Electrically Responsive Coarse Filters Endowed by High-Dielectric-Constant Surface Coatings toward Efficient Removal of Ultrafine Particles and Ozone

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ABSTRACT: Ambient ultrafine particles (UFPs) cause severe health threats, and UFPs can transport in indoor environments through building envelopes. In building HVAC (heating, ventilation, and air conditioning) systems, fibrous filtration has been extensively used to resist particulate matter. However, there is an intrinsic conflict between fibrous filters’ high filtration efficiency and low air resistance. Herein, we designed a dielectric-coated fiber structure and fabricated electrically responsive coarse filters to remove UFPs by utilizing the Coulomb force between the charged particles and the polarized fibers. The electrical responsiveness endowed the filters with an excellent performance for the removal of UFPs. With the synergistic improvement of particle precharging and filter polarization, the UFPs’ removal efficiency of the coated filter increased from ~0% to the highest of 90.50%, with ultralow ~13.0 Pa air resistance at 1 m/s face air velocity, which ensured a considerable filtration performance even under large face air velocity. Furthermore, the combination of MnO₂ and polyurethane (PU) filters performed with the ability to remove the unwanted ozone generated by particle precharging. The promising features give these filters excellent potential for use in energy-efficient electrostatic air-cleaning devices, thus achieving a healthy and sustainable environment.

KEYWORDS: Indoor air quality, EAA filtration, Coulomb force, Ultrafine particles, Ozone removal

1. INTRODUCTION

Nowadays, there exists a growing demand for healthy indoor air because investigations have found that people spend over 80% of their lifetime indoors.1,2 With the development of transportation, industry, and the overcombustion of fossil fuel, hazardous pollutants come into being, broadly exist in the atmosphere, and then transport into indoor environments through building envelopes. Moreover, pollutants released from indoor sources are also worthy of attention. Indoor airborne pollutants, especially particulate matter (PM), have emerged as an imminent health threat to the public. It has been demonstrated that PM exposure poses negative health consequences, leading to skin inflammation,3,4 respiratory diseases,5,6 cardiovascular diseases,7 cancer,8 and even DNA damage.9 Research has determined that most ambient PM (73%) is in the ultrafine fraction,10 which consists of particles with an aerodynamic diameter of less than 100 nm. Ambient ultrafine particles (UFPs) mainly come from urban traffic emissions,11,12 industrial production emissions,13,14 combustion sources,15 and the nucleation by chemical reactions between gaseous pollutants.13 Thus, UFPs are mainly composed of organic compounds, potentially toxic elements, nitrates, sulfates, and trace metals.16 Due to a larger surface area and a tinier size,17 UFPs could penetrate the bronchi more easily than larger PM18,19 and further enter the vascular space to be digested directly into the blood, causing an even more severe health threat.20 Human exposure to particles of outdoor origin often occurs indoors due to the entry of UFPs into buildings.21 UFPs penetrate into indoor environments by air infiltration and ventilation through opening envelopes.22 Therefore, measures should be carried out to facilitate UFP removal devices in buildings for the sake of human health. Furthermore, when applied in the heating, ventilation, and air conditioning (HVAC) systems in buildings for large-scale purification, the removal setups should still be efficient under a large airflow rate and face air velocity.

Fibrous filtration is a noticeable method for airborne UFP removal.23 Generally, there are five basic mechanisms for a particle depositing onto a fiber in fibrous filtration: interception, inertial impaction, diffusion, gravitational settling, and...
and electrostatic attraction. The first four mechanisms are called mechanical collection effects. However, the notable problem is that there is an intrinsic conflict between fibrous filters’ high filtration efficiency and low air resistance. Researchers have usually fabricated denser and thicker fibrous filters to achieve a stronger mechanical collection effect. However, high-efficiency air filters accompany a large resistance to airflow, possibly costing up to a quarter of power consumption in large commercial HVAC systems. Additionally, as time goes by, the filter will gradually become clogged by the PM coating, so the air resistance further rises steeply. Fabricating nanofiber filters is an ideal way to reduce air resistance. The possibility of particle capture increases due to the larger surface area of fibers on the nanometer scale, ensuring effective PM removal with a much thinner filter. Besides, lower air resistance occurs when the nanofiber diameter is comparable to or smaller than the mean free path of air molecules, thus introducing a slip effect. However, nanofiber filters are usually made into membranes, which is not suitable for facilitation in building ventilation systems. Besides, in these works, the performance toward UFPs was not specifically mentioned.

Utilizing the electrostatic effect is another choice for capturing small airborne particles. A high capture efficiency was achieved when using electrospun electret fibers, electrostatic aerogel filters, triboelectrically activated fibers, corona charged dielectric fibers, and so on. With the electrostatic enhancement, most of these works are available to perform a lower airflow resistance based on medium-high-efficiency filters. However, the electret charges will degrade rapidly, leading to a relatively short lifetime. Moreover, in these works, either filter medium or particles carry charges to trigger a monopole attraction rather than a dipole attraction between them, so the particle capture efficiency could be limited.

