Environmental storage conditions influencing the filtration behavior of electret filters with repeated use

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Abstract
Worldwide attention has been paid to effective protection strategies against the COVID-19 pandemic. Filtering masks are generally kept for a certain period of shelf-life before being used, and frequently, they are used repeatedly with recurrent storages. This study investigates the effect of storage temperature and humidity on the structural characteristics and charges of an electret filter, associating with the filtration performance in terms of efficiency and pressure drop based on a practical use-storage scenario. For the repeated use conditions with recurrent storage, humid storage conditions significantly deteriorated the filtration efficiency as hygroscopic particles quickly wetted the surface and masked the surface charges. The high temperature rapidly deteriorated the filter charges and caused a lowered electrostatic filtration efficiency. In a heated condition, the web became fluffer, yet it did not directly affect the pressure drop or mechanical filtration efficiency. The approach of this study is progressive in that rigorous analysis was performed on examining the particle morphology and internal structure of filter media with varied storage conditions to link with the filtration performance and the effective lifetime. This study intends to provide a scientific reference guiding a desirable storage condition and replacement cycle of filtering masks considering the actual use habits and storage environment.

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Keywords
electret filter, storage, repeated use, filtration, structure, charge, effective lifetime

Introduction

Wearing a filtering mask is straightforward and basic protection from the exposure to COVID-19, and the use of mask has become a daily necessity in the pandemic situation. With the prevalent use of filtering masks, concerns over the shortage of masks were addressed, especially in the earlier period of the COVID-19 outbreak.1,2 Practically, masks are stored on a shelf for a while before being used, and the same mask is used repeatedly for an extended period of time with recurrent exposures to environmental contaminants.3 Regarding this usage pattern, critical concerns are brought about on how storage condition between the recurrent uses affect the filtration performance, but there is no standard method to test the performance in such a use scenario.4,5 The US National Institute for Occupational Safety and Health (NIOSH) standard is designed to simulate the use scenario where the filters are continuously exposed to particles/aerosol up to 200 mg of aerosol mass.6–8 Without prior knowledge on the effect of storage condition on the filtration performance, it needs clarifications of how such use habits affect the physical and electrical properties of filtering masks.

The filtration characteristics in terms of pressure drop and efficiency are affected not only by the filter material characteristics, which is well studied elsewhere,9–19 but also by the use habits such as repeated wearing. Regarding the environmental storage condition, the preconditioning of filter material at high temperature and/or high humidity resulted in a significant reduction of filtration efficiency,20–27 and the implication of this result can be associated with the shelf-life storage condition. Notably, previous studies by Motyl et al.24 and Liu et al.25 reported that filter storage at high temperature and humidity, prior to use, may cause charge decay of an electret filter, reducing the filtration efficiency. Lee and Kim26 suggested that the moisture absorption of a filter material critically affected the filtration efficiency after an electret filter was exposed to a high humidity environment. Yang et al.27 investigated the influence of indoor humidity and temperature during the filtration test, and reported both temperature and humidity interactively affected the filtration efficiency. While those studies reported the relation of environmental temperature and humidity with filtration performance, most of the studies conducted the environmental treatment on the unused filters, lacking the observation for the environmental effect during the repeated usage. The implication here is that for the recurrent usage, the characteristics of particles that are loaded on filter media may be changed by the storage temperature and humidity.28 Thus, it is necessary to investigate the effect of storage conditions on the filtration performance with the repeated recurrent use scenarios.

The tendency of pressure drop and efficiency with repeated and recurrent use was previously investigated,29,30 in which the humidity was reported as a factor affecting the filtration characteristics. Unlike the continuous loading where sodium chloride particles built a significant pressure drop, the intermittent exposure with humid storage conditions did not show a rapid increase of pressure drop. While Lee et al.29 reported about the
filtration tendency with the intermittent use habits, the information is limited to the mechanical filtration mechanism. However, both electrostatic and mechanical filtration would be affected by the storage conditions, and it needs an extended study to consider both aspects in determining the effective lifetime. Also, a thorough analysis is needed to probe the effect of storage conditions on the morphology of filter materials and the resulting mechanical filtration.

