

# Phenol Removal from Water with Potassium Permanganate Modified Granular Activated Carbon

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## ABSTRACT

In order to improve adsorption capacity of granular activated carbon (GAC), potassium permanganate was used to react with GAC to change the surface properties and improve the adsorption capacity of GAC. By batch experiments, improvement of adsorption capacity of potassium permanganate modified GAC ( $GAC_M$ ) was studied. The influence of adsorption time, temperature, ratio of phenol with GAC/ $GAC_M$ , initial concentration of phenol and pH on adsorption efficiency of  $GAC_M$  was studied. The results showed that modified by potassium permanganate, the adsorption capacity of GAC improved to a higher level. The removal efficiency of phenol increased to about 20%.

**Keywords:** Modified Granular Activated Carbon; Potassium Permanganate; Phenol; Adsorption

## 1. Introduction

With the development of chemical industry, more and more organic compounds were disposed into water and aggravated water pollution. Adsorption is an effective technology in removing organic pollutants from water [1-3]. In which the number of adsorbents, activated carbon has been seen as one of the main and effective adsorbents. Activated carbon has high surface area, large pore volume, tunable pore size, chemical stability, high hydrophobicity and interconnected frameworks [4-8]. While, pore structure and surface properties affect the adsorption capacity of activated carbon [9,10]. To improve the adsorption efficiency and save dosage, in recent years, many researchers have dedicated on researches of modification of activated carbon [11-18]. The use of oxidizing agents is the most common methodology to modify AC surface [19-22]. Some researchers have found that carboxylic functional groups can be introduced upon oxidizing the surface of carbon materials by various oxidation agents such as concentrated nitric [23], ozone [24], ammonium [25] and  $H_2O_2$  [26], which could help to change the inert and hydrophobic nature of carbon materials and enhance the wettability for polar solvents and make the surface active for the immobilization of organic compounds via adsorption [27]. For example, Gil *et al.* [28] studied the oxidation of  $HNO_3$  and found that under different temperature, oxidation of  $HNO_3$  would

affect the macro and micro porous structure of activated carbon. Liu *et al.* [7] found that modification of air oxidation improved the wettability and the adsorption properties of pitch-based spherical activated carbon as the increase of the oxygen-containing groups. Yin *et al.* [29] reviewed the chemical, physical and biological modification techniques of activated carbon, and found that the beneficial effects of specific modification techniques on activated carbon adsorption of targeted contaminant species from aqueous solutions were profound. Utrilla *et al.* [18] compared the advantages and disadvantages of different methodologies to modify the surface of activated carbons and found that modified by using  $H_2O_2$ ,  $O_3$  and  $HNO_3$ , acidic surface functional groups increase in the AC surface as the reduce of the AC surface area; and the nitrogenation or sulfuration of the AC surface can increase its basicity favoring the adsorption of organic compounds.

As an oxidant, potassium permanganate has been widely used in water treatment [30-35]. In this paper, activated carbon was modified with potassium permanganate, and with representative organic matter of phenol, the impact of modification on adsorption capacity of activated carbon was studied.

## 2. Materials and Methods

### 2.1. Pretreatment of Granular Activated Carbon

The particle size of granular activated carbon was 10 - 20

mish. After repeatedly soaked and washed with deionized water, the activated carbon was dried to constant weight at 105°C for over 24 h.

## 2.2. Modification Method of Activated Carbon

Modification of GAC was conducted under neutral condition. The activated carbon was immersed in pollution of potassium permanganate (controlled the ratio of concentration of potassium permanganate to activated carbon (L/M) to a certain value) for a certain period of time and slow stirring with constant temperature oscillator, then separated and washed repeatedly with deionized water to no color, then dried to constant weight at 105°C for 24 h.

## 2.3. Adsorption Studies

Adsorption experiments were performed to study the effects of important parameters such as effect of temperature, amount of adsorbent, time of contact and pH, ratio of phenol with GAC/GAC<sub>M</sub>, initial concentration of phenol in water was 250 mg/L. 250 ml of water (contaminated with phenol) was taken into 500 ml volumetric flask, then 2 g of adsorbents (GAC/GAC<sub>M</sub>) was put into the flasks. The flask was then subjected to shaking for proper adsorption on magnetic stirrer for a certain time. After the fixed time of contact, these solutions were filtered with filter (whatman filter size 0.45 µm PES filter media ) and the amount of the phenol adsorbed was analyzed.

