

## Performance Study of Composite Air Filters Using Heterogeneous Fibers

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**ABSTRACT:** Recently, the worldwide demand for disposable masks has increased due to COVID-19 infections and severe air pollution. Personal masks should reduce breathe resistance while maintaining filtering performance. In this study, a solution blowing process is used to produce composite nanofiber filters to co-spin two polymers at once. The manufacture process of the various fiber diameter filter was designed, and the filtration performance and differential pressure of the prepared filter was investigated. Poly vinylidene fluoride-hexafluoropropylene (PVDF-HFP) and Polylactic acid (PLA) fibers were chosen to be entangled together in a layer with a diameter of 1.05  $\mu\text{m}$  and 0.33  $\mu\text{m}$ . Composite nanofilters showed up to 87% filtration efficiency and 32 Pa differential pressure.

**Key Words:** Air filter, Composite, Heterogeneous fibers, Solution blow spinning

### 1. INTRODUCTION

The coronavirus disease (COVID-19) pandemic and severe air pollution, such as fine dust and yellow dust, have increased demand for disposable masks since the end of 2019. Using medical masks like N95 efficiently protects against infectious droplets and air pollution [1-3]. As a result, wearing a disposable mask for an extended period is essential to successfully prevent the spread of viruses or inhalation of particulate matter (PM). However, if a disposable mask is used for an extended period, it can cause breathing difficulties depending on the wearer's health condition [4]. Therefore, public attention is centered on how comfortable it is to wear and how easy it is to breathe. Microdiameter fibers produced by meltblowing PP (Polypropylene) or PET (Polyethylene Terephthalate) polymers are being used in disposable masks. Since large-diameter fibers have relatively large holes, the filter thickness is raised to achieve infection-prevention filtration performance, or several filters are employed [5,6]. A disadvantage exists in the case of a disposable mask with a nanofiber filter: the high differential pressure caused by small pore diameters disturbs the user's breathing. Furthermore, when

the pores are small, the differential pressure rises rapidly because fine dust gathered in a mask clog the pores between the fibers when worn for an extended period. These issues contribute to a disposable mask's service life being cut short. Therefore, recent studies [7-9] have proposed a hybrid structure mixing large and small fibers or layering to solve each problem. This suggests that it may be useful in lowering pressure difference while retaining the filtering performance of a filter.

Roh *et al.* investigated the role of intra and interlayer space of filter media on the pressure drop development with continued particle loading. The intermingled structure of micro-fiber and nanofiber has higher porosity and improved quality factor (QF) than nanofiber [10]. ZheWang investigated multi-layered filters with varying fiber diameter and fabricated a filter with high filtration performance and low pressure difference [7]. Yang *et al.* conducted an experiment to lower differential pressure by employing a fiber bead and securing a gap layer between fibers [9]. Shichao Zhang developed single-layer filters with varied fiber diameters using electrospinning [11]. Solution blow spinning (SBS) is a method used to produce fibers with nano-to-micro diameters. Concentric nozzles are used in SBS, with an inner nozzle extruding a polymer solution and an

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outer nozzle discharging high pressure gases (air, nitrogen, argon, etc.). The polymer solution forms droplets by being forced inflowing at a constant feed rate through an internal nozzle. High-pressure compressed gases passing through the external nozzle deform the polymer solution into a cone shape. When the surface tension of the solution is overcome, a solution jet is ejected and accelerated toward the target to be collected. During an ejection, the solvent is rapidly evaporated and polymer fibers are finally collected on a collector [12].

The SBS method has advantages of high yield, short preparation time, and high practicality. Because the SBS method is not limited by static electricity or the solution dielectric constant, it is suitable for voltage insensitive polymers, and the mass production of nanofibers and large-scale industrial production is possible. Accordingly, the SBS process has been receiving attention gradually recently as a new spinning technology. The SBS method has been widely used in various fields such as biopharmaceutical [13-16], energy materials [17-20], sensors [21,22], and filters [23-25]. The SBS process is used in this study to produce single-layer composite nanofiber filters with varied fiber diameters in a short period. The surface potential of the filter manufactured in the experiment was measured, and the spinning shape was compared to the performance efficiency of the filter.