This study investigates the influence of storage conditions on the filtration behavior with one-time continuous use and repeated recurrent use, employing an electret polypropylene meltblown media and NaCl nanoparticles (count median diameter $\sim$75 nm). The focus of the study is to reveal how the storage temperature and humidity leads to morphological changes and the distribution of particulate mass retained in filters, and the findings are associated with filtration performance and effective lifetime. Particularly, this study intends to reveal the effect of storage conditions on the filtration performance with repeated recurrent use scenario. The analytical emphasis lies in the detailed 3D observation of fiber morphology, particle distribution in the filter media, and dimensional change in the web with varied storage conditions. It is anticipated that the result of this study provides a scientific reference to guide appropriate use habits and storage conditions. Figure 1 illustrates the schematic diagram of the research overview.

**Materials and methods**

**Materials**

A commercially available charged polypropylene (PP) meltblown media was obtained from UnionFilTech (South Korea) and used as received. The filter media had the basis weight of $32.1 \pm 0.2 \ g \ m^{-2}$, the thickness of $229 \pm 17 \ \mu m$, the porosity of $86.5 \pm 0.8\%$, and mean fiber diameter of $5.1 \pm 2.2 \ \mu m$. 

![Figure 1. Schematic diagram on the research process.](image-url)
**Particulate filtration tests**

Filtration test was conducted using an automated filter tester (TSI 8130, TSI Inc., USA) with sodium chloride aerosol. An aqueous solution of 2% (w/w) NaCl (Daejung, Republic of Korea) was prepared for generating NaCl aerosol (count median diameter, CMD \(\sim 0.075 \pm 0.020 \mu\text{m})). The charges of NaCl aerosol were equilibrated to have zero net charge in Boltzmann distribution. In TSI8130 filter tester, there are built-in light scattering laser photometers. The photometer measures the amount of light scattered by the particles, and calculates the upstream and downstream particle concentrations simultaneously. The penetration (%) of particles is determined by the ratio of downstream to upstream particle concentrations measured by the photometer.

The environmental effect on filtration efficiency was investigated in two different approaches: (1) continuous storage (for 37 days) and continuous particle exposure (up to 40 mg, \(\sim 3\) h testing), (2) repeated and recurrent particle loading (2.2 mg, 9 min testing) and recurrent storage in between the particle loading (for all day except the particle loading time and 30 min pre-conditioning time) every day, up to 37 days. Environmental storage conditions were determined based on practical applications such as: (1) 23°C, 50% RH simulating common office condition; (2) 40°C, 75% RH simulating a hot and humid weather; (3) 40°C, \(\sim 0\)% RH simulating a hot and dry shelf-life; (4) 90°C, \(\sim 0\)% RH, simulating a disinfection method in oven for reuse of respirator.17

Face velocity of 2 cm/s was used for the test, assuming that an adult breathes through a facepiece in 250 cm\(^2\) at 30 L min\(^{-1}\) (normal breathing condition). The following calculation was used to determine the face velocity: \((30 \text{ L/min} \times 1000 \text{ cm}^3/\text{L} \times 1 \text{ min}/60 \text{ s}) = (250 \text{ cm}^2 \times 2 \text{ cm/s})\). For the continuous filter test, a filter media area of 100 cm\(^2\) was exposed to NaCl aerosol in the mass concentration of 20.0 ± 5.0 mg m\(^{-3}\) at the face velocity of 2 cm s\(^{-1}\). The penetration and the pressure drop were monitored through the continued mass loading until the penetration was nearly reached to zero (about 40 mg NaCl loading for about 3 h).

For the simulated repeated use-storage test, the filter media was challenged to 2.2 mg of NaCl aerosol per day. The following scenario was considered to determine the exposure mass per day. If a 250 cm\(^2\) filtering facepiece is used in a harsh particulate level of 300 \(\mu\text{g m}^{-3}\) for 10 h with 30 L min\(^{-1}\) of inhalation rate, a total aerosol mass of 22 \(\mu\text{g cm}^{-2}\) would be challenged to a filtering mask every day.\(^{35,36}\) For 100 cm\(^2\) filter area of this study, 2.2 mg would be challenged every day. The aerosol concentration of the test equipment was fixed in 20 mg m\(^{-3}\), thus it took about 9 min to challenge 2.2 mg of NaCl mass at the face velocity of 2 cm s\(^{-1}\). Immediately after the aerosol exposure, the filter media was stored in the predetermined environmental condition until the next day test. After filters were stored in those conditions, samples were kept in an ambient condition (23°C, 50% RH) for 30 min before the next aerosol test to challenge 2.2 mg of NaCl. The sequence of aerosol exposure and storage was conducted at an interval of 24 h, and this sequence was repeated until the total mass of 80 mg was challenged to a filter media.
Characterizations

