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# Facile synthesis of hierarchical porous carbon material by potassium tartrate activation for chloramphenicol removal

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## ABSTRACT

Potassium tartrate as activator was used to produce porous carbon materials (PCMs) by activating petroleum cokes (PC), fallen leaves (FL), chicken feathers (CF) and *Enteromorpha prolifera* (EP) under the identical conditions (activation temperature of 750 °C, impregnation ratio of 2.5:1 and activation time of 1 h). Potassium tartrate can also be self-activated to facilitate synthesize PCMs simultaneously. The PCMs synthesized through various precursors showed significant discrepancies in morphologies and the porous structure was characterized by SEM and N<sub>2</sub> adsorption-desorption isotherm. The BET surface areas of FL-PCM (1721 m<sup>2</sup>/g), CF-PCM (1819 m<sup>2</sup>/g), and EP-PCM (2151 m<sup>2</sup>/g) were larger than that of PC-PCM (256 m<sup>2</sup>/g) and the microporosity of EP-PCM (48%) was much lower than those of others. In addition, the adsorption properties of the four PCMs under various conditions were detected by selecting chloramphenicol (CAP) as a target adsorbate. The kinetics and isotherms of adsorption processes were well fitted by pseudo-second-order and Langmuir model for four PCMs. Meanwhile, EP-PCM exhibited high adsorption capacity of 892.86 mg/g.

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## 1. Introduction

Porous carbon is a classical material applied as high efficient adsorbent, electrode and catalyst carrier et al. The main characteristics of PCM are physical properties (surface area, pore volume and pore-size) and chemical properties (functional groups), which are principally affected by precursor, activator and activated condition [1]. There are various precursors to produce PCMs mainly containing petrochemicals [2], lignocellulosic biomass [3], keratoprotein [4] and algae [5] et al. The representatives of these categories are petroleum cokes (PC), fallen leaves (FL), chicken feathers (CF) and *Enteromorpha prolifera* (EP), respectively. PC is an unavoidable by-product with high carbon content (about 90 wt %) and low ash [2,6]. PC is a traditional material for producing PCM with low-cost. FL falls to the road especially in autumn and is traded as rubbish. FL contains various carbonaceous compounds and is almost free, accessible and renewable [7]. As a staring material, it can realize resource utilization and endow FL with addition value. CF is mainly treated as waste by landfill without sufficient use [4]. The principal ingredient of CF is keratin which is an excellent precursor to

prepare N-doped PCMs [4,8]. CF has been proved to be a fine material to prepare PCMs in previous studies [4]. EP, a type of green alga, is a product due to marine eutrophication, and composed of polysaccharose, protein and fat et al, which are easily pyrolyzed [9]. Hence, choosing these stuffs as the typical precursors can compare their effects for preparing PCMs.

KOH is a high-efficiency activator to produce PCMs with super-large surface area, but there still exists some deficiencies during the activating while the most prominent one is high corrosive level [7]. Hence, exploiting sylvite as activator is imperative to lower the deficiencies. Potassium tartrate is an organic sylvite which has been used to produce porous carbons with 1018 m<sup>2</sup>/g surface area by immediate carbonization previously [10]. Therefore, potassium tartrate could activate precursors and be self-activated to synthesize PCMs with high surface area. In addition, the secondary pollution caused by potassium tartrate was lighter than that of KOH, because the anion (tartrate anion) could work as carbon sources and it's consumed to synthesize the porous carbon. Based on the above, potassium tartrate was used to activate PC, FL, CF and EP, and self-activate to produce PCMs in this study.

With the vast usage of antibiotics, the harm to the environment caused by antibiotics has drawn more attention in recent years. Antibiotics are overused in stock farming due to lack in establishing relevant regulations [11]. Approximately 75% of administered

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drugs by animals is not assimilated and excreted through feces or urines into the environment [12]. Bacteria resistance for antibiotics has been an intractable puzzle and has caused enormous threat to human health [13]. Therefore, many technologies have been applied to remove antibiotics, such as photocatalysis [14], advanced oxidation [15] and PCMs adsorption [16]. Due to the high adsorption capacity, low cost and eco-friendly et al, PCMs adsorption is a valid method to remove the antibiotic wastewater [17].

Combined with the above viewpoints, primary targets of this study were to probe into the feasibility that potassium tartrate could activate precursors (PC, FL, CF and EP) and be self-activated to prepare PCMs, and to investigate the differences between PC-PCM, FL-PCM, CF-PCM and EP-PCM in characteristics of pore structure and morphology. Meanwhile, the activated mechanism of potassium tartrate involved in the preparation process was explored. In addition, the adsorption contrasts of CAP as a model antibiotic were investigated as a function of adsorption time, initial concentration and solution pH.

## 2. Materials and methods

### 2.1. Materials

All the chemical reagents were analytically pure and distilled water was used in the whole experiment. Potassium tartrate and chloramphenicol (CAP) were provided by Aladdin (Shanghai, China). The four waste materials were obtained locally, washed and smashed to powder.

### 2.2. Preparation of PCMs

The four raw materials were carbonized under 500 °C for 1.5 h using a carbonization furnace (KSY-4D-16) with an enclosed chamber to get char. 10 g char was mixed with 25 g potassium tartrate (an impregnation ratio of 2.5:1). Then the mixtures were put into a tubular resistance furnace (SKQ-3-10) where samples were heated to a requested temperature 750 °C with a constant heating rate of 10 °C/min in the existence of N<sub>2</sub> and then maintained for 60 min. When cooled down to room temperature, the products were washed by HCl, and distilled water until the value of pH kept constant. Finally, the samples were dried, ground and sieved to 100–200 mesh (0.074–0.15 mm).

