Building and Environment 125 (2017) 60–66

Contents lists available at ScienceDirect

Building and Environment

journal homepage: www.elsevier.com/locate/buildenv

Carbon nanotubes / activated carbon fiber based air filter media for simultaneous removal of particulate matter and ozone

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ARTICLE INFO

Article history:
Received 2 July 2017
Received in revised form
2 August 2017
Accepted 19 August 2017
Available online 24 August 2017

Keywords:
Particulate matter
Ozone
Carbon nanotubes
Activated carbon fiber
Air cleaning

ABSTRACT

Indoor environment is faced with complex pollution by particulate matter (PM) and ozone. In this study, we proposed to use carbon nanotubes/activated carbon fiber (CNTs/ACF) to remove both PM and ozone with high removal efficiencies, high quality factor and low pressure resistance. The CNTs/ACF filter media were fabricated through growth of CNTs upon pristine ACF using chemical vapor deposition method. The PM filtration efficiencies of the CNTs/ACF and pristine ACF were measured for different particle sizes. The quality factors of these two media were calculated and compared. The ozone removal efficiency of the CNTs/ACF and ACF were tested as well. Results indicate that growth of CNTs on ACF elevated the media for PM filtration, and increased the quality factor by 48%. The CNTs/ACF presented >99% ozone removal, as high as the ACF. Compared with other CNTs-based filter media that have been proposed in previous studies, the CNTs/ACF exhibited higher quality factor, much lower pressure resistance, higher ozone removal efficiency, and upper-middle PM filtration efficiency.

1. Introduction

Increasing ambient air pollution has raised great concerns globally, and particularly in developing countries [1]. Exposure to ambient air pollution is significantly associated with mortality and morbidity [2]. Among major ambient air pollutants, particulate matter (PM) and ozone attract special attention, since air pollution caused by these two pollutants is visible as haze and photochemical smog, which both historically and presently have aroused public anxiety [3,4]. Adverse effects on human health resulting from PM and ozone exposure include respiratory symptoms, cardiovascular effects, and even cancer [5,6]. Outdoor PM and ozone can enter indoor environments via ventilation and infiltration [7]. Additionally, indoor pollutant sources, such as cooking for PM and electrostatic air cleaners for ozone [8], may lead to elevated indoor concentrations, even higher than outdoor concentrations in some scenarios [9,10]. Furthermore, because humans spend most of their time indoors, exposure to indoor PM and ozone may be a more serious issue [11–13]. Moreover, indoor ozone-initiated surface and gaseous reactions can lead to secondary emissions of PM [14–16] and volatile organic compounds (VOCs) [17–19] that may be more harmful than ozone itself [20]. Hence, it is necessary to remove PM and ozone from indoor environments.

High-efficiency particulate air (HEPA) filters and activated carbon (AC) filters are widely used to deal with indoor PM and ozone pollution, respectively. These two filters are often placed back to back in building ventilation systems and individual air cleaners to remove PM and ozone from indoor environments. Most commercial HEPA filters are made of glass fiber and expected to have a filtration efficiency >99.97% for 0.3 μm airborne particles [21] through direct interception, Brownian diffusion, inertial impact, gravitational settling, or electrostatic deposition [22]. AC filters can remove ozone effectively [23,24] through reactions with oxide groups on the surface of the AC [25,26] and the catalytic action of AC in ozone decomposition [27,28]. AC combined filters, consisting of fiber media and AC, were used for simultaneous particle and ozone removal as well [29–32]. Though the use of such filters has been shown to remove PM and ozone in common indoor conditions, several problems still exist: a sharp decrease in removal efficiency resulting from high humidity [33], high energy consumption resulting from the high pressure resistance of HEPA filters particularly for situations in demand of maintained airflow [34,35], and secondary pollutants resulting from ozone reaction on the filters [36]. Therefore, there is constant demand to develop new

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http://dx.doi.org/10.1016/j.buildenv.2017.08.040
0360-1323/

approaches for improved PM and ozone removal.

Carbon nanotubes (CNTs) have been widely applied since their discovery, due to their superior properties [37–39]. In recent years, CNTs were introduced to the field of PM and ozone removal. Previous studies have proposed several structural designs of CNTs-based air filters for PM filtration [35], such as CNTs coating [40], free-standing CNTs film [41], aligned CNTs sheet [42], hierarchical CNTs structure, and agglomerated CNTs fluidized bed [43]. Among these designs, hierarchical CNTs structure took advantage of macroporous structure and high structural stability of the porous substrate by simple fabrication methods, providing good application prospects. Studies have treated micron-sized fibrous metal filters [44], glass fiber filters [45] and ceramic filters [46] as substrates for CNTs growth to obtain hierarchical CNTs-based air filters.

Addition of CNTs has been shown to increase PM filtration efficiency to some extent. However, filtration efficiencies at the most penetration particle size (MPPS) of these filters were lower than 80%, which is not sufficient to control indoor PM levels. Other researchers have studied micromachined Si/SiO_2 [47] and quartz fiber [48] as substrates, obtaining filters with >99% filtration efficiency at MPPS. However, the high pressure resistance of these filters may limit their applications.

In our previous study, the CNTs/quartz fiber film presented effective removal of ozone with a higher removal efficiency than that of granular AC and KI solution with the same weight [49]. Nevertheless, high pressure resistance of the filter medium was a major problem and can hamper commercial applications. Hence, it is of great interest to develop improved CNTs-based air filter media with high PM and ozone removal efficiencies but low pressure resistance.

