High fidelity simulation of ultrafine PM filtration by multiscale fibrous media characterized by a combination of X-ray CT and FIB-SEM

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ABSTRACT

Air filtration mechanisms in the composite filter media used in practical applications are important and challenging to understand because the component fibers could have various size scales and morphologies. In this work, a three-dimensional digital model of nanofiber-based filter media was reconstructed for the first time based on the X-ray tomography data for the cellulose substrate and the Focused Ion Beam-Scanning Electron Microscope (FIB-SEM) image analysis for the several micrometers thick (3.82–7.90 μm) electrospun polyvinylidene fluoride (PVDF) nanofiber membrane. Besides the high-resolution model where the details of the fibrous structures were fully resolved, another low-resolution model with approximated unresolved structures was also established. Filtration simulations utilizing these models were conducted considering the drag force, Brownian diffusion and aerodynamic slip. The simulated filtration efficiencies agreed well with the experiments for particles of 70–400 nm, including the most penetrating particle size (MPPS, 100–200 nm). Moreover, the structure-resolved models had higher accuracy but higher computational costs, while the unresolved simulations saved much running time but over-predicted the filtration efficiency, especially for smaller particles (<100 nm). Our study presents a comprehensive strategy for investigating the composite filter media with multiscale complex structures using a combination of advanced characterization technologies and modular simulation models.

1. Introduction

The safety and environmental issues have recently attracted broad attention due to the continuously deteriorating environment, air pollution and outbreaks of the respiratory infectious diseases [1], which drives the investigation of air filtration technology and application [2]. As a recognized cost-effective material to capture the suspended particles in air, fibrous filter media with extensively fiber networks and tortuous interconnected pores have been widely used in air filtration [3], especially for PM_{2.5} particles which may contain various viruses and bacteria, threatening human health [4]. Fibrous filter media utilize different types of fibers, which can be classified by sources, e.g. natural fibers, synthetic polymer fibers, inorganic/mineral fibers and biofibers. As one of the most rapidly growing technology over the last 20 years, nanofibers have been widely adopted in various air filtration applications due to the superb performance in the interception and inertial impaction regime, and their flexibility to be fabricated from different types of materials [5–10]. However, the thin nanofiber membrane (NFM) often has limited mechanical properties [11,12]. To achieve balance between the high filtration efficiency and the structural integrity during applications, a nanofiber membrane is usually fabricated as a layer or coating to a micro-scale fiber based substrate e.g. cellulose and spunbond substrate [13], whereby the nanofiber improves the filtration efficiency and the substrate as the support structure enhances the mechanical properties and enlarges the dust storage space. Therefore, it is important to investigate the structure and filtration mechanisms of these composite filter media which were close to the filters used in practical applications.

The detailed analysis for the structures of multiscale composite filter media is the indispensable information to better understand their filtration mechanisms. However, the accurate structures are difficult to obtain due to the numerous fibers with various size scales and...
morphologies. The filter media composed of micro-scale fibers can be measured using the standard techniques e.g. digital volume imaging [14], magnetic resonance imaging [15], laboratory or synchrotron X-ray tomography [16–20]. A typical laboratory X-ray computed tomography instrument (CT) has a resolution of 5 μm, and the submicron CT or nano CT could measure down to 0.5 μm [21]. Some synchrotron radiation tomographic microscopy performed at high energy beamline could reach an effective pixel size of about 0.15 μm [22], but the typical minimum object size should be at least 3 voxel size wide for a quality scan [23]. The above examples indicate that it is hard to completely reconstruct membranes containing fibers below 300 nm by using these standard techniques. Thus, the scanning electron microscope (SEM) is commonly utilized to obtain the key factors, including diameter, length, curvature of actual fibers of the nanofiber membranes [24–28]. For composite filter media, Zhang et al. reported a single layer PEO@PAN/PSU composite membrane structure model, reconstructed by using SEM images to obtain the pore structure parameters and the average diameters of PAN and PSU fibers, which were about 270 nm and 1.3 μm, respectively [29]. Zhang et al. studied the particle capture performance of a gradient-structure filter media composed of PSU fiber layer (diameter around 1 μm), PAN fiber layer (diameter around 200 nm) and PA-6 fiber layer (diameter around 20 nm), and a 3D digital structure model was generated with the parameters measured by experiments and SEM images [30]. Dusan et al. built up idealized 3D models for filter media with PVDF nanofibers (diameter around 100 nm) layered on melt blown substrate (diameter around 1 μm) based on the fiber features obtained from SEM images [31]. However, the slip effect around the nanofibers was neglected in the flow simulations of these studies, which played an important role in reducing the drag force due to nanofibers on the airflow [32]. Wang et al. used a 2-D structure model representing filter media with a nanofiber layer on a substrate, where the nanofiber layer was assumed as a layer with single-fiber thickness consisting of uniformly distributed circular fibers, and the substrate was simplified as a porous jump boundary ignoring the micro-structure inside it [33]. All of these structure models relied on substantial assumptions. The thickness of the nanofiber membranes was often ignored or derived from empirical equations since the cross-section of the nanofiber membranes could be heavily deformed during the cutting process for SEM images [34]. These idealized models used for nanofiber membranes were insufficient to represent the actual structure of the fibers in substrates, especially the cellulose fibers with hollow structure, where the inner diameter could be different due to the difference in sources and treatment [35], and thus could compromise the quality of numerical calculations for the filter performance. It is difficult to reconstruct the composite filter media with single characterization method. Moreover, the flow regimes in the substrate and nanofiber membrane were different due to the various fiber scales [36]. It remains unclear whether the slip model used at the surface of the nanofiber in numerical simulations could be suitable for composite filter media.