## 2.4. Analytic Methods

The concentration of phenol was analyzed with liquid chromatography. The chromatography column was Century SILC<sup>18</sup>-EPS (200 × 4.6 mm, 5 µm), made in Germany. The mobile phase was acetonitrile 50%-water 50% of volume proportion. Column temperature was 20°C, and column pressure was 11.0 MPa. The flow rate of mobile phase was 1.0 mL/min. The detector was UV detector, and with detection wavelength of 270 nm.

## 3. Results and Discussion

### 3.1. Effect of Potassium Permanganate Concentration on Adsorption Efficiency of GAC<sub>M</sub>

2 g GAC was modified with 20 mL different concentration of Potassium permanganate solution of 10.0, 15.0, 20.0, 25.0, 30.0, 40.0, 50.0, 60.0 mg/L for 90 min previously. The initial phenol concentration was 250 mg/L. The adsorption time of GAC<sub>M</sub> for phenol was controlled for 4 h. Temperature was 20°C. Effect of potassium permanganate concentration on adsorption efficiency of

GAC<sub>M</sub> for phenol was shown in **Figure 1**.

It could be seen that modified with different concentration of potassium permanganate, the adsorption efficiency of GAC<sub>M</sub> for phenol differed. At beginning, the removal efficiency of phenol increased with the increase of concentration of potassium permanganate. When concentration of potassium permanganate increased to 30 mg/L, the removal efficiency of phenol increased to the highest, and then decreased with the increase of concentration of potassium permanganate.

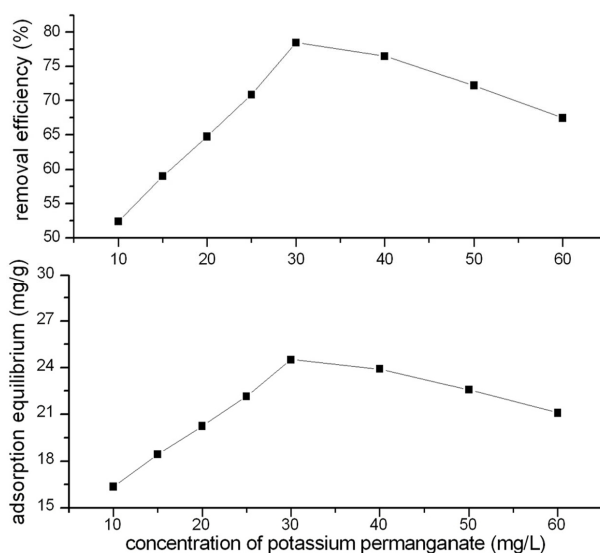
This could be because that with the increase of concentration of potassium permanganate, there are more and more potassium permanganate react with GAC and oxidized the surface of GAC, and formed more pore structure and functional groups in GAC surface, which would increase the adsorption capacity for phenol. But when concentration of potassium permanganate increased to a higher level, excessive potassium permanganate would react with the surface function groups, which would decrease the adsorption capacity of activated carbon otherwise.

From results of **Figure 1**, it could be concluded that for 2 g GAC, the optimum concentration of potassium permanganate was 30 mg/L.

### 3.2. Effect of Modification Time on Adsorption Efficiency of GAC<sub>M</sub>

In process of modification, the concentration of potassium permanganate solution was 30 and 40 mg/L, and the dosage of GAC was 2.0 g. Modification time of GAC with potassium permanganate was controlled at 10, 30, 60, 90, 120, 150, 180, 210 min respectively.