Applying a continuous electrostatic force to both fibers and particles has been proposed as a promising method to overcome the limitations of the above studies. Choi et al. conducted a particle precharger before an Al-coated conductive fibrous filter for an efficient UFP removal. By the electrostatic force between the charged fibers and particles, the particles 30–400 nm in size were captured with a distinguishing removal efficiency of ~99.99%, and the initial air resistance was 0.6 Pa at 0.025 m/s face air velocity. Sambudi et al. used a unipolar corona charger to continuously charge the particles and the electret filters, which can overcome the charge degradation of fibers. When fresh and neutralized electret filters were used with the unipolar charger (electric field: 1.9 kV/cm), the highest particle collection efficiency for 70 nm particles was kept at 99.8% and 98.2%, respectively. Li et al. reported active-poled filtration with both the fibers and particulates being polarized under the external electric field. At an airflow velocity of 0.21 m/s and a poling voltage of 2 kV, the filters’ removal efficiency for PM2.5 reached 98.72%, and the air resistance was 82 Pa. Tian and Mo demonstrated a new strategy for the electrostatically assisted air (EAA) coarse filter. In the EAA filtration device, corona charging to the particles and polarizing to the filters are synergized to realize a higher filtration efficiency. This structure increased the single-pass filtration efficiency for 0.3–0.5 μm particles of a polyethylene terephthalate (PET) coarse filter from 0.4% to 99.0% with 21.0 Pa air resistance at 1.2 m/s filtration velocity.

EAA filtration is an effective way to significantly augment the removal efficiency based on a low-efficiency coarse filter without deteriorating air permeability when applied under a large flow rate, thus achieving high efficiency, low air resistance, and large particle loading capacity at the same time. However, stronger corona charging of the particle achieved by a higher external voltage will lead to unsafe air breakdown and generate ozone. In addition to that forming in the filtration devices, ambient ozone can also be a non-negligible source for indoor pollutants and penetrate the indoor environment by air exchange, causing health risk and economic loss. As a result, electrically responsive filters are expected to achieve a higher filtration efficiency without further lifting external voltages and ozone generation, so the principle of EAA filtration is worthy of discussion and verification. Besides, limited research has paid attention to removing UFPs by utilizing the electrostatic effect, so the EAA performance to remove UFPs is also worth investigating.

Herein, we designed a dielectric-coated fiber structure and fabricated electrically responsive coarse filters to remove UFPs by utilizing the Coulomb force between the charged particles and the polarized fibers. A normalized factor $E/E_\infty$ was proposed to measure the dielectric response for different kinds of fibers under a certain external electric field in a finite element model. Further, we coated functional dielectric materials (manganese dioxide and activated carbon) to the polyurethane (PU) filters to improve the electrostatic filtration efficiency of UFPs and ozone simultaneously. PU substrates are available commercial filters with excellent toughness, chemical resistance for recycling use, and dielectric properties. The electrically responsive coatings endowed the filters with an excellent performance for the removal of UFPs, and the combination of MnO$_2$ and polyurethane (PU) filters performed with the ability to remove the unwanted ozone generated by particle precharging. The filters in the EAA device also performed with low air resistance and energy consumption under a large airflow rate and face air velocity and thus has a good application prospect for achieving a healthy and sustainable environment.

2. THEORETICAL ANALYSIS

In a normal coarse fibrous filtration, the gap between fibers (~μm) is significantly at least 1000X larger than the scale of the particle diameter (~μm), thus achieving low air resistance. However, low air resistance means a limited mechanical filtration effect to achieve high filtration efficiency. Introducing the electrostatic effect between fibers and particles should be a solution to solve this problem.

In EAA filtration, the polluted airflow will be first charged by the pin-to-plate module and then go through a polarizing module. The dielectric fibrous filter will be induced to charge by the external polarizing electric field, and the charged particle will deposit onto a polarized fiber through the Coulomb force between them. The Coulomb force $F_i$ (N) could be calculated by eqs 1 and 2.

$$F_i = E_i q$$

(1)

$$E_i = E_p \left[ 1 + \left( \frac{e_L - 1}{e_f + 1} \right) \frac{d^2}{4r^2} \right]$$

(2)

where $q$ is the charge (C) on the particle; $E_i$ is the induced electric field intensity (V/m); $E_p$ is the external polarizing field.
moves toward them. 如此，我们可以提出一种方法来改变介质的表面电介质特性，而无需改变其原始的材料表面修饰，其中材料的表面修饰率、纤维直径（m）。

ε是空气的相对介电常数，q是电荷（C）在粒子上的电荷；μ是空气的黏度（N s/m²）；U0是空气流速度（m/s）；εf是纤维表面的相对介电常数（F/m）；d2是纤维直径（m）；dp是粒子直径（m）。

图像1展示了(a)未涂层单纤维和(b)电介质表面涂层单纤维在极化电场中时，带电粒子向它们移动。

图2。示意图展示了制备PU滤芯时表面涂层的方法。

强度 (V/m)；εr是纤维的相对介电常数；d2是纤维直径（m）；r是距离中心线的纤维和粒子的距离（m）。

 meanwhile，当带电粒子在相邻的中性电介质纤维上移动，并不使用外部极化，局部电场由粒子生成并引电荷在纤维中。故此，其效果将取决于粒子和纤维间的相互作用，其可由图1(b)中等式（3）来表述，该等式为

\[ N_I = \left( \frac{\epsilon_r - 1}{\epsilon_r + 1} \right) \frac{q^2}{12\pi^2 \mu_0 \epsilon_r d_2^2 d_p} \]  

where \( \epsilon_r \) is the relative dielectric constant of the fiber; \( q \) is the charge (C) on the particle; \( \mu \) is the air viscosity (N s/m²); \( U_0 \) is the airflow velocity (m/s); \( \epsilon_0 \) is the vacuum dielectric constant (F/m); \( d_2 \) is the fiber diameter (m); and \( d_p \) is the particle diameter (m).

The image force \( F_I \) can be further calculated by eq. 4

\[ F_I = \frac{1}{16\pi\epsilon_0} \left( \frac{\epsilon_r - 1}{\epsilon_r + 1} \right) \frac{q^2}{r - \frac{d_2}{2}} = \frac{3\pi\mu_0 d_2^2 d_p}{4\left(r - \frac{d_2}{2}\right)^2} \]

where \( \epsilon_0 \) is the vacuum dielectric constant, 8.854 × 10⁻¹² F/m.