This work studied the particulate filtration process of nanofiber-based composite media with experimental and simulation methods. The filter media composite #1, #2 and #3 were respectively created based on the X-ray to reconstruction membranes containing fibers below 300 nm by using these standard techniques. Thus, the scanning electron microscope (SEM) is commonly utilized to obtain the key factors, including diameter, length, curvature of actual fibers of the nanofiber membranes [24–28]. For composite filter media, Zhang et al. reported a single layer PEO@PAN/PSU composite membrane structure model, reconstructed by using SEM images to obtain the pore structure parameters and the average diameters of PAN and PSU fibers, which were about 270 nm and 1.3 μm, respectively [29]. Zhang et al. studied the particle capture performance of a gradient-structure filter media composed of PSU fiber layer (diameter around 1 μm), PAN fiber layer (diameter around 200 nm) and PA-6 fiber layer (diameter around 20 nm), and a 3D digital structure model was generated with the parameters measured by experiments and SEM images [30]. Dusan et al. built up idealized 3D models for filter media with PVDF nanofibers (diameter around 100 nm) layered on melt blown substrate (diameter around 1 μm) based on the fiber features obtained from SEM images [31]. However, the slip effect around the nanofibers was neglected in the flow simulations of these studies, which played an important role in reducing the drag force due to nanofibers on the air flow [32]. Wang et al. used a 2-D structure model representing filter media with a nanofiber layer on a substrate, where the nanofiber layer was assumed as a layer with single-fiber thickness consisting of uniformly distributed circular fibers, and the substrate was simplified as a porous jump boundary ignoring the micro-structure inside it [33]. All of these structure models relied on substantial assumptions. The thickness of the nanofiber membranes was often ignored or derived from empirical equations since the cross-section of the nanofiber membranes could be heavily deformed during the cutting process for SEM images [34]. These idealized models used for nanofiber membranes were insufficient to represent the actual structure of the fibers in substrates, especially the cellulose fibers with hollow structure, where the inner diameter could be different due to the difference in sources and treatment [35], and thus could compromise the quality of numerical calculations for the filter performance. It is difficult to reconstruct the composite filter media with single characterization method. Moreover, the flow regimes in the substrate and nanofiber membrane were different due to the various fiber scales [36]. It remains unclear whether the slip model used at the surface of the nanofiber in numerical simulations could be suitable for composite filter media.

