



Antibacterial action of silver-doped activated carbon prepared by vacuum impregnation

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ABSTRACT

Silver-containing activated carbon (Ag/AC) exhibiting controlled release of silver and antibacterial action was prepared by vacuum impregnation using acetate silver as a precursor. The antibacterial activity toward *E. coli* and resistance to water erosion were investigated with the view of water purification. N₂ adsorption at 77 K, scanning electron microscopy and X-ray diffraction were used to characterize the surface morphology and crystalline properties of the Ag/AC samples. As the concentration of CH₃COOAg increased, the samples change from exhibiting no antibacterial activity to inhibition of bacteria growth and then to antibacterial activity because of the higher silver content and smaller size of the silver particles. The Ag/AC composites showed a lower release rate of silver than that of a composite prepared by a traditional AgNO₃ impregnation method, which suggests a strong interaction between the silver particles and carbon. Because the Ag particles block the pores of AC, the BET surface area, total pore volume and average pore diameter of the Ag/AC samples decreased as the concentration of the CH₃COOAg solution increased. The higher antibacterial activity and controlled release of silver by Ag/AC containing 1.65 wt % silver means that it shows promise for purification of drinking water.

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1. Introduction

Activated carbon (AC) has been widely used in water treatment to remove organic and inorganic pollutants because of its high surface area, high amount and rate of adsorption, and specific surface reactivity [1]. However, when AC is used to purify drinking water, bacteria preferably adhere to it because of its good biocompatibility [2–4]. Bacteria may then breed on the AC during the purification process so that the AC itself becomes a pollutant [2,5,6]. The World Health Organization recommends that any water intended for drinking should contain fecal and total coliform counts of 0 in any 100 mL sample [1]. To achieve this, antibacterial ACs are required.

The antibacterial effects of metallic silver have been known for centuries. This beneficial property originates from silver ions dissolving from the surface of bulk silver [7–9]. The oxidation of metallic silver to active silver ions proceeds through the interaction of silver with water molecules [10–12]. Silver ions possess significant antiseptic properties [13–15], with only a few types of bacteria being intrinsically resistant to this metal.

Silver-based antibacterial composites have to release silver ions into a pathogenic environment to be effective [3,7]. To use silver/activated carbon (Ag/AC) to purify drinking water on a

practical scale, three factors need to be optimized [2–4]: surface area, antibacterial activity and the amount of silver lost. To date, most ACs loaded with silver have been prepared by impregnation with AgNO₃, which involves soaking AC in a solution of AgNO₃ followed by reduction with agents such as formaldehyde and/or hydrazine or simply by heating [2]. For Ag/ACs prepared by this method, large silver particles are usually distributed unevenly on the surface of the AC [2,16,17]. When used for water purification, the Ag content of Ag/AC decreases remarkably as the contact time with water increased. Such drastic removal of silver is mainly because of the low resistance of the Ag/AC to attrition, which caused silver particles to dissociate from the carbon surface. As a result, the antibacterial activity of the Ag/AC will decrease over time. Also, it is difficult to use bulk silver in industrial or domestic applications because of its high price and low rate of ion release. Recently, the use of silver nanoparticles has allowed a new approach for the application of silver antimicrobial agents [18–20]. The rate of release of silver ions is proportional to the surface area of the exposed silver nanoparticles. Compared with silver ions, silver nanoparticles are long lasting, and are subject to controlled release [19]. In addition, they are cheap and allow controlled rate of release of silver ions compared with bulk silver [18,20,21]. Therefore, to obtain high performance antibacterial carbon materials, it is extremely important that the size and morphology of loaded silver particles can be controlled. The design of silver materials with long-term dispersion stability and excellent antimicrobial efficiency has become a primary challenge for Ag/AC preparation.

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Thermal decomposition of metal carboxylates is now widely used for the preparation of highly dispersed metals and metal oxides; synthesized metals can be used as catalysts. Nanoparticles of metal can be obtained in such processes because of rather low temperature of this intramolecular reduction of metals [22].

Vacuum impregnation (VI) of a porous product consists of exchanging the internal gas or liquid occluded in open pores for an external liquid phase by the action of a hydrodynamic mechanism [23]. Compared with traditional impregnation methods, VI can realize the uniform deposition of silver on both the internal and exterior surfaces of porous refractory catalyst supports [24,25]. Such supported silver products are effective as a catalyst for the direct oxidation of ethylene to ethylene oxide.

In this work, Ag/AC containing loaded silver nanoparticles and showing a controllable release rate of silver ions and antibacterial behavior was prepared by VI using silver acetate as a precursor. The antibacterial activity of the Ag/AC sample toward *E. coli* and resistance to water erosion were investigated from the perspective of water purification.

2. Experimental

2.1. Preparation of Ag/AC

Commercial coconut AC ($S_{\text{BET}} = 876.2 \text{ m}^2 \text{ g}^{-1}$, particle size 2–4 mm) prepared by a traditional two-step physical activation was used. The AC was designed specifically for water treatment, and was generously supplied by Jianxin Company (Tangshan, China). Prior to use, the carbon sample was washed with distilled water and dried in an oven at 105°C for 24 h; this sample is referred to as AC_{raw} .