This study intends to introduce activated carbon fiber (ACF) as the substrate for CNTs growth to obtain better removal performance for both PM and ozone. The CNTs/ACF was fabricated through growth of CNTs upon pristine ACF using the chemical vapor deposition method. The PM filtration efficiencies of the CNTs/ACF and pristine ACF were measured at different particle sizes. The ozone removal efficiencies and pressure resistances of these two materials were measured as well, and their quality factors were calculated and compared.

2. Materials and methods

2.1. Fabrication of the CNTs/ACF filter media

The pristine ACF filter media (Lianbin Environmental Protection Company, China) were used directly as the substrate for in-situ CNTs growth using the floating catalyst chemical vapor deposition method. In a typical process, an ACF filter (50 mm diameter and 2.94 mm thickness) was placed in the middle of a quartz tube (38 mm inner diameter and 180 cm length). Ferrocene (purity > 99.99%, Tianjin Damao Chemical Reagent Factory, China), acting as the catalyst, was introduced at the entrance side of the quartz tube inside a heater (TF55030C-1, Thermal scientific, USA). With the protection of argon at 600 mL/min (purity >99.999%, Beiwen Gas Co., Ltd., China) and hydrogen at 100 mL/min (purity >99.999%, Beiwen Gas Co., Ltd., China), the temperature inside the quartz tube was increased to and maintained at 740 °C. After heating, the ferrocene slowly evaporated and the gaseous ferrocene adhered to the pristine ACF medium. Next, ethylene (purity >99.5%, Beiwen Gas Co., Ltd., China) was used at 200 mL/min for 40 min as the carbon source for CNTs growth on the catalyzed ACF medium. The heater was then turned off and cooled to room temperature in argon and hydrogen atmosphere, and the CNTs/ACF was finally obtained.

2.2. Measurement of physical properties

The morphology of the pristine ACF and the CNTs/ACF filter media were characterized by a scanning electron microscope (JSM7401F). The structure of the two materials were inspected with a Raman spectrometer (Horiba HR 800, 532/633 nm). The Brunauer–Emmett–Teller (BET) specific surface areas (S_{BET}) of these two filter media were examined using a Quadra Sorb Station (Quantachrome Instruments Corp., Florida, USA) through the nitrogen adsorption-desorption method. The weights were measured using an electrical scale (HZT-A1000, Hz & Huazhi, USA).

2.3. Experimental setting

The pristine ACF and the CNTs/ACF filter media were placed in a fixture (xx5004700, Millipore) with 32 mm inner diameter at 6.21 cm/s filtration velocity when measuring PM and ozone removal efficiencies. The PM filtration efficiency tests followed a previously published procedure of our group [48]. PM were generated by atomizing 0.01 g/mL NaCl solution using an atomizer (TSI 3076) with nitrogen as carrier gas. PM concentration and size distribution at the inlet and exhaust of the fixture were measured by a scanning mobility particle sizer (SMPS, TSI 3936), consisting of an electrostatic classifier (TSI 3080), a long differential mobility (TSI 3081) and an ultracondensational particle counter (TSI 3775 low). The particle size distribution of generated NaCl aerosols in triplicated experiments was presented in Fig. 1. As shown, the particle generation exhibited good consistency, and particles ranging from 20 nm to 300 nm dominated the distribution. Ozone removal efficiency tests followed the procedure in our previous study [49]. Ozone of 300 ppb net inlet concentration was generated by an ultraviolet (UV) lamp with synthetic air. An ozone monitor (model 205, 2B Technology) was used to record ozone concentration every 1 min for 10 h. Before each experiment, the ozone monitor and the particle sensor were calibrated by Beijing Institute of Metrology, China to ensure accuracy of the sensors. A differential gage (P3000T, BESTACE, USA) was used to measure pressure resistances for the pristine ACF and the CNTs/ACF at different airflow rates, varying linearly from 0.5 L/min to 5.0 L/min.

The pollutant removal efficiency, η_i, was calculated according to equation (1):

$$\eta_i = \left(1 - \frac{C_{out,i}}{C_{in,i}}\right) \times 100\% \tag{1}$$

where the subscript i stands for PM or ozone, $C_{in,i}$ is the inlet

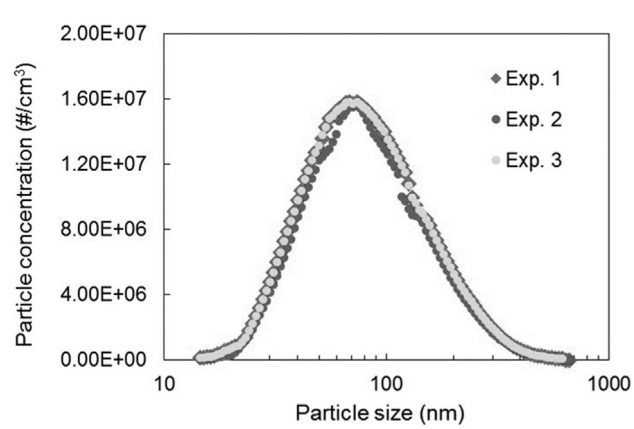

Fig. 1. Particle size distribution of inlet NaCl aerosols in three independent experiments.