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Highly Efficient, Flexible, and Recyclable Air Filters Using Polyimide Films with Patterned Thru-Holes Fabricated by Ion Milling

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Abstract: We present the first demonstration of the environmental application of a polyimide film (Kapton) with patterned thru-holes as a novel, efficient, flexible, recyclable, and active particulate matter (PM) air filter. The Kapton air filter captures PM using micro-sized thru-holes and the strong electrostatic force because of its high work function. It is highly efficient, transparent, flexible, and heat-resistant. Furthermore, it can be recycled simply by washing it with tap water. The proposed PM filter is a promising candidate for use as a highly efficient and economical recyclable air filter for home appliances, such as air-purifiers, air-conditioners, and humidifiers, as well as industrial filtration systems.

Keywords: polyimides; air applications; properties & characterization; filter

1. Introduction

Recently, emerging environmental pollution issues—especially, air pollution produced by emissions from residential energy use (heating, cooking), traffic, and power generation—have become serious problems worldwide. Such air pollution has serious long-term effects on the global atmosphere and public health.

In recent years, there has been growing concern about the harmful effect of very fine particulate matter (PM) in the air as the PM can be absorbed into and eventually settle in the human body [1–7]. For example, PM with a diameter that is smaller than 10 µm (PM10) can pass through human tissue and cause lung disease [8]. Furthermore, PM with a diameter that is smaller than 2.5 µm (PM2.5) leads to increased plaque deposition in arteries, causing vascular inflammation and atherosclerosis, which can lead to heart attacks and other cardiovascular problems [9].

Conventionally, PM10 is filtered using general thread filters, and the smaller PM2.5 is filtered using high-efficiency particulate arresting (HEPA) filters. In modern air-conditioning systems, HEPA filters are generally made from a fabric of tangled, warped fibers and they work by pulling or pushing the air through a filter screen that catches the PM. The fibers are randomly arranged and these randomly oriented individual fibers force the air through an indirect, non-linear path. As the particles pass through the filter, they are caught in the fabric maze by four mechanisms: impaction, sieving, interception, and diffusion.

However, the conventional PM filter systems have many inherent problems. First, the bulky structure of the HEPA filter requires a large working space and causes a large pressure difference between the inlet and outlet of the filter, limiting the efficiency of air circulation [10,11]. Second, HEPA filters cannot be cleaned, allowing potentially hazardous micro-organisms to accumulate and prosper in the filters. Lastly, the HEPA filters are not recyclable and must be discarded after use, causing environmental pollution.

In order to remove these limitations, various PM filter systems based on new materials and production technologies have been developed. Polar, polymeric nanofiber networks based on surface chemistry, using polyacrylonitrile, nylon, and polyimide, have been proposed as potential transparent PM filters with high efficiency [12–15]. However, such filters are single-use and not reusable. Wang et al. examined hot-gas filters composed of a polyimide nanofiber membrane sandwiched between woven-carbon fabrics that can be recycled by back-air flushing [16]. Recently, Jeong et al. employed metal nanowire percolation networks to develop a transparent, reusable, and active PM filter, with a high efficiency and small volume, using a hierarchical Ag nanowire percolation network on a nylon mesh [17]. Their filter can be reused multiple times only after cleaning with certain agents, such as ethylene glycol. Furthermore, the use of clay nanoparticles [18] and biopolymers [19] has been suggested for adsorption of pollutans.

In this research, our aims in developing a new air filter can be summarized as follows. First, we aimed to minimize the pressure difference between the inlet and outlet while maintaining the performance of the filter in order to allow a less powerful fan to be used, thus lowering the power consumption. Second, the new filter should be easily cleanable to prevent the accumulation and growth of molds of fungus of micro-organisms in the filter, and be reusable. Third, the large-area filter should be made from only one material and its fabrication process should be as simple as possible. Lastly, the filter should be flexible and heat-resistant. The filter is not required to be rectangular, allowing innovative designs and applications. The filter should have adequate thermal stability.