This work studied the particulate filtration process of nanofiber-based composite media with experimental and simulation methods. The filter media composite #1, #2 and #3 were prepared by electrospinning PVDF nanofibers on the cellulose fiber-based substrate with different fabrication durations, which were 1.0, 1.5 and 2.0 h, respectively. The average diameter of PVDF nanofibers was around 309 nm, and the diameter of the fibers in the substrate was around tens of micrometers. The 3D digital structure models of the three nanofiber-based composite filter media were first developed by a new method which combined X-ray tomography and FIB-SEM. The 3D voxel-based virtual substrate and NFMs were respectively created based on the X-ray tomography (CT) slices and the FIB-SEM images analytic data. Then the voxel sizes of the substrate and NFM were unified to be assembled together to obtain the complete composite structures. In addition to the high-resolution model in which the details of the fibrous structures were fully resolved, another low-resolution model with approximated unresolved structures was also established. Particular filtration simulations based on the resolved and unresolved models were carried out considering the air drag force, Brownian diffusion and slip effect around the surface of fibers. The obtained results from numerical simulations such as pressure drop and filtration efficiency were analyzed and compared with the corresponding experimental data. The strategy forms the basis of a powerful tool for innovative multi-layer composite media design and optimization. The various composite filter media models proposed here can provide reference for further investigation of the dynamic evolution of the microstructure and the macroscopic filtration characteristics of composite filter media in the long-term particle loading process.

2. Experiment

2.1. Material and samples preparation

The substrate composed of micro-scale cellulose and polyester (PET) fibers was prepared with the wet-laid method, provided by Fibrweb (Guangzhou, China). Then a PVDF nanofiber membrane was fabricated on the substrate by electrospinning. For the fabrication of NFM, polyvinylidene fluoride (Mw = 15,000, Sigma-Aldrich Co., Ltd) powders were dissolved in N,N-dimethylformamide (DMF)-Acetone (Sigma-Aldrich Co., Ltd) (1:1 wt%), and the solution was stirred for over 12 h at room temperature. A multi-jet electrospinning machine (NaBond Technologies Co., Ltd) equipped with three spinnerets and a rotating drum substrate was used to produce the double layer composite filter media. The detailed parameters for the electrospinning process were summarized in Table 1.

2.2. Filter media characterization

2.2.1. Fiber size distribution and thickness

The fiber diameter distribution was determined by measuring over 50 fibers in the SEM images using an image analyser (Image J, NIH) [37]. Different methods were utilized to characterize the thickness in this study due to the dual-layer structure of the filter media containing fibers with different scales. The thickness of the substrate was directly determined by SEM visualization with normal sampling method. Since the NFM was too weak and thin to keep the cross-section from deforming during the normal cutting process, a combined FIB-SEM (Helios NanoLab DualBeam, USA) was used to measure its thickness. The cross-section was exposed by a FIB milling and imaged in SEM secondary electron (SE) mode at a sample tilt of 52°. After tilt correction, the thickness of NFM was determined.

2.2.2. Air permeability and solidity

The air pressure drops of the medium were measured at different face velocities from 16.67 to 100 mm/s. Specially, the pressure drop (Δp) of the NFM was estimated by subtracting the pressure drop of substrate Δp of the overall pressure drop of composite filter media. A gas pycnometer (AccuPyc II 1340, Micromeritics) based on the gas displacement method was applied to determine the skeleton density of the substrate and the analysis gas was helium. Then the solidity ϕ can be derived by Equation (1).

$$\phi = \frac{\rho_s}{\rho}$$  

(1)

where ρs was the skeleton density; ρ1 was the macro density of material defined as the ratio between the mass and the total volume of the substrate. Since the solidity of NFMs could not be determined with the gas pycnometer directly, a method by fitting simulated results with measured values was used [38]. Air flow simulations were conducted on the generated 3D virtual nanofiber structures with predefined solidities (the generation and modeling process to be discussed).
can be found in the previous work [40]. The filtration efficiency was in agreement with the experimental requirement. More details of the setup were set before DMA to ensure the ratio between the sheath flow and the aerosol flow entering the DMA was around ten [39]. Two condensation particle counters were installed upstream and downstream of the filter holder to measure the particle concentrations. Two mass flow control aerosol flow and makeup air flow rates in the monodisperse sodium chloride nanoparticles were utilized in the tests. The dry and clean compressed air was delivered to a lab-made aerosol generator to generate polydisperse sodium chloride particles by atomizing 1 wt% NaCl solution. The generated aerosol went through a diffusion dryer for evaporating the water, and then neutralized by a Kr-85 bi-polar charger to reach the Boltzmann equilibrium charge distribution. The monodisperse particles with mobility diameters of 50, 70, 90, 100, 200, 300, 400, 500 nm were respectively selected by a differential mobility analyser (DMA, model 3081, TSI). A laminar flow meter was set before DMA to ensure the ratio between the sheath flow and the aerosol flow entering the DMA was around ten [39]. Two condensation particle counters were installed upstream and downstream of the filter holder to measure the particle concentrations. Two mass flow controllers (MFC) were used to keep the aerosol and makeup air flow rates in agreement with the experimental requirement. More details of the setup can be found in the previous work [40]. The filtration efficiency was defined by the percentage of the particle collection as in Equation (2).