According to the computations presented in the Supporting Information, when a single bare fiber is polarized, the Coulomb force will be a dominant factor in influencing the movement of particles. Furthermore, \( \epsilon_r \) is a crucial variable for enhancing both the Coulomb force and the image force. Higher- \( \epsilon_r \) fibers lead to a larger surface charge density when they are polarized, thus attracting the charged particles in the airflow and improving the efficiency of EAA filtration. From the perspective of materials, surface modification is an effective way to alter the surface dielectric properties of the filter medium without changing the original fiber structure. When higher- \( \epsilon_r \) materials are coated on the fiber surface, a stronger Coulomb attractive force will form. For example, as shown in Figure 1a, when a single bare fiber is polarized, the induced electric field will be circumscribed by the limited surface charge density, so the charged particles will slip over the fiber easily. Conversely, as shown in Figure 1b, when the bare fiber with a dielectric surface coating is polarized, the induced electric field will be enhanced significantly. Then, the charged particle will directly deposit on the fiber surface due to the fortifying Coulomb force. In other words, the invisible induced electric field filled the spatial "gap" between fibers in a coarse filter to achieve an efficient filtration, and the air permeability would be maintained at the same time. Moreover, if the coating materials consist of a certain catalyst or absorbent, the modified filters will be expected to remove ozone, the byproduct generated by the external charging in EAA filtration.

3. METHODS

3.1. Electrostatic Simulation. The induced electrical field intensity for fibers with different materials and structures under an external polarizing field was simulated using a commercial finite element software solver, Ansys Electromagnetics Suite 19,2, developed by Ansoft Inc. The electrodes for polarizing and the dielectric fiber were constructed to be a two-dimensional model from an overhead view. According to microscope observation, the cross-section of the single fiber was simplified as a circle area of 100 μm in diameter. The fiber was placed at the center of two 0.1 mm × 12 mm parallel stainless-steel electrode plates. Voltage was applied to one of the plates, and the other plate was connected to the ground. The distance between the two plates was 12 mm, and the simulating region was filled with an air medium.

In the simulation, we also designed a composite structure including the fiber and the coating materials with different \( \epsilon_r \) ranging from 5.1 to 1450 (see Figure 3). The coating materials were located uniformly onto the fiber surface with a 10 μm
diameter. The $\varepsilon_r$ values for different coating materials are given in Table S2.

3.2. Materials. Polyurethane (PU) filters (thickness, 8 mm; porosity, 35 and 50 ppi) were purchased from Dongguan Dongchi Sponge Products Co. Ltd. Manganese dioxide (MnO$_2$) powder and activated carbon (AC) powder were obtained from Zhongshan Topg Ecology Tech Co. Ltd. The ethyl vinyl acetate adhesive (EVA) and sodium carboxymethylcellulose (CMC-Na) were purchased from Meryer Chemical Technology Co., Ltd.

3.3. Fabrication of the Filters. The fabrication progress of the filters is as follows (see the overview in Figure 2). (1) Sieve the powders of MnO$_2$ and AC using 200-mesh sieves to eliminate larger granules. (2) Prepare the coating adhesive gel by mixing the sieved powders, ethyl vinyl acetate (EVA, as the adhesive), and sodium carboxymethylcellulose (CMC-Na, as the thickener) with different mass ratios shown in Table S4. (3) Stir the gel with the speed of 200 rpm for 10 min. (4) Pour the gel made of different proportions onto the PU filters and then use a roll-to-roll device to squeeze the overflowing coatings out of the substrate. The roll-to-roll distance was set to 2 mm to ensure the same squeezing force on each filter. In this way, the gel could be loaded uniformly on the filter surface. (5) Dry the squeezed PU filters in an oven under 70 °C for 120 min. The coating gel is insoluble in water, so the filters can be washed for recycled use. Different filters were named by the porosity and the adhesive proportion of the gel (see Table S4).

3.4. Characterization. The morphologies of the fibers were investigated by using a field emission scanning electron microscope (SEM, Merlin VP compact, Carl Zeiss AG) coupled with energy dispersive X-ray spectra (EDS). The $\varepsilon_r$ values of the materials were obtained by using a precision impedance analyzer (Agilent 4294A, Agilent Technologies, Inc.), and the detailed process is shown in the Supporting Information.

3.5. Performance Test. The filtration performance tests were conducted in an acrylic air duct with an inner diameter of 50 mm, as shown in Figure S2a. Ambient air with airborne particles was driven through the module in the sequence of a steady flow plate, a pin-to-plate charging module, the polarizing module including the filter to be tested, and an axial flow fan. The face air velocity across the filters was set at 1 m/s if it is not mentioned specifically. The detailed experimental methods are presented in the Supporting Information.

4. RESULTS AND DISCUSSION

4.1. Electrostatic Simulation. Polarization of the dielectric medium will lead to the separation of negative and positive charges.
positive charges, forming a local reinforcement of the electric field. As for a single cylindrical fiber (projected to a circle on a two-dimensional plane), the induced enhancement in the radial direction could be expressed by a normalized factor, $E/E_{\infty}$, where $E$ is the intensity of the induced field, and $E_{\infty}$ is the intensity of the external polarizing field. As shown in Figure S1, the induced field intensity around the polarized fiber will be enhanced ($E/E_{\infty} > 1$) owing to the dielectric effect. Furthermore, the degree of the enhancement is determined by the dielectric constant of the fiber, implying that, in the EAA filtration, filters with different $\varepsilon_r$ values could lead to differences in the filtration performance.

