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Short Communication

Fabrication of a multi-walled carbon nanotube-deposited glass fiber air filter for the enhancement of nano and submicron aerosol particle filtration and additional antibacterial efficacy

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ABSTRACT

We grew multi-walled carbon nanotubes (MWCNTs) on a glass fiber air filter using thermal chemical vapor deposition (CVD) after the filter was catalytically activated with a spark discharge. After the CNT deposition, filtration and antibacterial tests were performed with the filters. Potassium chloride (KCl) particles (<1 μm) were used as the test aerosol particles, and their number concentration was measured using a scanning mobility particle sizer. Antibacterial tests were performed using the colony counting method, and *Escherichia coli* (*E. coli*) was used as the test bacteria. The results showed that the CNT deposition increased the filtration efficiency of nano and submicron-sized particles, but did not increase the pressure drop across the filter. When a pristine glass fiber filter that had no CNTs was used, the particle filtration efficiencies at particle sizes under 30 nm and near 500 nm were 48.5% and 46.8%, respectively. However, the efficiencies increased to 64.3% and 60.2%, respectively, when the CNT-deposited filter was used. The reduction in the number of viable cells was determined by counting the colony forming units (CFU) of each test filter after contact with the cells. The pristine glass fiber filter was used as a control, and 83.7% of the *E. coli* were inactivated on the CNT-deposited filter.

1. Introduction

When a human inhales, particles in the air enter the body. Micron-sized particles are intercepted by nostril hairs, but submicron or nano-sized particles can reach the lungs and subsequently reside in the alveoli (Mitsakou et al., 2007). Nanofibers have emerged as a promising filtration media because they can provide greater filtration efficiency and higher performance than conventional fibers (Gradoń et al., 2006; Barhate and Ramakrishna, 2007; Przekop and Gradoń, 2008). However, filters composed entirely of nanofibers are not commonly used as air filters because of the high pressure drops across their surfaces. In an attempt to avoid this pressure drop, nanofiber-coated filters have recently been introduced. By coating nanofibers onto a micron-sized filter, the filtration efficiency is improved without introducing a significant difference in the permeability of the filter. Graham et al. (2002) showed that a coating of nanofibers on the surface of conventional micron-fibrous filters produced using the electrospinning method can significantly improve filter performance. Wang et al. (2008) investigated filters that were composed of a layer of nanofibers on a substrate of micrometer fibers, and compared the performance of such nanofiber media to that of conventional micrometer fibrous filters. Their results showed that when the solidity of the nanofiber increased, the particle filtration efficiency also increased.

Bioaerosols are airborne particles with biological origins; these include viruses, bacteria, fungi, and a variety of living materials. Indoor bioaerosols accumulate in large quantities on filters where they are able to multiply under certain conditions, especially if a large amount of moisture is present on the filter (Maus et al., 2001). Moreover, the organic and inorganic materials deposited onto the filter medium after the air filtration contribute to microbial growth. This inevitably leads to an increased pressure drop across the filter and probable deterioration of the filter, with the eventual release of microorganisms. To avoid these problems, an antibacterial agent coating process is required. Recently, carbon nanotubes (CNTs) have been used as antibacterial agents. Kang et al. (2008a) showed that direct cell contact with CNTs seriously affected cellular

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membrane integrity, metabolic activity, and the morphology of bacteria due to stress related to cell membrane damage and oxidative stress caused by the CNTs. Recently, single-walled CNT (SWCNT) filters were successfully created and used to remove biological agents from water with high filtration efficiency (Brady-Estévez et al., 2008). However, SWCNTs appear to have greater toxic effects on cultured human fibroblasts than multi-walled CNTs (MWCNTs). MWCNTs have fewer toxic effects on human fibroblast cells than activated carbon and carbon black (Tian et al., 2006). Moreover, MWCNTs are significantly cheaper than SWCNTs, and are now available at a reasonably low price (~100 US$/kg), making them practical for large-scale applications. Recently, MWCNTs have been used as a new type of filter media. Srivastava et al. (2004) developed MWCNT filters that removed micron-scale and nano-scale contaminants from water and heavy hydrocarbons from petroleum.

Even though MWCNT filters have been extensively studied for water filtration, few reports are available in the literature on MWCNTs used for air filtration. Park and Lee (2006) constructed a metal MWCNT filter in which the carbon nanotubes were grown directly on the micron-sized metallic fibers of the air filter using the thermal chemical vapor deposition (CVD) method. The metal MWCNT filter had higher filtration efficiency than conventional filters without a significant increase in pressure drop. Guan and Yao (2010) investigated the removal efficiencies of CNT filters for a collection of aerosolized biological and non-biological particles. Their results revealed that particle types and CNT loading had greater effects on the efficiencies than the membrane types and pore sizes tested.

In this study, samples of glass fiber filter media were catalytically activated with iron (Fe) nanoparticles generated by a spark discharge method. The catalytically activated filter samples were placed in a thermal CVD reactor in which MWCNT fibers were grown on the activated spots after acetylene (C_2H_2) gas was carried into the reactor. Using the synthesized MWCNT-coated samples, particle filtration efficiency and antibacterial efficiency were tested using an aerosol number counting technique and a colony counting method, respectively.

