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# Regeneration of activated carbon saturated with chloramphenicol by microwave and ultraviolet irradiation

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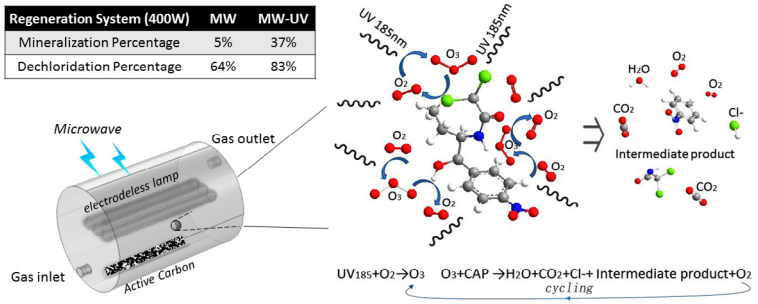
## HIGHLIGHTS

- Secondary pollution from the adsorbent regeneration process was reduced.
- Regeneration process was evaluated by mineralisation and dechlorination percentage.
- Chloramphenicol degradation included pyrolysis on GAC surface and degradation in air.
- Ultraviolet radiation is a key part to complete the mineralization process.
- In-situ production of ozone avoided nonuniform mix of organic compounds with ozone.

## GRAPHICAL ABSTRACT

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## ABSTRACT

To reduce the secondary pollution of volatile organic compounds generated from adsorbent regeneration, a regeneration method for granular activated carbon (GAC) saturated with chloramphenicol is studied. The saturated GAC is regenerated by microwave and ultraviolet irradiation which radiated from electrodeless lamps activated by microwave. The existence of ultraviolet rays can affect the degradation of chloramphenicol, and it is analyzed by adsorption capacity recovery percentage, mineralisation percentage and the amount of inorganic chloride. The saturated GAC is regenerated in microwave reactor at 2450 MHz for 10 min. The mineralisation percentage increases to 37% from 5% when adds the electrodeless lamp into the regeneration reactor. Besides, 83% of the total chloride in chloramphenicol can transform into inorganic chloride. Add ultraviolet radiation in microwave regeneration reactor can enhance the oxidizability of microwave regeneration method. Furthermore, the adsorption ability of GAC can maintain at a high level after five absorption/regeneration cycles. These results show a great potential use of microwave and ultraviolet irradiation to regenerate activated carbon saturated with chlorinated organic compound. This regeneration method can reduce the risk of secondary pollution of chlorinated organic compound during adsorbent regeneration.



## 1. Introduction

Adsorbent is widely used in wastewater treatment and air pollution control as its easy operation and environmentally friend [1,2]. These used adsorbents are always incinerated or discarded in a landfill after exhausted [3,4]. However, if not properly treated

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these adsorbents which exhausted with dangerous pollutants may result secondary pollution [5]. For economical and environmental consideration, it is necessary to regenerate the exhausted adsorbents. The regeneration of exhausted adsorbents such as activated carbon is widely studied [6–9]. As the regenerated by-products in regeneration process can result in secondary pollution. More attentions should be attracted on the elimination of the pollutants adsorbed on adsorbents and by-products in regeneration process.

At the present time, microwave irradiation has attracted many attentions on regenerating adsorbents, for its nature of molecular level heating which offers advantages such as less regeneration time and no thermal inertia [10]. Many researchers have investigated the microwave regeneration method to reduce regeneration time [8] or focus on energy conservation [7,11].

Some researches investigate the degradation of adsorbed organic pollutants by microwave, which focus on the relationship between microwave and organic pollutants. Xitao Liu [12] regenerated activated carbon by microwave, investigated the decomposition of pentachlorophenol and also identified the dechlorination products. Erdan Hu [13] investigated the degradation of atrazine sorbed in micropores of zeolites. As there are no improvements on the microwave regeneration equipment, the degradation of adsorbed pollutants only relates to the nature of organic pollutants. However, the by-products generated at the regeneration process may be more dangerous than the initial pollutants, especially if the organic pollutants contain chlorine [14,15]. For example, the intermediate products of chloramphenicol during degradation process using γ-irradiation are more toxic than chloramphenicol itself [16]. For these reasons, the development of environmentally friendly adsorbent regeneration techniques should be concerned.

Electrodeless lamp is a kind of ultraviolet lamp, which can be activated by microwave. It is easy to operate and have long service life for its electrodeless. Besides, ultraviolet rays are widely used in wastewater and air treatment [17,18]. Hence, the electrodeless lamp was added in the microwave regeneration reactor that would enhance the oxidizability of microwave regeneration method.