## 2. EXPERIMENT

### 2.1 Materials

For this study, Polylactic acid (PLA) was purchased from Rainbox Filament by Hanil Industrial Co., Korea. Poly vinylidene fluoride-hexafluoropropylene (PVDF-HFP; Mw = 400 kDa) was purchased from Sigma-Aldrich, and acetone was purchased from SK chemical Co., Ltd, Korea.

### 2.2 Preparation of Solution Blown Filter Media

Each 12 wt% solution of PLA and PVDF-HFP was prepared in acetone at 35°C on a hot plate with magnetic stirring for 6 h. Two-stream solution blowing was performed for SBS by simultaneously solution blowing of PVDF-HFP solution and PLA solution from two perpendicular capillaries, as shown in

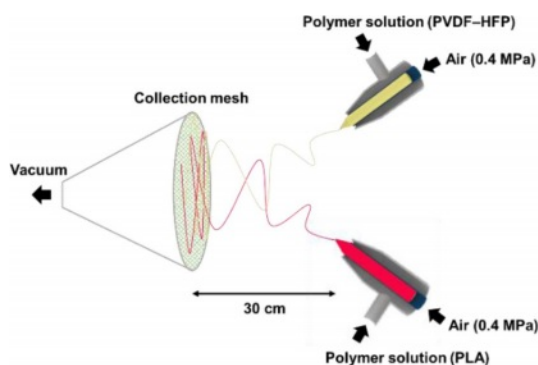


Fig. 1. Schematic of SBS simultaneous spinning

Fig. 1. The polymer solution jet was released using an 18-gauge needle at a pumping rate of 30 mL/h. For single SBS, PVDF-HFP and PLA, air was released at a pressure of 0.4 MPa for 4 min. In the case of dual SBS, PVDF-HFP/PLA, air was released at a pressure of 0.4 MPa for 2 min. A vacuum was coupled to the bottom of the poly-propylene mesh to collect the blown fibers. The distance between vacuum-connected collection mesh and nozzles was 30 cm. The entire spinning process was conducted at 20 °C and the relative humidity of 30%.

### 2.3 Characterization

Scanning electron microscopy (NovaNanoSEM450, FEI, USA) at a voltage of 20 kV was used to examine the morphology of the fibers and filtered PM. The as-spun fibers' pore size distribution was evaluated using a capillary flow porometer (CFP-1500Ae, Porous Materials Inc., USA) at 5 psi. Individual fiber diameters were measured from at least 100 points and averaged. Additionally, the surface potential of each sample was measured over time, as this affects the filter performance.

### 2.4 Filter test of Solution Blown Filter Media

All samples are exposed to ambient air for 24 h before the filter test. The PVDF-HFP and PLA sample were lightly clapped 20 times each other, and the non-rubbed cases were measured as well. The cases in which the sidewalls of the spun fibers were rubbed 20 times and the non-rubbed case was measured in the case of samples in which PVDF-HFP and PLA were simultaneously spun. All samples were cut in a 50 mm diameter.

The efficiency of as-spun fibers in filtering of aerosols was investigated by measuring the size distribution of aerosol at inlet and outlet of tested filters using a scanning mobility particle sizer (Model 3926, TSI, USA). Aerosol was generated by atomizing 1.0 wt% distilled water solution of potassium chloride (KCl). The distribution of particle sizes is presented in

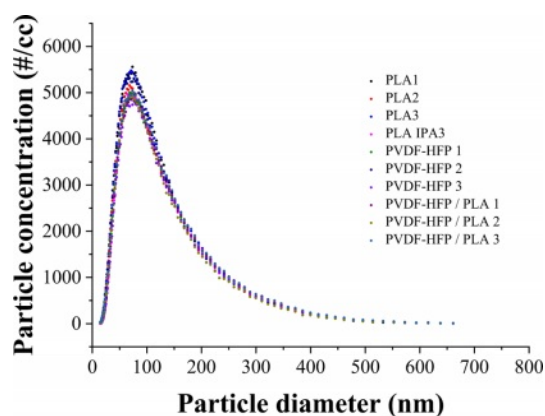


Fig. 2. Size distribution of KCl particles used in aerosol filtration test

Fig. 2. The size distribution of the KCl particles at the inlet was almost kept same in all the tests. The surface area of the respirator was approximately 25 cm<sup>2</sup>, and the face velocity at 4.0 LPM corresponds to approximately 5.3 cm/s. The inlet/outlet concentration for the filtration efficiency was measured in the order of “IN-OUT-IN” at 3.7 LPM for each sample, and the inlet concentration was averaged as following Eq. (1).