The composite structure consisting of the fiber and coating materials emerged a much larger induced electric field locally (for instance, less than ~20 $\mu$m to the fiber surface) compared to the bare fiber (see Figures 3 and 4a). In the region where the field is enhanced, a stronger Coulomb force exists between the charged particle and the polarized fiber, showing that the surface coatings to the bare fiber will be an effective way to improve the performance of EAA filtration.

However, as illustrated in Figure 4b, filters with higher-$\varepsilon_r$ coatings did not reveal a significantly stronger electric field enhancement even in a short distance (5 $\mu$m from the fiber surface), as the $\varepsilon_r$ varied from 5.1 to 1450, from manganese dioxide (MnO$_2$) to the barium titanate (BaTiO$_3$) in the simulation. There are two possible reasons to account for this phenomenon. (1) The scale of the coating material is much tinier than the fiber substrate. Thus, in the local region for the fiber scale, field intensity would not appear as a qualitative change despite the increasing $\varepsilon_r$ of the coating materials. (2) According to eq 2, in the dielectric single-fiber model, the electric field will not enhance steeply indeed along with the increase of $\varepsilon_r$. In a particle filtration process, experiment results proved that the surface coating itself rather than the coating materials was the crucial point to improve the filtration performance of the EAA filter, indicating that the surface coatings of the fiber do not need an extremely large $\varepsilon_r$ to achieve high efficiency. Besides, materials with a large $\varepsilon_r$ have the potential to reveal a surface microdischarge due to an overaccumulation of the charges under the polarized electric field, even causing hazardous ozone production. As a result, the selection of the coating materials on the fiber surface to strengthen EAA filtration needs to be considered cautiously.

In EAA filtration, charged particles will be accelerated by the electrostatic attraction and move toward the dielectric fiber. In this work, we simulated the assumed particle migration velocity toward polarized PU fibers with different coatings (MnO$_2$, ZnO, TiO$_2$, BaTiO$_3$) based on the electrostatic simulation results. The charge to mass ratio of particles was evaluated as $1 \times 10^{-6}$ C/kg, and the initial velocity of particles from 20 $\mu$m toward fiber surfaces was set as 0.8 mm/s according to previous observations. The polarizing electric field intensity was $4.17 \times 10^5$ V/m when the potential difference between the two plates was 5 kV. A detailed calculation procedure is shown in the Supporting Information.

As seen in Figure 4a, compared to the bare PU fiber, particles will move to the BaTiO$_3$@PU fiber at a higher velocity (4.5 mm/s compared to 3.9 mm/s at a 2 $\mu$m distance from the fiber surface), because of the local enhancement of the $E/E_{\infty}$ (1.66 compared to 1.03). Due to the effect of the

**Figure 5.** SEM images of (a1–a3) Bare-35; (b1–b3) 35-M-3; and (c1–c3) 35-C-3. Codes for different filters represent the bare 35 ppi PU filter, 35 ppi MnO$_2$@PU filter, and 35 ppi AC@PU filter fabricated by different proportions of adhesives. (See the detailed fabricating parameters in Table S4.)
fortified Coulomb force, the randomly diffused particles will be accelerated when moving toward and eventually depositing on the composite fiber, like a disoriented bug captured by a powerful intangible tentacle. Tian et al. observed the particle-capturing behaviors through an in situ CCD (charged coupled device) camera device, finding that the smoke particles would quickly deposit on a polarized fiber with BaTiO3 coatings compared to the unpolarized one. The particle migration velocity increased from 0.6 to 4.1 mm/s, while the particles were moving toward the fiber from ~20 to ~8 μm away. The calculations match the observation results, implying that the Coulomb force between the polarized fiber and the charged particles is the dominating factor in deciding the particle movement in the local region. As a result, it is promising that coating dielectric materials to the surface of the coarse filter will enhance the EAA filtration performance.

4.2. Characterization. Figure 5 and Figures S3–S9 show the optical photographs and scanning electron microscopy (SEM) images of the Bare-35, Bare-50, MnO2@PU, and AC@PU filters. Compared with the bare PU filter, the powdered coatings were adhered to and uniformly distributed on the surface of PU fibers. Samples coated with the gel composed of a larger adhesive proportion emerged with less powder on the surface. The size of the MnO2 and AC coatings is approximately 10 and 2 μm, while the fiber diameter is 100–150 μm. The gap between the fibers is ~1 mm as shown in the images, indicating that the surface modification will not significantly change the air permeability of the bare PU filters. Figures S3–S9 reveal that different coating powders/adhesive
mass ratios did not make much difference in the morphologies of filters. Also, we weighed and calculated the net coating amounts of the gel on the fabricated filters (as presented in Table S4) and found no significant difference, either.

Figure S10 shows the energy dispersive spectroscopy (EDS) elemental mappings of Bare-35, MnO2@PU, and AC@PU filters. MnO2 and AC were distributed uniformly on the fiber surface. The Mn element appeared on the MnO2@PU mapping image, and the surface contents of C and O elements of MnO2@PU and AC@PU filters increased compared to those of the bare PU. Figure S11 also presents the EDS element spectrogram of the tested filters.

The relative dielectric constants (\(\varepsilon_r\)) of MnO2 and AC, which indicate how easily the materials can become polarized by an external electric field, were tested at 10.85 and 24.25 at 1 MHz, respectively. Besides, as a mixture compounds, the \(\varepsilon_r\) of the coconut-shell-based AC used in this work was reported in the range 5–20 at 1 GHz \(^{61-63}\) (the value will be even larger at 1 MHz), which is much larger compared to the PU substrate (\(\varepsilon_r = 1.1\) at 1 MHz). As a result, it is reasonable that the MnO2 and AC coatings will be more responsive than the bare PU under the external electric field, and the coated filters will achieve a higher electrostatic effect in capturing airborne particles.

