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Study of degradation of Phenol by Fenton Reagent under Sunlight

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Abstract: Degradation of phenol was studied in the presence of Fenton Reagent. The initial concentrations of $C_2O_4^{2}$, H_2O_2 , Fe^{2+} and pH value on the reaction were investigated. The optimum conditions to degrade phenol from water were determined when initial phenol concentration was 500mg/L, the concentration of $Na_2C_2O_4$, H_2O_2 and Fe^{2+} were 0mmol/L, 300mg/L and 60mg/ L respectively, and pH value was 3. Under the optimum conditions, phenol degradation and mineralization rates could reach 80% and 50%, respectively when the degrade time lasted 10 min.Reference to other literature of sodium oxalate to join can effectively improve the utilization of the ultraviolet and visible light,thus enhancing the effect of high concentrations of phenol wastewater removal, but this experiment reflects the high $C_2O_4^{2-}$ will play an inhibitory effect .

Introduction

Phenol is an important chemical raw material , widely used in medicine, dyes, paper and other industrial fields .Fenton reagent can effective oxide removal of refractory organics which can not be removed by conventional wastewater treatment technology,and its essence is H_2O_2 in the catalytic effect of Fe^{2+} to generate highly reactive hydroxyl radical (• OH) which oxidation potential of up to 2.8V , oxidation of most of organic matter broken down into small molecules .Meanwhile,Fe^{2+} is oxidized to Fe^{3+} to produce coagulation and sedimentation,removal of a large number of organic compounds[1]. UV-Fenton also called photo-Fenton,which is an advanced oxidation technology by the formation of introduce UV to the Fenton system , can improve the utilization of H_2O_2 at the same time reduce the Fe^{2+} dosage[2].

At present , most of the study of the photochemical oxidation of wastewater treatment in the artificial light source , will undoubtedly increase the operating costs , thus limiting the large-scale application of the method in actual wastewater treatment . Sunlight is clean , inexpensive , easy to get the energy , sunlight instead of artificial light source , which can save energy , reduce wastewater treatment costs . Sunlight be used in wastewater photochemical oxidation process ,has begun to attract the attention of scholars at home and abroad , abroad already existing Solar photocatalytic treatment of wastewater engineering applications [3] .

This paper tested the factors of $Na_2C_2O_4$ concentration , H_2O_2 dosage , Fe^{2+} dosage and reaction pH impact of phenol the sunlight Fenton oxidation degradation .

Analysis and Test Methods

Determination of the concentration of phenol : preparation of the concentration of 500mg / L of phenol in wastewater , adding a certain amount of Sodium oxalate , ferrous sulfate and hydrogen peroxide solution , shaking mixed in a volumetric flask , hydrochloric acid and sodium hydroxide to adjust pH , sunlight exposure . Taking some water samples in the distillation flask , adding glass beads and two drops of methyl orange indicator , adjusting to pH 4 with phosphoric acid , adding 5mL of copper sulfate solution to distill . Taking distillate liquid for the determination of phenol concentration to calculate the degradation rate[4] .

$$D = (A_0 - A) / A_0 \times 100$$

Where , D is the phenol degradation rate , A_0 is sample absorbance before it is degradation of processing , A is sample absorbance after degradation treatment .

Determination of COD : measure the water sample irradiated by sunlight , accurate to add 10mL potassium dichromate into conical flask , mixed shake , adding 0.4g of mercury sulfate and 30mL sulfuric acid sulfuric acid silver , reflux condensation for 2 h , cooling . Rinsing condenser wall with

90ml water , removing the flask is again cooled , adding 3 drops of test the ferrous spiritual indicator , titrated by standard solution of ferrous ammonium sulfate , the titration end point is the solution color from yellow to blue-green to reddish-brown , according to the amount of ferrous ammonium sulfate standard solution to calculated COD .

$$COD_{cr}(O_2,mg/L) = 8 \times 1000 \times C \times (V_0 - V_1) / V$$

Where , C is the concentration of ferrous ammonium sulfate , V_0 is consume the volume of ferrous ammonium sulfate for the blank test , V_1 is consume the volume of ferrous ammonium sulfate in water samples , V is the water sample volume .

