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Enhanced nano-aerosol loading performance of multilayer PVDF nanofiber electret filters

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ABSTRACT

Aerosol loading behavior of PVDF nanofiber electret filters using neutrally charged nano- and submicron aerosols was investigated experimentally for the first time. The loading behavior include variations of filtration efficiency and pressure drop and distribution of deposited aerosols in the filters all having the same fiber basis weight (3.060 gsm). Through the filtration efficiency variations of uncharged/charged, single-/multi-layer filters with aerosol loading, it was observed that mechanical PVDF filters had continuously increasing filtration efficiency, while PVDF electret filters had initially decreasing and subsequently increasing filtration efficiency until reaching 100% due to diminishing electrostatic effect and enhancing mechanical effect.

By combining the pressure drop evolution of different filters during aerosol loading and detailed SEM images of the loaded filters, we have demonstrated that multilayer PVDF filters, especially the electret ones, could significantly slow down the pace of filter clogging (skin effect) and increase significantly the aerosol holding capacity during depth filtration. Generally, the multilayer nanofiber filters received the most aerosol deposit during depth filtration, whereas the single-layer nanofiber filters with the same basis weight of fibers received the most deposit during cake filtration. The multilayer nanofiber filters had approximately 70% aerosol deposit in the filter during depth filtration fully utilizing the full filter thickness, especially for the electret filters that had charged fibers, and only 30% deposit in the cake. On the contrary, the single-layer uncharged/charged nanofiber filters were exactly the reverse due to persistency of the skin effect with only 30% deposit in the filter mostly located in the upstream layer, yet 70% deposit in the cake. During depth filtration, the pressure drop per added mass deposit for the multilayer electret filter was very low at 11 Pa gsm⁻¹, which was at least twice below any other nanofiber filters. This was all attributed to the uniform capture of aerosols by electrostatic effect across the entire filter depth from the upstream to downstream layers of the multilayer electret filter. The above conclusion was confirmed by the detailed SEM images taken across the different filter layers for the multilayer filter configuration. The 4-layer electret nanofiber filter with a 3.060-gsm basis weight has 4 times more aerosol holding capacity than the single uncharged/charged nanofiber filter with the same fiber basis weight in depth filtration. Based on the standpoint of highest efficiency and capacity with maximum pressure drop 800 Pa imposed on the filtration operation, the 4-layer nanofiber electret was the best among all 4 filters. It had 52% more aerosol holding capacity than the single layer uncharged nanofiber filter and 38% more capacity than the charged single-layer and the uncharged multilayer nanofiber filters. The multilayer PVDF electret filters have excellent filtration performance for long-term aerosol filtration and also great potential applications in the fields of personal health care and environmental protection.

1. Introduction

Aerosol among many is one of the bad elements in air pollutants. In particular, aerosols less than 1 μm (PM1) can penetrate deep into the alveoli of our lung causing chronic respiratory and breathing disorder. Aerosols less than 100 nm (PM0.1) are referred to as nano-aerosols, nano-particles or ultrafine particles. By virtue of their tiny size, the nano-aerosols can easily diffuse into our blood stream surrounding the lung. Our circulation system can carry these aerosols to different parts of our body, including our brain, leading to various long-term harmful effects. In addition, airborne viruses, with sizes in the nano-aerosol range, can also lead to acute sickness. The recent novel coronavirus that has infected many people, as well as the previous SARS and MERS viruses, is also of size approximately 100 nm. Conventional filters made
of microfibers with fiber diameter of 2–20 μm are ineffective in capturing these nano-aerosols. Nanofibers with fiber diameter of 200–600 nm are effective in filtering the nano-aerosols (less than 100 nm) by diffusion and aerosols greater than 100 nm by interception. On the other hand, aerosols larger than 1 μm can be captured effectively by inertia impaction. Here, we consider a special nanofiber filter that is electrostatically charged – nanofiber electret filter. In particular, we will study the behavior of these filters as they are loaded with the nano-aerosols. We have broadened up the size range in our study to not only the nano-aerosols size less than 100 nm but extending all the way to 500 nm. In general, we will refer hereafter to this size range as just aerosols or particles in short.

The merits of multilayer PVDF nanofiber electret filters have been well-proven by their great filtration properties, both in efficiency improvement and pressure drop reduction, compared with their 1-layer counterpart [1]. Nevertheless, as a filter is continually challenged with aerosols, both the aerosol penetration (i.e. inverse of the efficiency) and the filter resistance (i.e. pressure drop) gradually change while the filter becomes increasingly loaded [2,3]. Regardless of the usage in real applications, it is the long-term performance that is more valued when health impact, financial cost or energy consumption are considered.

In terms of filtration efficiency, as a departure from conventional uncharged filters which show increasing values throughout the loading process, the efficiency of electret filters initially decreases to a certain point before increasing with loading. This phenomenon is due to the shielding of electrostatic effect by the aerosol deposition and the enhanced mechanical capture due to deposit that can be considered as artificial fibers. Ultimately, an aerosol cake forms on the filter surface which becomes the effective surface filter media [2,4–8]. In the early stage of filtration using an electrically active fibrous filter, electrostatic mechanism, including Coulombic force collection for charged particles and induction charging collection for uncharged particles, plays the dominant role in aerosol capture. With continuous filtration, the initially captured particles act as collection sites for the latter particles and gradually form dendritic structures. The adjacent dendrites gradually get connected together, which initiates the formation of a “cake” on the upstream end of the filter. During the dendrites/cake growth stage, the electrostatic effect diminishes because of the “shielding effect” on the charge fibers by deposited particles, while the mechanical collection enhances contributed by the dendrites/cake. The filtration efficiency keeps decreasing until the reduction in electrostatic efficiency is equally compensated by the improvement in mechanical efficiency, at which point the filtration efficiency falls to its minimum value before increasing thereafter. On forming a complete cake layer (cake filtration stage), particle penetration falls to zero (or filter efficiency reaches 100%), where electrostatic effect is fully shielded and the future incoming particles are solely collected via the mechanical effect of the cake layer, i.e., the aerosol capture has totally transferred from depth filtration to surface filtration.

As for pressure drop, electret filters have generally similar increasing trend to conventional filters [4–8]. In the early stage, particles deposit on the surface of individual fibers across the porous filter depth. Due to the exceedingly limited space occupied by the captured particles, the fluid flow is only slightly affected and the pressure drop increases at rather low rates. With dendrites/cake forming and the filter more and more clogged, the pressure drop keeps rising at an accelerating rate. When the cake is completely formed, the increase rate of pressure drop reaches its peak value, after which the rate of pressure drop increases linearly with the loading mass, indicting a constant cake resistance growth with additional aerosol deposit.