### 2.3. Characterization methods

To understand the weight loss of different precursors during activation process, the four kinds of mixtures (potassium tartrate and char) were characterized by thermogravimetric analysis (TGA), (SHI-MADZU, TGA-50) and heated from room temperature to 750 °C with a rate of 10 °C/min and then kept at 750 °C for 60 min under N<sub>2</sub> atmosphere.

The pore structure properties of PCMs were measured by N<sub>2</sub> adsorption-desorption isotherms under 77 K using a surface area instrument (JW-BK 122W, Beijing JWGB Sci. & Tech. Co., Ltd., China). Meanwhile, the surface morphologies of the four PCMs were characterized by SEM (FEI, NOVA NANOSEM450, USA). X-ray photoelectron spectroscopy (XPS, ESCALAB 250) was used to investigate the surface chemical properties of PCM. X-ray diffraction (XRD, Rigaku D/MAX-YA) was employed in the 2θ range of 10–80°.

### 2.4. Adsorption experiments

In adsorption experiments, the influencing parameters of adsorption time, concentration and pH were explored for the four PCMs. For each sample, 0.050 g of PCM accurately weighed was added to 50 mL solution of CAP in 250 mL triangle bottle for each

sample, which was shaken in a constant temperature bath oscillator with a speed of 150 rpm. In kinetics studies, the samples owed 1000 mg/L initial concentration of CAP and 0.050 g PCM, were taken out according to a pre-set time interval. The residual CAP was segregated with PCMs by membrane-filter and its concentration was measured at 277 nm (maximum absorption wavelength of CAP) by an apparatus UV-vis spectrophotometer (UV-754, Shanghai). In thermodynamics experiments, the different initial concentrations (300–1000 mg/L) of CAP were adsorbed by the four PCMs under 293 K and natural pH for 24 h. To study the impact of pH, the pH of CAP solution (800 mg/L) was adjusted from 2 to 12 by adding the 0.1 mol/L HCl or NaOH solutions. The quantity of adsorbed CAP at adsorption equilibrium,  $q_e$  (mg/g) was calculated via the following equation:

$$q_e = \frac{(C_0 - C_e)v}{w} \quad (1)$$

where  $C_0$  and  $C_e$  (mg/L) are the original and equilibrium CAP concentration, respectively.  $v$ (L) is the CAP solution volume and  $w$  (g) is the PCMs weight.

## 3. Results and discussion

### 3.1. Thermal analysis

By TGA and DTG (differential TGA), the pyrolysis processes of four mixtures (potassium tartrate: char of precursor=2.5:1) are described in Fig. 1. The weight-loss showed the analogous tendency in the four compounds during pyrolysis from Fig. 1(a), which can be divided into three stages. The first section was before 300 °C, and the mass losses of PC-PCM, FL-PCM, CF-PCM and EP-PCM were 16.59%, 21.77% 20.81% and 23.17%, respectively, which was ascribed to the moisture losses (adsorbed and bound water) and degradation of low-boiling compounds [18]. When the temperature reached 750 °C, there was a section of weight losses for each sample (17.46% of PC-PCM, 15.78% of FL-PCM, 19.48% of CF-PCM and 17.04% of EP-PCM). There were numerous degradation reactions between four crude materials and potassium tartrate forming the cross-linked structure. In the third portion, the quality-losses after activating 60 min at 750 °C were 11.15%, 25.26%, 19.18% and 23.02% for PC-PCM, FL-PCM, CF-PCM and EP-PCM, respectively, which produced more porous structures by the catalysis of potassium tartrate. The total weight losses were 45.2%, 62.81%, 59.47% and 63.23% for PC-PCM, FL-PCM, CF-PCM and EP-PCM, respectively. The weight loss of PC-PCM was much less than those of others, which may be caused by the firm construction of PC and difficult degradation even in the existence of activation agent [1]. Meanwhile, the DTG curves have a similar trend in Fig. 1(b) except that a rapid weight loss appeared around 50 °C and no peak occurred in 227 °C for EP-PCM.

The synthesis mechanism of potassium tartrate activating PC, FL, CF and EP exhibited certain similarities with the activated process of directly carbonized organic salt or KOH chemical activation with comparison and analogy method [19,20]. When the temperature reached about 450–650 °C, the organic fraction of precursors was degraded and K<sub>2</sub>CO<sub>3</sub> was generated, meanwhile, the organic moiety of activated agent (tartrate) worked as carbon source. The existence of K<sub>2</sub>CO<sub>3</sub> was proved by XRD texting when potassium tartrate was directly carbonized at intermediate temperature of 650 °C (Fig. S1). Above 700 °C, K<sub>2</sub>CO<sub>3</sub> were transformed into K<sub>2</sub>O and CO<sub>2</sub>, and the gas of CO<sub>2</sub> reacted with C to form CO. This reaction can contribute to generate more pores via C gasification. Meanwhile, a vital reaction could generate K over 700 °C between K<sub>2</sub>O and C. K in the form of vapor possessed a significant function to insert into carbon matrix to form more porous structure by enlarging microstructure. Referred chemical reactions were listed as equations as followings:

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