\[ E_{\text{total}} = 1 - \frac{C_{\text{down}}}{C_{\text{up}}} \]  

where \( C_{\text{down}} \) and \( C_{\text{up}} \) were respectively the up- and down-stream particle number concentrations.

### 3. Numerical simulation

#### 3.1. Reconstruction of filter media

**3.1.1. Substrate model**

The top-view and cross-sectional SEM images of substrate was showed in Fig. 1. The cellulose fibers in the substrate had irregular shapes and hollow structure, often twisted and entangled with each other, which made it difficult to reconstruct the internal micro-structure by using the method to randomly generate fiber structures with only parametric constraints such as mean fiber size and porosity. An X-ray tomography-based method was used to build the digital structure of the cellulose substrate. Fig. 2 showed the reconstruction processes, including slices acquisition, image processing. Here, the air flow direction was aligned with the z-axis, while the surface of membrane was defined parallel to the x-y plane. The slices acquisition was conducted by using a tomography instrument (RX Solution Easy Tom XL) equipped with 160 kV and 230 kV X-ray sources. The tube with 40 kV was used for electrons striking to produce the X-ray cone beam in this study, and the tube current was 160 μA. Raw X-ray digital images were captured by a high resolution and high-speed CCD detector. The distance between the X-ray source and specimen was 7.109 mm, and the X-ray source to detector distance was 610.65 mm. The frame rate of the imager was 1 fps. After being filtered by a Tukey window, 1352 slices taken in the vertical (x-z) plane with 1845 × 1845 voxels were obtained as shown in Fig. 2a, and the effective voxel size was 1.48 μm. Then the slices were imported to the ImportGeo-Vol module in the GeoDict software package for 3D processing (Fig. 2b). The following procedures were utilized to build the 3D digital structure model: 1) permute the axis and rotate the images to align the cross-sections and surface of substrate to the x-z and x-y plane, respectively (Fig. 2c); 2) crop the slices to 700 × 700 voxels to avoid the edge effects and cut the domain to 225 voxels with 333 μm thickness according to the measured values (Fig. 2d); 3) de-noise the images by using the non-local means filter (Fig. 2e–f); 4) separate the pore space from solid (Fig. 2g). The segmentation was performed by using the single threshold filter. The threshold value was set according to the peak position in the gray value map and the measured solidity. The resulting substrate medium was shown in Fig. 2h, and the effective voxel size was 1.48 μm. In contrast to the digital model from CT slices, a random parametric model based on the component information provided by the producer and fiber orientation from the analysis of above structure was created, and the other geometric parameters were the same as those of reconstructed model from CT data. Specially, the

![Fig. 1. a) Top-view SEM images of substrate, b) cross-sectional SEM images.](image-url)
hollow structure of the cellulose fibers (see Fig. 1b) was considered by introducing a parameter called the inner diameter fraction, defined as the ratio of inner diameter to outer diameter of hollow structural section. The inner diameter fraction was also used as the fitting parameter to match the pressure drop measured in experiment. The creation process was similar to our previous work (see supporting Text S1) [41].

3.1.2. NFM model

Fig. 3a–c showed the surfaces of the NFM on top of the composite #1 to #3. The measured diameters of the PVDF nanofibers were from 0.130 to 0.891 μm. Since the minimum size of detectable object in the specimen should be 3 times larger than the resolution of CT instrument, fibers with diameters around 0.1 μm were too small for imaging [21]. Moreover, the PVDF nanofibers were almost straight and much more regular than cellulose fibers in morphology. Therefore, a random structure based on the parameters, i.e. fiber size distribution, thickness, and solidity, was generated to represent the NFM. The log-normal distribution was applied to simulate the size distribution of nanofibers in the membranes according to the measurements, and the count median diameter (CMD) and geometric standard deviation (GSD) were 270.65 nm and 1.64, respectively. The cross-sectional SEM images were shown in Fig. 3d–f, and the determined thicknesses of the NFM of composite #1 to #3 were 3.82, 6.42 and 7.90 μm, respectively. The voxel size was 50 nm. Three repeat structure models were created for each case by changing the random seed to adjust the inner structure.