The adsorption experiment was conducted in 250 mL



**Figure 1.** Effect of concentration of potassium permanganate on adsorption capacity of GAC<sub>M</sub>.

water. Temperature was 20°C. The phenol concentration was 250 mg/L, and the dosage of GAC<sub>M</sub> was 2.0 g (that is the ratio of phenol with GAC<sub>M</sub> was 62.5:2.0 mg/g), and adsorption time was 4 h. **Figure 2** showed the effect of modified time on adsorption capacity for phenol of GAC<sub>M</sub>.

It could be seen that the removal efficiency for phenol increased with the increase of modification time of GAC<sub>M</sub>. At beginning, the removal rate increased very fast, and when modification time increased to 90 min, the removal rate increased gentle. At the same time, it could be seen that when concentration of potassium permanganate was 30 mg/L, the removal efficiency of phenol was higher than that of 40 mg/L. So on later, for 2.0 g GAC, the concentration of potassium permanganate was controlled at 30 mg/L, and the modification time was 90 min.

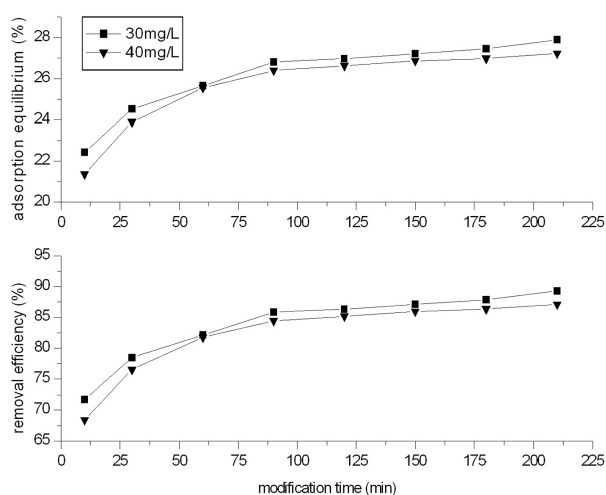
### 3.3. Effect of Time on Adsorption Capacity of GAC/GAC<sub>M</sub>

Temperature is 20°C. Ratio of phenol with GAC/GAC<sub>M</sub> was 62.5:2.0 mg/g. Results of adsorption capacity versus time were shown in **Figure 3**.

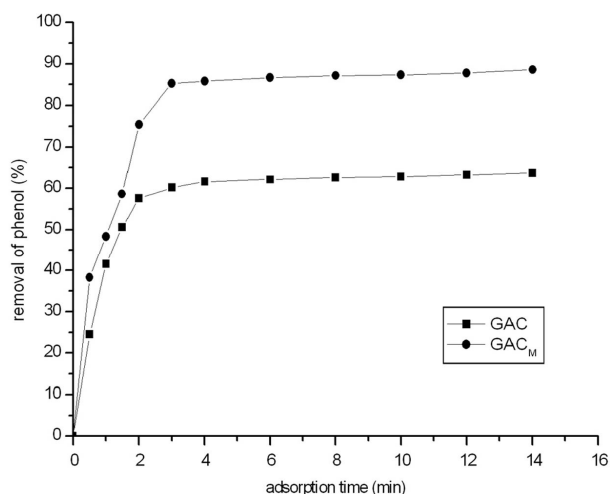
It could be seen that for GAC and GAC<sub>M</sub>, the adsorption capacity for phenol increased with the increase of adsorption time to about 4 hours, and then were nearly at constant. This showed that the adsorption for phenol of GAC/GAC<sub>M</sub> was reached equilibrium at nearly 4 hours. At the same time, it could be seen that the equilibrium of GAC<sub>M</sub> was about 28 mg/g, while of GAC was 20 mg/g. This showed that the adsorption capacity for phenol of GAC<sub>M</sub> was greater than that of GAC.

### 3.4. Effect of Temperature

The temperature was adjusted to 20°C, 25°C and 30°C re-



**Figure 2.** Effect of modification time of GAC on phenol removal rate by GAC<sub>M</sub>.



**Figure 3.** Effect of adsorption time.

spectively. In 250 mL water, the dosage of GAC/GAC<sub>M</sub> was 2.0 g, and the concentration of phenol was adjusted to 50, 100, 150, 200, 250, 300, 350, 400 mg/L to satisfied the ratio of phenol to GAC/GAC<sub>M</sub> was 12.5:2.0, 25.0:2.0, 37.5:2.0, 50.0:2.0, 62.5:2.0, 75.0:2.0, 87.5:2.0, 100.0:2.0 mg/g. Adsorption time was 4 h. The effect of temperature on removal of phenol at different ratio of phenol to GAC/GAC<sub>M</sub> was shown in **Figure 4**.

