Eco-Friendly Poly(Vinyl Alcohol) Nanofiber-Based Air Filter for Effectively Capturing Particulate Matter

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Abstract: Due to the increasing use of polypropylene-based nonwoven dust masks and air filters, environmental problems that occur due to the plastic pollution resulting from the disposal of these materials have also increased. Hence, an eco-friendly air filter based on PVA nanofibers (NFs) was fabricated by electrospinning on a nonwoven fabric, and its performance was evaluated as a filter capable of blocking or capturing particulate matter. The quality factor of the optimized PVA NF-based air filter was found to be 0.010606 Pa−1, which is lower than that of a HEPA filter (0.015394 Pa−1), but higher than that of a cabin air filter (0.010517 Pa−1) and a dust mask (0.009102 Pa−1). The contamination level of the PVA NF-based filter was analyzed by optical and structural analyses of the filter surface. Finally, the filter was soaked in water to selectively remove the contaminated PVA NF layer, and the remaining nonwoven fabric was able to be reused to make the filter.

Keywords: eco-friendly air filter; electrospinning; nanofiber; poly(vinyl alcohol); air purifier

1. Introduction

Recently, the use of polypropylene (PP)-based nonwoven face masks and filters has increased significantly due to air pollution (particulate matter, harmful gases, etc.) and respiratory viral infections (coronavirus, Spanish flu, etc.) [1–4]. PP is an extremely useful plastic because of its physical and thermal properties, such as light weight and heat resistance. It does not leach or decompose in hot water due to its excellent heat resistance [5]. Additionally, it is approved for food contact by the United States Food and Drug Administration (US FDA) [6]. However, the PP-based nonwoven fabric for filters cannot be recycled. Additionally, when incinerated, it generates dioxins, a group 1 carcinogen, which fatally affects humans and the natural ecosystem [7,8]. To solve this problem, studies on reusable (or washable) filters and the use of environmentally friendly polymers as filter media have been conducted [7]. Ko et al. proposed a reusable silver nanowire percolation-network-based air filter for face mask applications. This filter worked with the assistance of a negative ionizer [9]. Jang et al. reported a recyclable boron nitride nanotube (BNNT)-based membrane to capture nanoparticles. This BNNT membrane could be reused following heat treatment at ~450 °C for 6 h [10]. Recently, Kim et al. introduced a reusable polyvinylidene difluoride (PVDF) nanofiber-based air filter for face masks. However, to
fabricate clean PVDF nanofiber-based face mask filters, toxic organic solvents such as dimethylformamide (DMF), dimethyl sulfoxide (DMSO), and ethanol are required [11]. Park et al. proposed photocatalytically degradable poly(vinyl alcohol)/titanium dioxide nanofilters for capturing particulate matter (PM). These nanofilters were decomposed by UV irradiation (0.68 W/m²) for 9 days [7]. As such, various studies have been conducted on environmentally friendly materials and reusable filter technology that can replace the existing PP-based nonwoven filters. However, to enable these technologies to be applied on a daily basis, further research is required.

In this study, we introduce a polymer nanofiber-based air filter that can be disposed of in an environmentally friendly manner using water. To this end, an air filter with a two-layer structure (PVA nanofiber/PP-based nonwoven fabric) was fabricated by electrospinning a PVA solution onto a nonwoven fabric. Electrospun PVA nanofibers (NFs) and nonwoven fabrics are significant materials and reusable substrates for eco-friendly air filters, respectively. The physical properties and filter performance (filtration efficiency and pressure drop) of the prepared PVA NF-based air filter were compared with those of a commercial air filter. Finally, it was confirmed that the PVA NF layer of the used filter can be easily disposed of by soaking in water, and the remaining nonwoven fabric can be reused as a receiver to form an NF layer.

