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Preparation of hierarchical porous carbons from sodium carboxymethyl cellulose *via* halloysite template strategy coupled with KOH-activation for efficient removal of chloramphenicol

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ABSTRACT

The wide applications of antibiotics in human and veterinary medicines have attracted ever-increasing attention due to their potential adverse effects for ecological environment, therefore, it is urgent and important to efficiently remove antibiotics from water. In this paper, we prepared hierarchical porous carbons from sodium carboxymethyl cellulose (CMCHPCs) *via* halloysite template and KOH-activation method for efficient removal of chloramphenicol. The physicochemical properties of CMCHPCs are analyzed by means of different characterizations. The adsorption properties of CMCHPCs are evaluated by equilibrium, kinetic, and thermodynamic adsorption experiments. In addition, the effects of ionic strength, solution pH, and humic acid on adsorption are studied. The results indicated CMCHPCs exhibit higher maximum monolayer adsorption capacity up to 769.95 mg/g and shows good regenerability and adaptability. Importantly, the adsorption mechanisms of chloramphenicol adsorbed onto the CMCHPCs are also discussed.

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1. Introduction

The widespread applications of pharmaceuticals in human and veterinary medicines have drawn considerable attention because of their potential adverse effects in the long-term for ecological environment, such as leading to emergence of antibiotic resistant genes, effects in aquatic organisms [1–3]. Therein, antibiotics have received ever-increasing concern due to a wide usage in global scale. Chloramphenicol (CAP), a broad-spectrum antibiotic for Gram-positive and Gram-negative cocci and bacilli with excellent antibacterial properties, has been widely used in the treatment of human and animal diseases [4]. Generally, many antibiotics cannot be completely metabolized by organisms, which are mostly discharged into the environment through feces and urine maintaining biological activity. Recently, more and more antibiotic residues have been detected in sewage wastewater, hospital wastewater, even drinking water [5]. The antibiotic residues in water environment will lead to serious threat to the ecosystem and human health. Therefore, it is urgent and important to efficiently remove antibiotics from water.

Considerable efforts have been devoted to treat antibiotic wastewater, such as advanced oxidation method [6], photocatalytic degradation [7], adsorption [8], and bio-degradation [9]. Among them, adsorption method is favored by researchers because of economic and environmental considerations. The adsorbents with excellent adsorption capacity are crucial for removal of contaminations. Compared with other adsorbents, porous carbon materials possess excellent chemical and thermal stability, high specific surface area, which are increasingly used in different fields including electrochemical, catalysis, and energy storage [10–12]. Notably, the porous carbon materials are also used as potential adsorbents of high binding affinity and capacity for antibiotics from water environment owing to their remarkable physicochemical properties.

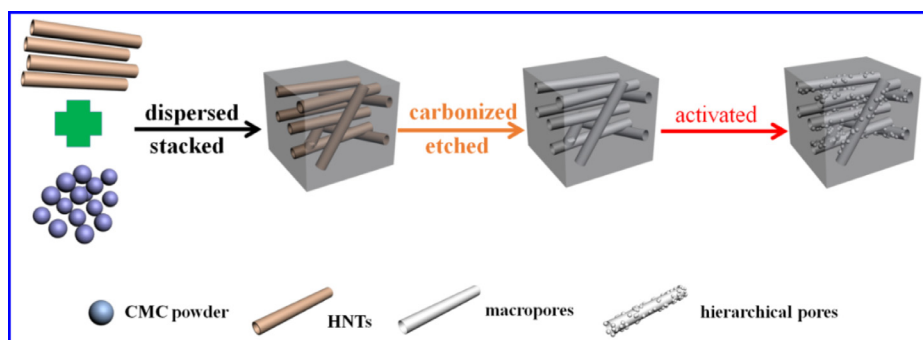
Currently, researchers increasingly prefer to prepare porous carbon materials using low-cost materials as carbon precursor for removal of organic pollutants. Such as, Fan et al. [13] reported the adsorption of chloramphenicol in wastewater on the renewable bioresources of bamboo charcoal. Qin et al. [14] reported porous carbon derived from sustainable shrimp shell for high-performance removal of sulfamethazine and chloramphenicol. Gundogdu et al. [15,16] produced activated carbon by chemical activation using zinc chloride from tea industry wastes. This activated carbon shows good adsorption capacity for phenol molecules from aqueous

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Scheme 1. Schematic illustration of the fabrication of hierarchical porous carbons from sodium carboxymethyl cellulose via halloysite template strategy coupled with KOH-activation.

solution. Habila et al. [17] reported activated carbons from waste (palm, paper, and plastic wastes) as an efficient adsorbent for removal of malathion from environmental samples. These economic porous carbon materials show great promising application in the removal of organic pollutants from water. Thus, it is of great significance to prepare porous carbon materials using low-cost and renewable biomass materials. Sodium carboxymethyl cellulose (CMC) is a nontoxic and renewable material derived from natural cellulose. CMC has been used in different fields including water treatment, pharmaceuticals release, etc. for their rich sources, excellent biodegradability, biocompatibility, bioactivity, and environment friendliness [18]. However, there are no literatures about the preparation of porous carbon materials using CMC as carbon precursor. Based on this point, rational utilization of cellulose-based materials is very significant in energy and environment fields.