The results showed that the removal efficiency for phenol decreased with the increase of temperature. When temperature increased from 20°C to 30°C, the highest removal efficiency for phenol decreased to 50.86% from 61.47% with GAC, and decreased from 78.46% to 61.02% with GAC<sub>M</sub>. That is to say that the adsorption for phenol with GAC/GAC<sub>M</sub> is exothermic, and the lower temperature is favorable for adsorption for phenol by GAC/GAC<sub>M</sub>.

At the same time, it could be seen that the effect of temperature on adsorption for phenol with GAC<sub>M</sub> was as the same as that with GAC, but at same temperature, the adsorption capacity of GAC<sub>M</sub> was higher. On later experiments, temperature was 20°C.

### 3.5. Effect of Dosage of Activated Carbon on Adsorption Capacity

The concentration of phenol in water was 250 mg/L, and the dosage of GAC/GAC<sub>M</sub> was adjusted to satisfy the ratio of phenol to GAC/GAC<sub>M</sub> was 62.5:0.5, 62.5:1.0, 62.5:1.5, 62.5:2.0, 62.5:2.5, 62.5:3.0, 62.5:3.5, 62.5:4.0 mg/g. Previous modification time of GAC<sub>M</sub> was 30 and 90 min respectively. Adsorption time was 4 h. Result of **Figure 5** showed the effect of dosage of GAC/GAC<sub>M</sub> on removal efficiency for phenol from water by adsorption of GAC/GAC<sub>M</sub>.

It could be seen that at the same initial concentration of phenol, the removal efficiency for phenol increased

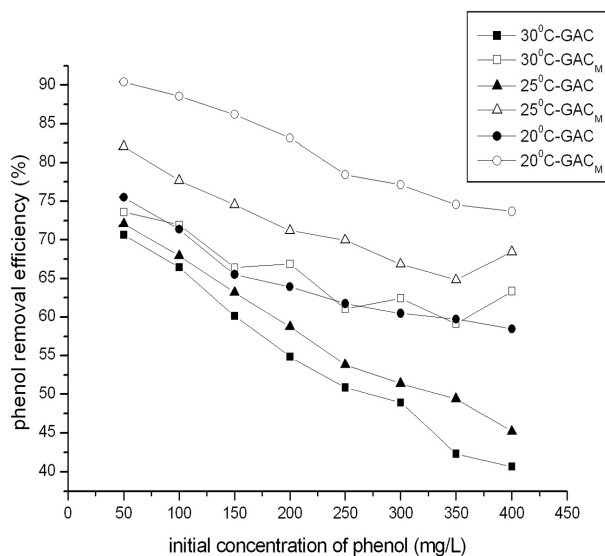


Figure 4. Effect of temperature on the adsorption rate for phenol by GAC/GAC<sub>M</sub>.

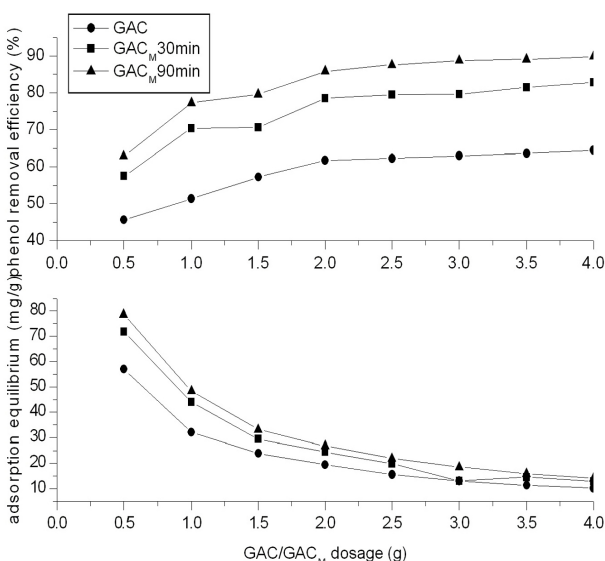


Figure 5. Effects of dosage of carbon on the removal rate.