3.1.3. Composite model

Since the created digital structures, both for substrate and NFM, were voxel-based models, the voxel size of them need to be unified for assembling. To balance the computation cost and simulation accuracy (see Fig. S2), four substructures with a domain size of 100 × 100 × 225 voxels were cut from the reconstructed substrate from CT slices. Then the voxel length of each substructure was refined from 1.48 μm to 105.6 nm, i.e., each original voxel was divided into 14 × 14 × 14 grids. The domain of each substructure was turned into 1400 × 1400 × 3150 voxels. The composite filter media were generated by putting the NFM with the same surface area and voxel size on the rescaled substrate. Note that, up to this step, the voxels constituting the virtual fiber structures were considered as solid, and the other voxels were treated as empty, and these digital composite media were noted as resolved models. In contrast, the unresolved composite models were created, where the detailed structure of NFM was ignored and replaced by a porous jump plane composed of porous voxels, which were simplified computational grids representing the average properties of the NFM. The virtual substrates were still from the CT slices. The porous jump plane was modelled as a homogeneous material with given thickness and permeability from experiments, and the pass-through model (the details to be discussed) was used for describing the probability that a particle passed through the filter media. The domain size of the unresolved model for the entire composite was 700 × 700 × 226 voxels, and the voxel size was the same with the reconstructed substrate, which was 1.48 μm.
3.2. Numerical simulation

3.2.1. Air flow

A well-known integrated software package GeoDict (Math2Market, Germany) was used in this study for the air flow and particle filtration process simulations. The fluid inside the virtual structures was assumed viscous, incompressible and steady. Conservation equations of mass (continuity equation) and momentum (Navier-Stokes equation) governing fluid flow in differential form can be given as follows:

\[ \nabla \cdot \vec{v} = 0 \]  (3)

\[ \rho (\vec{v} \cdot \nabla) \vec{v} = -\nabla p + \mu \nabla^2 \vec{v} + \vec{f} \]  (4)

where \( \rho \) is the fluid density, \( \vec{v} \) was the flow velocity, \( p \) is the pressure, \( \mu \) was the fluid density, and \( \vec{f} \) was a force density, which was zero here. In the case of fibrous filter media considered here, the Reynold number was sufficient small (\( Re \approx 1 \)), the conservation momentum can be simplified into Stokes equation as Equation (5):

\[ -\mu \Delta \vec{v} + \nabla p = \vec{f} \]  (5)

The slip effect around the nanofibers was also considered. In this work, the Knudsen number (\( Kn \)), i.e. the ratio of the mean free path of air molecule to the average fiber diameter, was 0.21, indicating that the air flow around the nanofibers was mainly in the slip or early transition flow regime (0.1 < \( Kn < 0.4 \)). Kirsch et al. stated the slip model may be applied up to \( Kn \approx 1 \) [42]. Thus, the rarefaction effect along the nanofibers could be described as a finite partial slip using Maxwell’s velocity slip as Equation (6) [43].

\[ \nu_w = \frac{2 - \sigma_v}{\sigma_v} \frac{dv}{dn} \]  (6)

where \( \nu_w \) was the tangential velocity of the flow at the wall, \( \sigma_v \) was the tangential momentum accommodation coefficient, which was taken as unity here [28], \( \lambda \) was the mean free path of air molecules and it was 66 nm, and \( \frac{dv}{dn} \) was the strain rate in the normal direction.

The standard LIR (left-identity-right) solver of Geodict, using a very memory efficient adaptive grid structure, was employed to solve the partial differential equations describing the fluid dynamics [44]. The error bound stopping criterion was set to 0.01. The configurations of boundary conditions were shown in Fig. S3. Periodic boundary conditions were applied for the inlet, outlet and boundaries parallel to the flow directions. An implicit in-flow and an out-flow region were added, which were both 200 voxels thick, to avoid the flow channel closure.