An X-ray computed tomography (Xµ-CT, Xradia 510 Versa, Carl Zeiss Inc., Germany) was employed to analyze the 3D internal structure of filter media. The X-ray source was operated at a voltage of 60 kV with a power of 5.0 W. The field of view was 700 μm, and the corresponding voxel sizes were 0.7 μm. The reconstructed Xµ-CT (3D internal structure) dataset was imported to ORS Dragonfly Pro software (ORS Inc., Canada) for image processing.

The morphology of filter media and captured particles were observed by the field-emission scanning electron microscope (FE-SEM, SM-7800F Prime, JEOL Ltd., Japan), with prior coating with Pt (108auto, Cressington Scientific Inc., UK). An Environmental SEM (ESEM, Quattro S, FEI, USA) was used to observe the morphological change of the deposited particles on the filter media in humid conditions. The particle-loaded filter sample was put on a stage and observed at 2°C and 100% RH.

The tensile test was performed on a rectangular sample of 50 mm × 150 mm using a tensile tester (5 ST, Tinius Olsen Ltd., UK) operated at a crosshead speed of 10 mm min⁻¹. The surface potential of the meltblown was measured by a non-contacting electrostatic voltmeter (Model 542A, Trek, Lockport, NY, USA). The surface potential probe was placed 2 cm above the web surface, moving across different filter area in 50 cm distance.

Results and discussion

Filtration characteristics with varied environmental storage conditions

Figure 2 examined how the storage temperature and humidity affected the aerosol penetration and pressure drop. The filters were treated at the pre-determined storage conditions for 37 days and were tested for continuous particle loading (Figures 2(a) and (b)). The pressure drop and penetration during the continuous aerosol loading, whether the samples were pretreated or not, depicted typical plots showing a maximum penetration as the accumulated aerosol mass increased; after reaching the maximum penetration, the penetration began to decrease as the pores of the filter were clogged. The pressure drop and the penetration of filter media pretreated at 40°C condition were similar to those stored at the ambient condition (23°C, 50% RH). However, 90°C temperature and high humidity (40°C, 75% RH) conditions produced increased penetration (lowered efficiency) and lowered pressure drop compared to the ambient storage condition.

As the environmental temperature and humidity could affect the charge stability of electret filter media during the storage, the surface potential of filter media was investigated with different storage conditions (Figure 3). It should be noted that the measurement of surface potential may not directly reflect the surface charges, because an electret filter commonly hold both positive and negative charges, compensating each other. Thus, the observation was focused on the extent of potential variations in estimating the relative charges of the material. For a highly charged media, the surface potential measurement showed large variations, while the surface potential profile was flattened as
the charging was deteriorated. The surface potential was measured from 2 cm away from the sample surface, probing across the filter area within 50 cm.

The result in Figure 3 is the surface potential of filter media that were stored for 37 days. The surface potential was significantly deteriorated with 40°C, 75% RH condition, indicating that the electrostatic filtration became ineffective as the storage progressed. The pressure drop after 37 days of storage at this condition showed large

**Figure 2.** Filtration characteristics of an electret filter media with the conditioned storage for 37 days (a) Pressure drop and (b) penetration with continuous test after 37 days pretreatment. (c) Pressure drop and (d) penetration with recurrent exposure during 37 days.

**Figure 3.** The surface potential of filter media stored for 37 days with varied storage conditions.
variations, thus the aging effect at this condition on the pressure drop is inconclusive (Figure 2(a)). It is argumentative whether the environmental moisture affects the filtration efficiency of polypropylene (PP) electret filter media, where the moisture absorption of PP is negligible. From Lee et al. ‘s study, storage at 20°C, 65% RH for 2 days hardly affected the filtration performance of PP filter media. On the other hand, Motyl and Lowki reported that storage at 24°C and 78% RH may affect the surface charges of electret filter, but in different extents depending on the filter type. Yet the detailed information is not available on which factors of an electret filter affect the humidity-dependence in surface charges and filtration efficiency. The result in Figure 3 shows that an extended storage at high humidity (75% RH) and warm temperature (40°C) deteriorated the electret charges leading to the reduced filtration efficiency.