From the description above, it seems that an electret filter only has a minor advantage of initially higher filtration efficiency over a conventional filter regarding aerosol loading process because it basically works as a surface filter once being fully clogged. However, due to the big role played by electrostatic mechanism in the early stage of loading, it is naturally expected that for an electret filter, the deposition properties of particles, including the capture position, dendrite/cake formation rates and cake structures, are different from a pure mechanical filter. Therefore, the electret filter may show a different pressure drop increase characteristic in the subsequent cake filtration. In fact, the lower clogging rates of some electrically active fibrous filters have already been reported in the literature and the authors attributed the improved holding capacities to more uniform distribution of deposited particles. Nonetheless, without investigating the real morphology of particle deposition, such as use of SEM images on deposition pattern in the filter, previous researches merely qualitatively drew conclusions based on the difference in pressure drop evolutions between electret and mechanical filters [4–6]. Moreover, the electret filters in these researches usually had large thicknesses within just one single layer and multiple layers of electret have not been studied. Further, it has been well proved that “skin effect” exists in aerosol filtration using a fibrous filter, where much more particles deposit at the face region than at the back side [9,10]. For an electret filter, that means when a complete cake is already formed on the surface of the filter, the charged fibers downstream of the surface fibers are far from being fully utilized and wasted.

Filtration efficiency has been confirmed to improve by redistributing charged fibers from a single layer to multiple thinner layers of nanofibers stacking one on top of each other [1]. The objective of this study was to study the aerosol loading behaviors of the multilayer PVDF filters, especially of the electret ones. In fact, loading of multilayer nanofiber filters, uncharged or charged, has never been done before. Even loading of a charged electret 1-layer nanofiber filter has not been reported, which in our study serves as a basis of comparison as well.

It was hypothesized that compared to a 1-layer filter, a multilayer filter with an identical total fiber basis weight but a much lower packing fraction has a higher resilience to clogging and a larger aerosol holding capacity. Due to the higher porosity and smaller thickness of each layer in the multilayer filter, it will take a longer time for the dendrites to grow to a significant size to block the aerosol flow. Thus, the pace of skin layer formation slows down and aerosol capture across the filter layers is more uniform. Specifically, for the charged multilayer PVDF filters, the charges on the fibers of downstream layers are more effectively being utilized. Further, on the level of individual fibers, the captured particles are expected to be more uniformly distributed circumferentially and lengthwise along the fibers.

In this study, the filtration efficiency variation of uncharged/charged, single-/multi-layer filters with aerosol loading were compared for the first time. Moreover, the pressure drop evolution of different filters were analyzed in accordance to their physical properties. Besides, the deposition of captured aerosols on the filters, both in amount and distribution, was illustrated using SEM images. Ultimately, we want to address the efficiency, pressure drop and aerosol capacity for the 4 types of test filters in various combinations (uncharged/charged, single-/multi-layer filters) under loading starting from a clean filter, progressing to depth filtration and subsequent cake filtration.

2. Experimental

2.1. Fabrication of filters

The general fabrication methods of PVDF nanofiber filters by electrospinning and corona discharge were similar to that in our earlier work [1]. PVDF solution properties and electrospinning conditions were finely tuned to get continuous, straight and defect-free fibers [11,12]. To alleviate the nonuniform distribution of fibers, the collecting substrate was horizontally inverted at intervals during each batch of electrospinning. The fiber basis weight was closely monitored throughout the spinning process until the target value was reached. Multilayer filters were prepared by stacking up a certain number of 1-layer filter modules with identical physical properties. In this study, 4-layer PVDF filters were used to distinctly show the different loading
behaviors from the 1-layer counterparts having the same basis weight of nanofibers. Based on charging state, the filters could be divided into uncharged and charged (electret) filters, with the former prepared by discharging the pristine filters using isopropanol while the latter was obtained by subsequent charging the “uncharged” filters using corona discharge. Each filter was named according to its configuration (S for single layer and M for multiple layers), layer basis weight (the same as filter basis weight for 1-layer filters) and charging state (U for uncharged and C for charged). For instance, S-3.060-U refers to the uncharged 1-layer PVDF filter with the layer basis weight of 3.060 gsm, while M4-0.765-C denotes the charged 4-layer PVDF filter with the layer basis weight of 0.765 gsm for each layer, but the total basis weight for all 4 layers added up to 3.060 gsm. In case that parallel groups of filters were used to repeat the test run, different numbers were tagged at the end of each filter for making differentiation among the different filters in the repeated tests.

2.2. Filtration performance evaluation of filters

Before loading up a filter, it is essential to test the filtration performance in a clean (i.e. unloaded) state. The experimental set-up for clean filter tests is shown in Fig. 1a. The same test method as used in our earlier work was adopted to determine filtration efficiency (η), pressure drop (∆p) and quality factors (QF) of each filter [3]. Within the upper detection limit of condensation particle counter (CPC), concentration of each particle size was ensured to be sufficiently high to reduce errors of measurement.

2.3. Aerosol loading behavior evaluation of filters

As shown in Fig. 1b, a similar set-up to that of clean filter test was applied when the behavior of a filter during aerosol loading was to be studied except for the position of the differential mobility analyzer (DMA). After the first neutralizer, the polydisperse aerosols were directly fed to the test column to challenge the filter. The reason for the modification was that it would take impractical long time to load up the filter. Every ten minutes, the aerosol flow upstream of the filter was sampled via the DMA to get the size distribution of the particles used for filter loading, coupled with the CPC to measure particle concentrations channel by channel. Note that in the set-up for filter loading, the real concentrations shown in aerosol size distribution were obtained by automatically calculating the corresponding values measured by the CPC which only counted the relatively low fractional particles exiting the DMA. At prescribed intervals, the set-up was switched back to that of Fig. 1a for testing the filtration efficiency with loading mass until the values all increase approximately to 100%, after which the efficiency testing (i.e. Fig. 1a) was stopped as a cake has already formed on the filter surface and surface filtration was the only mechanism for aerosol capture. The set-up for aerosol loading was not used to determine filtration efficiency because of the difficulty in tuning the equipment response time for polydisperse aerosols, which could bring inaccurate and misleading results. Throughout the loading process, the pressure drop was monitored at regular intervals, which was expected to show a linear trend after the formation of the full cake. The loading was terminated until the pressure drop reached an arbitrary limit which had been far past the starting point of complete surface filtration. In this study, the pressure drop limit was set at about 800 Pa.

Fig. 2a and b show the typical aerosol size distributions of set-ups for filtration efficiency tests and filter loading, respectively. The maximum concentration at particle size of 80–90 nm (in nano-aerosol range) for the former set-up was approximately 6.0 × 10^5 cm^-2 while for the latter one above 3.9 × 10^6 cm^-2 which is almost 62 times higher than the former case. Therefore, the occasional efficiency tests had little interference in the loading behaviors of filters. The loading period could be further shortened by using NaCl solution with a higher concentration. Nevertheless, apart from the risks of clogging and corrosion, higher concentrations were not chosen with concern that the filtration efficiency would change too quickly during the efficiency test.

Combining particle size distribution, filtration efficiency with loading time, face velocity of the aerosol stream and filter surface area, the captured aerosol mass during any time period and the cumulative mass at any time point could be calculated. In the literature, some researchers weighed the filters before and after aerosol loading and took the weight differences as the loaded aerosol masses [13]. This method was not used in this study mainly due to two reasons. For one reason, removing the filter can easily break the structures formed by captured aerosols, e.g. dendrites and cake, and cause loss in deposited aerosols. For another reason, the weight of deposited aerosols is often too light to be detected accurately, which can bring significant errors to the calculations.

