Technical Note

Antimicrobial Air Filter Fabrication Using a Continuous High-Throughput Aerosol-Based Process

Joon Sang Kang1, Hanna Kim2, Jeongan Choi1, Hak Yi3, Sung Chul Seo4, Gwi-Nam Bae1*, Jae Hee Jung1**

1 Center for Environment, Health and Welfare Research, Korea Institute of Science and Technology, Seoul 136-791, Korea
2 Department of Mechanical Engineering, Seattle Pacific University, Seattle, WA 98119, USA
3 Department of Mechanical Engineering, University of California, Los Angeles, CA 90095, USA
4 Department of Environmental Health, College of Medicine, Korea University, Seoul 136-701, Korea

ABSTRACT

A continuous high-throughput aerosol-based method for fabrication of antimicrobial air filters using natural antimicrobial nanoparticles was developed. We used the nebulization and electrospray methods for deposition of nanosized antimicrobial substances on pristine filter media. The roll-to-roll process was introduced for high-throughput fabrication of antimicrobial filters, and electrospray generation and dispersion equipment were used for high performance. The present method covers a filter area of 4500 mm$^2$ at one time with uniform deposition. The characteristics of the airborne particles generated by nebulization and the electrospray method were evaluated using a scanning mobility particle analyzer (SMPS) and scanning electron microscopy (SEM). Furthermore, filter performance, such as the pressure drop and antimicrobial efficiency, was examined. The pressure drop of the antimicrobial filter showed a general increasing trend with amount of deposited antimicrobial particles. When 2.64 and 3.52 µg mm$^{-2}$ of antimicrobial particles were loaded on pristine filter media, the measured antimicrobial efficiency of the filter was over 99.5% based on a 24-h contact time. This study provides useful information for the development of a high-throughput production process for antimicrobial air filtration systems.

Keywords: Antimicrobial filter; Antimicrobial natural product; Antimicrobial nanoparticle; Air filtration; Nebulization; Electrospray.

INTRODUCTION

Bioaerosols, such as airborne viruses, bacteria, fungi, and pollen, are widespread in the air, and can cause various adverse health effects when inhaled, such as infectious diseases and allergy (Bush and Portnoy, 2001; Douwes et al., 2003; Lee and Liao, 2014). In particular, microscopic bioaerosols, such as bacteria and viruses, have low settling velocities due to their small particle size, and could therefore be suspended in the atmosphere for longer than other particles with larger diameters, resulting in a higher likelihood of inhalation (Hinds, 1999; Hong et al., 2015).

There have been many attempts to develop effective means of eliminating or inactivating microorganisms to prevent infection. The most common methods include sterilization with UV, removal of microorganisms using an electric precipitator, and elimination of microbes with antimicrobial filters (Lin and Li, 2002; Mainelis et al., 2002; Yu et al., 2008). Antimicrobial filters are used most commonly due to their simplicity and affordability (Devinny et al., 1998; Choi et al., 2015). There have been many studies of the use of antimicrobial filters in air purifiers, air conditioners, and ventilation systems (Simmons and Crow, 1995; Maus et al., 2001; Miller, 2002).

Previous studies have proposed two kinds of representative method for the antimicrobial treatment of filters: a liquid dip-coating process and an aerosol-based process. Liquid dip-coating is generally used for antimicrobial filter production. This method, which involves soaking air filters in liquid containing antimicrobial substances, is relatively fast and easy, and is therefore the most common production method. However, the application of this method to the antimicrobial treatment of high-efficiency particulate air (HEPA) filters should be performed with caution because HEPA filters can be damaged by physical stress or

* Corresponding author.
Tel.: 82-2-958-5676; Fax: 82-2-958-5805
E-mail address: gnbae@kist.re.kr

** Corresponding author.
Tel.: 82-2-958-5718; Fax: 82-2-958-5805
E-mail address: jaehee@kist.re.kr
deformation. Additionally, as their pore size is small, the filter pores can easily be blocked by deterioration during the liquid dip-coating process, which results in an increased pressure drop. Aerosol-based processes, in which airborne antimicrobial particles are loaded onto pristine filters to achieve antimicrobial ability, are potentially useful candidates (Park et al., 2013). Aerosol-based processes work according to the same principle as the conventional air filtering mechanism, in which airborne particles are collected on filter medium. However, in these methods, solid antimicrobial airborne particles are used to coat pristine air filters.