At 40°C dry condition, the charge decay was not observed as much as 40°C, 75% RH condition. As a result, the filter efficiency stored at this condition was comparable to that stored at room temperature and humidity at 23°C, 50% RH. The surface potential of the filter media kept at 90°C was nearly zero with little variations, indicating that the reduction of electrostatic charges occurred significantly. Thus, the penetration of the filter stored at 90°C considerably increased for both continuous and intermittent exposures. After the charge decay, the electrostatic particle capture mechanism such as Coulombic attraction and induced polarization can no longer contribute to the overall filtration efficiency; instead, the filtration occurs mostly by the mechanical particle capture mechanism such as diffusion, interception, and inertial impaction. The result implicates that for about a month or longer period of shelf-life, the storage condition needs to be carefully chosen to maintain the filter performance of electret filter.

For the recurrent aerosol exposure and storage conditions (Figures 2(c) and (d)), the pressure drop did not increase as much as the continuous exposure, and it remained below 30 Pa. Especially for the repeated use with storage at 40°C, 75% RH, pressure drop did not increase with days of repeated particle exposure, maintaining the pressure drop almost the same. On the other hand, the pressure drop during the 1-day exposure (2.2 mg of challenging) showed a slight increase. The recurrent test with an ambient storage (23°C, 50% RH) was additionally conducted, and it was confirmed that the trend of filtration performance at the ambient condition was similar to that of 40°C, 75% RH condition. For all tests of recurrent aerosol exposures, a leap in the penetration was observed between the daily test intervals (after storage), implying that the loaded aerosol caused the performance deterioration during storage.

For the repeated exposure-storage test with 90°C storage condition, overall penetration increased steeply with a leap between the testing intervals (Figures 2(c) and (d)). At around the accumulated exposure of 50 mg, there was a sudden discontinuity in the plots both for pressure drop and penetration. This discontinuity in the graph is the result of the fiber breakage occurred at 50 mg exposure (Figure 4(b)). From the result of the tensile test (Figure 4(a)), the stress of filter media that was continuously aged (without particle loading) at 90°C for 25 days decreased from 83 gf cm⁻² to 21 gf cm⁻², showing that the extended storage at 90°C caused the loss of mechanical strength of material. From the repeated recurrent particle test, the filter media could not withstand the weight of the loaded particles at about 50 mg accumulated exposure, causing the breakage of fibers in
the web (Figure 4(b)). The test was repeated three times, and the fiber breakage occurred rather consistently at around 50 mg of particle exposure in the recurrent test. The penetration (%) of particles was calculated as a percentage of downstream concentration divided by upstream concentration using a photometer. Thus, the abrupt penetration rise at 50 mg mass exposure is due to the particle leakage through the torn filter. The filtration test was stopped from this point because testing with a torn sample was meaningless.

**Morphology of filter media with varied storage conditions**

Any evidence of morphological changes in internal structures of filter media with 37 days of conditioned storage was investigated by Xµ-CT analysis. Fibers and pores are distinguished by the X-ray intensities, and image processing was performed to reconstruct 3D images of the filter sample. After the reconstruction of 3D image in 500 μm × 500 μm × 500 μm, morphological parameters including filter thickness, fiber diameter, and pore distribution in the web were analyzed. The thickness of filter media after 37 days of storage was the following: filters stored at ambient condition (23°C, 50% RH), 229 ± 17 μm; filters treated at 40°C, 75% RH, 283 ± 17 μm; filters treated at 40°C, 298 ± 12 μm; filters treated at 90°C, 285 ± 14 μm (Figures 5(a) and (b)). The filter media became slightly thicker after every storage condition for 37 days, probably because heat treatment (40°C or 90°C) eliminated the internal stress of the filter media that was imposed during the meltblown manufacturing process.25 Thus, after heat treatment, the filter media became fluffier.37 However, this structural change did not have direct influence on the pressure drop and the penetration. Fiber diameter and pore size were further analyzed by Xµ-CT images. The filter media, regardless of environmental storage conditions, displayed their fiber diameter distributions from 1.5 μm to 15 μm, and the average diameter was 5–6 μm (Figures 5(a) and (c)). There was no observable difference in the fiber diameter for different pretreatment conditions, and the fiber diameter was not a cause for the change of filtration performance during the conditioned storage.