The loaded filters were sampled for SEM characterization to study the aerosol deposition properties of uncharged/charged, single-/multilayer filters. Specially, the back side (i.e. downstream side) of the loaded filters were also observed under SEM to investigate whether the nanofibers and nanofibrous layers were more fully utilized after charging or distributing into multiple layers.

3. Results and discussions

First before aerosol loading, the multilayer PVDF nanoﬁber electret filter was compared to two conventional microfiber electret filters made of polypropylene with all filters having the same range of pressure drop from 23 to 26 Pa and being challenged by sodium chloride aerosols with a face velocity of 5.3 cm s^-1. The efficiency and quality factor are compared between these filters in Appendix A. As shown in Figs. A1 and A2, both efficiency and quality factor at clean state for the PVDF nanoﬁber electret filter are much higher than those of the two conventional microfiber electret filters. For example, for the 300-nm aerosols, the efficiency of the multilayer was 87.5% on average while the multilayer electret nanoﬁber filter achieved 98.3% which is a 12% increase. The quality factor of the two conventional microfiber electret filters made of polypropylene achieved a quality factor of 0.085 Pa^-1 on average, while the multilayer electret nanoﬁber filter achieved 0.155 Pa^-1 which is a whopping 82% increase over the conventional microfiber electret filters.

To investigate the differences in loading behaviors between uncharged and charged filters, and 1-layer and multilayer filters, filters with same average fiber diameter of 525 nm and same total basis weight of 3.060 gsm were used, including uncharged and charged 1-layer filters, and uncharged and charged 4-layer filters. Filters of 4-layer were used in this research because they were expected to have distinctly different behavior as compared to that of the 1-layer filters. The SEM images of S-3.060 and a typical layer of M4-0.765 are shown in Fig. 3a and Fig. 3b, respectively. It is clearly shown that the fibers in S-3.060 are much more densely packed than that in a single layer of M4-0.765.

3.1. Comparison of loading behaviors between 1-layer and multilayer

3.1.1. Variation of filtration efficiency and pressure drop of uncharged PVDF filters with loaded mass

The filtration efficiency curves of S-3.060-U and M4-0.765-U with loading time/loaded mass are shown in Fig. 4a and b, respectively. Generally, the two uncharged filters show similar trends with the efficiency increasing with loading time/loaded mass, which has been widely confirmed in the literature [9,10,14]. The deposited aerosols, including individual particles, dendrites and cake, could all act as new aerosol collection sites, or as additional ‘artificial fibers’, thus improving the filtration efficiency, as well as increasing the pressure drop across the filter [15–17]. It is also interesting to note that the most penetrating particle sizes (MPPS) of both filters gradually decreased
with loading mass—from 250 nm to 80 nm for S-3.060-U and from 280 nm to 80 nm for M4-0.765-U as shown in Fig. 5. Similar phenomenon was also observed by Leung et al. [9,10]. This is due to the more prominent interception effect than the diffusion effect of the deposited aerosol agglomeration, with the two effects being the main mechanisms for filtration efficiency improvement before surface filtration took place. However, regardless of the similarities, the efficiency of S-3.060-U apparently increased at a much higher rate than that of M4-0.765-U. In clean state, the two filters had similar efficiency curves. After 2-h loading, the efficiency of S-3.060-U at the MPPS (120 nm) drastically increased to 91.9% from the clean filter efficiency of 45.6%, while that of M4-0.765-U at 280 nm increased only from 40.1% to 43.8%. Overall, it merely took only 4 h for S-3.060-U to reach approximately the efficiency of 100% for all challenging particles in contrast to 14 h for M4-0.765-U. Due to the constantly higher filtration efficiency, more aerosols deposited on S-3.060-U than M4-0.765-U at

Fig. 1. Schematic diagrams of set-ups for (a) aerosol filtration tests and (b) aerosol loading of filters (Adapted from [1]).
the same loading time. For example, after 2-h loading, S-3.060-U had a loaded mass of 2.25 gsm, which was 0.67 gsm higher than 1.58 gsm for M4-0.765-U. Corresponding to filtration efficiency, the pressure drop of S-3.060-U rose much faster than that of M4-0.765-U. Before loading, the pressure drop values were 20.8 Pa and 16.9 Pa for S-3.060-U and M4-0.765-U, respectively. After 2 h loading, the pressure drop for S-3.060-U increased by 42.8 Pa while only 1.5 Pa for M4-0.765-U. Barely 4 h elapse, S-3.060-U has reached 180.5 Pa. In contrast, 12 h elapse was required for M4-0.765-U to reach 189.3 Pa.

Contributed by the slower growth of dendrites/cake, the filtration efficiency of M4-0.765-U increased at lower rates than that of S-3.060-U with loading time. Moreover, benefitting from the higher uniformity of aerosol deposition across the filter, M4-0.765-U had much lower increase in pressure drop per loaded aerosol mass than S-3.060-U. In other words, multilayer filters could mitigate the clogging by aerosol accumulation and enhance the aerosol holding capacity. As can be seen in Fig. 4a-b, when the filtration efficiency approached near 100%, M4-0.765-U had an aerosol loaded mass of 16.76 gsm (at 14 h) while S-3.060-U only 6.48 gsm (at 4 h). The only drawback for M4-0.765-U is that the efficiency before approaching the deposit of 6.1 gsm (8 h) was less than 80% whereas S-3.060-U with about 1-gsm deposit (1–2 h) could achieve 80% efficiency so that loss of uncaptured aerosols was
the different filter structures and aerosol deposition properties. S-3.060-U, with only one layer, had a high fiber packing fraction and a low porosity. The space between individual fibers was so small that dendrites formed and grew considerably fast into a cake on the filter surface with the high concentration of challenging aerosols. The result was the rapid increase in both filtration efficiency and pressure drop and the insufficiently utilized nanofibers downstream of the filter surface [9,10]. By redistributing the same amounts of fibers from the initial single layer to 4 layers, with each layer of M4-0.765-U consisting of much loosely packed fibers, the challenging aerosols could penetrate to the downstream layers of the filter before dendrites and cake grew to an extent that seriously blocked the incoming aerosol stream. This hypothesis was confirmed by SEM images of the loaded filters. Figs. 7–9 are SEM images which depict the aerosol deposition on the front or back side of loaded S-3.060-U and each layer of M4-0.765-U, respectively. The surface of the loaded S-3.060-U is covered by a cake (Fig. 7a–b); therefore, it is difficult to infer the aerosol profile within the filter. However, from the back side of the loaded S-3.060-U as shown in Fig. 7c–d, it can be observed that not many particles are present across the filter depth especially at the downstream end of the filter on the back side.

On the contrary, aerosol deposits more uniformly across the four layers of M4-0.765-U (Figs. 8 and 9), though the loaded amount decreases with the layer depth. When a multilayer filter is used for aerosol filtration, a portion of the aerosols is filtered out by the first layer upstream directly facing the challenging stream, leaving the rest of the aerosols challenging the downstream layers. The result is that fewer aerosol particles are available for the downstream layers. If every layer has similar structures and thus similar filtration efficiency in clean state, more aerosol will deposit on the layer more upstream at any given time, making its filtration efficiency and pressure drop always higher and increase at faster rates than the layers downstream. This is actually the very reason for “skin layer” effect which was also observed by Leung et al. [9,10]. In Figs. 8e–h and 9e–h, nevertheless, quite a large number of particles were still deposited on the last two downstream layers in M4-0.765-U, benefiting from the high permeability of each layer.