The electrospray method, which is an aerosol-based antimicrobial filter production method, generates monodispersed micro/nanosized droplets with a unipolar charge, which are therefore easy to control and create micro/nanostructures (Kusdianto et al., 2014). Micro/nanostructured antimicrobial substances can markedly increase the antimicrobial efficiency of filters because of their very large surface area, leading to a large contact area between the antimicrobial substance and microorganisms (Karabacak et al., 2014). Use of the electrospray method for antimicrobial filter production, however, results in low productivity due to the low flow rate, and the production of filters with satisfactory antimicrobial performance requires a long time. This represents a major limitation for commercial production of antimicrobial filters using only the electrospray method. For a manufacturer to be able to produce aerosol-based antimicrobial filters, a high-throughput production method using processes that can cover a large surface area is required.

To overcome the drawbacks outlined above, this study was performed to develop a fabrication method for aerosol-based HEPA antimicrobial filters with high throughput, thus enabling economically viable mass production. This technique uses nebulization and electrospray methods simultaneously to deposit antimicrobial particles on a large filter surface area to maintain the advantages and avoid the disadvantages of the electrospray method. The nebulized antimicrobial particles are mixed with electrosprayed particles in our device. The mixed substance is transferred to the aerosol disperser and particles are deposited evenly onto the pristine filter medium. After completion of mixing and dispersion, a roll-to-roll machine is used to move the filter at a constant speed (~2 mm s⁻¹) for continuous high-throughput filter production.

MATERIALS AND METHODS

Antimicrobial Filter Production Using a Roll-to-Roll Device

For aerosol-based high-throughput and high-quality antimicrobial filter production, we developed a technique involving a roll-to-roll device, nebulization, and the electrospray method. The apparatus for this method is shown schematically in Fig. 1(a).

We coated pristine HEPA filter media (fiberglass filter, China Textile Filters, Shenzhen, China) (fiber porosity: 0.3 µm; thickness: 2.5 mm; grade: H13) with antimicrobial substances using a roll-to-roll device. The filtration efficiency of the HEPA filter was measured using *Staphylococcus epidermidis* (KCTC 1917) bacterial bioaerosols, which have a size range of 0.5–2 µm. This Gram-positive bacterium is commonly used in bioaerosol research because staphylococci are associated with serious health risks to humans and animals (Jung et al., 2011a; Leppänen et al., 2014). The filtration efficiencies of the coated filters were calculated using the following equation:

$$\eta = 1 - \frac{C_{\text{outlet}}}{C_{\text{inlet}}}$$  

where $C_{\text{inlet}}$ and $C_{\text{outlet}}$ represent the particle concentrations (particles cm⁻³) of the bacterial aerosol at the inlet and outlet of the filter, respectively. In this study, the filtration efficiency of the control filter was measured to be > 99.9%.

Recently, extracts of natural products with antimicrobial activity have been considered as novel, efficient, and cost-effective materials for the development of antimicrobial filter media (Dixon, 2001). Plant extracts, such as *Melaleuca alternifolia* (tea tree), *Eucalyptus*, and *Sophora flavescens*, in particular, can be used as a coating for filters to inactivate fungal spores, bacteria, and influenza viruses (Pyankov et al., 2008; Huang et al., 2010; Pyankov et al., 2012; Hwang et al., 2015a, b). The treatment of filter surfaces with nanosized particles of a natural product is an effective method for enhancing their antimicrobial activity, because the nanosized natural products provide the maximum possible specific surface area to contact surrounding agents. In this study, the antimicrobial substance used for coating was a natural product, *Sophora flavescens* ethanolic extract, which has been shown previously to have outstanding antimicrobial activity (Jung et al., 2013). To prepare the antimicrobial substance, *S. flavescens* was dissolved in ethanol (Merck, Darmstadt, Germany) at a concentration of 0.625% (w/v) using a vortex mixer (Scientific Industries, Bohemia, NY) for 3 minutes and placed in a sonicator (Branson, Danbury, CT) for 20 minutes. Then, the undissolved residue was removed using syringe filters with a pore size of 0.40 µm. The *S. flavescens* thus prepared was applied to the nebulizer and syringe for nebulization and electrospray, respectively. The nanostructured antimicrobial substance was generated by the electrospray method and additional nanosized antimicrobial substances were generated by nebulization to increase productivity.