Hierarchical porous carbons (HPCs), a family of carbon materials combined advantages of different pore types, which are facilitated to the adsorption of adsorbates [19]. Currently, template method has been widely used in preparation of HPCs. Halloysite nanotubes (HNTs), a class of regular nano-tubular structure natural mineral, are considered as ideal hard template candidate for preparation of HPCs because of their cost-efficient, environmentally friendly and abundant reserves [20]. Thus, HNTs was chosen as sacrificial template for construction of HPCs considering their availability in large-scale.

Herein, reported is a mineral template and KOH-activation strategy to prepare the CMC-based hierarchical porous carbons (CMCHPCs) for efficiently removing CAP antibiotic from water environment, in which the CMC served as biomass carbon precursor, HNTs as sacrificial template and KOH as activating agent, the fabrication process as shown in Scheme 1. In fabrication process, the HNTs-template simply serves as a scaffold around CMC via hydrogen bonding for existing hydroxyl onto surfaces of two materials. The HNTs-template directs the formation of pores during the carbonization process. The carbonaceous material formed after carbonization turns into a continuous carbon framework, while the space once occupied by the host template is transferred into the macropores. The KOH attached on the inner cavity of macropores will react with the carbon atom to produce micropores or mesopores. The CMCHPCs exhibits a hierarchical pore structure, and their high specific surface area and surface groups lead to an enhanced adsorption capacity and adsorption kinetics.

2. Experimental

2.1. Materials

HNTs were received from Zhengzhou Jinyangguang Chinaware Co., Ltd. (Henan, China) with purification procedure by grinding, sieving, sedimentation and pickling. Potassium hydroxide (KOH,

AR) and hydrochloric acid (HCl, 36–38 wt%) hydrofluoric acid (HF, AR, $\geq 40\%$) were provided by Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). Ammonium hydroxide ($\text{NH}_3 \cdot \text{H}_2\text{O}$, 25–28%), sodium carboxymethyl cellulose (CMC, 300–800 mPa s, USP) and CAP (AR, 98%) were purchased from Aladdin Reagent Co., Ltd. (Shanghai, China). All antibiotic solutions in the entire experiment were prepared using deionized water.

2.2. Instruments and characterization

The carbonization and activation process use a tube furnace (SK-GO6123K, TIANJIN ZHONGHUAN Experiment Electric Furnace CO., LTD., China) in N_2 atmosphere. The microstructure of samples was observed by field emission scanning electron microscope (EF-SEM; S-4800, Hitachi, Japan) and transmission electron microscopy (TEM; JEM-2100, JEOL, Japan). X-ray diffraction was conducted on specimens using an X-ray diffractometer (XRD, D8 CEVANCE, Bruker AXS, Germany) operating at 40 kV and 40 mA. Nickel-filtered Cu K α radiation was used in the incident beam. Raman spectroscopy was examined with a Laser Raman Spectrometer (DXR, ThermoFisher, USA). The specific surface area and pore size distribution of samples were calculated from the N_2 adsorption-desorption isotherms at 77 K by multi-point BET and DFT method using a BELSORP instrument (BELSORP, BEL Japan Inc., Japan). X-ray photoelectron spectroscopy (XPS; AXIS ULTRA DLD, SHIMADZU, Japan) analysis was carried out in a Kratos Axis Ultra DLD spectrometer, using monochromated Al K α X-rays, at a base pressure of 1×10^{-9} Torr. Survey scans determined between 1000 and 0 eV revealed the overall elemental compositions of the sample and regional scans for specific elements were performed. The peak energies were calibrated by placing the major C1s peak at 284.5 eV. Samples were prepared identical to those of the batch experiments.

2.3. Preparation of CMCHPCs

The typical procedure is as follow: CMC (4 g) was added in deionized water (200 mL) under magnetic stirring at 45 °C to form transparent and uniform solution, and then HNTs (8 g) was added to the solution with vigorous stirring for 30 min to evenly disperse. The mixed solution was poured into the suction bottle vacuum degassing for 30 min, then, it was slowly poured into a dish and evaporated at 100 °C to get solid mixture (CMC-HNTs). After that, right amount of the CMC-HNTs was treated at 500 °C in a tubular furnace for 2 h with a ramp of 5.0 °C/min under N_2 flow. The products were soaked in HF solution (5.0 wt%) for removing HNTs template, the carbonized CMC (CCMC) was obtained after washing several times with deionized water to neutral and drying at 60 °C. Subsequently, CCMC and KOH were evenly grinded with mass ratio

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