It is also worth noting that few particles can be seen on the back of individual fibers in both S-3.060-U (Fig. 7c–d) and M4-0.765-U (Fig. 9). Theoretically, particles can deposit on the back of a fiber through Brownian diffusion. However, due to the large fiber diameter of the filters used in this research (525 nm), Brownian diffusion was just a minor mechanism whereas interception played the major role for the uncharged filters, leaving aerosols mainly depositing on the front or side surfaces of uncharged fibers [2].

3.2. Loading behaviors of 1-layer PVDF filters—effects of charging

3.2.1. Variation of filtration efficiency and pressure drop of 1-layer PVDF electret filters with loaded mass

For electret filters, filtration efficiency has been observed by many researchers to first decrease to some extent before gradually increasing to 100% when a cake completely formed. This was attributed to the variations of two opposing effects—the reduction in electrostatic filtration due to charge shielding by the deposited aerosols and the increase in mechanical filtration from the continuous growth of dendrites and cake [4–7]. Fig. 10a shows the filtration efficiency evolution of S-3.060-C-1. Contrary to the expectation, the efficiency increased all the time till surface filtration was reached, despite that the clean filter exhibited a typical efficiency pattern of charged PVDF nanofiber filters, with the value increasing from 80.7% at 50 nm to 88.0% at 500 nm. After 1-h loading, mechanical filtration was the dominant mechanism indicated from the efficiency curve, and the efficiency was 92.6% at MPPS (particle with the lowest grade efficiency) of 250 nm. Gradually, the efficiency approached approximately 100% and the MPPS dropped to 80 nm (Fig. 5). Correspondingly, the pressure drop of S-3.060-C-1
increased sharply by 146.7 Pa from the initial 21.1 Pa after 3-h loading with a deposited mass of 6.80 gsm.

Given that the efficiency tests were only carried out intermittently with loading time, there was a possibility that the efficiency initially dropped during the first half hour. To prove this point, another charged filter (525-S-3.060-C-2) with similar physical properties was used to repeat the test under the identical experimental condition except that two more efficiency tests were carried out, respectively, at 10 min and 20 min. As can be seen from Fig. 10b, similar to S-3.060-C-1, the pressure drop of S-3.060-C-2 rose from 24.5 to 170.4 Pa with 7.45 gsm of deposited mass after 3 h. However, the filtration efficiency did decrease in the first 10 min and increase in the next 10 min. Subsequent loading time followed similar trend as that of S-3.060-C-1. Nonetheless, the largest reduction in efficiency of particles was only 2.7% which was too small to draw a convincing conclusion. It is also worth mentioning that S-3.060-C-1 and S-3.060-C-2 behaved a little differently concerning the variation of filtration efficiency, pressure drop and loaded mass, which was because of the randomness and inhomogeneity of fibers and charge distributions on the filters [18].

Considering the high concentration of aerosol flow for loading up the filter, it was hypothesized that, once the loading began, the mechanical effect strengthened so fast that the mechanical efficiency improvement trumped the electrostatic efficiency loss. Therefore, in another test an aerosol flow with much lower concentrations (shown in Fig. 11) was applied for 3 h to load up a third similar charged filter (525-S-3.060-C-3). After the first 3 h, the aerosol concentration was raised to the value used for loading up S-3.060-C-1 and S-3.060-C-2, which was aimed to further confirm the influence of loading aerosol concentration qualitatively. As shown in Fig. 10c, after 0.25-h loading, there were remarkable decreases in filtration efficiency for all-sized particles, especially for those with larger sizes. Particles of 350 nm (MPPS at 0.25 h as shown in Fig. 5) experienced a reduction of 22.4% from 77.6% to 55.2%. After further reductions ranging from 0.8% to 4.3% at 0.5 h, slowly the efficiency values increased and the MPPS decreased, with 60.7% of filtration efficiency at an MPPS of 220 nm. Note that for extremely small particles, the variations in filtration were insignificant. For instance, the efficiency for 50-nm particles slightly varied between 74.9% and 82.4%. This should result from the different mechanisms dominating filtration by electret filters for neutralized aerosols with different sizes [2,3]. In this research, particles were brought to a Boltzmann charge distribution before challenging a filter, with most of them being zero-charged so induction charging followed by electrostatic attraction (i.e. dielectrophoretic filtration) were the main mechanisms for electrostatic filtration. Compared with larger particles, small particles were less prone to induction effect as the positive/negative charge separation distance is also smaller, therefore they were less affected when the charge on filters were shielded by aerosol deposition. It should be pointed out that from 0.5 h to 1 h, the filtration efficiency of particles with sizes between 40 nm and 220 nm decreased while that of the rest increased. The reason might be that during this period, the decrease in electrostatic efficiency was not fully compensated by the increase in mechanical efficiency, leading to the further efficiency reduction for smaller particles. While for larger particles, they might be trapped due to the increasingly strong surface filtration effect. During 1–3 h, the filtration efficiency increased with slow paces and the average value remained low, corresponding to the gradual increase of loaded aerosol mass from 1.35 to 3.82 gsm.

Compared with the former two counterparts, with similar loaded mass, S-3.060-C-3 always had much lower pressure drop. For instance, the pressure drop of S-3.060-C-3 with 3.82 gsm of deposited aerosols was only 28.7 Pa, while that of S-3.060-C-1 and S-3.060-C-2 with fewer loaded aerosols (2.46 and 3.03 gsm, respectively) reached 64.8 and 73.4 Pa, respectively. The much alleviated air flow resistance of S-3.060-C-3 was attributed to the low aerosol loading concentration, which prevented the fast formation of dendrites and cake on the filter surface and the downstream fibers and fibrous layers became more accessible. Similar conclusion was obtained for microfiber electret filter by Tang et al., where higher particle concentrations led to higher increase rates of pressure drop and lower clogging points from the faster formation of dendrites and cake [8]. This point was further proved by the characterization of the deposited particles. As shown in SEM images in Fig. 12a–b, more particles were present on the interior fibers of S-3.060-C-3 than S-3.060-C-1 observed from the back side of loaded filters. In contrast to 1–3 h, from 3 to 4 h, there was a great surge in
filtration efficiency, which was in accordance with the significant increase in the concentration of loading aerosols. Afterwards, the filtration efficiency of S-3.060-C-3 increased with a trend similar to the aerosol loading during 1–3 h of S-3.060-C-1 and S-3.060-C-2. Corresponding to the filtration efficiency, the pressure drop during 3–7 h increased with significantly higher rates than that during 0–3 h, indicating the much faster growth of dendrites. Because more aerosols deposited within the filter, when the filter efficiency reached 100%, S-3.060-C-3 had 8.86 gsm of loaded aerosols (at 136.6 Pa). This was higher than both S-3.060-C-1 (6.80 gsm at 167.8 Pa) and S-3.060-C-2 (7.45 gsm at 170.4 Pa). Through the loading tests for S-3.060-C, aerosol concentrations in the challenging airflow were shown to affect the loading behaviors of a filter. Nevertheless, the major aim of this study was to investigate the differences in aerosol holding capacities among various test filters. Therefore, aerosols with similar size distribution were used for most loading tests and no further experiments using varied aerosol concentrations were conducted.