As shown in Fig. 1(a), compressed air was passed into a six-jet Collison nebulizer (BGI Inc., Waltham, MA) via a flow controller at a flow rate of 5 L min⁻¹ to provide sufficient pressure for nebulization. A diffusion dryer was inserted to remove the moisture of the antimicrobial particles and droplets from the antimicrobial suspension generated by the nebulizer. The antimicrobial substance then formed solid nanoparticles, which were delivered into the mixing chamber and blended with particles generated by the electrospraying process. The mixing chamber was cylindrically-shaped with a diameter of 8 cm and a height of 12 cm. The mixing chamber had two inlets: one inlet delivered antimicrobial particles generated by nebulization and the other delivered compressed air. The antimicrobial substance was sprayed using a stainless steel nozzle with inner and outer diameters...
Fig. 1. (a) Schematic diagram of the high-throughput filter fabrication method. (b) Roll-to-roll device. The aerosol disperser is equipped with a Z-stage and continuously sprays antimicrobial particles onto the filter. The pristine filter moves at a constant rate, and the suction pump is situated below the moving stage to allow aerosol deposition on the pristine filter. (c) Aerosol disperser. The aerosol enters through two side inlets and is uniformly dispersed by a small hole array (500 holes) in the disperser. Scale bar: 1 cm.

of 200 µm and 1/16 inch, respectively (Index Health & Science, Oak Harbor, WA) in the top of the mixing chamber, as shown in the inset of Fig. 1(a). A high voltage, 8.5 kV, was applied to the nozzle for spraying. The antimicrobial substance dissolved in ethanol was delivered to the nozzle through the syringe pump (KD Scientific, Holliston, MA) at a flow rate of 1 mL h⁻¹. An aluminum extractor (ring-type, 1 cm in thickness) was placed 2 cm away from the nozzle to provide sufficient potential to the particles. To monitor the stable spray mode of the system, transparent viewing ports were inserted on opposite sides of the mixing chamber. One was to allow the introduction of light into the chamber (MLC-150; Motic Instruments Inc., Richmond, BC, Canada), and the other was for observing the shape of the electrospray liquid meniscus using a CCD digital camera (MARIN F-145C2; Allied Vision Technologies Inc., Stadtroda, Germany) (Jung et al., 2009a, b; Hwang et al., 2015b). We carefully checked the stability of the electrospray mode in all experiments.

The mixture of antimicrobial particles generated by electrospraying, nebulization, and the compressed air passed through the outlet of the mixing chamber. The synthesized antimicrobial particles from the chamber were delivered to the aerosol disperser to uniformly coat the pristine filter medium. Fig. 1(c) shows the aerosol disperser that was used to evenly coat the filter with antimicrobial particles, which had dimensions of 150 mm × 30 mm × 70 mm (width × length × height). The aerosol disperser had 500 holes, each 1 mm in diameter (50 holes horizontally and 10 holes vertically) to provide a uniform pressure drop and to prevent a preferential flow path. The viscous flow through these holes in the disperser damps out all fluctuations, which reduces the swirl and level of turbulence (Fay, 1994). Thus, a smooth, uniform flow was formed by the disperser. Antimicrobial particles suspended in the air were sprayed on the filter through the holes and were uniformly deposited on the 150 mm × 30 mm (width × length) area simultaneously (same dimensions as the aerosol disperser). The area covered was about 40 × greater than in the previous method (Jung et al., 2013). As shown in Fig. 1(b), the aerosol disperser was equipped with a z-stage that could be moved up and down. The rolled pristine filter medium was constantly released and the antimicrobial aerosol was continuously sprayed by the aerosol disperser, and finally the antimicrobial filter was re-rolled on the opposite side of the roll-to-roll device. A suction pump was used to allow deposition of sprayed particles onto the filter substrate.