2. Materials and Methods

2.1. Preparation

PVA powders (Mw: 85,000–124,000, 87–89% hydrolyzed, Sigma-Aldrich, St. Louis, MO, USA) were dissolved in deionized (DI) water, and a 7 wt% PVA solution was stirred for 12 h at 90 °C. As shown in Figure 1a, the PVA solution was loaded into a syringe and fed at a rate of 0.5 mL/h using a syringe pump (KD Scientific, Holliston, MA, USA) to a stainless-steel needle connected to a high-voltage direct current (DC) power supply (Nano NC, Seoul, Korea). For electrospinning, an optimized high voltage (12.5 kV) was applied between the needle and grounded collector (the distance was 100 mm), and the electrospun PVA NFs were continuously accumulated on the nonwoven fabric substrate (thickness: 0.07 mm, HANSONG. Co., Ltd., Eumseong-gun, Korea). The thickness and pore size of the PVA NF-based filter were controlled by adjusting the electrospinning time. The synthesized PVA NF-based filters were dried in a forced convection oven for 3 h at 50 °C before the filtration test.

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**Figure 1.** (a) Schematic of electrospinning; (b) photograph of a poly(vinyl alcohol) nanofiber (PVA NF)–based filter fabricated by electrospinning on a nonwoven fabric substrate; (c) surface FE-SEM images of the nonwoven fabric substrate and PVA NF–based filter.
2.2. Characterization

To investigate the structural properties, field emission scanning electron microscopy (FE-SEM; Sirion FEI, Hillsbro, OR, USA) images were obtained at an accelerating voltage of 10 kV. To obtain the exact thickness of PVA NF-based filter media, the total thickness of ten filter media was measured using Vernier calipers (CD-15CPS, Mitutoyo, Kawasaki, Japan), and then the average thickness of one filter medium was calculated. To investigate the optical properties, the reflectance spectra of the PVA NF-based filters were obtained using a UV-VIS-NIR spectrophotometer (Cary 5000 UV-Vis-NIR, Agilent, Santa Clara, CA, USA).

2.3. Testing

The performance test (filtration efficiency and pressure drop) of the PVA NF-based air filter was conducted using a handmade air filtration evaluation system by benchmarking previously reported research papers (see Figure S1) [12–14]. The filtration efficiency of the air filter was calculated by comparing the PM concentrations before and after filtration. For this test, the smoke generated by incense burning was used as the PM source. According to literature, the average diameter of PM particles generated while burning incense is less than 1.0 µm [15–17]. The PM concentrations before and after filtration were monitored using PM sensors (PMS A003, Plantower Co., Ltd., Beijing, China), and the pressure drop in the filter medium was measured using a differential pressure meter (Testo 510, Testo Inc., Lenzkirch, Germany). The filtration efficiency and pressure drop of the air filter media were measured using a vacuum pump at a flow rate of 22 L/min. To evaluate the overall performance of all filters in terms of efficiency and pressure drop, a quality factor (QF), \( Q_f \), was used as an indicator. \( Q_f \) is defined as [18,19]:

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Q_f = -\ln(1 - E)/\Delta P
\]

where \( E \) is the PM filtration efficiency and \( \Delta P \) is the pressure drop across the filter.

To check the applicability of the PVA NF-based filter as a filter for an air purifier, we monitored the change in PM concentration in an airtight chamber while running the fan of the air purifier equipped with a PVA NF-based filter (see Figure S2). The volume of the chamber was ~400 L, and the initial PM concentration in the chamber was ~800 µg/m³. A direct current (DC) voltage of 12 V was applied to the fan using a DC power supply (E3648A, Agilent, Santa Clara, CA, USA), and the change in PM concentration was measured using a portable air quality monitor (BR-SMART 126, BRAMC, Beijing, China). For comparison, the performance of commercial air filters was evaluated using the same method.

3. Results and Discussion

Figure 1b shows a photograph of the synthesized PVA NF membrane peeled off from the nonwoven fabric substrate (here, the electrospun PVA NF-based filter was not dried). Figure 1c shows the surface FE-SEM images of a nonwoven fabric substrate and a synthesized PVA NF-based filter. As shown in the figure, the pore size was effectively reduced by the formation of PVA NFs with an average diameter of ~150 nm on a nonwoven substrate with an average diameter of ~20 µm. This means that the pore size of the PVA NF-based filter could be adjusted by controlling the electrospinning time; thus, the limit of the PM size collected by the filter can be reduced (see Figure S3) [1,13].

