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# Regeneration of exhausted activated carbon by electrochemical method

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**Abstract**

The regeneration of coconut shell activated carbon contaminated with phenol was systematically investigated by electrochemical method under different operating conditions. The effects of several operating parameters on the electrochemical regeneration efficiency were measured at room temperature. The experimental results show that the electrochemical method can be used to regenerate the activated carbon exhausted with phenol. The electrochemical regeneration efficiency depends on several operating variables such as electrolyte concentration, regeneration current intensity and regeneration time. The residual phenol concentration in solution was much lower and the regeneration efficiency can reach 85.2% in a stirred electrochemical reactor after regeneration for 5 h. © 2002 Elsevier Science B.V. All rights reserved.

*Keywords:* Activated carbon; Electrochemical regeneration; Adsorption

## 1. Introduction

Activated carbon has been widely used to remove organic pollutants from industrial and municipal wastewater. In view of the high production cost and consumption of resources, the way how to regenerate the exhausted activated carbon effectively remains a key problem. A variety of regeneration techniques [1–9] for exhausted activated carbon, especially carbon saturated with organic pollutants such as phenol, are suggested, e.g. thermal regeneration and chemical regeneration.

The thermal regeneration techniques are widely used with high efficiency. But they are also characterized by 5–10% carbon loss due to oxidation and attrition, and by high energy consumption to keep the temperature between 800 and 850 °C. Meanwhile, the exhausted activated carbon need to be delivered through a long distance to a special production plant to be generated which will results in a higher transportation cost.

By chemical methods, the exhausted activated carbon may not be returned to the manufacturer to regenerate and carbon loss could be neglected. The regeneration efficiency by chemical methods depends on the types of organic pollutants. It is a very important process to treat tap water and wastewater contaminated with phenol by activated carbon. The research [1–16] on how to regenerate this kind of exhausted activated carbon is very active. The granular activated carbon saturated with phenol is usually regenerated by chemical methods such as using Lewis base desorption and electrochemical oxidation. It is relatively difficult to make the regeneration efficiency of activated carbon over 70%. The regeneration efficiency by electrochemical methods can be conveniently operated in-situ, and reach as high as 80–95% which depends on specific regeneration methods and operating conditions; in principle, it is possible for the regeneration efficiency to reach 95% on bench-scale experimental conditions. The electrochemical regeneration technique was suggested by Narbaitz [10]. The electrochemical regeneration mechanism was investigated by different researchers [10–16]. It is necessary to understand how to scale up this method and develop an in-situ electrochemical regeneration technique based on the understanding of the process mechanism of electrochemically regenerating exhausted activated carbon. The research in this project focuses on several parts. First, the regeneration mechanism for the exhausted activated carbon that is regenerated by fixing activated carbon on electrodes under different operating conditions is investigated. Then the regeneration was carried out in a stirred electrochemical reactor, fixed bed reactor and so on. The research in this paper is part of this project development.

## 2. Materials and methods

The granular activated carbon used here was made from coconut shell and supplied by Baoding Activated Carbon Factory (China). Its iodine adsorption value is 1000 mg/g,

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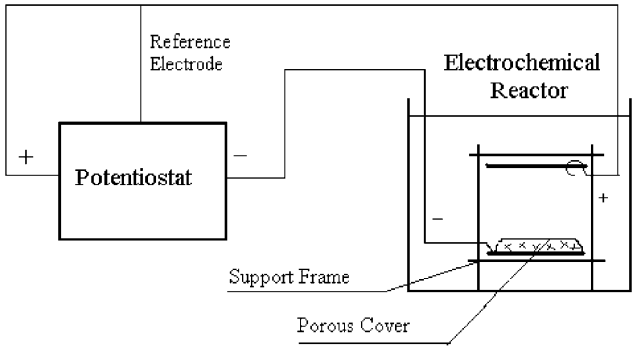


Fig. 1. The electrochemical reactor of regenerating activated carbon.

the carbon tetrachloride adsorption value is 58%, its hardness is 95%, its particle size is 10 × 20 mesh. The wastewater containing phenol in the experiments was prepared from the chemical agent and purified water.