4.3. Performance Test. 4.3.1. Performance for Particle Removal. In this work, we defined UFPs to represent the particles with an aerodynamic diameter ranging from 11 to 115 nm to fit the scanning range of the particle size. Moreover, the particles with a diameter of 0.3–0.5 \(\mu\)m (specially denoted by PM0.3–0.5 in this work) were also measured, as it is most possible for the minimum filtration efficiency to appear at the 0.3 \(\mu\)m fraction for the fibrous filters.\(^{64}\) Figure S12 shows a typical size distribution of the particles in the ambient air during the test.

The removal efficiency \(\eta\) of particles with a certain size of \(d_p (\mu m)\) was calculated by

\[
\eta(d_p) = \left( \frac{C_{up}(d_p) - C_{down}(d_p)}{C_{up}(d_p)} \right) \times 100\%
\]

where \(C_{up}(d_p)\) and \(C_{down}(d_p)\) are the particle counts (cm\(^{-3}\)) of a certain diameter of \(d_p (\mu m)\) in a sampling period upstream and downstream of the filter, respectively.

Figure 6b,c and Figure S14a,b show the removal efficiency for UFPs and PM0.3–0.5 of different filters, while the charging voltage (\(U_p\)) and the polarizing voltage (\(U_p\)) were set at different values in the EAA device. Obviously, when no electric field was applied, all filters operated nearly \(\sim0\%\) removal efficiency for ultrafine particles due to their considerable pore size (\(\sim mm\)) compared to the particle diameters (\(\sim \mu m\)). Further, with the existence and the increase of the charging voltage, the particles were charged in stepwise degrees, and the efficiency was accordingly enhanced. When a charged particle migrated to the neutral fibers, the fiber surface would be induced to carry an opposite charge, hence forming an image force between the particle and the fiber. In this scenario, the scale of the image force is determined by the dielectric constant of the fiber, so the filters with higher-\(\varepsilon_r\) coatings performed with a better removal efficiency compared to the bare PU filters. For instance, under a charging voltage of 10 kV, the average UFP removal efficiency of the filter 35-C-3 was 84.43\%, obtaining a 173% enhancement based on the bare 35 ppi PU fiber (\(\sim 48.60\%\)). On top of that, only applying a polarizing voltage to the filter could further elevate the efficiency by 5\%, indicating an even stronger effect from the Coulomb force instead of the image force. In the theoretical analysis, we raised that the polarization applied to the dielectric medium would lead to local reinforcement of the electric field, thus accelerating the particle migration velocity toward the fibers. It is implied that the charging and polarizing effect are synthesized to strengthen the performance of the dielectric coarse filters in EAA filtration. Besides, it is worth mentioning that the charged particles would attach to the polarized fiber surface one after another and be a part of the fiber–particle binary system. The whole system would then be polarized again to form a dipole, thus attracting the following charged particles rather than repelling them.\(^{57}\) It was also found that, based on the existing research toward the removal of PM,\(^{65-66}\) EAA filtration is even more efficient for UFP fraction removal, as the electrostatic effect on capturing airborne particles increases with a reduction of the particle size. Further, a higher efficiency could be achieved by lowering the airflow velocity (see Figure S18). We also compared the performance of the coated filter with some similar electrostatic filters in the Supporting Information. Detailed results can be seen in Table S6.

Different coating materials did not significantly influence filtration performance. Overall, the PU filters coated with AC performed slightly better compared to MnO2 in the UFP removal, although the \(\varepsilon_r\) of AC (24.25) is much larger than that of MnO2 (10.85). The coating amount did not significantly influence the filtration performance, either. Although the filters were coated with gels composed of different proportions of ingredients, the net loading amounts of the powders on the filters were close to each other (see Table S4). As for loaded filters fabricated with different proportions of powders and adhesives, the average efficiency for UFPs was tested to be 90.50\%, 87.08\%, and 86.68\% for 50-M-3, 50-M-4, and 50-M-5, respectively, and 89.77\%, 90.47\%, and 90.30\% for 50-C-3, 50-C-4, and 50-C-5, respectively, as seen in Figure S16. All of the high-\(\varepsilon_r\) coated filters demonstrated a dramatically lifting efficiency compared to bare substrates.

In addition to endorsing an electrical responsiveness to the filters, the coatings also altered the filter surface roughness to improve the filtration performance. According to Figure S5 and Figures S3–S9, the MnO2 and AC powders formed peaks and gaps on the coated filters, leading to a much rougher surface than that of bare PU filters. If a charged particle moves close to the fiber surface, the dominant factor is the electrostatic effect, and if the particle adheres to the fiber, the combined factor of the van der Waals force and the electrostatic force from the external electrostatic field makes the particle attach firmly on the surface. On one hand, the surface coating formed a micro-/nanostructure roughness, which has been proved to enhance the surface dipolar charge of the fiber effectively.\(^{43,67}\) On the other hand, rougher surfaces provided a stronger van der Waals force for capturing particles.\(^{68-70}\) Thus, rough surfaces performed with higher filtration efficiencies through a binary effect of an electrostatic mechanism and mechanical mechanism.

4.3.2. Air Resistance and Quality Factors. The MnO2@PU and the AC@PU filters can perform a considerable UFP removal efficiency with an excellent airflow permeability. It was noteworthy that, with the surface coatings on the low-resistance 35 ppi PU coarse filters, the air resistance merely increased from 8.7 to 11.8 Pa (35-M-3) and 11.0 Pa (35-C-3)
under a 1 m/s face velocity. As for the denser 50 ppi substrate, the highest air resistance just reached 13.0 Pa for the 50-M-5 filter. Although some filters were coated with gels composed of different proportions of ingredients (for example, 35-M-3, 35-M-4, and 35-M-5), their air resistances were close to each other, in the range 11.7–11.9 Pa. According to the surface morphologies observed by SEM (Figure S1 and Figures S3–S9), the scale of the coating materials is much tinier than the fiber substrate, so the loaded thin films attaching on the surface will not significantly alter the air permeability for the coarse filter. The detailed results are summarized in Figure 6d.