Performance Test of the Filter

We evaluated the size characteristics of airborne antimicrobial particles using a scanning mobility particle analyzer (SMPS; TSI Inc., Shoreview, MN) to determine the aerosol size distributions produced by each of the nebulization, electrospray, and mixed spray methods. The average particle charge was measured by the SMPS and an aerosol electrometer (3068; TSI). Scanning electron microscopy (SEM; XL30 ESEM-FEG; Philips Electron Optics, Eindhoven, The Netherlands) was used to examine the morphology of the antimicrobial particles on the filter medium. The pressure drop of the antimicrobial filter was tested using a pressure gauge (Dwyer, Michigan City, IN) by measuring the pressure difference between the inlet and outlet of the filter holder. Pressure drops, related to the amount of antimicrobial substance loaded (0.44, 1.76, 2.64, and 3.52 µg mm⁻²), were measured for a range of face velocities from 1 to 7 cm s⁻¹.
S. epidermidis was used to test the antimicrobial performance of the filter by counting the bacterial colony forming units (CFUs). Test bacteria of $1 \times 10^6$ cm$^{-3}$ were placed in contact with the antimicrobial filter for 1, 10, and 24 h at room temperature. After each designated time, the filters were submersed in 10 mL of phosphate-buffered saline (PBS; pH 7.4) with 0.05% Tween 80, and the bacteria were separated from the filter using a vortex mixer for 2 min and a sonicator for 10 min. The separated S. epidermidis was then cultured on nutrient agar (NA; Becton Dickinson, Franklin Lake, NJ, USA) and incubated for 24 h (37°C). A pristine HEPA filter was used as a control filter and was also tested to compare the effectiveness of the antimicrobial filter. The relative microbial survival (i.e., viability) was calculated using the following equation:

$$\text{Relative microbial survival} = \frac{\text{CFU}_a}{\text{CFU}_c}$$

where $\text{CFU}_a$ is the CFU of the antimicrobial substance-coated filter and $\text{CFU}_c$ is that of the pristine filter.

**RESULTS AND DISCUSSION**

**Antimicrobial Substance Particle Size and Shape**

The concentrations and diameters of antimicrobial S. flavescens nanoparticles were measured with SMPS at the upstream of the aerosol disperser to evaluate the amount of antimicrobial particles loaded onto the filter by nebulization and electrospaying. Three different cases, i.e., particles generated by nebulization, electrospaying, and by both methods, were examined and the results are shown in Fig. 2. The number distribution of particles generated by nebulization showed a unimodal distribution with a peak at 110 nm for a concentration (dN/dlogdp) of $1.4 \times 10^7$ cm$^{-3}$. In comparison, the particles formed by electrospaying showed a bimodal size distribution with peak diameters of 40 nm and 220 nm and number concentrations of $6.0 \times 10^6$ cm$^{-3}$ and $1.3 \times 10^6$ cm$^{-3}$, respectively, because the electrospaying method generates both quasi-monodisperse droplets and satellite droplets (Jung et al., 2011b; Kang et al., 2013). Fewer particles were generated by electrospaying compared to nebulization. When we generated antimicrobial aerosols using a combination of both electrospaying and nebulization, the particle distribution was not the same as a superposition of the two individual methods. The electrospaying peak at 40 nm was almost eliminated in the mixed spray method, but the concentration at 40 nm was still higher compared with nebulized particles. These observations indicate that nanoparticles generated by the electrospaying method aggregated with nebulized particles. As electrospaying causes particles to become unipolar (+3.4 of the average charge per particle with a high positive polarity), while nebulized particles are electrically neutral (−0.06 of the average charge per particle with a very low negative polarity) (Jung et al., 2011a), the particles generated by the two methods are easily aggregated. The peak diameter and concentration of the mixed spray method were 130 nm and $1.4 \times 10^7$ cm$^{-3}$, respectively. The peak diameter of particles produced by the mixed spray method was slightly larger than that of particles produced by nebulization, which represents an agglomeration effect. Additionally, the average charge of the particle was measured as approximately +0.54 with a positive polarity.