The potential threat of pharmaceuticals and personal care products to ecosystem is becoming a hot issue in recent years [19,20]. Chloramphenicol, a broad-spectrum antibiotic isolated from Streptomyces venezuelae in 1947 is widely used to treat human and animal diseases [21]. However, this use for diseases treatment has been restricted in China as it could cause fatal bone marrow depression and aplastic anemia [22]. Chloramphenicol is stable and could inhibit the activities of microbial, lead it not suitable for biological treatment. Therefore, the chloramphenicol is chosen as target pollutant for adsorption and degradation experiments. Activated carbon was used in the experiments for its microwave absorbing property and excellent adsorption capacity [23].

The widely used technique to regenerate activated carbon is under steam or an inert atmosphere. The purpose of this study is to improve the microwave regeneration method with ultraviolet rays, and regenerate activated carbon by microwave-ultraviolet under air atmosphere to oxidate the adsorbed organic pollutant. The effect of ultraviolet rays on the degradation of organic pollutant was also investigated, and results from the experiments are expected to be helpful for the secondary pollution decrease during activated carbon regeneration.

## 2. Experimental

### 2.1. Materials

Chloramphenicol (99%) was purchased from Aladdin Industrial Corporation, Shanghai, China, without further purification. Commercial coconut shell granular activated carbon (GAC) purchased from Xilong Chemical Company, Guangdong China, was used in this study. The GAC was immersed in 5% sodium hydroxide for 24 h to modify the GAC [24], then washed by deionized water to remove the sodium hydroxide. The GAC was dried at 105 °C for 24 h in Vacuum drying box before the experiment. Distilled deionized water was used throughout the experimental program. Air was used during the regeneration process. The airflow was pass through desiccant before pumped into reactor, the flow rate in regeneration experiments was 0.016 $m^3$/h.

### 2.2. Adsorption and regeneration

The adsorption experiments were conducted in screw-cap vials containing 1 g GAC and 200 mL chloramphenicol solutions. The screw-cap vials were shook at room temperature for 24 h. The concentration of chloramphenicol was 500 mg/L to ensure the active sites on GAC inner surface fully occupied (adsorption isotherm in Fig. SI2). The concentration of chloramphenicol was measured by ultraviolet–vis spectrophotometer (JENWAY-UV 6505). The saturated GAC was dried at 80 °C for 24 h after the adsorption experiment, and then regenerated the saturated activated carbon in microwave-ultraviolet reactor. The chloramphenicol will not evaporate and desorb from GAC at this temperature from the drying experiments. 5 g activated carbon was regenerated during regeneration experiment at 400 W microwave power supply. Microwave-ultraviolet reactor was made by quartz, and an electrodeless lamp was put into the reactor. To warm up microwave oven, microwave oven was turned on for 5 min before regeneration experiment. The microwave power used in experiments is continuous. Every parameter that could be affected by environment was evaluated by performing blank experiments (e.g. $M_t$). The amount of chloride ions generated in regeneration experiment was measured by Inductively Coupled Plasma-Atomic Emission Spectrometry. Carbon dioxide was detected and quantified by $Ba(OH)_2$ solution [25]. The concentration of ozone was detected by the ozone detector (GM Pro) purchased from Anshan Anseros Environmental Protection Co., Ltd. The surface groups of GAC were estimated by boehm method described in Ref. [26]. The determination of $pH_{pzc}$ was conducted by adjusting the pH of 50 ml 0.01 M NaCl solution to a value between 2 and 12. 0.05 g of activated carbon was added and the final pH was measured after 48 h under agitation. The $pH_{pzc}$ is the point where $pH_{initial} - pH_{final} = 0$. The adsorption capacity recovery percentage (δ), weight loss percentage (η), mineralisation percentage (ε) and the dechlorination percentage (ζ) were calculated according to following equations:

$$\delta = \frac{A_t}{A_0} * 100\% \quad (1)$$

$$\eta = \frac{W_t}{W_0} * 100\% \quad (2)$$

$$\varepsilon = \frac{Mt}{Mo} * 100\% \quad (3)$$

$$\zeta = \frac{Cl_t}{Cl_o} * 100\% \quad (4)$$

where δ is the adsorption capacity recovery percentage of the regenerated GAC; $A_0$ and $A_t$ are the adsorption capacity of GAC before and after the regeneration respectively. η is the weight loss percentage of GAC; $W_0$ and $W_t$ are the weight of GAC before and after regeneration respectively. ε is mineralisation percentage of adsorbed chloramphenicol; $M_t$ is the mol of $CO_2$ detected in regeneration process. $M_0$ is the mol of carbon atom in adsorbed chloramphenicol; ζ is dechlorination percentage of adsorbed chloramphenicol; $Cl_t$ is the mol of chloride ions detected in regen-