Comparing S-3.060-C-1 (Fig. 10a) with S-3.060-U (Fig. 4a), the charged filter always had higher filtration efficiency than the uncharged filter with loading time, benefiting from the pronounced

Fig. 8. SEM images of (a) and (b) the first, (c) and (d) the second, (e) and (f) the third and (g) and (h) the fourth layers of loaded M4-0.765-U seen from the front side.
electrostatic effect. With the same loading time and aerosol concentrations, the aerosol deposition amount was higher for the charged filter owning to the constantly higher filtration efficiency. For instance, at 2 h, 3.69 gsm of aerosols were loaded on S-3.060-C-1 while 2.25 gsm on S-3.060-U. Moreover, the pressure drop of S-3.060-C-1 increased at slightly lower rates than the uncharged filter with loaded mass before reaching 100% filter efficiency, see Fig. 13. For example, S-3.060-C-1 and S-3.060-U had pressure drop of 64.8 and 63.6 Pa with loaded masses of 2.46 and 2.25 gsm, respectively. This was attributed by the more uniform distribution of captured aerosols on individual charged fibers than on uncharged fibers [2,5,16]. That is, more aerosols could be captured on the back side of the charged fibers due to dielectrophoretic filtration effect, resulting in slower dendrites formation within S-3.060-C-1. In fact, the slope of the linear portion of the pressure excursion curve indicates the pressure drop from the additional cake deposited on the nanofiber filter. With reference to Fig. 13, the S-3.060-C-1 has a lower slope (33.3 Pa gsm⁻¹), thus a lower pressure drop than that of S-3.060-U (40 Pa gsm⁻¹). The charged filter

Fig. 9. SEM images of (a) and (b) the first, (c) and (d) the second, (e) and (f) the third and (g) and (h) the fourth layers of loaded M4-0.765-U seen from the back side.
represents a 17% reduction as compared to the uncharged one. This may have to do with the more porous cake formed on the electret nano fiber media as captured aerosols were distributed on both front and back faces of the fibers as well as distributed further into the filter than the uncharged counterpart. In other words, the skin effect was somewhat reduced; however, this advantage would have been much fully utilized for the multilayer charged filter which will be investigated in the next section.

The pressure drop evolution of S-3.060-C-2 with aerosol loaded mass is also shown in Fig. 14. The two 1-layer electret filters had similar behaviors suggesting the repeatability of the filter performance. The slope for S-3.060-C-2 is even lower at 29 Pa gsm$^{-1}$. As compared to 40 Pa gsm$^{-1}$, the reduction in pressure drop of 28% was even more significant.

3.2.2. Deposition of aerosols on 1-layer PVDF electret filters

As described in the above section, dielectrophoretic filtration effect facilitated the deposition of aerosols on the back side of individual fibers. This can be proved by the SEM images of loaded filters shown in Fig. 15c–d where aerosols were present on the back side of fibers of S-3.060-C-1, yet almost little to no aerosols for S-3.060-U were observed from the downstream of loading flow in Fig. 7c–d.

Given the lower pressure drop for surface filtration of a single-layer electret nanofiber filter as compared to the uncharged single-layer nanofiber filter, the issue of interest is whether a different arrangement of the charged fibers can further facilitate more uniform capture of aerosols upstream of the filter that provides a more permeable cake to be formed? As we have seen, if the fiber packing density was not sufficiently low, a cake could form after a short loading period, leaving the charged fibers downstream of the "skin layer" insufficiently utilized. As shown in Fig. 15c–d, not many aerosols can be seen within the loaded S-3.060-C-1 looking through the filter toward upstream from the back side. Owing to the electrostatic effect, a fraction of the particles penetrating S-3.060-C-1 were attracted by the charged fibers on the back side surface. Nonetheless, most particles had already been trapped in the "skin layer" due to the low porosity and the high initial filtration efficiency, thus only a small number of particles can be observed on the back side of S-3.060-C-1. Based on this, the multilayer electret filter was investigated to explore if the filter can be fully utilized across the entire filter thickness from upstream to downstream by redistributing the charged fibers in the filter.

3.3. Enhanced loading performance of multilayer PVDF electret filters due to both multilayering and charging

3.3.1. Variation of filtration efficiency and pressure drop of multilayer PVDF electret filters with loaded mass

Since the advantages of multilayering have been shown for the uncharged 4-layer filter in easing the pace of clogging and improving the capability of aerosol holding, compared to the uncharged 1-layer filter with the same basis weight, it is intuitive that the charged
multilayer filter would show similar properties. Fig. 16a shows the filtration efficiency variation of M4-0.765-C-1 with loading time, where the filter exhibits a clearer trend of an initial decrease and a latter continuous increase of efficiency till reaching nearly 100%. From 0 h to 2 h, the efficiency values gradually dropped. More reduction was observed for large particles while insignificant changes for very small ones, which was due to the more prominent electrostatic induction effect towards aerosols with larger diameters [2,3]. After 3 h aerosol loading, the filtration efficiency curve transferred to a typical shape of a mechanical filter from the curve after 2 h, which depicts a strong electrostatic mechanism. This means mechanical filtration, mainly by the growing dendrites and cake, began to play a remarkable role in trapping aerosols, especially for the smaller ones (40–60 nm) for which the efficiency increased while that of others were still decreasing during 3–4 h. From 4 h on, the enhancement of mechanical filtration exceeded the decline of electrostatic filtration. Meanwhile, the filtration efficiency increased at high rates with the slowly decreasing MPPS (Fig. 5) from 80.0% at 280 nm to 99.1% at 150 nm.