Charged particles can be attracted not only to oppositely-charged fibers by coulombic attraction, but also to neutral fibers at close range by image forces, which are weaker than coulombic forces. An image force between a charged particle and a neutral fiber is generated when the charged particle induces an equal and opposite charge in the fiber surface, thereby creating its own field of attraction. These charged particles would be self-dispersing in space, and deposited uniformly onto the filter surface, as shown in Fig. 3. This characteristic particle deposition might be the primary cause of the improved antimicrobial activity of the natural-product nanoparticle-deposited filters. However, further research is required to obtain more definitive explanations.

The mass distributions of S. flavescens particles produced by the different methods are shown in Fig. 2(b). The
Fig. 3. Scanning electron microscopy (SEM) images of aerosolized *S. flavescens* particles deposited on pristine high-efficiency particulate air (HEPA) filters with different particle generation methods. (a) Nebulization only. (b) Electrospray only. (c) Combination of both the nebulization and electrospray methods.

The electrospray method had bimodal peaks of 60 nm and 300 nm, but the mass concentration at 60 nm was very small (337 µg m⁻³) compared with the other peak at 300 nm. The nebulization method did not show a clear peak. The large particle size but low concentration may have an effect on the mass distribution. The mixed spray method showed a unimodal mass distribution with a peak diameter and concentration of 310 nm and 4.6 × 10⁴ µg m⁻³, respectively. The difference in mass distribution between the nebulization and mixed spray methods around the 300-nm region was due to the electrospray method.

To quantify the deposition of particles deposited on the filter medium by each spray method, we defined the mass deposition rate (MDR) per unit time (minute), and unit area (mm²):

\[
MDR \mu g mm^{-2} s^{-1} = \sum \frac{\pi d_{p,i}^3 \rho_p N_i Q_a}{6 \eta A_d}
\]

where \(d_{p,i}\) is the representative diameter of each SMPS channel, \(\rho_p\) is the particle density, \(N_i\) is the number concentration of particles from each SMPS channel, \(Q_a\) is the aerosol flow rate, \(A_d\) is the coated area, and \(\eta\) is the collection efficiency of the pristine HEPA filter. As the gap distance between the disperser and filter is small (< 5 mm), we assumed the value of \(A_d\) to be the area of the aerosol disperser when the roll-to-roll device was stationary. The density of *S. flavescens* was measured at 0.9 g cm⁻³, \(A_d\) was 4500 mm², \(Q_a\) was 10 L min⁻¹, and \(\eta\) was 0.99. Table 1 shows the total number concentration and MDR of the nebulization, electrospray, and combined spraying methods. The particles produced by nebulization had a capacity of 90% of the combined method and that of the electrospray method was 43% based on number concentration. The actual number concentration was 33% lower than the ideal, which was assumed to be because the particles generated by electrospraying and nebulization aggregated with each other. Using Eq. (2), the MDR calculated for nebulization was 0.065 µg min⁻¹ mm⁻², that for electrospraying was 0.016 µg min⁻¹ mm⁻², and that for the combined method was 0.088 µg min⁻¹ mm⁻². Thus, the combined method showed a 10 times higher MDR value than the electrospray method alone, as described previously (Jung et al., 2013).

**Pressure Drop and Antimicrobial Efficiency**

Pressure drop is one of the most important characteristics of air filters, and that of filters made using the combined electrospray and nebulization method was measured using a pressure gauge. The pressure drop of filters after deposition of different amounts of antimicrobial substance was measured by changing the face velocity to 1, 3, 5, or 7...
Table 1. MDR and total number concentration of aerosolized S. flavescens with different particle generation methods as determined by SMPS.

<table>
<thead>
<tr>
<th>Method</th>
<th>Total number conc. of particles (cm⁻³)</th>
<th>MDR (µg min⁻¹ mm²)</th>
<th>Contribution ratio based on number concentration a</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nebulization</td>
<td>7.39 × 10⁶</td>
<td>6.52 × 10⁻²</td>
<td>0.9</td>
</tr>
<tr>
<td>Electrospray</td>
<td>3.52 × 10⁶</td>
<td>1.61 × 10⁻²</td>
<td>0.43</td>
</tr>
<tr>
<td>Nebulization + electrospray</td>
<td>8.17 × 10⁶</td>
<td>8.75 × 10⁻²</td>
<td>1</td>
</tr>
</tbody>
</table>

a Contribution ratio based on number concentration = (Total number concentration generated by nebulization or electrospray method)/(Total number concentration generated by combined nebulization and electrospray methods).