Generally, for fibrous filters, there exists a conflict between the efficiency and air resistance, so the quality factor (QF) is usually calculated to assess their trade-off properties:

$$QF(d_p) = \frac{-\ln(1 - \eta(d_p))}{\Delta P}$$

where QF represents the QF (Pa⁻¹) for particles of a certain diameter of $d_p$ (μm), $\eta(d_p)$ is the removal filtration efficiency of particles with a certain size of $d_p$, and $\Delta P$ is the air resistance (Pa).

We use $QF_{UPP}$ to represent the quality factor toward the UFP removal and $QF_{E/E}$ to represent the quality factor toward the 0.3–0.5 μm particle removal. EAA filtration is an effective method to improve the quality factor of the coarse filters. For example, the $QF_{UPP}$ of the bare 35 ppi PU filters increased from 0.0037 to 0.0879 Pa⁻¹ thanks to the electrostatic enhancement when applying a 10 kV charging voltage and 20 kV polarizing voltage. Moreover, as shown in Figure 6e, filters with higher-$\varepsilon_r$ coatings exhibited an even more remarkable $QF_{UPP}$. For example, compared to the 35 ppi bare PU, the $QF_{UPP}$ of the 35-C-4 filter enlarged by 52 times from 0.0037 to 0.1906 Pa⁻¹, due to both the electrostatic effect and the coatings. Overall, the $QF_{UPP}$ values of the 50 ppi filters were lower than those of the 35 ppi filters, owing to a larger air resistance and relatively less efficiency lifting. The corresponding results for $QF_{E/E}$ are presented in the Supporting Information.

Further, the idea of using the coarse filter in EAA filtration is to provide a high particle removal efficiency with low pressure drop, which can save energy for the driven fans. However, the charging and polarizing electric power supply also consumed energy. We calculated the fan power ($P_{fan}$) and the power dissipation ($P_{polar}$) of this work in the Supporting Information.

The results showed that the power consumption of the electric power in EAA filtration was lower than that of the driven fans. Detailed calculations can be obtained in Table S5.

### 4.3.3. Performance for Ozone Removal

Some concerns remain to be addressed in that EAA filtration has the potential to generate ozone because of the air breakdown driven by the high-voltage corona discharge, especially at the charging pin cusps, the roughness of the polarizing meshes, and the fabric surfaces. In the fabrication, the filters with surface coatings of MnO₂ (act as a catalyst) and AC (act as an absorbent) were enabled to remove ozone.

Figure 7a presents the ozone concentration at three sampling positions when $U_c$ and $U_p$ were set at 10 and 20 kV, respectively. When using the bare-35 PU fibers, the average ambient inlet ozone was 24.7 ppb (part per billion), and downstream of the charging module and the polarizing module, the average concentration was up to 84.7 and 95.6 ppb, respectively, indicating that the charging pin and the polarizing module were both able to generate ozone by the discharge. By installing the filters fabricated in this work, the net ozone production of the whole EAA devices decreased. The concentration at the downstream decreased to 11.0, 17.8, 44.3, and 38.3 ppb, performing a single pass efficiency of 82.5%, 77.4%, 40.4%, and 52.2% for 35-M-3, 50-M-3, 35-C-3, and 50-C-3, respectively. Figure 7b shows the net ozone production for the filters investigated. As for the filters coated with MnO₂, the net ozone production even decreased to minus values, suggesting that these filters were even capable of removing the ambient ozone.

### 5. Conclusions

In this work, we proposed a single-fiber simulation model to evaluate the performance of fibers with different dielectric properties in EAA filtration, and further to give instructions to the design of the fiber structure. We proposed a normalized factor $E/E_\infty$ to measure the local reinforcement of the electric field. By coating higher-$\varepsilon_r$ materials onto the bare fiber surface, the induced field intensity around polarized fibers will be enhanced, so charged particles will move to and eventually deposit on the fiber owing to the effect of the fortified Coulomb force. Compared to the bare PU fiber, particles will move to the high-$\varepsilon_r$-coated fiber at a higher velocity (4.5 mm/s compared to 3.9 mm/s at a 2 μm distance from the fiber surface), because of the local enhancement of the $E/E_\infty$ (1.66...
compared to 1.03). Based on that, we coated MnO$_2$ and AC onto the bare polyurethane (PU) filters by adhesion to simultaneously improve the electrostatic filtration efficiency for UFPs and the removal performance for ozone. By adhering higher-$E$ coatings and applying charging and polarizing electric fields to a coarse PU filter, the highest removal efficiency for UFPs was dramatically improved from $\sim$0% to 90.8% under 1 m/s face velocity. A higher efficiency could be achieved by further lowering the airflow velocity. The filters were found capable to remove the unwanted ozone from the EAA filtration, or even effective to remove the ambient ozone. Meanwhile, these efficient filters also achieved an outstanding air permeability, owing to the ultralow air resistance of $0.4$ at a large filtration velocity at 1 m/s. We expect this work to have good application prospects for low-cost and energy-efficient electrostatic air-cleaning devices, thus achieving a clean, healthy, and sustainable environment.

**ASSOCIATED CONTENT**

**Supporting Information**
The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsestengg.1c00186.

Additional data and figures including photographs, schematics, SEM images, EDS element mapping, EDS element spectrum, PM size distributions, filters’ removal performances and electricity dissipation (PDF)

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**Notes**
The authors declare no competing financial interest.

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**REFERENCES**

(1) Duan, X. Exposure factors handbook of Chinese population; Environmental Science Press: Beijing, 2013.