The regeneration experiments of granular activated carbon were carried out in an electrochemical batch reactor that consisted of two platinum electrodes shown in Fig. 1, one is anode and another is cathode, and a reference electrode submerged in electrolytes. Both anode and cathode have an area of 6 cm × 8 cm, and there was a fixed distance of 1.8 cm between anode and cathode in those experiments. An amount of 0.3 g exhausted activated carbon was fixed on the electrodes by using a porous cover which made activated carbon fixed and the gases from the electrochemical reaction release easily. The activated carbon was used to adsorb phenol so as to be saturated at 25 °C. Then the saturated activated carbon was put onto the selected electrode to be regenerated in the reactor. The regenerated activated carbon was saturated with equilibrium adsorption of phenol and regenerated again and again under the same conditions.

Analysis of phenol in the solution was done by a type 721 spectrophotometer using 4-amino-antipyrine–ferricyanide photometric method [17], which is a standard analysis for phenol in China.

The saturated adsorption values of activated carbon in this paper were denoted by the equilibrium adsorption values of phenol on activated carbon under a 1.0 g/l phenol solution concentration except for the adsorption isotherms. The saturated adsorption values of phenol before and after regeneration were analyzed to calculate the regeneration efficiency. The regeneration efficiency was computed comparatively to the saturated adsorption of fresh activated carbon under the same equilibrium solution concentration.

## 3. Experimental results and discussions

The regeneration efficiency of activated carbon adsorbed with phenol depends on electrolyte type, electrolyte concentration, regeneration current and time and the electrode for activated carbon to be placed which are the most important variables affecting regeneration efficiency. The variables were investigated in this paper to assess the electrochemical regeneration process so as to improve the operation performance and understand the regeneration mechanism.

### *3.1. The adsorption equilibrium isotherm of phenol on activated carbon*

The adsorption equilibrium of phenol on activated carbon was measured at room temperature. In order to measure the adsorption equilibrium isotherm of phenol in activated carbon, N bottles with solutions of equal volume and different phenol concentrations were used. A certain amount of activated carbon with same weight was put into the bottles for 24 h so that the adsorption equilibrium could be reached. All bottles were put in the thermostat water bath at 25 °C. The adsorption equilibrium values can be calculated according to the concentrations of phenol in the solutions before and after adsorption. The effect of NaCl on the adsorption equilibrium was also measured because the electrochemical regeneration made use of 2% NaCl solution as an electrolyte. The experimental results are shown in Fig. 2. NaCl can increase the equilibrium adsorption capacity of activated carbon for phenol, but the increasing capacity of equilibrium adsorption is <5%. The adsorption equilibrium isotherm of phenol in activated carbon was analyzed by different isotherm equations such as Langmuir, Freundlich, etc. The regression results show that Freundlich isotherm equation has a much better regression precision, the regression result for the phenol solution without NaCl is $q = 28.66C^{0.27}$, where equilibrium adsorption capacity of phenol in activated carbon and the concentration phenol in solution are denoted by $q$ and $C$, respectively.

### *3.2. Regeneration on cathode and anode*

Spent activated carbon saturated with phenol was respectively placed in cathode or anode zone to be regenerated with different regeneration currents and 1.8 cm distance between electrodes. The electrolyte was sodium chloride and its

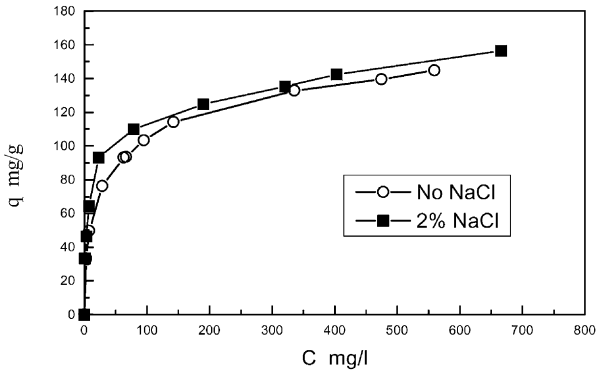


Fig. 2. Adsorption equilibrium of phenol on activated carbon.