cm s⁻¹, and the results are shown in Fig. 4(a). Regardless of the amount of antimicrobial substance sprayed, the pressure drop increased as the face velocity increased. In addition, the pressure drop increased with the amount of antimicrobial substance deposited. Based on a face velocity of 7 cm s⁻¹, when 0.44 µg mm⁻² of antimicrobial substance was loaded, the pressure drop was 36% higher than for a pristine filter. The pressure difference increased as the amount of antimicrobial substance loaded increased, with coatings of 1.76 µg mm⁻², 2.64 µg mm⁻², and 3.52 µg mm⁻² increasing the pressure difference by 69%, 78%, and 98%, respectively. As mass loading increased, the pressure difference for a HEPA filter with 3.52 µg mm⁻² of antimicrobial substance increased to twice that of the pristine filter. The pressure drops were measured at different positions on the filter to verify the uniformity of the coating. When the face velocity was 1 cm s⁻¹, the average pressure drop from five different positions was ~14.55 mmH₂O with a coefficient of variation (CV) of 0.027, as shown in Fig. 4(b). The pressure drops increased evenly in terms of the different positions compared to the pressure drop of the control filter (8.33 mmH₂O). This result indicates that our aerosol disperser is suitable for uniform particle deposition.

Fig. 5 shows the antimicrobial efficiency of the filter produced by the method developed in this study. The antimicrobial efficiencies of filters loaded with different amounts of antimicrobial substance were tested by calculating the viability of S. epidermidis as a model microorganism using Eq. (1). The viability of microorganisms decreased with increasing contact time. With 0.44 µg mm⁻² of the antimicrobial substance loaded on the filter, 80% of the test microorganisms were inactivated within 1 h, and the inactivation ratio reached 98% within 10 h. However, the inactivation gradient became low with long contact times. Similar results were observed for all four cases, as shown in Fig. 5, but the inactivation ratio within 1 h of contact showed marked differences between groups. These observations indicate that the main effect of differences in the quantity of the test antimicrobial substance was within the initial period. On the other hand, the inactivation gradient of all four cases was similar after 1 hour. This means that the intrinsic antimicrobial ability of loaded particles mainly contributes at later time points. The percentages of antimicrobial efficiency with a contact time of 24 h were 98% for 0.44 µg mm⁻², 99% for 1.76 µg mm⁻², 99.5% for 2.64 µg mm⁻², and 99.6% for 3.52 µg mm⁻².

As shown in Figs. 4 and 5, deposition of larger amounts of antimicrobial substance was associated with a greater contact area, resulting in higher antimicrobial efficiency. However, the optimal amount of antimicrobial substance per pristine filter should be determined taking the pressure drop into consideration. The amount should be increased when antimicrobial efficiency has priority over the pressure difference, but the amount sprayed should be reduced when the pressure drop is more important than the antimicrobial efficiency. In this study, we obtained a 99% antimicrobial efficiency.
efficiency and $-14$ mmH$_2$O for the pressure drop (1 cm s$^{-1}$ of the face velocity) when the amount of antimicrobial substances deposited was 1.76 µg mm$^{-2}$ (contact time of 24 hours).

CONCLUSIONS

In the present study, we developed a high-throughput fabrication process for antimicrobial filters with high antimicrobial efficiency using nebulization and electrospray methods. This process overcomes the disadvantages of previous methods, including low throughput and discontinuous treatment. The method developed here can cover an area of 4500 mm$^2$ at any one time which is the same as the area covered by the aerosol disperser. This coverage area is much larger than previous methods. Also, the current method can be applied to continuous processing with high productivity. Concurrent use of nebulization and electrospraying evenly coated pristine filters with large amounts of an antimicrobial substance. The electrospray chamber and aerosol disperser were built to combine the two types of particles generated by each spraying method and to uniformly coat the filter.

The method presented here showed 10 times higher throughput than the electrospray method for antimicrobial filter production, while retaining similar antimicrobial capability. The amount of antimicrobial substance coating the filter is proportional to the pressure drop, but it improves the antimicrobial ability, so control of the optimal amount to prioritize either antimicrobial ability or pressure difference is required.

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