$$\text{Filtration efficiency (\%)} = \frac{C_i - C_0}{C_i} \times 100 \quad (1)$$

### 3. RESULTS AND DISCUSSIONS

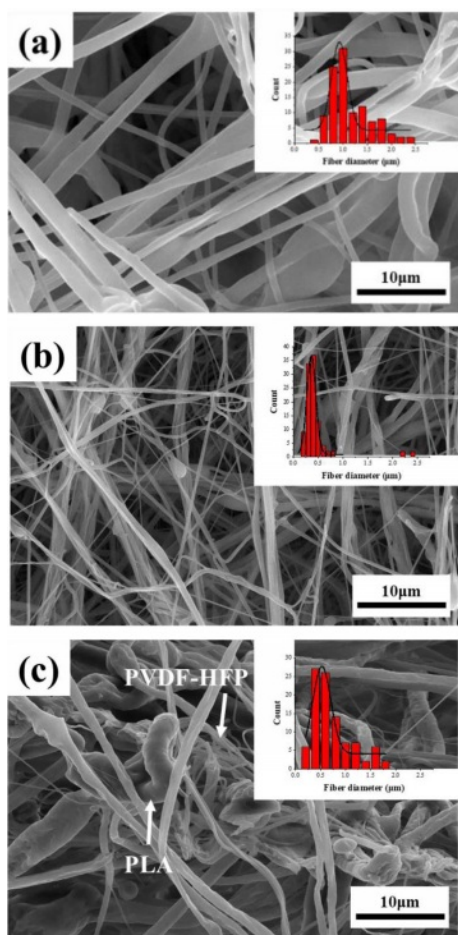
#### 3.1 Morphology of Filter Media

The images of the as-spun fibers are summarized in Fig. 3. Fibers were prepared with 12 wt% PLA and PVDF-HFP solution, spun at 0.4 MPa and 30 cm distance for 4 min in single spinning, and 30 mL/h of flow rate for 2 min in simultaneous spinning. The diameters of the spun fibers were measured at 100 points and averaged. The distribution of fiber diameters is summarized as a histogram in Fig. 3. The average diameters of

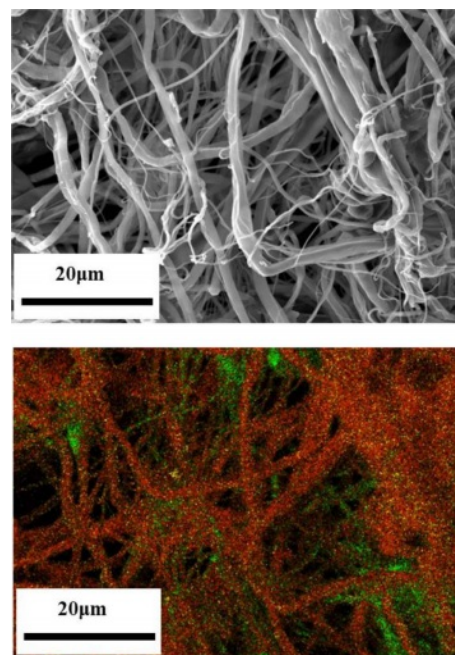
the PLA, PVDF-HFP, and PVDF-HFP/PLA were  $1.05 \pm 0.42$ ,  $0.33 \pm 0.12$ , and  $0.60 \pm 0.40$  μm, respectively. Despite the same solution concentration, PLA had a thicker fiber diameter than PVDF-HFP, and in the case of simultaneous spinning, fibers of various diameters could be confirmed through the diameter distribution diagram to be mixed.