The removal efficiency of the system of CODcr indicates that the mineralization rate .

Results and Analysis

The impact of the concentration of $Na_2C_2O_4$

Phenol concentration of 500mg / L , Fe^{2+} concentration of 30mg / L , pH value is 3 , sunlight 10min , the phenol degradation in different $Na_2C_2O_4$ concentration as follows:

Table 1 Phenol degradation in different $Na_2C_2O_4$ concentration

sodium oxalate(mmol/L)	0	3	6	9
Phenol degradation rate	63.8%	52.6%	51.5%	51.1%
COD degradation rate	33.3%	30.3%	24.2%	15.2%

In the Fenton system sodium oxalate can improve the rate of degradation of phenol , because of the reaction process of the formation of ferric oxalate complexes have strong ability of competition for the UV and absorption of light in a wide wavelength range , light solution . Fe^{3+} with $C_2O_4^{2-}$ can be formed three kinds of stable ferric oxalate complexes of $Fe(C_2O_4)^+$, $Fe(C_2O_4)_2^-$ and $Fe(C_2O_4)_3^{3-}$, they have photochemical activity , which $Fe(C_2O_4)_3^{3-}$ has the strongest photochemical activity , play a major role in water treatment . When $Na_2C_2O_4$ lower concentrations , $Fe(C_2O_4)_3^{3-}$ to generate a small amount and the • OH less in the system ; but the $Na_2C_2O_4$ concentration is too high , it will inhibit the $Fe(C_2O_4)_3^{3-}$ Photodissociation , and it will also increase in the concentration of HCO_3^- and CO_3^{2-} in the system which occur side reactions with • OH , • OH is eliminated[7] .

By comparing and analyzing , finding that excessive amount of sodium oxalate added at the trial of this article , and the obtained results speak volumes for that too high sodium oxalate will disincentive to the reaction , reducing the effect of degradation of phenol .

The impact of the concentration of H_2O_2

H_2O_2 is the main to produce • OH and the amount of H_2O_2 will directly affect the • OH production rate and formation , thereby affecting the degradation rate and degradability of phenol [8] . Phenol concentration of 500mg / L , $Na_2C_2O_4$ concentration of 0 mmol / L , Fe^{2+} concentration of 30mg / L , pH value is 3 , sunlight 10min , different H_2O_2 solution concentration on phenol degradation shown in Figure 1.

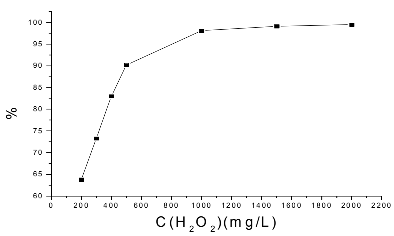

Fig. 1 Phenol degradation in different H_2O_2 concentration

The figure shows the degradability of phenol increased with increasing concentration of H_2O_2 . When the dosage of hydrogen peroxide to a value (1000 mg / L), phenol degradation rate increased slowly . This is because the H_2O_2 is not only • OH generation agent, but also a scavenger . The low concentration of H_2O_2 make the • OH generate less , this is not conducive to the degradation of organic matter ; the high concentration of H_2O_2 make the • OH scavenging effect strengthen to play an inhibitory effect on the degradation rate[9] . H_2O_2 dosage should be appropriate in this system , the most appropriate concentration of 1000 mg / L . In practical applications , not only to consider the effect of oxidant wastewater treatment , also need to consider the cost of wastewater treatment , therefore , in this experiment to select the 300mg / L .