3.2.2. Particle transport

The movement of the particle during the filtration process was simulated by using Lagrangian Particle Tracking method. The dispersed two-phase flow was assumed to be dilute, i.e. the distance between the particles in the air was large enough to ignore the particle-particle collisions, and the flowing particles have no effect on the flow. The sodium chloride particles were simplified into spherical particles, and the motion of each particle was influenced by the combination of the fluid drag force and Brownian motion. Then the trajectory of each particle can be obtained by solving the equations as follows:
\[
\begin{align*}
\frac{d u_c}{dt} &= 3 \mu_p D C_c \left( u_c - u_p \right) + \sqrt{\frac{2k_B T}{\tau} \frac{d W(t)}{dt}} \quad (7) \\
\frac{dx_p}{dt} &= u_p \quad (8)
\end{align*}
\]

where \( m \) was the particle mass (kg), \( u_c \) was the air flow velocity (m/s), \( u_p \) was the particle velocity (m/s), \( D \) was the particle diameter (m), \( k_B \) was the Boltzmann constant (J/K), \( T \) was the temperature (K), \( \gamma \) was the friction coefficient, \( dW \) was 3D Wiener measure (\( \sqrt{s} \)), responsible for the continuous-time stochastic process. As the diameters of NaCl particles we investigated were down to nanoscale, the slip condition was needed. \( C_c \) was the Cunningham slip corrections factor which can be given as Equation (9) [45].

\[
C_c = 1 + \frac{\lambda}{D} \left( 2.34 + 1.05a^{0.304} \right) \quad (9)
\]

The computation domain and boundary condition configurations for particle transport were the same as those in the air flow simulation. The particle initial position was located in the added inflow region, which was 1 \( \mu m \) downstream from the inlet, to avoid escaping particles across the inlet due to Brownian motion. The diameters of the simulated particle were in the range of 10–1000 nm, and 1000 particles of each diameter were tracked during the filtration process. For the simulation of resolved models, particles were assumed captured after first touching the fibers both in the NFM and substrate, while for the unresolved models, particles were assumed captured after first touching the random parametric substrate model. The red cylinders represented PET fibers, while the curved green and yellow objects with hollow structure represented two types of cellulose fibers in the actual substrate.

Fig. 4d compared the measured and simulated pressure drops across the substrate at different face velocities from 16.67 to 100 mm/s. As in the air flow simulation process, the Stokes equation was used to simulate the flow field, leading to the linear relation between the pressure drop and the surface velocity. The pressure drop from experiments was in agreement with the simulated results. Then the inherent permeability \( k \) was derived by applying Darcy’s law expressed as Equation (11).

\[
v = \frac{-k \Delta P}{\mu \cdot t} \quad (11)
\]

where \( v \) was the face velocity; \( \Delta P \) was the pressure drop; \( t \) was the thickness of the media. The permeabilities of the actual filter media, digital model from the CT slices and geometric parameters were respectively 1.47 \( \times 10^{-11} \), 1.57 \( \times 10^{-11} \) and 1.50 \( \times 10^{-11} \) m\(^2\). It revealed that the porous structures of digital models were nearly the same with the actual media. Fig. 4e presented the velocity magnitude inside the digital substrate from CT slices at the flow velocity of 5.35 cm/s. The porous structure inside the substrate formed the flow channels for the air passing through the substrate, and the uneven pore sizes led to the inhomogeneously distributed flow velocities.

Before the simulation of air flow inside the NFMs, the first order Maxwell model used at the surface of fibers were evaluated (see supporting Text S2 for detailed discussions). The simulated pressure drops with no slip and slip boundary conditions were also compared and showed in Fig. 5ac. As expected, the simulated pressure drops of substrate with slip and no-slip boundary conditions were in good agreement because the \( Kn \) was close to 0, while the pressure drops across the NFMs using slip boundary condition were much lower than using no-slip boundary condition.

The comparisons of pressure drop vs. face velocity curves of the NFMs from experiments and simulations were shown in Fig. 5a. Since the input solidity was tuned to fit the pressure drop measurement results, the pressure drops from simulations agreed well with the experimental results. The derived solidity values of the NFMs corresponding to composite #1 to #3 were respectively 4.10%, 6.25% and 7.00%. Fig. 5b–d showed the final 3D digital structures of the NFMs, the thickness of the virtual NFM increased with the electrospinning duration.