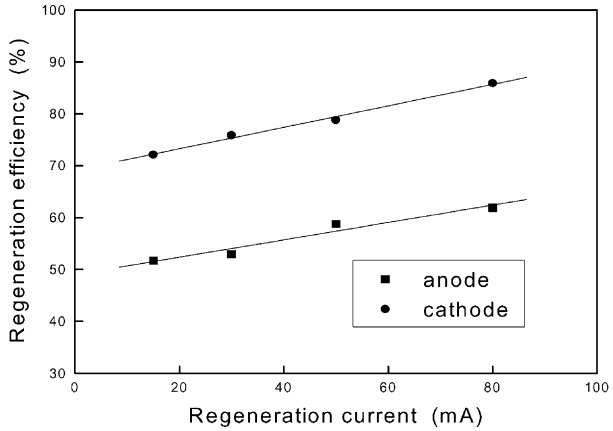


Fig. 3. Regeneration efficiency of activated carbon in different electrodes.

concentration was 1% in the solution of 800 ml, the regeneration time was 5 h. The regeneration efficiency of activated carbon was computed according to its saturated adsorption capacity before or after regeneration. The experimental result is shown in Fig. 3.

Obviously, the regeneration efficiency of activated carbon in cathode is higher than in anode by an average of 20%. The higher the regeneration current, the higher the regeneration efficiency in both electrodes will be. When regeneration was processed in cathode, a large amount of fog-like gases are formed to gather on the surface of electrolyte solution. Bubbles raised mainly from the cathodic plate like air mist in the liquid, and the electrolyte solution turned into dark brown with uncomfortable smell. Residual phenol could be detected after 5 h. However, on the other hand, such phenomenon did not appear when carbon in anode, the electrolyte solution was transparent without phenol left. More experiments showed that whether activated carbon was put on cathode or anode, the color of electrolyte solution would fade away in the end if the regeneration time was prolonged or electrolyte concentration was increased, e.g. 2% NaCl. All the next regeneration experiments of activated carbon saturated with phenol were to put it in cathode considering its high regeneration efficiency. The research results [11–15] for the reaction mechanism of phenol oxidation by electrochemical oxidation indicated that during the reaction processes, under the influence of the electric field, the phenol first became yellow *p*-benzoquinone then was oxided into carbon dioxide and water, accompanied with a small part of polymers to form at the same time. The experimental phenomena were conformable to the above-mentioned results. The electrochemical regeneration mechanism [8] of activated carbon saturated with phenol shows that the phenol is firstly desorbed from the activated carbon surface, then oxidized by electrochemical active chloride or oxygen [14–16]. Under the influence of the electric filed, the $Na^+$ ions move to cathode to make phenol in activated carbon be easily desorbed in the cathode zone because the $Na^+$ ion reacts with phenol into sodium phenate which is very difficult to be adsorbed in activated carbon. Meanwhile the pH value at the cathode zone increases with the movement of the $Na^+$ ions to the cathode. The equilibrium adsorption capacity of phenol in activated carbon decreases with the increase of the pH value in basic solution [8]. The equilibrium adsorption capacity of phenol in activated carbon increases in acidic solution. The desorbed phenol is easily oxidized in the anode by electrochemical active chlorine or oxygen. So the concentration polarization can take place because of mass transfer resistance. The effect of concentration polarization can be decreased in the compulsive stirring to enhance mass transfer, this can be proved by the following experimental results in a stirred electrochemical reactor.

Table 1
The effects of different electrolytes on regeneration efficiency

| | NaCl | $Na_2CO_3$ | $NaHCO_3$ | $Na_2SO_4$ |
|---|---|---|---|---|
| Regeneration efficiency (%) | 78.8 | 66.5 | 57.7 | 65.5 |
| Residual phenol concentration (mg/l) | 11.8 | 24.3 | 22.6 | 28.4 |

### 3.3. Electrolyte type

Several kinds of electrolytes such as sodium chloride, sodium carbonate, sodium bicarbonate and sodium sulfate with the same concentration of 1% were tested. In each test, the spent activated carbon was regenerated in electrolyte solution of 800 ml with 50 mA current intensity. The fixed distance between electrodes was 1.8 cm. The experimental data are presented in Table 1. The result demonstrates that sodium chloride solution is the best electrolyte in that it has high regeneration efficiency and low residual phenol among the limited examinations in the research.

