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# Preparation and properties of Ag-coated activated carbon nanocomposites for indoor air quality control

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ABSTRACT

Activated carbon (AC) has been widely used in indoor air quality (IAQ) control for removal of hazardous volatile organic compounds (VOCs). A detrimental effect of this adsorption technology is that bacteria multiplied on AC may deteriorate IAQ. In this paper, antibacterial AC nanocomposites with well-dispersed silver nanoparticles (Ag/ACs) were prepared by the attachment of $Ag^+$ on the functionalized AC surface via ion–dipole interactions and the subsequent *in-situ* reduction of $Ag^+$. The surfaces and microporous structures of the obtained Ag/ACs were analyzed by means of scan electron microscope (SEM) and pore size surface area analysis. Antibacterial tests were performed using *Escherichia coli* (*E. coli*) and *Staphylococcus aureus* (*S. aureus*) as model bacteria. Antibacterial activity against airborne bacteria and toluene adsorption capacity of AC nanocomposites were further evaluated. It was found that the introduction of Ag nanoparticles significantly improves antibacterial effect of AC but slightly reduces toluene adsorption ability. Ag/ACs can efficiently kill bacteria within 100 min without decreasing adsorption ability toward toluene.



## 1. Introduction

Indoor air quality (IAQ) remains a very important issue today due to the fact that most people spend an average of 90% of their time in enclosed buildings. Volatile organic compounds (VOCs) released from building materials and furniture are regarded as major source of indoor air contaminants, which significantly impact indoor air quality and pose a health threat. A long-term exposure to VOCs will be detrimental to human health causing sick building syndrome (SBS) such as headaches, dizziness, nausea, or allergic reaction [1,2]. There are a number of technologies available for VOCs abatement, among which, the adsorption-based air cleaning technology has been employed in various applications [3–6].

Activated carbon (AC) is the most widespread adsorbent to eliminate VOCs because of its large surface area and outstanding adsorption capacity [7–9]. Although AC cannot effectively collect airborne microorganisms, some fraction of airborne bacteria can deposit on the AC. The deposited bacteria are easy to multiply on the AC surface because carbon materials have high biocompatibility [10]. As a result, indoor airborne bacteria accumulate in large quantities on its surface and consequently deteriorating IAQ because AC itself can become a source of bacterial contamination. On the other hand, VOCs produced by bacterial metabolism can be emitted from the contaminated AC. This inevitably brings the secondary indoor air pollution. Therefore, antibacterial AC, which can remove VOCs but also kill bacteria, is required for good IAQ.

Silver is well known as a potential antimicrobial agent because of its broad-spectrum antibacterial activity [11–13]. Many silver-containing carbon composites have been developed in antibacterial application using sol–gel method and Ag-coated method [14–20]. For Ag-coated method, Ag nanoparticles are loaded on AC by directly mixing Ag nanoparticles solution with AC or depositing gas-phase Ag nanoparticles. Differently, Ag ions are loaded on AC, and subsequently in-situ reduced to Ag nanoparticles for sol–gel method. Although these materials cannot collect airborne microorganisms, they can still exhibit good antibacterial effect for deposited bacteria. Major limitations for preparation of these composites are aggregation of Ag particles and tedious reaction process. The aggregation of Ag particles may reduce antibacterial activity since homogeneous dispersion is a prerequisite for sufficient contact and interaction between Ag and microbial species [11]. Besides, the large-size Ag particles generated from aggregation can block micropores and reduce the adsorption capacity. Currently, challenging issues for silver-containing carbon composites are the uniform and well-adherent coating of Ag particles on

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**Table 1**
Preparation parameters of Ag/ACs.

| Additive/wt% | Ag/AC-1 | Ag/AC-2 | Ag/AC-3 | Ag/AC-4 |
|---|---|---|---|---|
| $AgNO_3$ | 0.5 | 1 | 2 | 4 |
| $Na_3Ct$ | 0.75 | 1.5 | 3 | 4.5 |
| DMEA | 0.03 | 0.06 | 0.12 | 0.18 |

carbon surface to achieve highly efficient antibacterial effect and good adsorption capacity.

In this paper, Ag-coated AC nanocomposites (Ag/ACs) with well-dispersed silver nanoparticles were prepared by the attachment of $Ag^+$ on the functionalized AC via ion–dipole interactions and the subsequent *in-situ* reduction of $Ag^+$. In this approach, oxygen-containing groups on the functionalized AC surface serve as the dispersing and stabilizing effect of $Ag^+$, thus suppressing the coagulation and overgrowth of Ag particles in the next reduction step. Hence, Ag nanoparticles can be well distributed on AC surface. The investigation of antibacterial activity and adsorption capacity for toluene as model VOCs show that the introduction of uniform Ag nanoparticles on AC not only can provide highly efficient antibacterial activity, but also can maintain high adsorption capacity.

## 2. Experimental

### 2.1. Materials

Coconut shell activated carbon (AC) was purchased from Tianjin Kemel Chemical Co. Ltd., China. Silver nitrate ($AgNO_3$), trisodium 2-hydroxypropane-1, 2, 3-tricarboxylate hydrate ($Na_3Ct$), and dimethylethanolamine (DMAE) were from Sinopharm Chemical Reagent Co. Ltd., China. Typtone, agar, and yeast extract were obtained from Guangzhou Jianyang Biotech. Co. Ltd., China. Deionized water was used throughout the experiments.