![Figure 4.](image_url)

(a) Tensile test after aging 25 days at heated storage conditions. (b) The test sample was torn at around 50 mg NaCl loading for the repeated test with particle exposure and 90°C storage condition.
Figure 5. Morphological analysis by Xμ-CT. (a) 3D morphology with web thickness and fiber diameter, (b) web thickness, (c) fiber diameter distribution, (d) distribution of pore diameter, (e) cross-section of the web in YZ and XY axis.
In Figure 5(d), pore diameter was analyzed to better understand the pore distribution with varied storage conditions. Overall, the pore size distributions of filter media that were treated in different storage conditions were not considerably different. Figure 5(e) shows the cross-sectional image of the web in the varied axis, which analyzes the distribution of fibers and pores in the cross-section. The fiber region was colored in gray, and the pores were colored from yellow to purple depending on the distance from the pore to fibers. For the filter media kept at ambient condition, the yellow color was not seen between fibers in YZ plane and XY plane, meaning that fibers are packed rather tightly with fewer neighboring pores. On the other hand, 90°C pretreated sample showed larger pores (yellow) acting as tortuous channels for direct air flow, and these enlarged pores may affect the mechanical filtration efficiency and pressure drop.

Particle loading characteristics in the filter media

To confirm the accumulated NaCl particle morphology on the filter media, SEM and Xμ-CT images were examined. As particles were continuously challenged to filter media at once, the particles were mostly stacked up as a thick layer at the surface of filters regardless of preconditioning (Figure 6). Since the densely packed NaCl particles blocked the air flow, the pressure drop rose quickly. Unlike the continuous exposure to aerosol, samples with recurrent exposure and storage showed NaCl particles distributed deeper down in the filter media (Figure 7). It seems that the recurrent and repeated air flow self-propelled the particles and enhanced transport through the pore, delaying the clogging.38

For recurrent use conditions (Figure 7), the particles appear throughout the entire depth of the filter media. As a result, the pressure drop was lowered at the expense of increased penetration. In the storage condition at 40°C, 75% RH, the pressure drop maintained almost consistent, and this is attributed to the deliquescence of captured NaCl particles. During the humid storage, large NaCl aggregates of crystals were formed because of the hygroscopic property (Figure 7(d)). The hydration layer on the NaCl surface could have acted as a liquid bridge, and this liquid bridge could induce particle agglomeration.39,40 In humid conditions, wet NaCl particles spread on the filter fibers, quickly masking the filter

Figure 6. SEM and Xμ-CT images of 80 mg NaCl loaded filter media; one-time continuous filtration test with filter media pretreated for 37 days in (a) 23°C, 50% RH (ambient condition), (b) 40°C, 75% RH, (c) 40°C, (d) 90°C.
charges like liquid aerosol; as a result, the filtration performance may deteriorate rather rapidly in this condition.

ESEM analysis was carried out to observe the effect of humidity on NaCl loading characteristics. From ESEM in Figure 7(d), hygroscopic NaCl deposited on filter fiber was wetted slowly in humid condition, and began to swell, which will lead to masking of charged surface. The filtration efficiency was rapidly deteriorated after NaCl-loaded filters were stored in 75% RH, as shown from the steep increase of penetration (Figure 2(d)). For the thermal storage condition either at 90°C or at 40°C, the captured particles appeared as dendrites and the pressure drop showed an overall inclination with repeated exposure to the aerosol.

**Effective lifetime of filter media**

The effective lifetime of filter media can be limited by the build-up of pressure drop, deteriorated filtration efficiency, and mechanical damage of material. When filters are not properly replaced at the right time, it may cause a significant build-up of pressure drop or an increased penetration of pollutants due to the charge decay and/or damaged material. Figure 8 depicted the mass of particles deposited/retained in the filter media.
versus the mass challenged to the filter media. The plots filled with dark and light colors indicate that particle mass retained in the filter media and the mass penetrated through the filter, respectively. For the continuous exposure test, the pressure drop increased with a steep slope as the deposited mass increased, while the efficiency was nearly 100% throughout the mass accumulation.