To achieve these goals, we present a demonstration of the application of a Kapton film—with micro-sized, patterned thru-holes—to air filtration. Various micro machining processes (wet/dry etching, laser drilling, and so on) for polymer films have been proposed for making patterned thru-holes using laser beam, chemical etching and dry etching processes for use as a shadow mask or a peel-off stencil mask in production of optoelectronic devices [20–22]. However, it has been challenging to make tiny thru-holes with small (under 10 μm) distances between adjacent holes in polymer films due to the weak heat resistance of polymer materials during dry etching process. The laser drilling technique may not be appropriate to produce large area films economically. In the case of wet etching, the distance between holes cannot be minimized due to the isotropic etching property. To the best of our knowledge, it has been almost impossible to realize thru-holes on polyimide films with the ion ethching process. The ion collision process generates the heat, which causes a change in the polymer film during the process. In addition, the thin polymer film cannot be easily attached at the bottom of the sample holder during the process. Even if the film can be attached by special equipment (i.e., electrostatic chuck), the damage of high cost bottom holder cannot be ignored. Nano-porous polymeric films and membranes having holes with diameters of few tens of nanometers that were etched out selectively by acid hydrolysis after self-assembly of the block copolymers were also proposed for nanomaterial synthesis, size-selective catalyst supports, and advanced separation [23].

In this study, we report the realization of Kapton films with patterned thru-holes by ion etching process and the application of the Kapton air filter for use as an active air filter by exploiting its own superior properties of flexibility, transparency, heat resistance, mechanical durability, and reusability. We developed a new ion punching machine and process for the polymer film. The characteristics can be summarized as follows: (1) the process temperature is low, which does not elevate the temperature of the sample and (2) the adhesive material, which is soluble in acetone, is inserted between a polymer film and a sample holder to attach the film during the process. The small thru-holes effectively filter PM10 and the high electrostatic field of the Kapton film effectively captures PM2.5. The intrinsic characteristics of the film (tough, washable, and heat resistant) allow it to maintain its initial performance after a simple cleaning process using tap water. Furthermore, the thinness of the single layer Kapton films and 25% opening ratio significantly reduce the pressure difference between the inlet and outlet, allowing effective air flow through the filters.

2. Materials and Methods

A 25 μm film-type photoresist (PR;RY-3625, HITACHI Chemical, Tokyo, Japan) was tightly attached to a 5 μm-thick Kapton film by lamination. The prepared film was then exposed using an i-line contact aligner and developed with 1 wt% $NaCO_3$ solution at 20 °C. The photolithographed film was machined by ion milling according to the following protocol. A specifically designed ion milling machine with an ion-gun coverage area of 400×500 mm^2 was manufactured by Vault Creation Corporation. The etch rate was 7 Å/s and the film was attached to an electrostatic chuck, which was also manufactured by Vault Creation Co. (Seoul, South Korea). The thru-hole diameter was 15 μm and the interhole distance was 30 μm. A schematic of this fabrication process is shown in Figure 1. Smaller thru-holes do not allow enough flow for efficient air filtration in practice. The Kapton air filters with a large area can be produced massively and reproducibly.