To ensure that the two polymer fibers were uniformly mixed in the simultaneous-spun PVDF-HFP/PLA fibers, an F (fluorine) element mapping was done using EDS. The results are shown in Fig. 4. The green in the image showed the F element present in the PVDF-HFP. The PVDF-HFP fibers could be confirmed to be entangled with the PLA fibers, and the space between the PLA fibers was occupied by PVDF-HFP fibers making it better filter performance.

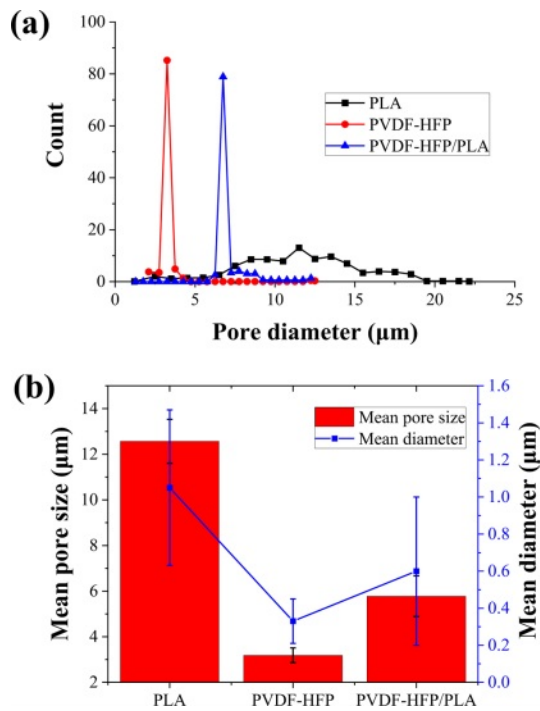
A pore distribution was measured examine the average pore size and pore distribution of the two types of spun fibers. The pore sizes of each fiber were  $12.57 \pm 0.96$ ,  $3.1 \pm 80.32$ , and  $5.78 \pm 0.89$  μm for PLA, PVDF-HFP, and PVDF-HFP/PLA, respectively, and the pore size distribution is shown in Fig. 5a. By forming different diameters, the two types of polymer solutions influence pore sizes. When seen in the pore size distribution diagram, the PLA with a large fiber diameter case could be confirmed of distributing over a wide range, and larger pore size than cases in spun with the PVDF-HFP only and spun simultaneously with the PVDF-HFP and PLA. Furthermore, as shown in Fig. 5b, the diameter and pore sizes could be seen as having the same tendency. PVDF-HFP/PLA is in between PLA and PVDF-HFP. As seen in the previous SEM (Scanning Electron Microscope) images, this was because the PLA fiber with a large diameter made a large space



**Fig. 3.** SEM image and fiber diameter distribution of each as-spun fiber (a) PLA, (b) PVDF-HFP, (c) PVDF-HFP/PLA



**Fig. 4.** Elemental mapping image of PVDF-HFP/PLA (X 1500, green: F)



**Fig. 5.** (a) Pore size distribution of each spun fiber, (b) mean pore size and mean diameter

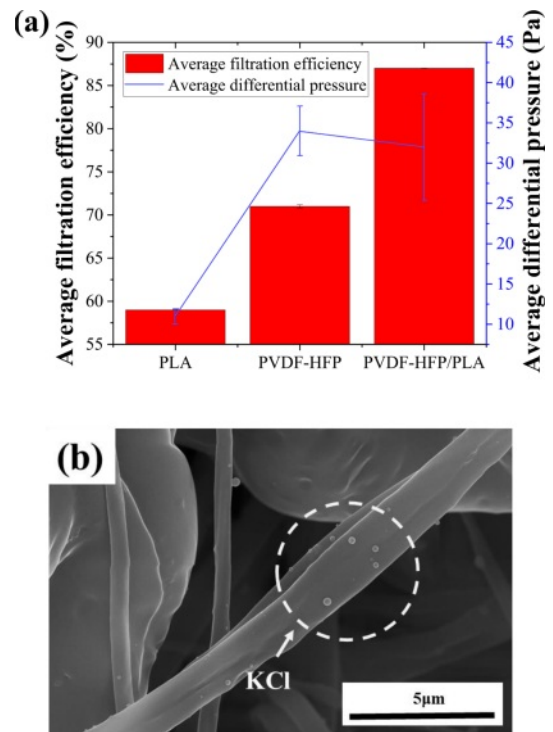
between the fibers and thin PVDF-HFP fiber occupied the space. Thus, it was confirmed that the pores sizes could be controlled by SBS of the two fibers concurrently.