Similar to S-3.060-C-1, benefiting from the electrostatic effect, M4-0.765-C-1 had continuously higher filtration efficiency and deposited aerosol mass than its uncharged counterpart M4-0.765-U before 100% of filtration efficiency. However, the pressure drop of M4-0.765-C-1 was significantly lower than that of M4-0.765-U with similar loaded mass, especially at higher deposition amounts. M4-0.765-U and M4-0.765-C-1 had pressure drop of 45.9 and 25.7 Pa with 6.11 and 6.28 gsm of loaded masses, whereas 337.0 and 150.5 Pa with 16.76 and 18.30 gsm, respectively. The prominently alleviated air flow resistance shows that the advantage of charged fibers of uniform aerosol deposition was greatly enhanced through redistributing the same amount of nanofibers of S-3.060-C-1 into the 4 layers of M4-0.765-C-1. Though S-3.060-C-1 always filtered out more aerosols than M4-0.765-C-1, the captured aerosols accumulated so fast on the surface of the 1-layer filter that it progressed to cake filtration in an extremely short time. M4-0.765-C-1 and S-3.060-C-1 had pressure drop values of 18.1 Pa and 21.1 Pa in clean state, respectively. After 1-h loading with similar aerosol loaded masses (1.32 and 1.28 gsm, respectively), the pressure drop of the former filter increased to 20.8 Pa, less than half that of the latter which increased to 43.2 Pa. With an additional 1-h loading, M4-0.765-C-1 had an increase of only 2.2 Pa in air flow resistance with 1.84-gsm added deposit, in contrast to the huge increase of 50.9 Pa for S-3.060-C-1 with an additional 2.41 gsm. When complete filtration was reached on each filter, it took 12 h for the 4-layer filter with 26.09 gsm of loaded aerosols, 9 h longer and 19.29 gsm more than its 1-layer counterpart. The much slower saturation rate of aerosols in the filter demonstrates the merits of multilayering for electret filters. During the loading process of S-3.060-C-1 with a high fiber packing density, dendrites and cake shielded the filter surface in such a short period that charges as well as fibers within the filter were far from being sufficiently used [8]. In contrast, by separating fibers to different layers, as the surface of the upstream layers was porous enough, the charges on both sides of the fibers in the downstream layers were more accessible to aerosols, thus trapping more aerosols while maintaining a significantly lower increase rate of pressure drop. Also, as a departure from M4-0.765-U, each layer of M4-0.765-C-1 experienced an initial drop of filtration efficiency. This facilitated the penetration of more aerosols through the upstream to downstream layers and consequently

Fig. 12. SEM images of the back side of loaded (a) S-3.060-C-1 and (b) S-3.060-C-3.

Fig. 13. Evolution of pressure drop ($\Delta p$) of S-3.060-U and S-3.060-C-1 with loaded aerosol mass ($M_{dep}$).

Fig. 14. Evolution of pressure drop ($\Delta p$) of S-3.060-C-1 and S-3.060-C-2 with loaded aerosol mass ($M_{dep}$).
more uniform aerosol distribution across the electret filter. It is also worth noting that the filtration efficiency kept at relatively high values for particles of all sizes, indicating that aerosol penetration in the initial loading stage was not significant and the filter has a potential for real application.

The pressure drop variation of M4-0.765-C-1 with aerosol deposition mass is depicted in Fig. 17. The charged 4-layer filter had a much lower pressure drop than that of the uncharged 4-layer filter and the charged 1-layer filter, respectively. This can be explained by the more efficient use of electrostatic effect of the charged filter downstream of the filter surface after multilayering. With this merit, not only aerosols could be deposited on the back of fibers but also distribute more uniformly across the entire filter. Therefore, the formation and growth rates of dendrites and cake, which catalyzed the formation of skin layer, were reduced at the upstream layer.

To make a convincing conclusion, another charged 4-layer filter prepared from the same batch was used to repeat the loading process. As shown in Figs. 16b and 18, a similar result to M4-0.765-C-1 was obtained, indicating the merits of multilayering and charging were reproducible instead of being purely random.

3.3.2. Deposition of aerosols on multilayer PVDF electret filters

The hypothesis of more sufficiently use of charged fibers in the multilayer PVDF electret filter was further proved by SEM images of loaded filters. Figs. 19 and 20 respectively show different layers of the loaded M4-0.765-C-1 seen from the front and the back sides of the filter. By comparison, more aerosols deposited on both sides of the fibers of the two middle layers of M4-0.765-C-1 (Figs. 19–20) than the fibers deep in S-3.060-C-1 (Fig. 15c–d), revealed more effective use of charged fibers across the filter. Compared with M4-0.765-U (Fig. 8), the aerosol deposition across the four layers of M4-0.765-C-1 was more uniform, which was attributed to the role of electrostatic mechanism in aerosol capture [2,5,16,19]. Moreover, many particles can be seen adhering to the back side of individual fibers in all the layers of M4-0.765-C-1 (Fig. 20), resulting from the charges uniformly distributed on the fibers in these layers. In contrast, nearly no particles can be observed at the similar positions of M4-0.765-U (Fig. 9). Besides, the main size of particles seems to decrease with layer depth for M4-0.765-C-1 while this phenomenon was not found for M4-0.765-U. This can be explained by the relation between the size of uncharged particles and the strength of induction effect by charged fibers. As is well known, large particles are prone to charge induction than small particles, therefore the dipoles for larger particles are stronger, and electrostatic mechanism is more pronounced for the capture of bigger size particles [2,3]. During the loading process of M4-0.765-C-1, larger aerosols were removed more by the first layer and smaller aerosols were more penetrating, resulting in relatively high fraction of smaller-sized aerosols in the more downstream layers. Prominently, there was a classification effect on particle size with the multilayer electret nanofiber filter.

4. Theoretical analyses of filter efficiency, capacity and pressure drop

There are three merits for a loaded filter – high efficiency, low pressure drop, and high aerosol storage capacity per unit filter area. Here, we denote single-layer uncharged nanofiber filter as SU, single-layer charged nanofiber filter as SC, 4-layer/multilayer uncharged nanofiber filter as MU, 4-layer/multilayer charged nanofiber filter as MC, and all 4 filters have a total basis weight of 3.060 gsm fibers.

4.1. Efficiency

The efficiency of the SU and MU filters were at 40+% efficiency during the initial period until a cake was formed at which the efficiency reached 100%. The SU could attain 80+% efficiency after 1 h of aerosol loading while the MU needed 8 h of loading to attain 80+. On the other hand, all the charged filters SC and MC started at much higher efficiency of 85–90% and dropped shortly to 80+ due to shielding of the charges but quickly restored back to 100% efficiency when a cake formed on the filter surface.
4.2. Loading capacity and pressure drop

It is desirable for a filter to have large aerosol storage capacity but with low incurred pressure drop. The typical pressure drop versus loading for a filter is depicted in Fig. 21.

With reference to the schematic Fig. 21, starting from a clean filter with initial pressure drop \( \Delta p_m \), there are two regimes of operation: depth filtration followed by cake or surface filtration. During depth filtration, aerosols are trapped and deposited in the filter until it reaches Point 1, where the filter efficiency of all challenging aerosols (i.e. all sizes) reaches 100%, the total deposited mass reaches \( \Delta M_D \), and the pressure drop has escalated to \( \Delta p_1 \). This point marks the ending of depth filtration and the beginning of surface filtration, where the challenging deposit are captured and forms a cake on the filter surface. Thereafter, the cake becomes the effective filter media and its thickness keeps growing with more aerosol deposition. The pressure drop, \( \Delta p \), continues to build up and eventually a ‘structured cake’ forms on the filter surface with approximately constant permeability or resistivity, which can be determined from the slope of the linear portion of the \( \Delta p \) escalation curve. Once the pressure drop of the entire filter reaches an arbitrary set maximum pressure drop, \( \Delta p_2 \) (Point 2 in Fig. 21) the filtration operation stops and the loaded filter needs to be replaced or regenerated. The following interesting issues arise:

a. Which filter among the four filters has the maximum aerosol deposit at the end of the filtration as imposed/set by the maximum pressure drop \( \Delta p_2 \)?

b. For each filter, how much aerosol is deposited during depth and cake filtration, respectively?

c. What are the pressure drop in depth and cake filtration, respectively?

d. What is rate of change of pressure drop with additional mass deposit during cake buildup in surface filtration [20]?