The effective lifetime of filter can be determined by a certain level of efficiency and breathing resistance, which are measured by % penetration and pressure drop. In this study, performance levels of 5% of penetration and 12 Pa of resistance (25–30% increase from the initial pressure drop) were used as example criteria of effective lifetime of a well-functioning filter. For the repeated exposure cases with humid storage conditions (40°C, 75% RH storage), the pressure drop maintained to be low, so that the filter users may not notice the increase of breathing resistance. On the other hand, filtration efficiency reached over 5% after 5-days’ use in this storage condition, losing a considerable extent of efficiency. In such a condition, the users may not recognize the deteriorated performance nor the breathing resistance when they use filters repeatedly.

With the repeated use with 90°C storage condition, the penetration went up over 5% in 5 days with a considerable build-up of pressure drop. In this case, users may replace the

Figure 8. Evolvement of pressure drop and filtration efficiency with accumulated particle mass versus challenged particle mass; one-time continuous filtration test for the filter media stored in (a) ambient condition (23°C, 50% RH), repeated filtration test for filters with deposited mass (b) 40°C, 75% RH, (c) 40°C, (d) 90°C.
filter as they sense the significantly increased breathing resistance compared to the initial resistance. At 90°C recurrent storage condition, the pressure drop was drastically reduced on 21st day because of the air leakage through the torn part of the filter. The mechanical property of PP filter media was deteriorated with the repeated thermal storage at 90°C, and at 21st day, the filter media could not withstand the mass of accumulated particles, being torn. This result implies that the facepiece user needs to be careful about storing the facepiece under high temperature.

For the repeated use with 40°C dry storage, the filter performance was effective (penetration <5%) until day 7, but the build-up of pressure drop was a dominant factor influencing the useful lifetime. For the repeated use with recurrent storages, the deposited mass of aerosol was not directly related with the effective lifetime; instead, storage condition, particularly humidity, played an important role affecting the performance and effective lifetime. If the masks were not timely replaced, contaminants may unexpectedly penetrate in quite large quantities that may threaten the user health. Therefore, understanding the filtration behavior with actual use scenarios is important for a proper protection.

**Conclusions**

The effect of storage temperature and humidity on filtration behavior in terms of penetration and pressure drop was investigated for (1) the unused filter that is conditioned for 37 days at a predetermined temperature and humidity and (2) the filters recurrently exposed to aerosols with conditioned storages between uses. The storage conditions included 23°C, 50% RH (ambient), 40°C, 75% RH, 40°C, ~0% RH, and 90°C, ~0% RH. The heated preconditioning (40°C or 90°C) made unused filter fluffier and thicker, however it did not directly affect the pressure drop or mechanical filtration efficiency. The filtration characteristics were investigated simulating the practical application of recurrent and repeated use of filters, compared to the continuous exposure to the aerosol. For the repeated exposure tests, the humidity during the recurrent storages significantly deteriorated the efficiency as the hygroscopic solid particles were wet during the storage, and quickly decayed the filter charges. In addition, a high temperature storage condition (90°C) quickly deteriorated the surface charges and aged the polypropylene fibers, causing the lowered efficiency and fiber breakage.

This study explored the effective lifetime of the filter with the particulate mass retained in the filter, in addition to challenged mass. For the continuous aerosol loading, a deposited mass was directly related to the pressure drop. For the recurrent and repeated exposure condition, pressure drop was hardly associated with the deposited mass; on the other hand, the recurrent storage conditions affected both the filtration efficiency and mechanical structure of filter material. As a result, the effective lifetime for the repeated use was largely influenced by the rapid deterioration of filtration efficiency. The approach of this study is progressive in that rigorous analysis was performed to investigate both particle distribution characteristics and structural changes of filter media in relation with the filtration performance and effective lifetime. This study provides a scientific reference that guides the desirable filter storage conditions and guides the filter replacement cycle in
association with use habits and storage environments to cope with overwhelming pandemic situations.

**Declaration of conflicting interests**

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