(2) Jones, A. P. Indoor air quality and health. Atmos. Environ. 1999, 33 (28), 4535–4564.

(3) Cheng, Z.; Liang, X.; Liang, S.; Yin, N.; Faiola, F. A human embryonic stem cell-based in vitro model revealed that ultrafine carbon particles may cause skin inflammation and psoriasis. J. Environ. Sci. 2020, 87, 194–204.

(4) Verdin, A.; Cazier, F.; Fitoussi, R.; Blanchet, N.; Vié, K.; Courcot, D.; Monsas, I.; Seta, N.; Achard, S. An in vitro model to evaluate the impact of environmental fine particles (PM0.3–2.5) on skin damage. Toxicol. Lett. 2019, 305, 94–102.

(5) Mikela, K.; Ollila, H.; Satinen, E.; Vuorinen, V.; Peltola, E.; Kaarteenaho, R.; Myllarniemi, M. Inorganic particulate matter in the lung tissue of idiopathic pulmonary fibrosis patients reflects population density and fine particle levels. Ann. Diagn. Pathol. 2019, 40, 136–142.

(6) Sandström, T.; Brunekreef, B. Traffic-related pollution and lung development in children. Lancet 2007, 369 (9561), S35–S37.

(7) Loxham, M.; Davies, D. E.; Holgate, S. T. The health effects of fine particulate air pollution. BMJ. 2019, 367, l6609.

(8) Wang, S.; Zhao, Y. Air pollution and lung cancer risks; Elsevier: Burlington, 2011; pp 26–38.

(9) Calderón-Garcidueñas, L.; Herrera-Soto, A.; Jury, N.; Maher, B. A.; González-Maciej, A.; Reynoso-Robles, R.; Ruiz-Rudolph, P.; van Zundert, B.; Varela-Nallar, L. Reduced repressive epigenetic marks, increased DNA damage and Alzheimer’s disease hallmarks in the brain of humans and mice exposed to particulate urban air pollution. Environ. Res. 2020, 183, 109226.

(10) Peters, A.; Wichmann, H. E.; Tuch, T.; Heinrich, J.; Heyder, J. Respiratory effects are associated with the number of ultrafine particles. Am. J. Respir. Crit. Care Med. 1997, 155 (4), 1376–83.

(11) Guo, S.; Hu, M.; Peng, J.; Wu, Z.; Zambra, M. L.; Shang, D.; Du, Z.; Zheng, J.; Fang, X.; Tang, R.; Wu, Y.; Zeng, L.; Shuai, S.; Zhang, W.; Wang, Y.; Ji, Y.; Li, Y.; Zhang, A. L.; Wang, W.; Zhang, F.; Zhao, J.; Gong, X.; Wang, C.; Molina, M. J.; Zhang, R. Remarkable nucleation and growth of ultrafine particles from vehicular exhaust. Proc. Natl. Acad. Sci. U. S. A. 2020, 117 (7), 3427.

(12) Wahlina, P.; Palmgren, F.; Van Dingenen, R. Experimental studies of ultrafine particles in streets and the relationship to traffic. Atmos. Environ. 2001, 35, 63–69.

(13) Lv, Y.; Chen, X.; Wei, S.; Zhu, R.; Wang, B.; Chen, B.; Kong, M.; Zhang, J. Sources, concentrations, and transport models of ultrafine particles near highways: a Literature Review. Building and Environment 2020, 186, 103725.

(14) Silva, L. F. O.; Hower, J. C.; Izaquiero, M.; Querol, X. Complex nanoreinernals and ultrafine particles assemblages in phosphogypsum of the fertilizer industry and implications on human exposure. Sci. Total Environ. 2010, 408 (21), 5177–5122.

(15) Shi, J. P.; Evans, D. E.; Khan, A. A.; Harrison, R. M. Sources and concentration of nanoparticles (<10nm diameter) in the urban atmosphere. Atmos. Environ. 2001, 35 (7), 1193–1202.

(16) Moreno-Rios, A. L.; Tejeda-Benitez, L. P.; Bustillo-Lecompte, C. F. Sources, characteristics, toxicity, and control of ultrafine particles: An overview. Geosci. Front. 2021, 101147.

(17) Wierzbicka, A.; Nilsson, P. T.; Rissler, J.; Sallsten, G.; Xu, Y.; Pagels, J. H.; Albim, M.; Österberg, K.; Strandberg, B.; Eriksson, A.; Bohgard, M.; Bergemalm-Rynell, K.; Gudmundsdottir, A. Detailed diesel exhaust characteristics including particle surface area and lung deposited dose for better understanding of health effects in human chamber exposure studies. Atmos. Environ. 2014, 86, 212–219.

(18) Clifford, S.; Mazariega, M.; Salini, F.; Ezr, W. N.; Veganeh, B.; Low-Choy, S.; Walker, K.; Mengersen, K.; Marks, G. B.; Morawski, L. Effects of exposure to ambient ultrafine particles on respiratory health and systemic inflammation in children. Environ. Int. 2018, 114, 167–180.

(19) Liao, C. M.; Chio, C. P.; Chen, W. Y.; Ju, Y. R.; Li, W. H.; Cheng, Y. H.; Liao, V. H. C.; Chen, S. C.; Ling, M. P. Lung cancer risk in relation to traffic-related nano/ultrafine particle-bound PAHs exposure: A preliminary probabilistic assessment. J. Hazard. Mater. 2011, 190 (1), 150–158.
Characterization and evaluate the efficiency of different filter media filtration properties.

F.; Ferreira, C.; Morawska, L. Ultrafine particles and children's operation condition. (1), 35472.