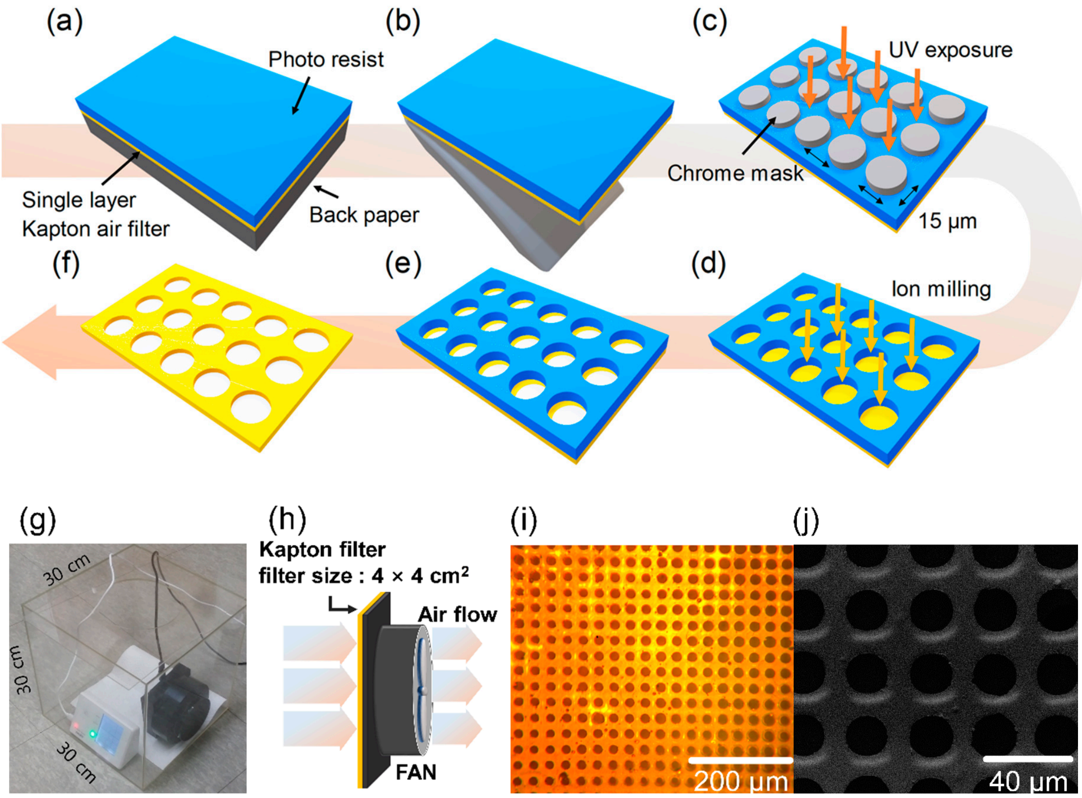

Figure 1. The process flow to produce a Kapton air filter. (**a**) Attach Photo resist (PR) on a Kapton film by laminating process. (**b**) Back paper was removed. (**c**) Photolithography obtained with a typical chrome mask and i-line exposure. (**d**) Etching process by ion milling to produce thru-holes in the Kapton air filter and PR. (**e**) Kapton film and PR with patterned thru-holes. (**f**) Kapton air filter with PR removed. (**g**) The closed chamber for measuring the performance of the Kapton air filter. The top hole can be closed by an another acryl plate. Mosquito pyrethrum coil was burned in the chamber for 1 min in a closed state to produce PM. (**h**) The schematics of the air cleaning system. (**i**,**j**) are optical microscopy (OM) and Scanning Electron Microscopy (SEM) images of a Kapton air filter as prepared, respectively.

3. Results and Discussion

The prepared Kapton film with thru-holes was tested by measuring their single-pass efficiency for filtration of dust-particle sizes ranging from 0.3 μm to 10 μm. After this, in order to investigate the performance of the prepared filters under realistic working conditions, a closed-chamber air cleaning system was used, which is shown in Figure 1g. The structure of the air cleaning system is simple but includes the necessary components (a filter and fan) to simulate commercial air purifiers (Figure 1h). A pyrethrum mosquito coil was burned in the chamber for 1 min to produce dust particles and the air condition was measured without the air cleaning system operating. After cleaning the chamber, another pyrethrum mosquito coil was burned in the chamber, and the PM density of the air with the air cleaning system was measured in order to allow for comparison of the two situations. A commercial PM sensor (Air Quality Monitor JSM-136, Shenzhen Aocome Tech. Co., Guangdong, China) was used to measure the PM2.5 density of the air, which is shown in Figure 1g.

Figure 1i,j show the optical microscopy (OM) and scanning electron microscopy (SEM) images of the as-prepared Kapton air filter, respectively. The SEM (Quanta 250FEG, ThermoFisher Scientific Co., Waltham, MA, U.S.A.) and energy-dispersive X-ray spectroscopy (EDS; Apollo X, AMETEK Materials Analysis Division Co., Mahwah, NJ, U.S.A.) measurements were conducted using an acceleration voltage of 20 kV and a working distance of 10 mm. Uniformly spaced pores with diameters of 15 μm are seen in the Kapton film. There are two notable observations. First, the polymer material was finely etched and micro-sized pores were bored without any burning. Second, the pores were not just formed on the surface but penetrated the film with an excellent aspect ratio.

PMs with various sizes were generated and passed through the filter once. The single-pass filtering efficiencies of the filter for different PM sizes (0.3–10.0 μm) were measured and listed in Table 1.

Table 1. The single pass efficiency of the Kapton filter according to size of dust particulates.

Particle Size (μm)	Initial Fractional Efficiency (%)
0.3~0.5	27.9
0.5~1.0	45.2
1.0~2.0	55.3
2.0~5.0	64.6
5.0~10.0	78.2

Although the diameter of the thru-holes is 15 μm, the Kapton air filter captures PMs with a diameter that is smaller than 10 μm while 0.3–0.5 μm sized PMs were also captured with an initial filtering efficiency of 27.9%. This single-pass efficiency is high enough to indicate that the repeated circulation of air through the filter will be able to effectively remove such particles. One of the mechanisms of capturing PMs was electrostatic force. Furthermore, the results imply that a strong electrostatic force is present at the surface of the Kapton film, which attracts PMs that are smaller than the thru-holes due to the electrostatic force created by friction from air flow. The high electrostatic force results from the large inherent work function of Kapton of 4.36 eV [24] as the large difference between the work functions of two materials produces a high electrostatic field at their interface. The electrostatic field is inversely proportional to the distance between the materials and the micro-sized thru-holes enhance the field effectively. A SEM image of the Kapton filter after capturing the dust particles is shown in Figure 2a–d.

