2 dosing time

Experimental conditions: Benzoic acid: 100mg / L; H_2O_2: 12.3mmol / L; catalyst: 1g / L; Ph=3

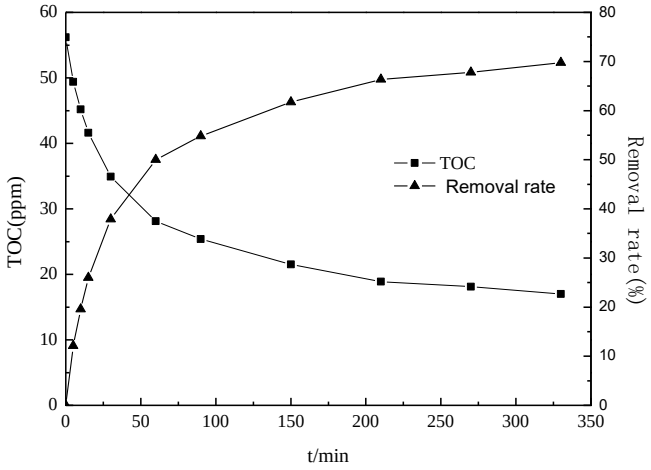

Fig.1 Effect of Fe / AC alone on benzoic acid adsorption

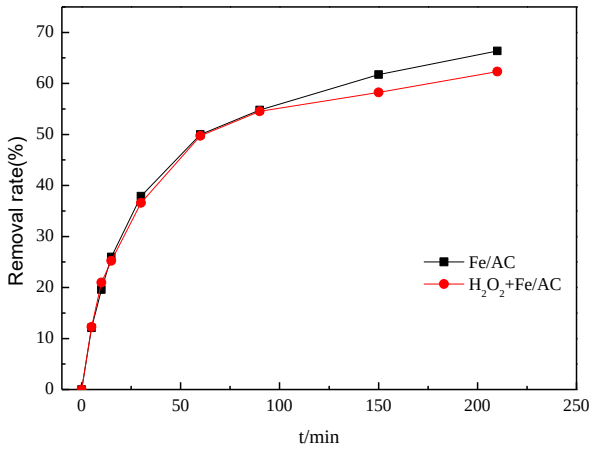

Fig.2 Effect of Fe/AC alone and Fe/AC + H_2O_2 system on benzoic acid removal

It can be seen from Fig. 1 that after adding Fe/AC, the TOC decreases with the adsorption time, and the corresponding removal rate increases with the change of the adsorption time. The adsorption rate of benzoic acid by the catalyst was the largest at first, and gradually decreased with the change of

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adsorption time. When the adsorption time reached 210 min, the adsorption rate of benzoic acid was basically 0, and the adsorption removal rate reached 66.37%. The subsequent change of the adsorption removal rate is small and can be ignored, and the adsorption saturation time of the catalyst is determined to be 210 min.

The time-dependent curve of the removal effect of benzoic acid by Fe/AC and Fe/AC + H_2O_2 systems over time is shown in Fig.2. The removal rates of TOC in solution by the two systems have the same trend over time. Both are gradually increasing. In the first 90 minutes, the two curves basically coincide. This is because during the reaction, the removal of TOC from the solution by the two systems mainly depends on the adsorption of the catalyst, and the catalytic degradation of benzoic acid by the Fe/AC + H_2O_2 system is not obvious on the curve. As the reaction precedes further, the removal rate of TOC in the solution by the Fe/AC system alone is always higher than that of the Fe / AC + H_2O_2 system.

This is because during the reaction, the Fe/AC + the H_2O_2 system continuously catalyzes the degradation of benzoic acid into small molecular substances. When the small molecular substance in the solution reaches a certain concentration, it will form an adsorption competition with the benzoic acid molecules, which makes the catalyst in the Fe / AC + H_2O_2 system adsorb and remove benzoic acid effect weakened. Because the contribution of benzoic acid molecules to TOC is larger than that of small molecules, the final TOC removal rate is reduced. Therefore, it is determined that the H_2O_2 addition time is when the catalyst has been saturated with adsorption.

3.2 Determination of response time

Experimental conditions: Benzoic acid: 100 mg/L; H_2O_2: 12.3 mmol/L; catalyst: 1g/L; pH = 3.The removal effect of TOC by two different systems of AC + H_2O_2 and Fe/AC + H_2O_2 is shown in Fig 3.

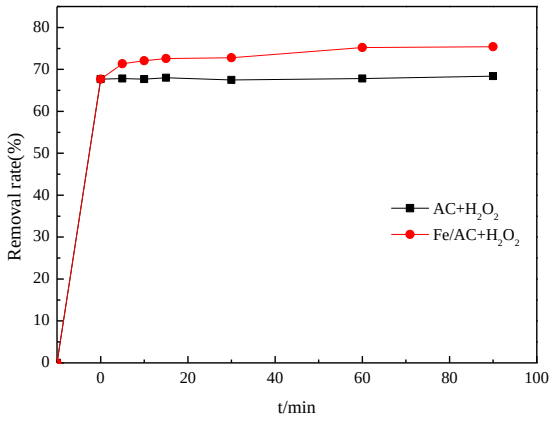

Fig. 3 The degradation effect of benzoic acid on AC + H_2O_2 and Fe/AC + H_2O_2 systems

The time-dependent change curves of the adsorption and degradation effects of benzoic acid on the two different systems of AC + H_2O_2 and Fe/AC + H_2O_2 are shown in Fig.3. As can be seen from the figure, the adsorption effects of benzoic acid by AC and Fe/AC alone are basically the same. Similarly, this shows that there is no significant change in the benzoic acid adsorption effect of AC after metal loading by the impregnation method. After adding H_2O_2, the AC + H_2O_2 system basically did not remove TOC in the solution, which indicates that the removal of benzoic acid by this system is mainly due to adsorption. The direct catalytic effect of AC on H_2O_2 to produce OH was not obvious, which may be the reason why the investigation time was relatively short (90min).Research by Ramirez et al. showed that activated carbon can catalyze the degradation of organic pollutants by H_2O_2, but the degradation rate is very slow. Compared with AC + H_2O_2 system, Fe/AC + H_2O_2 system is more effective in the degradation and removal of TOC, which indicates that Fe/AC catalyst prepared by impregnation method can effectively catalyze the formation of OH from H_2O_2 for Fenton reaction. It can also be seen from the figure that the removal rate gradually increases with time, the change is

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faster in the first 20 minutes, and basically does not change at 60 minutes, which indicates that as the heterogeneous Fenton oxidation process continues, H_2O_2 gradually decreases and eventually loses its ability to degrade. In order to fully investigate the degradation effect of heterogeneous Fenton on benzoic acid wastewater, the subsequent experimental investigation time was set at 90min.

4. Conclusion

(1) The length of adsorption time, the time point of hydrogen peroxide addition, the length of reaction time, acidity and alkalinity, the amount of hydrogen peroxide added, the amount of catalyst added, and the initial concentration of benzoic acid in the solution have a large effect on degradation.

(2) Adjust the initial pH of the solution to 3, add an appropriate amount of catalyst to a concentration of 1 g/L, and adsorb for 210 min under magnetic stirring, and then add 0.5 times the calculated theoretical amount of hydrogen peroxide (ie 0.5Qth = 6.15 mmol/L), and finally reached a TOC removal rate of 76.76% after 90 minutes of reaction at room temperature.

Acknowledgments

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