Chen, C.; Zhang, L. Effective removal of particles down to 15 nm using scalable metal-organic framework-based nanofiber filters. Energy and Buildings 2018, 158, 987–999.

Li, J.; Zhang, D.; Yang, T.; Yang, S.; Yang, X.; Zhu, H. Nanofibrous membrane of graphene oxide-in-polyacrylonitrile composite with low filtration resistance for the effective capture of PM2.5. J. Membr. Sci. 2018, 581, 85–92.

Liu, C.; Hsu, P. C.; Lee, H. W.; Ye, M.; Zheng, G.; Liu, N.; Li, W.; Cui, Y. Transparent air filter for high-efficiency PM2.5 capture. Nat. Commun. 2015, 6 (1), 6205.

Deng, N.; He, H.; Yan, J.; Zhao, Y.; Ben Ticha, E.; Liu, Y.; Kang, W.; Cheng, B. One-step melt-blowing of multi-scale micro/nano fabric membrane for advanced air-filtration. Polymer 2019, 165, 174–179.

Hassan, M. A.; Yeom, B. W.; Wilkie, A.; Pourdeyhimi, B.; Khan, S. A. Fabrication of nanofiber meltblown membranes and their filtration properties. J. Membr. Sci. 2013, 427, 336–344.

Zhang, P.; Zhang, S.; Wan, D.; Zhang, P.; Zhang, Z.; Shao, G. Multilevel polarization-fields enhanced capture and photocatalytic conversion of particulate matter over flexible schottky-junction nanofiber membranes. J. Hazard. Mater. 2020, 395, 122639.

Ligeris, D.; Dumée, L. F.; Al-Attabi, R.; Castenet, E.; Schütz, J.; Kong, L. Mixed matrix Poly(Vinyl Alcohol)-Copper nanofibrous anti-microbial air-microfilters. Membranes 2019, 9 (7), 87.

Shou, D.; Ye, L.; Fan, J. Gas transport properties of electrospun polymer nanofibers. Polymer 2014, 55 (14), 3149–3155.

Zhao, X.; Wang, S.; Yin, X.; Yu, J.; Ding, B. Slip-effect functional air filter for efficient purification of PM2.5. Sci. Rep. 2016, 6 (1), 35472.

Bian, Y.; Chen, C.; Wang, R.; Wang, S.; Pan, Y.; Zhao, B.; Chen, C.; Zhang, L. Effective removal of particles down to 15 nm using scalable metal-organic framework-based nanofiber filters. Applied Materials Today 2020, 20, 100653.

Wang, S.; Zhao, X.; Yin, X.; Yu, J.; Ding, B. Electret polyvinylidene fluoride nanofibers hybridized by polytetrafluoroethylene nanoparticles for high-efficiency air filtration. ACS Appl. Mater. Interfaces 2016, 8 (36), 23985–23994.

Liu, J.; Zhang, S.; Liu, L.; Yu, J.; Ding, B. High-performance PM0.3 air filters using self-polarized electret nanofiber/nets. Adv. Funct. Mater. 2020, 30 (13), 190554.

Oliveira, A. E.; Aguiar, M. L.; Guerra, V. G. Theoretical analysis of air filtration phenomena for a micro-fibrous filter medium enhanced with electrospun nanofibers. Aerosol Science and Engineering 2021, 5 (1), 81–92.

Kim, S. J.; Raut, P.; Jana, S. C.; Chase, G. Electrostatically active polymer hybrid aerogels for airborne nanoparticle filtration. ACS Appl. Mater. Interfaces 2017, 9 (7), 6401–6410.
(61) Shkal, F.; Lopez, S.; Slocombe, D.; Porch, A. Microwave characterization of activated carbons. *Journal of Computer and Communications* 2018, 6, 112–123.

(62) Atwater, J. E.; Wheeler, J. R. R. Microwave permittivity and dielectric relaxation of a high surface area activated carbon. *Appl. Phys. A: Mater. Sci. Process.* 2004, 79 (1), 125–129.

(63) Atwater, J. E.; Wheeler, R. R. Complex permittivities and dielectric relaxation of granular activated carbons at microwave frequencies between 0.2 and 26 GHz. *Carbon* 2003, 41 (9), 1801–1807.

(64) Lee, K. W.; Liu, B. Y. H. On the minimum efficiency and the most penetrating particle size for fibrous filters. *J. Air Pollut. Control Assoc.* 1980, 30 (4), 377–381.

(65) Tian, E.; Gao, Y.; Mo, J. Electrostatically assisted air coarse filtration for energy efficient ambient particles removal: Long-term performance in real environment and influencing factors. *Building and Environment* 2019, 164, 106348.

(66) Tian, E.; Mo, J.; Li, X. Electrostatically assisted metal foam coarse filter with small pressure drop for efficient removal of fine particles: Effect of filter medium. *Building and Environment* 2018, 144, 419–426.

(67) Wang, Y.; Xu, Y.; Wang, D.; Zhang, Y.; Zhang, X.; Liu, J.; Zhao, Y.; Huang, C.; Jin, X. Polytetrafluoroethylene/Polyphenylene sulfide needle-punched triboelectric air filter for efficient particulate matter removal. *ACS Appl. Mater. Interfaces* 2019, 11 (51), 48437–48449.

(68) Krupp, H. Particles adhesion theory and experiment. *Adv. Colloid Interface Sci.* 1967, 1, 111–239.

(69) Takeuchi, M. Adhesion forces of charged particles. *Chem. Eng. Sci.* 2006, 61 (7), 2279–2289.

(70) Gady, B.; Reifenberger, R.; Rimai, D. S.; DeMejo, L. P. Contact electrification and the interaction force between a micrometer-size polystyrene sphere and a graphite surface. *Langmuir* 1997, 13 (9), 2533–2537.