In order to test the performance of the filter in a realistic environment, a pyrethrum mosquito coil was burned for 1 min in a closed chamber. After burning and removing the coil, the measured PM density with and without the air cleaning system (the Kapton filter and fan; Figure 1h) was measured.

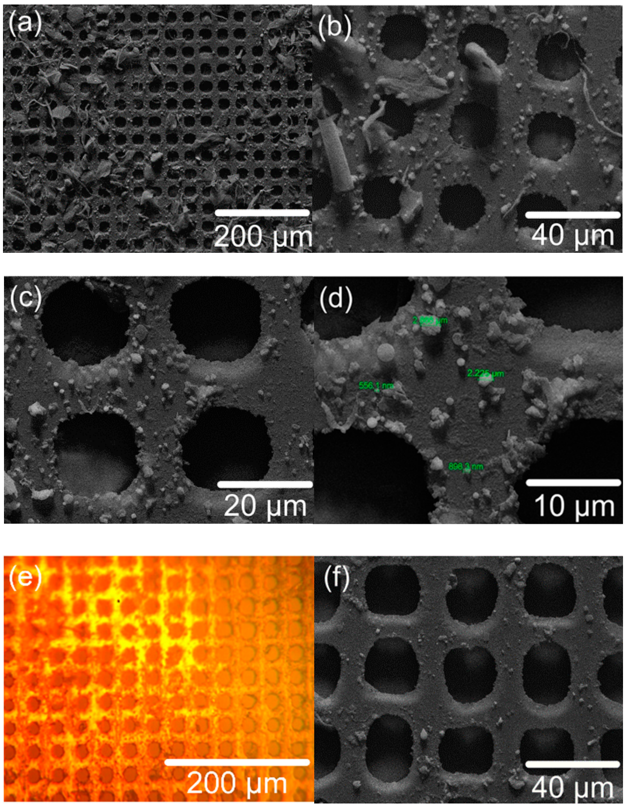

Figure 2. SEM images of a Kapton filter operated for 7 days. Micro dust is captured at the surface and around the pores. The magnifications are (**a**) 200×, (**b**) 900×, (**c**) 1500×, and (**d**) 2900×. Representative dimensions of the PM are shown in (**d**). (**e**) Optical microscope and (**f**) SEM images of a Kapton filter washed with tap water after filtration.

Figure 3a shows the evolution of the PM2.5 densities as a function of time, demonstrating the superior performance of the Kapton air filter. The error bars indicate the variation (±one standard deviation) of the repeatedly measured values by the particle detector. Only representative bars are displayed and the error for all data is the same. Without the operation of the air cleaning system, the PM density remained at a hazardous level (above the detection limit of 500 $\mu g/m^3$) for 400 min, as expected. On the other hand, when the simple air cleaning system was installed and operational, it took only ~23 min for the PM density in the closed chamber to be reduced to a safe level (below 50 $\mu g/m^3$). It should be noted that the dimensions of the Kapton filter were only 40 × 40 × 0.025 mm^3, which are much smaller than the filters in commercial air purifiers. The Kapton air filter used was a single layer and thus, a very low-power fan can be utilized.

The advantages of Kapton and polymer films include their water resistance, corrosion resistance, and high thermal stability. In order to exploit these advantages, we investigated the possibility of reusing the filter by washing it with tap water. Figure 2e,f show the surface morphology of the Kapton air filter after washing it with tap water. As mentioned for the single-pass filtering experiment, very fine particles were captured, which is shown in Figure 2f. It can also be seen that the surface of the Kapton filter has been effectively cleaned after being washed with tap water, compared to the SEM images in Figure 2a–d. No special cleaning agents or processes were needed as previously reported [14]. Thus, the filter can be recycled very easily and is very economical.

To investigate the durability and reliability of the filter, multiple washing processes were performed. Figure 3b shows the PM removal performance test after each cleaning cycle. Each filtration test was performed in the same conditions, which is shown in Figure 2. The PM2.5 density in all the experiments dropped to 40 $\mu g/m^3$ within 30 min. In this study, the proposed Kapton air filter demonstrates good PM removal performance and excellent durability and reliability for multiple uses after being cleaned with tap water.