2. Materials and methods

2.1. MWCNT synthesis on the fiber surface of the filter

We used a commercial medium filter (glass fiber filter media, Fabriano, Italy) composed of glass fibers as the substrate micron filter. Fig. 1 shows the two-step process of CNT synthesis on the fiber surface of the filter, including catalytic activation followed by the CVD process.

The catalytic activation process required a spark discharge that was formed in a pure nitrogen environment between two identical Fe rods in a reactor at standard ambient temperature and pressure (SATP). The flow rate of the nitrogen gas was controlled by a mass flow controller (3660, Kofloc, Japan) and was set to 5 L/min. When a voltage higher than the breakdown voltage was supplied to the metal electrodes, a spark was discharged across the electrode gap, thereby vaporizing material from the electrodes and producing primary particles a few nanometers in diameter via nucleation of the vapor (Byeon et al., 2006; Byeon et al., 2008). The generated particles were transported using a flow of nitrogen gas, and were deposited onto the surface of the sample filter for 10 min. The size distribution of the generated particles was measured using a scanning mobility particle sizer (SMPS; 3936N22 Custom, TSI Inc., USA) system consisting of a classifier controller (3080, TSI Inc., USA), a differential mobility analyzer (DMA; 3085, TSI Inc., USA), a condensation particle counter (CPC; 3022A, TSI Inc., USA), and an aerosol charge neutralizer (Soft X-ray Charger 4530, HCT Co., Ltd., Korea).

After catalytic activation, CNT synthesis was performed via thermal CVD in a furnace. The catalytically activated filter sample was loaded into the quartz tube of the furnace. As the temperature was increased to 650 °C, nitrogen gas was injected at 100 ml/min through the quartz tube to clean the inside of the tube. The pressure inside the tube was 2.7 to 4.5 Torr. When the temperature stabilized, the carrier gas was replaced by acetylene gas at 30 ml/min for 10 min. After the synthesis of the CNTs was finished, the quartz tube was cooled to room temperature in a nitrogen ambient environment. A field emission scanning electron microscope (FE-SEM; JSM-6500F, Jeol Ltd., Japan) was used to evaluate the morphologies of the CNTs grown on the surfaces of the glass fibers.

2.2. Particle filtration test with the MWCNT-deposited glass fiber air filter

The experimental setup for the particle filtration test is shown in Fig. 1. The system consisted of a test duct, a test particle generation system, and a measurement system. The test duct was constructed using acryl. A sample of the test filter was installed in the middle of the test duct. Two isokinetic stainless steel sampling probes were placed before and after the filter media for aerosol sampling. A differential pressure gauge (Magnehelic® 2000, Dwyer Instruments, Inc., USA) was used to measure the pressure drop across the test filter media sample. The temperature and relative humidity inside the test duct were 22.5 ± 3 °C and 10 ± 5%, respectively.

Potassium chloride (KCl) particles were used as the standard test particles, as recommended by ASHRAE Standard 52.2 (2007) and the ISO 11155-1 standard (2001). A constant concentration of KCl particles was supplied to the test duct. To determine particle penetration as a function of particle size, two aerosol generation systems were used to produce particles with different size distributions. A stream of particle-free compressed air from a dry-cleaned air supply system consisting of an oil trap, a diffusion dryer, and a high efficiency particulate air (HEPA) filter was delivered to an electrical tube furnace containing KCl powder at 720 °C (see A in Fig. 1). Particles from the tube furnace were passed through a neutralizer (Soft X-ray Charger 4530, HCT Co., Ltd., Korea) and delivered to the test duct after they were diluted by clean air. After using the tube furnace, another stream of clean air was delivered to a collision-type atomizer holding a solution containing 10 wt.% KCl (in water) (see B in Fig. 1) to generate the test particles. The aerosol particles from the atomizer were passed through a diffusion dryer for water removal, were charge neutralized (Soft X-ray Charger 4530, HCT Co., Ltd., Korea), and were delivered to the test duct. The number concentration of each test particle was measured using an SMPS (3936L22, TSI, USA). Fig. 2 shows the particle size distribution of the generated test particles. Another stream of clean air was delivered to the test duct and mixed with the particle-laden air flow. The mixture flow rate was maintained at 48 l/min to achieve a face velocity of 0.5 m/s for the filter media.

2.3. Antibacterial test with MWCNT-deposited glass fiber air filter

The process used in the antibacterial test is shown in Fig. 1. *Escherichia coli* (*E. coli*; ATCC 11775) was selected as the model bacteria. A bacterial suspension was prepared by incubating a 0.1 ml culture inoculated in 15 ml of nutrient broth for 24 h. The nutrient broth was prepared by dissolving 5 g of peptone and 3 g of meat extract in 1000 ml of sterilized deionized water, and then sterilizing the solution in an autoclave.

The antibacterial tests on the test filters were performed using the colony counting method. A pristine glass fiber filter, a catalytic activated filter, and a CNT-deposited filter were used as the test filters. Each test filter was divided into sixteen 10×10 mm^2 square pieces. The pieces were placed into a test tube with a 10 ml *E. coli* suspension (optical