Table 1 summarizes the essential results for the four filters – SU, MU, SC and MC, that address the above questions. Obviously, given the total pressure drop is set as \( \Delta p_2 = \Delta p_{max} \) it is desired to have a small pressure rise gradient in both depth filtration \( (\Delta p_1 - \Delta p_m)/\Delta M_D \) and in cake filtration \( (\Delta p_2 - \Delta p_1)/\Delta M_C = (\Delta p_C/\Delta M_C) \). Both are determined for the four filters in Table 1. Unfortunately, we can have a small slope only in one regime (e.g. depth filtration) but a much larger slope in another regime (e.g. cake filtration), or vice versa, but not in both. The goal is to maximize the total aerosol deposit in the filter in both depth filtration, \( \Delta M_D \), as well as cake filtration, \( \Delta M_C \). This will be apparent in the
4.2.1. SU and SC

Both SU and SC have a short, 3–4 h, depth filtration as a skin formed quickly upstream of the filter layer, leaving the downstream layers of the filter not fully utilized for aerosol capture. This has been discussed and confirmed previously by the SEM images. Interestingly, the total mass deposit in depth filtration from the charged filter SC was actually 6.8 gsm, 0.3 gsm more than the uncharged filter SU with deposit of 6.5 gsm; yet Δp1 was slightly lower at 168 Pa for SC versus 181 Pa for SU. This was all courtesy of the electrostatic mechanism in assisting capture of the aerosols forming a more porous aerosol deposit structure inside the filter, with perhaps lowered skin effect. In subsequent cake filtration, 17.5–19.6 gsm of aerosols were deposited in form of a cake, the higher deposit was attributed to SC despite there was no added electrostatic force due to complete shielding of the charged fibers in the filter by the cake, but the cake formed from the charged filter was found to have a more porous structure. The pressure drop for cake filtration

Fig. 19. SEM images of (a) and (b) the first, (c) and (d) the second, (e) and (f) the third and (g) and (h) the fourth layers of loaded M4-0.765-C-1 seen from the front side.
was higher for SC versus SU, 632 Pa versus 619 Pa, but the rate of increase of pressure drop per additional deposit, \( \frac{\Delta p}{\Delta M_C} \), is actually lower at 32 Pa gsm\(^{-1}\) for SC versus 35 Pa gsm\(^{-1}\) for SU. This indeed supports the assumption that the formed SC cake was more porous and permeable. A more direct measure on the pressure drop per deposit mass is obtained from the slope of the graph, Fig. 13. As seen in Table 1, SC gives a slope of 35 Pa gsm\(^{-1}\) while SU gives 40 Pa gsm\(^{-1}\). Based on these considerations in both depth and cake filtration, the SC filter is better than the SU filter. Interestingly, the clog point (Point 3 in Fig. 21) has been suggested for evaluating the filter clogging during cake formation. The clog point is a hypothetical point wherein the aerosol loading in the filter during depth filtration is replaced by a hypothetical clogging point corresponding to an initial deposition in the filter at equivalent zero pressure drop. Simply put, it is an extrapolation of the 'cake line' extending to zero \( \Delta p \). Based on this, the clog points for the 4 filters are determined in Table 1 for SC as 2.8 gsm and SU as 4.2 gsm.

Fig. 20. SEM images of (a) and (b) the first, (c) and (d) the second, (e) and (f) the third and (g) and (h) the fourth layers of loaded M4-0.765-C-1 seen from the back side.
From the clog point consideration, it appears that the SC filter is more clogged during depth filtration than SU which is incorrect! As can be seen in Table 1, the average \( \Delta p/\Delta M_0 \) is 22 Pa gsm\(^{-1} \) for SC which is lower than 25 Pa gsm\(^{-1} \) for SU. This casts a shadow of doubt on the use of the clog point as an index for comparison between filters. In fact, what is important is the rate of increase, i.e. \( \Delta p_c/\Delta M_c \), between the two filters as demonstrated herein.

### 4.2.2. MU and MC

The MU allows a better utilization of the filter trapping more aerosols not only in the upstream layers but also in the downstream layers as compared to SU. This is even more so when the nano filter is charged. As a result, from Table 1, 16.8 gsm of aerosols had been captured in depth filtration for MU during depth filtration. On the other hand, MC had 55% more trapped aerosols reaching 26.1 gsm. This is courtesy of the charged fibers that trapped aerosols by electrostatic interaction on all surfaces of the fiber in addition to interception mechanism. Further, all the layers across the entire filter depth from upstream to downstream were fully utilized for capturing aerosols. This provides a good combat against the skin effect which affects greatly the single layer charged/uncharged filters and also the multilayer uncharged filter to some extent. Given the aerosols are uniformly distributed across the filter, the pressure drop, \( \Delta p_1 \), was 6% lower at 316 Pa for MC as compared to 337 Pa for MU despite it had more aerosols trapped. In fact, the average \( \Delta p_c/\Delta M_c \) for MC is indeed very low at 11 Pa gsm\(^{-1} \) while that of MU is 19 Pa gsm\(^{-1} \). In subsequent cake filtration, the pressure drop rose very steeply with increasing deposition. Such steep rise was attributed to the fact that a cake was formed on a filter with both PVDF fibers as well as deposit trapped in the filter that acted also like artificial fibers [21]. This was equivalent to a filter with higher fiber packing density which resulted in an impervious cake with high pressure drop to fluid flow. Indeed, in surface filtration the MU filter had a further increase in \( \Delta p \) of 463 Pa for 9.6-gsm cake, while MC had an increase in \( \Delta p \) of 484 Pa for 10.4-gsm cake. In fact, from the slope of the curve from Fig. 17, \( \Delta p_c/\Delta M_c \) is 53 Pa gsm\(^{-1} \) for MU and is greater than 49 Pa gsm\(^{-1} \) for MC, given the skin effect for the latter was being reduced. Despite both pressure-drop increase was much higher than their counterparts – the single layer, cake filtration duration for the multilayer filters was shorter. From Table 1, only 28% of aerosol deposit was attributed to cake filtration for MC and 37% for MU. This demonstrates cake filtration was indeed short for the multilayer filters. This was actually favorable otherwise the pressure escalation could be extremely high in cake filtration taking away the earlier benefit of depth filtration where most aerosols were deposited.