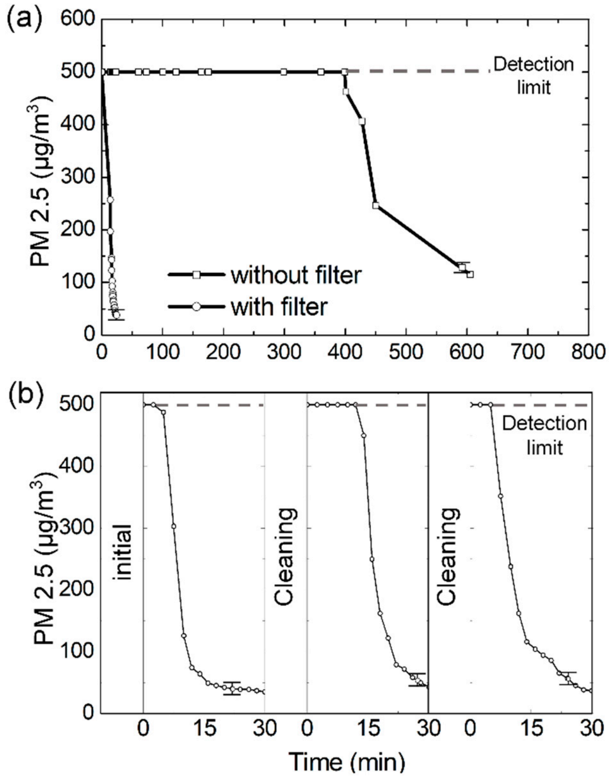

Figure 3. (**a**) Evolution of PM2.5 density as a function of time with and without the Kapton air filter. The detection limit was 500 μg/m^3. (**b**) The reliability and durability of the Kapton air filter after repeated washing.

Lastly, in order to verify the endurance of the Kapton films under high temperature conditions, such as gas outlets in factories and laboratories, the Kapton air filter was heated at 350 °C for 10 min. Figure 4 shows a SEM image of a Kapton film after heating. As expected, there has been no significant deformation or introduction of defects during the heating process. The decomposition temperatures (on-set temperature and 10% mass loss temperature) of the Kapton film determined by thermogravimetric analysis are 558.97 °C and 577.21 °C, respectively, and the glass transition temperature was determined to be 401.67 °C by differential scanning calorimetry analysis. To the best of our knowledge, no other commercial air filters can withstand such high temperatures. These results also indicate that the filter can easily be sterilized by being heated in home appliances, such as conventional and microwave ovens, preventing any micro-organisms from surviving. This is another notable advantage of the proposed air filter.

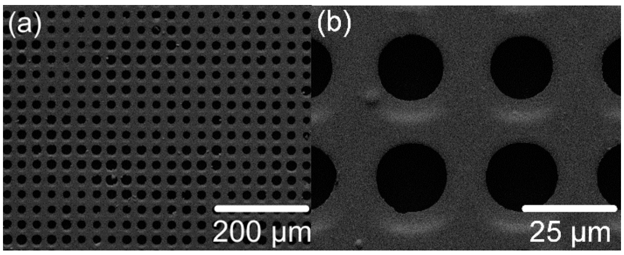

Figure 4. SEM images of a Kapton filter after heating at 350 °C for 10 min. The magnifications are (**a**) 200× and (**b**) 1500×.

4. Conclusions

We developed a Kapton film with patterned 20 µm-sized thru-holes by ion milling for use as a novel, efficient, flexible, recyclable, and active PM2.5 air filter. The single-layer Kapton air filter thru-holes effectively captured PM of various sizes (0.3–10.0 µm) through its micro-sized pores and strong electrostatic force (arising from its high work function). Although the diameter of the thru-holes is 15 µm, the Kapton air filter captures PMs with a diameter that is smaller than 10 µm. The single-pass filtering efficiency of 0.3–0.5 µm and 5.0–10.0 µm sized PMs are 27.9% and 78.2%, respectively, and these values will be significantly enhanced in repeated air circulation. Furthermore, the filter was reusable after multiple and simple washing cycles even with tap water and it was stable when heated at 350 °C for 10 min. Therefore, the proposed filter is a promising candidate for use as a highly efficient and economical recyclable air filter for various applications, including home appliances and industrial filtration systems.

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Conflicts of Interest: The authors declare no conflict of interest.

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