Figure 2a shows the reflectance spectrum in the visible region (wavelengths from approximately 400 to 800 nm) of the nonwoven fabric substrate before and after PVA NF formation. The inset shows the appearance of the samples. As shown in the graph, the reflectance of the nonwoven fabric substrate significantly increased after the formation of PVA NFs. In fact, the nonwoven fabric substrate appeared brighter after the PVA NF formation. This is because the PVA NFs accumulate on the nonwoven fabric substrate, reducing the pore size and increasing the physical thickness, resulting in a decrease in the amount of light transmitted through the sample [15,19].
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Table 1 shows the measured average fiber diameter, filtration efficiency ($E$), pressure drop ($\Delta P$), and calculated quality factor (QF) of the nonwoven fabric substrate, PVA NF-based filters, and commercial air filters used in this study. The average fiber diameter of the prepared PVA NF-based filter was just one-hundredth that of the commercial air filter media. This implies that the PVA NF-based filter has a much larger specific surface area than the commercial air filter media, which means that it can have a larger dust-holding capacity [16]. Additionally, the filtration efficiency and pressure drop of the PVA NF-based filter increased with increasing electrospinning time [13]. Here, the QF of the optimized PVA NF-based filter (electrospinning time: 300 s) was 0.010606, which is lower than that of a HEPA filter (0.015394), but higher than that of a cabin air filter (0.010517) and a dust mask (0.009102). This shows that the fabricated PVA NF-based filter has a performance level capable of practical use. Figure S4 demonstrates the use of a PVA NF-based filter to block high-concentration PM pollution. The right glass bottle contained a hazardous level of PM with a concentration higher than 1000 µg/m$^3$, and a PVA NF-based filter with a quality factor of 0.010606 Pa$^{-1}$ (filtration efficiency of ~86.81% and pressure drop of ~191 Pa) was placed between the two glass bottles. The PVA NF-based filter successfully blocked the PM moving from the right to left glass bottle. The PM concentration in the left glass bottle was safe (<30 µg/m$^3$). This shows that the PVA NF-based filter can be used as a dust mask and window screen that can block the inflow of PM [15,19].
Table 1. Measured average fiber diameter, filtration efficiency, pressure drop, and calculated quality factor of PVA NF-based filters and commercial air filters used in this study.

| Sample                              | Mean Fiber Diameter [nm] | E [%] | ΔP [Pa] | Qf [Pa⁻¹] |
|-------------------------------------|--------------------------|-------|---------|-----------|
| Nonwoven fabric substrate (without PVA NF) | ~20,000                  | 42.10 | 63      | 0.008674  |
| PVA NF-based filter (Electrospinning time: 60 s) | ~150                     | 50.41 | 79      | 0.008878  |
| PVA NF-based filter (Electrospinning time: 180 s) | ~150                     | 67.76 | 125     | 0.009056  |
| PVA NF-based filter (Electrospinning time: 300 s) | ~150                     | 86.81 | 191     | 0.010606  |
| PVA NF-based filter (Electrospinning time: 600 s) | ~150                     | 89.07 | 220     | 0.010062  |
| Cabin air filter                    | ~22,000                  | 40.27 | 49      | 0.010517  |
| Dust mask (KF80)                    | ~13,000                  | 85.61 | 213     | 0.009102  |
| HEPA filter                         | ~16,000                  | 94.55 | 189     | 0.015394  |

Figure 2b shows the test equipment used in this study for removing the PM particles in the chamber, comprising an air purifier equipped with a filter, and Figure 2c shows the experimental results. As shown in the experimental results, the performance of the air purifier with a PVA NF-based filter was superior to that of the dust mask filter and comparable to that of the commercial HEPA filter. This result shows that the PVA NF-based filter can be used as a semi-HEPA filter for air purifiers [13,16].