The impact of the concentration of Fe2 +

Fe^{2+} is a necessary condition for catalytic production of free radicals . Phenol concentration of 500mg / L , $Na_2C_2O_4$ concentration of 0 mmol / L , H_2O_2 concentration of 300mg / L , pH value is 3 , sunlight 10min , Effect of different Fe^{2+} concentration to the impact of phenol degradation shown in Figure 2

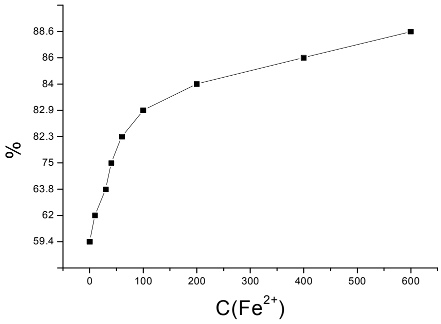

Fig. 2 Effect of different Fe^{2+} concentration on phenol degradation

The figure shows , the phenol degradation rate gradually increases with increasing concentration of Fe^{2+} . The reason is that : Fe^{2+} concentration gradually increased to promote the reaction Fe^{2+} + H_2O_2 Fe^{3+} + • OH + OH^- conduct , gradual strengthening of the decomposition of H_2O_2 ,improving the oxidative capacity of the system , so Phenol degradation efficiency is improved .

The impact of pH

Phenol concentration of 500mg / L , $Na_2C_2O_4$ concentration of 0 mmol / L , H_2O_2 concentration of 300mg / L , Fe^{2+} concentration of 60mg / L , sunlight 10min , the influence of pH on phenol degradation shown in Figure 3

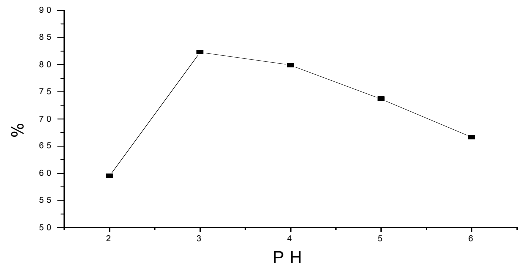

Fig. 3 Effect of different PH on phenol degradation

The previous results show that the optimum pH value of 3 to 5 in the Fenton reaction system[10]. The test results show that the optimal pH of sunlight Fenton oxidation of phenol is also within the acidic range of 3 to 4 . The figure shows when the pH value of 2 to 3 , the phenol degradation rate was significantly increased with the increase of pH value , and when greater than 4 , the phenol degradation rate but decreased . This is due to low pH values (pH <3) , the system H^+ concentration is too high , H^+ is the scavenger of • OH : H^++ • OH→H_2O , this is not conducive to the generation of • OH , degradation rate dramatic decline . But the high pH values (pH > 4) , Fe^{2+} is easy to form Fe (OH) 3 colloid or $Fe_2 0_3$ • nH2O amorphous precipitate , to cause the catalytic activity and photochemical activity of the system decreases or disappears , also not conducive to the generation of •OH , the degradation rate also decreased significantly[11]. So the pH has a great influence on the degradation of phenol , the trial suitable pH value is 3.0 to 4.0 .

Phenol mineralization

The process of Phenol wastewater degradation by sunlight -Fenton Reagent , wastewater quickly into a dark brown color (Color of soy sauce) from colorless , continuous dark brown after a few dozen minutes , faded to yellow , then to pale yellow . When the wastewater eventually becomes a very pale yellow (almost colorless, is the color of the solution containing a small amount of Fe^{2+} and Fe^{3+}), Phenol completely mineralized[12]. In this experiment (as illustrated below) , the color of Phenol is dark brown , but mineralization rate only reached 50% , description of phenol during the reaction is the formation of other organic complexes of benzoquinone and organic acids , does not completely mineralized into inorganic .

Conclusions

By experiment, confirmed the sunlight - Fenton treatment of phenol wastewater , fast and generally obtain better degradation results in a relatively short period of time , and the process is simple , this is a promising industrial organic wastewater treatment .

The optimal conditions are $Na_2C_2O_4$ concentration of 0 mmol / L, the concentration of H_2O_2 300mg / L , the Fe^{2+} concentration of 60mg / L , pH = 3 and reaction time 10min , phenol degradation rate is 80% and the mineralization rate is 50%.

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