### 4.2. Particle filtration efficiency

The initial filtration efficiencies of the filter media for the sodium chloride particles from 10 to 1000 nm with a face velocity of 5.35 cm/s were simulated using the reconstructed digital structure models (modelling process to be discussed in the experimental section and numerical models), with the same conditions of the experimental tests. Fig. 6a compared the simulated filtration efficiencies of the substrate models from CT slices and geometric parameters with the experimental results. The trends of the simulated results agreed well with the

| Table 2 | Parameters configured in the particle filtration simulation. |
|---------|---------------------------------------------------------------|
| Parameter | Values |
| Air density, \( \rho_a \) (kg/m\(^3\)) | 1.204 |
| Air dynamic viscosity, \( \mu \) (kg/(m s)) | \( 1.834 \times 10^{-5} \) |
| Particle density, \( \rho_p \) (kg/m\(^3\)) | 2165 |
| Particle diameter, \( D_p \) (\( \mu m \)) | 0.01–0.1, 0.1–1 |
| Face velocity, \( u_f \) (cm/s) | 5.35 |
experimental values. Good agreement was observed for particles smaller than 300 nm, while for particles larger than 300 nm, simulated efficiencies were slightly higher than the experimental values. This discrepancy can be related to ignoring the re-entrainment influence for larger particles in the numerical simulation [48]. The predicted values from parametric substrate models were higher than those from CT slices-based substrate, which may be caused by the insufficient ability of the idealized parametric models to consider the heterogeneity in the actual material [47]. But the parametric models were convenient for the design of the filter media by adjusting the input parameters such as diameter and length of fibers. As shown in Fig. 6b, the effect of hollow cellulose fibers on the filtration efficiency of substrate was studied. The increase in the inner diameter fraction of cellulose fibers led to the improvement of filtration efficiency. For a fixed solidity, the amount of fibers contained in the structures increased with the increase of inner diameter fraction of fibers, resulting in the increased effective filtration area.

The simulated filtration efficiencies of the NFMs were shown in Fig. 6c and compared with the corresponding experimental results. The filtration efficiencies of the NFMs from experiments were estimated based on the determined filtration efficiencies of the whole composite filter media ($E_{\text{total}}$) and the substrate ($E_s$) using the relation shown as Equation (12)

$$E_{\text{nl}} = 1 - \frac{1 - E_{\text{total}}}{1 - E_s}$$

The simulated results showed that the filtration efficiencies increased with the electrospinning durations from 1.0 to 2.0 h, which were consistent with the experimental values. As shown in Fig. 6c, the MPPS of the three NFMs were all in the range of 100–200 nm at face velocity of 5.35 cm/s. For particle sizes in the range of 70–400 nm including the MPPS, the simulated efficiencies agreed well with the measured values. It is also noted that the simulated efficiency curves started to deviate from the measured values when the particle sizes were smaller than 70 nm or larger than 400 nm. The slight over-predictions for the large particle sizes may be related to omission of effects from particle re-entrainment and particle rebound [49,50]. The minor over-predictions at small particle size range could be caused by the assumption of Equation (12) that the NFM and the substrate act independently during the filtration process. Due to the large diffusion coefficients of small particles (<70 nm) and the wide effective range of the diffusion mechanism, the filtration of small particles by the NFM and substrate may not be considered as independent [33].