with the increase of GAC/GAC<sub>M</sub> dosage. That is to say that when dosage of activated carbon was insufficient, the adsorption of activated carbon reached saturation easily, and the removal efficiency of phenol was low. With the increase of dosage of activated carbon, the removal efficiency of phenol increased. When dosage of activated carbon was smaller than 2.0 g, the removal rate for phenol increased fast with the increase of GAC/GAC<sub>M</sub> dosage. While when dosage of activated carbon was larger than 2.0 g, the removal rate for phenol increased slowly, and at that time, the equilibrium adsorption capacity was lower. This is because that when dosage of activated carbon was sufficient, the phenol was removed all the nearly. Increase of activated carbon

would increase the costs. Therefore, the dosage of activated carbon should be appropriate.

From results of **Figure 5**, it also could be seen that the optimum dosage of activated carbon has nothing to do with modification or not. At experiment condition, the optimal ratio of phenol to activated carbon was 62.5:2.0 mg/g. At the same time, the adsorption capacity of GAC<sub>M</sub> was higher significantly than that of GAC. For example, the removal efficient for phenol with GAC<sub>M</sub>, at ratio of phenol to activated carbon was 62.5:1.2 mg/g, was higher than that of GAC at ratio of 62.5:4.0 mg/g.

At the same time, modification time influence the removal efficiency for phenol by GAC<sub>M</sub>. With modification time increased to 90 min from 30 min, the removal efficiency for phenol increased to a higher level.

### 3.6. Effect of Initial Concentration of Phenol on Removal Efficiency

Adsorption time was 4 hour. The GAC was modified by potassium permanganate pollution of 30 mg/L, temperature was 20°C. Dosage of GAC/GAC<sub>M</sub> was 2.0 g, and the concentration of phenol in water was adjusted to satisfy the ratio of phenol to GAC/GAC<sub>M</sub> was 12.5:2.0, 25.0:2.0, 37.5:2.0, 50.0:2.0, 62.5:2.0, 75.0:2.0, 87.5:2.0, 100.0:2.0 mg/g respectively.

Results in **Figure 6** showed the effect of initial concentration of phenol on removal of phenol by adsorption of GAC/GAC<sub>M</sub>. It could be seen that with the increase of initial concentration of phenol, the equilibrium adsorption amount of phenol increased, while the removal efficiency of phenol decreased.

This is because that at a certain dosage of GAC, more and more phenol was absorbed by GAC, so the equilibrium adsorption amount of phenol increased with the

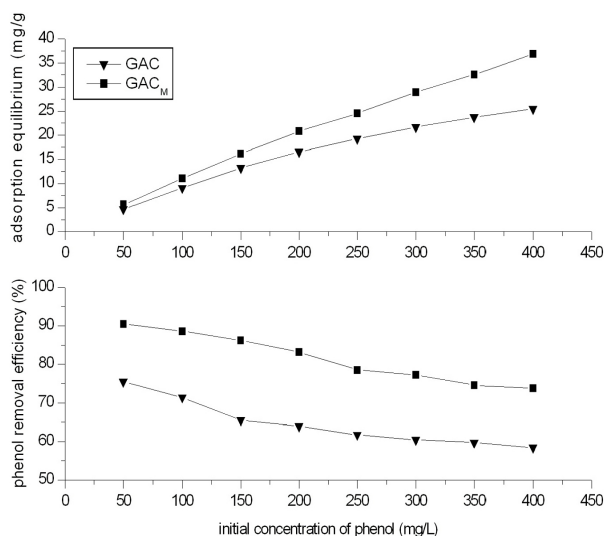


Figure 6. The effects of initial concentration of phenol the removal rate.