### 3.2 Filtration Performance and Differential Pressure

To confirm the filtration ability of heterogeneous fibers filters, a filtration test was performed. As shown in Fig. 6a, for the filtration performance, PLA, PVDF-HFP, and PLA/PVDF-HFP were measured by  $59 \pm 0.03\%$ ,  $71 \pm 0.18\%$ , and  $87 \pm 0.02\%$ , respectively, and the differential pressure by  $11 \pm 0.94$ ,  $34 \pm 3.09$ , and  $32 \pm 6.60$  Pa, respectively. When the results were compared, the PVDF-HFP sample showed higher efficiency and differential pressure than the other cases because the PVDF-HFP had smaller pores than the other cases. Compared to the other two cases, the PLA sample had the lowest filtration efficiency and differential pressure.

The previous findings showed that since the PLA had the largest fiber diameter and pores, particle matters could not be captured and escaped through the pores between the fibers.

In PLA/PVDF-HFP, the PLA fiber with a large diameter made a large space between the fibers and thin PVDF-HFP fiber occupied the space. that perhaps not only improve the capture ability of the particles but also prevent the penetration of air flow across the fibers. Thus, it is very important to optimize the structure, including through-pore size, and the combination form of fibers with different diameters, of composite membranes by dual SBS to gain high filtration efficiency with a relatively low pressure drop.



**Fig. 6.** (a) Filtration efficiency and differential pressure, (b) SEM image of PVDF-HFP/PLA after filter test (X8000)

**Table 1.** Parameters of commercial respirators

Sample	Pressure drop (Pa)	Filtration efficiency (%)	Reference
N95 grade	87	99.5	[26]
KF94 grade	82	99.5	[26]
UVEX-FFP2	80	$\geq 99$	[27]
3M 1860	88.26	$\geq 99$	[28]
3M 1870+Aura™	69.73	$\geq 99$	[28]

A surface analysis was conducted for the samples, which were proceeded adsorption experiments before and after the adsorption of PVDF-HFP/PLA samples. As can be seen from the SEM image in Fig. 6b, the round particles adhered to the fiber surface were KCl particles collected on the fiber during the filtration experiment.

Table 1 summarizes the differential pressure, and filtration performance of commercially available masks used to compare the performance of the PVDF-HFP/PLA sample with that of air filters employed. In the case of commercial masks, the surface exposed to the exterior and inner surface in touch with the respiratory organ were made of spun bonded fiber with 1 or 2 sheets of melt-blown fibers sandwiched in between. As a result, it was proven that the differential pressure in the overlapped filter form of many layers was more than that of the filter created in this study to achieve excellent filtration performance. Adjusting the filter thickness and



increasing the spinning time of the simultaneously spun sample could result in improved filtering performance.

## 4. CONCLUSIONS

The global demand for personal masks has risen sharply due to the covid-19 pandemic and air pollution. In this regards, many studies on a better mask that lowers breathing resistance while maintaining filter performance has been steadily progressing. In this study, we successfully composite two different polar polymers into nanofibers via the co-spinning of solution blowing. In particular, PLA and PVDF-HFP fibers were chosen to be entangled together in a layer with a diameter of 1.05  $\mu\text{m}$  and 0.33  $\mu\text{m}$ , respectively. The mean pore diameter of each fiber is 12.57  $\mu\text{m}$  and 3.1  $\mu\text{m}$ , but that of composite filter was measured to 5.78  $\mu\text{m}$ . The composite nanofilters made of two different polar materials showed filtration efficiency of up to 87% that is even higher than each PLA (59%) and PVDF-HFP (71%) while keep the differential pressure not to increases (32 Pa). From the aforementioned strategy, it is expected to improve the personal mask materials to reduce the breath resistance with high filtration performance.

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