### 3.4. Electrolyte concentration

As the concentration increases from 0.01 to 5%, the effect of the NaCl concentration on regeneration efficiency is significant. The regeneration time and regeneration current are 5 h and 50 mA, respectively. The regeneration efficiency increased radically when the electrolyte concentrations are changed from 0.01 to 1% (w/w) of NaCl in the solution. It nearly keeps the same after that. The color turns from brown into transparent during the 5 h of regeneration when the electrolyte concentration reached 2%. The experimental result is shown below in Fig. 4. The electrolyte concentration was kept the same as 2% in the following experiments.

### 3.5. Regeneration current intensity and time

The regeneration efficiency of activated carbon was measured at different regeneration current intensity regenerating for 5 h or at different regeneration time while keeping other experimental conditions the same as above-mentioned experiments. The results are shown in Figs. 5 and 6.

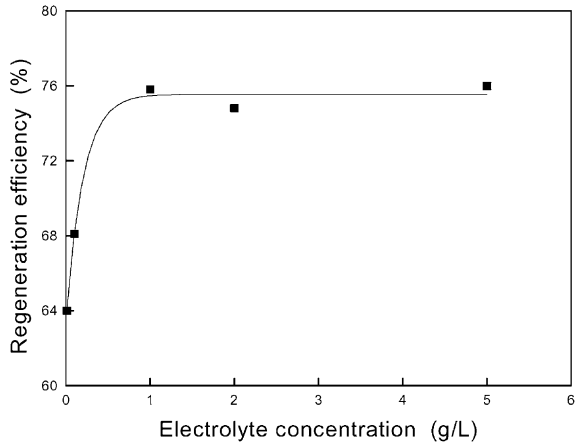


Fig. 4. The regeneration efficiency of activated carbon in different electrolyte concentration.

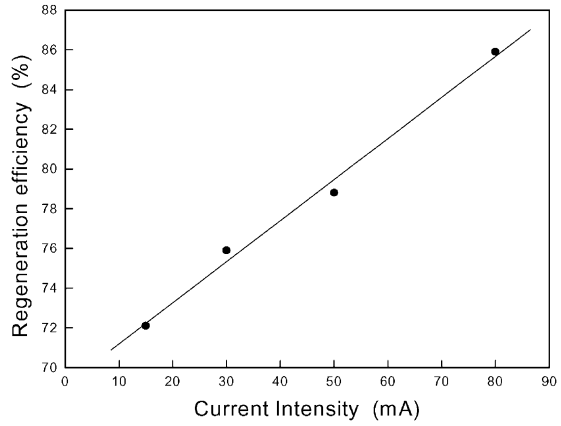


Fig. 5. Regeneration efficiency of activated carbon in different current intensity.

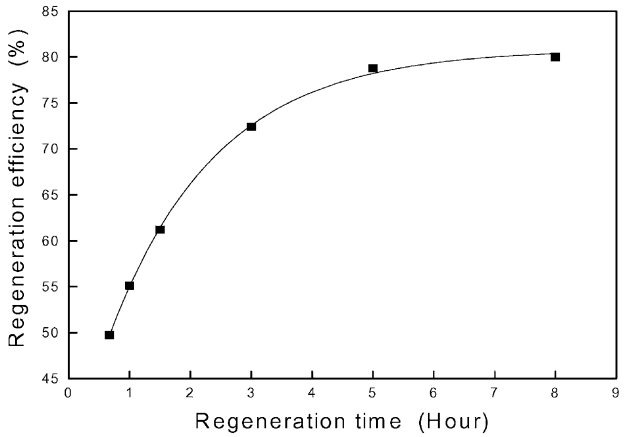


Fig. 6. Regeneration efficiency of activated carbon in different regeneration time.

The experimental result shows that the regeneration efficiency of activated carbon increases linearly along with the increase of the regeneration current intensity, this result is conformable with Faraday's law. It seems that the regeneration efficiency could increase and get better result if merely regeneration current intensity is increased. The regeneration current intensity of 50 mA was chosen in other experiments to be more suitable by assessing the regeneration cost of energy consumption, efficiency and other operative variables. At the same time, the regeneration efficiency also increases with regeneration time. The change of regeneration efficiency could be neglected after regeneration for 5 h which was chosen in other experiments.