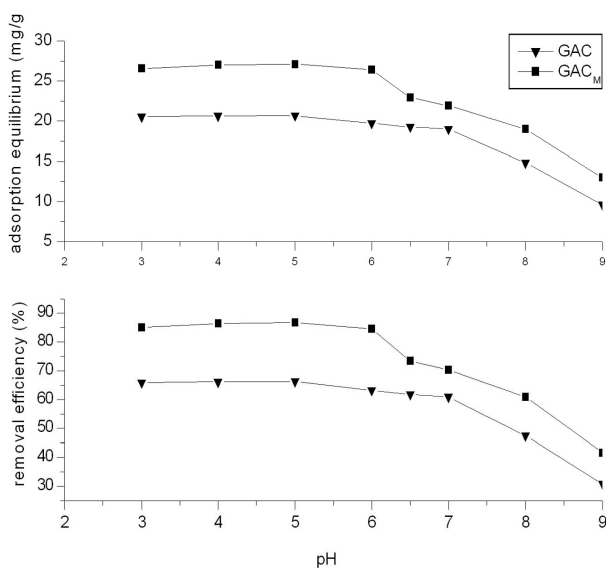
increase of initial concentration of phenol. On the other hand, the saturated adsorption capacity of GAC is limited. When the initial concentration of phenol is high, the phenol can't be absorbed by GAC completely, so the removal efficiency of phenol decreased instead. So, at a certain dosage of GAC, the most suitable initial concentration of phenol should be determined by removal efficiency of phenol not equilibrium adsorption amount only. At the same time, it could also be seen that compared with that of GAC, the removal efficiency and equilibrium adsorption amount of phenol by GAC<sub>M</sub> increased to a high level.

### 3.7. Effect of pH

Temperature was 20°C, phenol concentration in water was 250 mg/L, and the dosage of GAC/GAC<sub>M</sub> was 2.0 g (ratio of phenol to GAC/GAC<sub>M</sub> was 62.5:2.0 mg/g). Results of **Figure 7** show the effect of pH on adsorption efficiency of phenol by GAC/GAC<sub>M</sub>. It could be seen that in acidic and neutral condition, the removal efficiency is high. When pH value is in range of 3 ~ 6, pH has no effect on removal of phenol by GAC and GAC<sub>M</sub>. While with the increase of pH from 6 to 9, the removal efficiency of phenol decreased to about 30%, and the equilibrium adsorption amount decreased about 10 mg/g. This may be because that when pH is less than 7, phenol exists mainly in molecular form, which is slightly soluble in water, and easily be absorbed by GAC. While when pH is bigger than 7, phenol exists in ion form, which is soluble in water and is difficult be absorbed by GAC.

This result also showed that the removal efficiency of phenol by adsorption of GAC is obviously superior to phenol salt.

Compared with GAC, GAC<sub>M</sub> is superior for removal



**Figure 7.** Effects of pH on the removal rate.

of phenol. The removal efficiency increased about 25%, and the equilibrium increased about 7 mg/g.

## 4. Conclusions

In order to improve adsorption properties of GAC, modification for GAC was induced by solution of potassium permanganate. By batch experiment, the removal efficiency of phenol by modified granular activated carbon was studied. The results show that:

1) Modification with potassium permanganate improved the adsorption capacity of GAC. Compared with GAC, the adsorption efficiency of Phenol with GAC<sub>M</sub> increased 20%.

2) The adsorption equilibrium time of GAC<sub>M</sub> is about 4h, as same as that of GAC. For 2.0 g GAC, the optimum concentration of potassium permanganate was 30 mg/L, and modification time was 90 min.

3) The adsorption of GAC/GAC<sub>M</sub> is exothermic reaction. Modification affects the adsorption efficiency of GAC, but don't affect the influence of temperature.

4) The adsorption property of GAC<sub>M</sub> was better in acidic and neutral than in alkaline condition.

5) The ratio of phenol to GAC/GAC<sub>M</sub> affects the adsorption efficiency of phenol with GAC/GAC<sub>M</sub>. The optimum ratio of phenol to GAC/GAC<sub>M</sub> was about 62.5:2.0 mg/g.

## 5. Acknowledgements

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