Figure 3 shows the optical and structural changes in the PVA NF-based filter (electrospinning time: 300 s) during repeated chamber tests. The chamber test was repeated using an air purifier equipped with the PVA NF-based filter, and the changes in the reflectance spectrum and structural morphology of the filter surface were investigated. The inset in Figure 3a shows that the color of the PVA NF-based filter changed from white to dark brown as the PM capture time increased. As shown in Figure 3a, as the PM collection time increased, the reflectance of the PVA NF-based filter gradually decreased. In particular, the reflectance of the filter decreased significantly at wavelengths from 400 to 600 nm. This is because the PM captured by the PVA NFs selectively absorbs light at wavelengths from 400 to 600 nm [9,13].

The surface morphology of the PVA NF-based filter was investigated using FE-SEM to confirm the PM collection and mechanism. Figure 3b shows the PVA NF-based filter surface before capturing PM. The change in the filter surface morphology during continuous PM capture is shown in Figure 3c–e. Furthermore, the mechanism of the PM capture at different stages is described with the schematics shown in Figure 3f–i. At the initial PM capture stage, PM was captured by the PVA NFs and bound tightly to the NFs (see Figure 3c,g). As the duration of filter usage increased, more PM particles attached to the PVA NFs. The PM particles were able to move along the PVA NFs and aggregate to form larger particles. Additionally, the new incoming PM particles could directly attach to the PM that were already on the PVA NFs and then merge (see Figure 3d,h). As the PM capture continued, the surface of the PVA NF-based filter filled with large aggregated PM particles. The NF junctions accumulated more PM, which formed spherical particles of larger sizes (see Figure 3e,i) [20].
Figure 3. Optical and structural changes in PVA NF-based filter (electrospinning time: 300 s) during repeated chamber tests. (a) Changes in the reflectance spectrum of PVA NF-based filter according to repeated chamber tests; (b–e) surface RE-SEM images of PVA NF-based filters according to the amount of PM collected; (f–i) schematics showing the mechanism of PM capture by PVA NF-based filter at different time sequences.

Finally, as shown in Figure 4a, the filter was soaked in water to selectively discard the contaminated PVA NF layer. Here, the time required to completely remove the contaminated PVA NF layer from the nonwoven substrate was less than 5 s. The remaining nonwoven fabric was dried in an oven and reused as a substrate for the fabrication of a PVA NF-based filter. Figure 4b shows the surface FE-SEM images of the contaminated PVA NF-based filter observed during washing and recycling. In this process, there were no serious problems with the surface morphology and QF of the remade PVA NF-based filter. The result was equivalent to that of the chamber test with the first PVA NF-based filter (see Figure S5).
Figure 4. (a) Selective removal of PM−captured poly(vinyl alcohol) nanofiber (PVA NF) layer by immersion in water; (b) surface field-emission scanning electron microscopy (FE-SEM) images of the PVA NF−based filters during recycling.

4. Conclusions

In summary, eco-friendly air filters based on PVA NFs and nonwoven fabric were fabricated by electrospinning to overcome the limitations of the disposal of existing filters by incineration or in landfills, which cause environmental pollution. The quality factor of the optimized PVA NF-based filter was comparable to that of commercial air filters, such as dust masks, cabin filters, and HEPA filters. Additionally, by measuring the reflectance spectrum of the contaminated filter, it was found that the color of the PVA NF-based filter changed from white to dark brown due to the light absorption by the captured PM particles in the visible light region. Finally, the filter was soaked in water to selectively remove the contaminated PVA NF layer, and the remaining nonwoven fabric could be reused to make the filter. Based on the above experimental results, we conclude that the PVA NF-based filters fabricated in this study can be used as eco-friendly filters for effective PM capture. Furthermore, we expect that the results of this study can be used as a basic research data for the development of NF-based flexible printed electronic devices [21–23].

Supplementary Materials: The following are available online at https://www.mdpi.com/article/10.3390/app11093831/s1, Figure S1: Scheme of PM filtration set, Figure S2: PM removal device used in chamber test, Figure S3: Surface FE-SEM images of samples, Figure S4: Demonstration of the use of a PVA NF-based filter to block the PM entering from the right glass bottle to the left glass bottle, Figure S5: Comparison of chamber test results of a PVA NF-based filter by reusing nonwoven fabric substrate.

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