Ag nanoparticles were deposited on the porous AC using CH_3COOAg as a precursor by means of VI. The AC sample (5.0 g) was maintained at a pre-vacuum of 52 kPa for 60 min and then placed in an aqueous solution of CH_3COOAg (50 mL, concentration of 1.0, 2.0, 4.0 or 8.0 g L^{-1}) under atmospheric pressure at 25°C for 60 min to allow diffusion of the impregnation material. The obtained solid was washed with distilled water, vacuum dried at 60°C for 24 h, and then calcined at 500°C for 2 h under a nitrogen atmosphere. The prepared solids are denoted as Ag/AC_1 , Ag/AC_2 , Ag/AC_3 and Ag/AC_4 , which correspond to concentrations of CH_3COOAg of 1.0, 2.0, 4.0 and 8.0 g L^{-1} , respectively. Reference Ag/AC prepared by impregnation of AgNO_3 [2,26] was used for comparison and is named $\text{Ag/AC}_{\text{ref}}$.

2.2. Determination of silver content

Each Ag/AC sample (about 0.1 g) was dried under vacuum until it maintained a constant weight and then placed in a flask. Nitric acid (10 mL, 16 mol/L) was added to the flask and the mixture was boiled for about 10 min. The suspension was filtered and thoroughly washed with deionized water. The resulting solution was collected and diluted to 100 mL with deionized water. The concentration of silver in the obtained solution was determined using an atomic absorption spectrophotometer. The content of silver in each sample was then calculated according to the concentration of silver and weight of the sample.

2.3. Characterization

The Brunauer, Emmett, and Teller (BET) surface area (S_{BET}), total pore volume and pore size distribution (PSD) of the samples were obtained from N_2 adsorption–desorption isotherms measured at 77 K using a constant volume adsorption apparatus (Micromeritics 2020, Micromeritics, USA). Prior to measurement, the samples

were out-gassed at 200°C under a flow of N_2 for 2 h at a pressure in the range 10^{-6} – 10^{-5} Torr. The S_{BET} values of the samples were calculated by the BET method using adsorption isotherms in the range $0.05 \leq \text{relative pressure } (p/p_0) \leq 0.30$. The total pore volume of the samples was calculated from the volume of adsorbed N_2 at $p/p_0 = 0.99$. The PSDs of the samples were calculated by the Barrett, Joyner, and Halenda (BJH) method. The particle size and morphology of the resulting composites were characterized using an environmental scanning electron microscope (SEM, Quanta200, FEI, the Netherlands). X-ray diffraction (XRD) measurements were performed on a Rigaku D/max-rB diffractometer (Rigaku, Japan) using $\text{CuK}\alpha$ radiation ($\lambda = 1.54178 \text{ \AA}$).

2.4. Antibacterial activity

The antibacterial action of the samples was tested using the plate counting method. The Ag/AC sample was sterilized in an autoclave at 120°C for 20 min prior to starting the experiment. Ag/AC (2.0 g) was placed in distilled water (100 mL) containing about 10^7 CFU (colony forming units) mL^{-1} of *E. coli*. The mixture was aerobically incubated at 37°C under agitation. At predetermined intervals, 1 mL of the treated solution was removed and diluted with distilled water to a certain volume (to adjust the bacterial concentration of the solution to ensure that the bacterial colonies could be easily and correctly counted). The diluted solution was then spread on a Mueller–Hinton (MH) agar plate (15 mL) and the microorganisms were cultivated at 37°C for 24 h. The number of bacterial colonies on the plate was then counted.

2.5. Water erosion test

The resistance of the Ag/AC samples to water erosion was determined by an attrition test. Distilled water (100 mL) and an Ag/AC sample (2.0 g) was added to a 250 mL conical flask and then shaken at rate of 150 rpm at 25°C . The resistance of each Ag/AC sample to attrition was characterized by the change in the silver content before and after washing for a defined period.

3. Results and discussion

3.1. Pore structure and silver content

Fig. 1 shows adsorption–desorption isotherms of N_2 obtained for AC_{raw} and Ag/AC at 77 K. For AC_{raw} , Type I nitrogen adsorption–desorption isotherms were obtained, and the slope

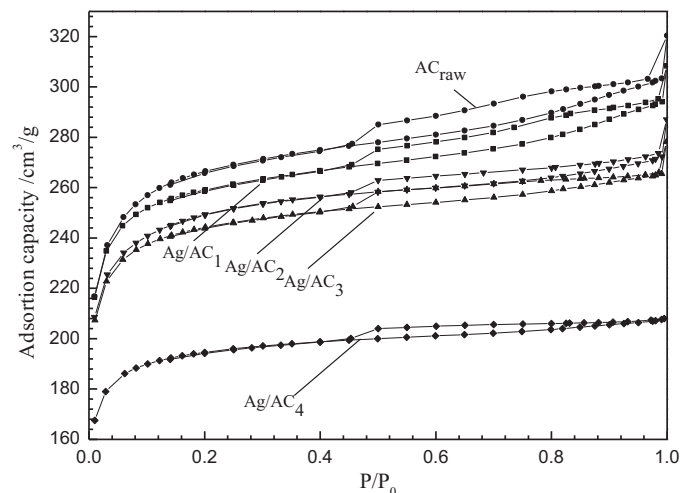


Fig. 1. N_2 adsorption–desorption isotherms of AC_{raw} and Ag/AC.

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