### *3.6. Regeneration cycle*

The activated carbon was used to adsorb phenol and was regenerated again and again under the conditions of 2% NaCl electrolyte solution, 1.8 cm distance of electrodes, 50 mA regeneration current intensity, regeneration time of 5 h. The regeneration efficiency of activated carbon, was respectively computed. Experimental result is shown in Fig. 7. The decrease of regeneration efficiency is not very clear that can almost be neglected except for first time when compared with the fresh carbon.

### *3.7. The regeneration in a stirred reactor*

In order to understand the possibility of electrochemically regenerating activated carbon, the experimental reactor was changed, that is, the exhausted activated carbon was not fixed onto the electrodes, the distance between two electrodes was adjusted to 9.5 cm, meanwhile, the solution including 0.3 g exhausted activated carbon was stirred by a stirrer so as to enhance mass transfer, all other experimental conditions like 50 mA regeneration current intensity, 2% NaCl and 25 °C were kept the same. The residual

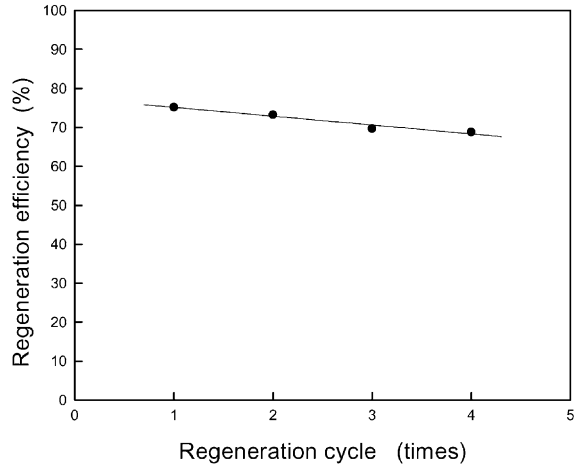


Fig. 7. Regeneration efficiency of activated carbon after different regeneration cycle times.

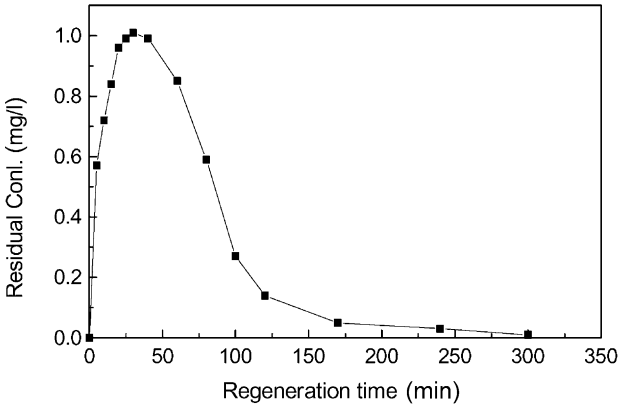


Fig. 8. The residual concentration of phenol at intervals in the reactor.

concentrations of phenol at intervals were measured and shown in Fig. 8. The result expressed that the compulsive stirring can enhance the regeneration efficiency of activated carbon. The regeneration efficiency can reach 85.2% after regeneration for 5 h and can be raised over 90% by prolonging the regeneration time. The residual phenol concentration in reactor can be decreased more quickly in this way than keeping activated carbon fixed on the electrodes. This result is conformable with the regeneration mechanism mentioned above.

## 4. Conclusions

The electrochemical method is very effective for the regeneration of exhausted activated carbon with phenol, and the regeneration efficiency can reach over 80%. Desorption and destruction of phenol adsorbed on granular activated carbon could be greatly enhanced in an electrochemical reactor. It is much more efficient for activated carbon to be put in cathode to regenerate than in anode. Sodium chloride is the best choice to be used as electrolyte compared with other electrolytes. The regeneration efficiency of activated carbon adsorbing phenol increases along with the increase of electrolyte concentration in the range of lower concentration. It almost keeps the same when the concentration reaches 1%. At the same time, the regeneration efficiency increases along with increasing regeneration current intensity to be a linear relationship. It also increases along with regeneration time, but it basically has no change after the regeneration time reaches 5 h. The decrease of regeneration efficiency is not very clear that it can almost be neglected, but the first time compared with the fresh carbon is an exception. The residual concentration of phenol is $<0.01$ mg/l and the regeneration efficiency can reach 85.2% in a stirred electrochemical reactor after regeneration for 5 h. This method indicates an increasing importance and wider application prospects.

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