On the contrary, it is the reverse for single-layer filter uncharged and charged. From Table 1, 74% of aerosol deposit was attributed to cake filtration for SC and 73% for SU. Therefore, majority of the aerosol deposit were made during cake filtration for the single-layer filters, uncharged and charged, as a skin formed very fast upstream of the filter for the single layer nanofiber filter (charged/uncharged) that quickly changed over to cake filtration. The multilayer filters (MC and MU) had a long depth filtration period with short steep pressure gradient in cake filtration. On the other hand, the single layer filters (SC and SU) had short depth filtration (not fully utilizing the filter downstream) with milder pressure rise in cake filtration. The behavior of these 4 filters were rather ‘polarized’ but the ultimate question is which filter had the highest capacity for trapping and storing aerosols when a pressure drop limit was imposed on the filtration cycle? In the present study, the pressure limit was set at 800 Pa which was similar in value to our early investigation [22]. Overall, with a maximum pressure drop of 800 Pa for the filter operation, MC had a total capacity of 36.5 gsm, both MU and SC had equal capacity of 26.4 gsm, and SU had 24 gsm. (Note the media resistance for all 4 filters in their clean state was within a narrow range between 16.9 and 21.1 Pa, see Table 1.) Both efficiency and capacity results are summarized for the 4 filters in Table 2 based on a maximum pressure drop of 800 Pa. In order of descending performance based on efficiency and capacity, MC is the best followed by SC, then MU, and finally SU. The two charged filters are better than their uncharged counterparts. MC has 52% more storage capacity than SU, and it has also 38% more capacity than both SC and MU. Note, the clog point that is used for assessing loading of filters do not provide meaningful interpretation on the capacity nor pressure drop in our present filter study despite we have also included their values in Table 1 for reference.

### 5. Conclusions

Comparative experiments were conducted to investigate the differences of aerosol loading behaviors between 1-layer and multilayer filters and uncharged and charged filters. Distinctive results were acquired and analyzed, which led to several conclusions listed below:

1. With loaded aerosol mass, the filtration efficiency of uncharged filters increased while that of charged filters initially decreased and then increased until reaching 100%. Nonetheless, the charged filters always had higher efficiency than the uncharged counterparts before a complete cake was formed on the filter.

### 5.1. Comparative Performance at Different Pressure Drops

Comparative experiments were conducted to investigate the differences of aerosol loading behaviors between 1-layer and multilayer filters and uncharged and charged filters. Distinctive results were acquired and analyzed, which led to several conclusions listed below:

| Filter | \( \Delta p_{in} \) Pa | \( \Delta M_0 \) gsm | \( \Delta p_1 \) Pa | \( \Delta p_c/\Delta M_c \) Pa/gsm | \( \Delta M_c \) gsm | \( \Delta p_c/\Delta M_c \) Pa/gsm | \( \Delta M_c/(\Delta M_0 + \Delta M_c)\% \) | \( \Delta p_c/\Delta p_{in} \) |
|--------|------------------|----------------|----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| SU     | 20.8             | 6.5            | 181            | 25              | 4               | 20              | 17.5            | 35              | 73%             | 79%             | 4.2             |
| SC     | 21.1             | 6.8            | 168            | 22              | 3               | 24              | 19.6            | 32              | 74%             | 81%             | 2.8             |
| MU     | 16.9             | 16.8           | 337            | 19              | 14              | 36.5            | 9.64            | 48              | 37%             | 59%             | 10              |
| MC     | 18.1             | 26.1           | 316            | 11              | 12              | 49              | 10.4            | 47              | 28%             | 62%             | 20              |
times more aerosol holding capacity than the single uncharged/charged nanofiber filter with the same fiber basis weight in depth filtration.

(7) In a filtration operation with 800 Pa maximum pressure, the 4-layer electret nanofiber filter with 3-gsm basis weight had 52% more aerosol holding capacity than the single uncharged nanofiber filter. It had also 38% more aerosol holding capacity than the 1-layer electret nanofiber filter as well as the 4-layer uncharged nanofiber filter.

To conclude, the multilayer PVDF electret filters possess excellent filtration performance for long-term aerosol filtration and have a great potential for commercial use in the fields of personal health care and environmental protection.

CRediT authorship contribution statement

Qiangqiang Sun: Investigation, Methodology, Software, Data curation, Formal analysis, Validation, Visualization, Writing - original draft. Wallace Woon-Fong Leung: Conceptualization, Formal analysis, Methodology, Supervision, Writing - review & editing, Funding acquisition, Project administration, Resources.

Declaration of Competing Interest

The authors declared that there is no conflict of interest.

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Appendix A

In this Appendix, the performance of the multilayer electret nanofiber filters with 4.59 gsm (6 layers, with each layer having 0.765 gsm) of nanofibers is compared with two conventional electret microfiber filters with 20–30 gsm of fibers. Fig. A1 shows a comparison of filtration efficiency versus aerosol or particle size from 50 to 500 nm for clean filter state before loading. As can be seen, the filtration efficiency is higher for the multilayer electret filter for all particle sizes tested. For example, at 300 nm, the two conventional microfiber electrets made of polypropylene achieve 87.5% on average while the multilayer electret nanofiber filter achieves 98% which is a 12% increase.

Fig. A2 shows a comparison of filtration efficiency versus aerosol/particle size from 50 to 500 nm for clean filter state before loading. As can be seen, the filtration efficiency is higher for the multilayer electret filter for all particle sizes tested. For example, at 300 nm, the two conventional microfiber electret filters made of polypropylene achieve a quality factor of 0.085 Pa^{-1} on average while the multilayer electret nanofiber filter achieves 0.155 Pa^{-1} which is a whopping 82% increase.

Table 2: Ranking of 4 filters based on maximum pressure, efficiency and capacity.

| Ranking | Filter | Efficiency (%) | Capacity (gsm) | Capacity ratio | Capacity ratio |
|---------|--------|----------------|----------------|---------------|---------------|
| 1       | MC     | 80–100         | 36.5           | 1.52          | 1.38          |
| 2       | SC     | 80–100         | 26.4           | /             | 1             |
| 3       | MU     | 40–100         | 24             | 1             | /             |

(2) For the charged filters during the initial stage of aerosol loading, the deposited aerosols influenced the capture efficiency of particles by the diminishing electrostatic effect and the growing mechanical filtration effect, with larger particles more affected by the variation of the former effect.

(3) In uncharged state, the 1-layer filter had a lower aerosol holding capacity than the multilayer filter with the same basis weights, due to the quick formation of “skin aerosol layer” from the low filter porosity and the initial high filtration efficiency.

(4) Combining the advantages of multilayering and charging, the charged multilayer filter had the best aerosol holding capacity and the highest aerosol distribution uniformity, which was further enhanced by the more sufficient use of charges on individual fibers across the entire filter depth/thickness. Under the same loading conditions with a maximum pressure drop of say, 800 Pa, filter performance based on efficiency and capacity is in the order of:

a. charged multilayer filter
b. charged 1-layer filter
c. uncharged multilayer filter
d. uncharged 1-layer filter

(5) The multilayer nanofiber had approximately 70% aerosols trapped in the filter and 30% trapped in the cake, while the single layer nanofiber had 30% aerosols trapped in the filter and 70% trapped in the cake.

(6) The 4-layer electret nanofiber filter with 3 gsm of basis weight has 4 times more aerosol holding capacity than the single uncharged/charged nanofiber filter with the same fiber basis weight in depth filtration.

Fig. A1: Comparing filtration efficiency (η) of multilayer nanofiber electret with two conventional microfiber electrets at 23–26 Pa of pressure drop and 5.3 cm s^{-1} of face velocity.
Appendix B. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.seppur.2020.116606.

Fig. A2. Comparing quality factor (QF) of multilayer nanofiber electret with two conventional microfiber electrets at 23–26 Pa of pressure drop and 5.3 cm s⁻¹ of face velocity.

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