### 2.2. Preparation of Ag-coated AC nanocomposites (Ag/ACs)

According to the described procedure in reference [21], AC was oxidized by concentrated $HNO_3$ (69 wt% $HNO_3$) to obtain functionalized AC (F-AC) with oxygen-containing groups. The obtained F-AC (1 g) was impregnated in 50 mL $AgNO_3$ solution and stirred for 1 h at room temperature. After filtration, AC loading $Ag^+$ ($Ag^+$/AC) was obtained. The resultant $Ag^+$/AC was added to 50 mL of trisodium citrate ($Na_3Ct$) solution. And then 1 mL dimethylethanolamine (DMEA) solution was added dropwise to the mixture, and the reaction system was stirred for 3 h at 90 °C. The solid was then separated by filtration and washed with deionized water. Subsequently, sample was dried under vacuum at 70 °C for 12 h to yield Ag-coated AC nanocomposites (denoted as Ag/ACs). Preparation parameters of Ag/ACs are listed in Table 1.

### 2.3. Characterization

The surface morphology and microscopic elemental analysis of Ag/ACs were examined using scan electron microscope (SEM, LEO 1530 VP) in combination with an energy-dispersive X-ray spectroscopy (EDS) probe for microscopic elemental analysis. The EDS line scans were performed in the range from 0 to 20 kev. Nitrogen adsorption isotherms measured at 77 K with ASAP 2010 instrument (Micromeritics Instrument Corporation, Norcross, GA, America) were used to determine BET surface area and pore volume of Ag/ACs. Ag content in the Ag/ACs samples was determined using a HITACHI Z-5000 atomic adsorption spectroscopy. The toluene concentration in chamber was determined by gas chromatography (GC) on an *Agilent* 7890A gas chromatograph spectrometer.

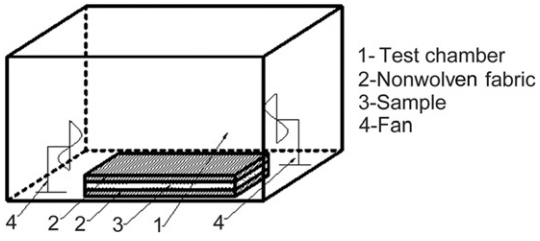


**Fig. 1.** Test rig for antibacterial activity against airborne bacteria.

### 2.4. Antibacterial activity

The antibacterial activities were investigated using a surface spread-plate method toward *Escherichia coli* and *Staphylococcus aureus* [21]. Control experiment was conducted to quantify the colony forming units (CFU) of bacteria strains in the blank samples. 100 mg of sample was dispersed in 30 mL containing $10^7$ colony forming units (CFU) $ml^{-1}$ of *E. coli* or *S. aureus*, and then shaken at 37 °C for the prescribed contact time of 0, 30, 60, 90, 120, 150, and 180 min 1 mL of bacteria culture was withdrawn and decimal serial dilutions with sterile phosphate-buffered solution were repeated with each initial sample. A 0.1 mL drop of the diluted sample was spread onto agar plate. After incubation of the plates at 37 °C for 24 h, the number of viable cells (colonies) on each plate was counted. All antibacterial tests were carried out in triplicate to ensure reproducibility.

### 2.5. Antibacterial activity against airborne bacteria

Fig. 1 shows a schematic diagram of experimental setup for antibacterial activity for airborne bacteria. As shown in Fig. 1, 5 g of sample (AC or Ag/ACs) sandwiched by nonwoven fabric was placed in the clean and airtight chamber (1 $m^3$) filled with the contaminated air. In view of Chinese health criterion of pollution air (1000 CFU/$m^3$), the moderately contaminated air with 1600 CFU/$m^3$ airborne bacteria was used in the experiment. The number of airborne bacteria of the contaminated air was kept 1600 ± 50 CFU/$m^3$ by mixing clean air and pollution air source. The air was fanned in order to keep uniformity of airborne bacteria in the air. The air was sampled and the airborne bacteria of samples were counted every 20 min. The number of airborne bacteria was counted using natural precipitation method on the basis of a closed air system [22]. Nutrient agar medium was exposed for 5 min in the contaminated air, and then cultured for 48 h at 37 °C. The number of bacterial colonies on each medium was counted. All tests were performed in triplicate. The antibacterial activity was represented by the time-related number of airborne bacteria in the presence of AC or Ag/ACs.

### 2.6. Toluene adsorption capacity test

Toluene adsorption capacity was measured by static adsorption experiment. A known mass of AC or Ag/ACs (0.5 g) was placed in the closed chamber (500 L) filled with gaseous toluene/$N_2$ under the conditions of room temperature and atmosphere pressure. The gaseous toluene was generated by bubbling liquid toluene at 0 °C using a nitrogen gas flow. The toluene concentration in the chamber was determined by gas chromatography (GC) and controlled at the level of 0.5 mg/L by diluting with $N_2$. Adsorption test was kept at room temperature for 2 days. After 2 days, sample was weighed every 6 h and until the mass of sample remains constant. When the adsorption equilibrium was reached, the amount adsorbed at equilibrium was calculated by the following equation:

$$q_e = \frac{m_e - m_0}{m_0} \quad (1)$$