The resolved model and unresolved model corresponding to composite #1 were shown in Fig. 7a(i, ii), respectively. The NFMs were rendered as red and located on the substrates, which were comprised of gray cellulose fibers. In the resolved composite model, the PVDF nanofiber and pore structure can be clearly observed, while there was no detailed structure inside the NFM for the unresolved cases. The filtration efficiencies of these structures were simulated separately. The particle movements during the filtration process were tracked and visualization of the captured particles was displayed in Fig. 7a(iii) and a(iv). It should be noted that the input filtration efficiencies for particles with different diameters used in the porous jump NFMs were from simulated results shown in Fig. 6c. The sizes of the deposited particles shown in the figures were enlarged for easy visualization. For the composite filter media,
most of the sodium chloride particles were captured by the NFM. As the filtration process progressed, part of the particles passing through the NFM deposited continuously on the fibers of substrate. The digital structures and deposited particles distribution of composite #2 and composite #3 can be seen in Fig. S5. Furthermore, Fig. 7 b and c showed the statistical summary of the number of deposited particles in the three composite filter media as a function of Z-coordinate for resolved and unresolved models, respectively. With longer electrospinning duration, more PVDF nanofibers were stacked on the substrate, leading to the increase in number of particles filtered by the NFM, which meant the improved total filtration efficiency of composite filter media. Then the number of particles passing through the NFM and deposited on the macro-scale fibers decreased (subplots on the right corner). The red curve representing the number of deposited particles in the substrate of composite #1 was higher than the other two cases. The comparison among the filtration efficiencies of the three composite media obtained from experiments and numerical simulation was presented in Fig. 8 a–c, respectively. Consistent with the statistical summary of the total captured particles, the filtration efficiencies of composite #3 were higher than those of composite #2 and composite #1 due to the thicker PVDF nanofiber membrane. The trends of simulated results from both resolved models or unresolved models were in good agreement with the tested values. In terms of resolved composite models, the simulated filtration efficiency vs. particle diameter curves agreed well with the experimental results for 70–400 nm particles. For large particle size (500 nm) or small particle sizes (<70 nm), the discrepancy appeared.

Fig. 5. a) Measured and simulated pressure drop through the substrate at different flow velocities. 3D virtual structures of PVDF nanofiber membrane on top of b) composite #1, c) composite #2 and d) composite #3.

Fig. 6. Comparison of filtration efficiencies for particles with different diameters of a) substrate and c) NFM from experiments and numerical simulations. b) Effect of inner diameter fraction of cellulose fiber contained in the substrate on the filtration efficiency.
One of the reasons was the accumulation of over-predictions from the NFM and substrate as we mentioned before. Another possible reason can be related to the inhomogeneity at the interface of NFM and substrate. In the early stage of the electrospinning process, the solidities or fiber sizes of the NFM in the gaps and on the surface of macro-scale fibers contained in the substrate could be inhomogeneous. This could cause the over-prediction of simulation results. On the other hand, the simulated efficiencies of the unresolved cases were higher than resolved models, especially for particles with small sizes. That was because in the pass-through model used in the porous jump NFM, the probability that a particle traveled in the membrane without getting captured was assumed as the set penetration as long as the displacement of this particle equaled the thickness of membrane. However, since the effect of Brownian motion on small particles was strong, the path lengths of these particles would be larger than the thickness, which could lead to the discrepancy. But for unresolved simulations, the computation time were much shorter than resolved simulations (see Table 3), which meant it could be an excellent choice for some time-consuming simulation e.g.
particle loading process. In addition, the calculated filtration efficiencies by using Equation (12) with the simulated values of NFM and substrate were compared with the above results from complete composite filter media. The good agreement between them depicted that the total filtration efficiency of the multi-layer filter media could also be evaluated based on the respective efficiencies of each layer, when the simulation for the complete structure was highly achieved.

5. Conclusion

In this work, the pressure drop and the particle filtration characteristics of dual layer filter media composed of a nanofiber membrane layered on the substrate were investigated with experimental and simulation methods. The composite filter media with three different thicknesses of nanofiber membrane were respectively prepared by fabricating PVDF nanofibers on the substrate with electrospinning method, and then the structure properties (i.e. solidity of substrate, thickness and fiber diameter distribution) and particle filtration performance for sodium chloride particles in the range of 30–500 nm were experimentally investigated. The digital models for the structures of substrate and nanofiber membranes were created separately, and then combined to form the composite filter media. The digital model of the substrate containing cellulose and PET fibers with dozens of micrometers diameter was reconstructed from the slices obtained from an X-ray tomography with sub-micron spatial resolution. The solidity, permeability and average PET fiber size of the reconstructed substrate were 26.10%, 1.57e-11 m

### Table 3

| Samples      | Computation time for resolved model (s) | Computation time for unresolved model (s) |
|--------------|----------------------------------------|------------------------------------------|
| Composite #1 | 11,983                                  | 1387                                     |
| Composite #2 | 13,636                                  | 1055                                     |
| Composite #3 | 13,890                                  | 1157                                     |

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.memsci.2020.118925.

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### References

[1] V.J. Munster, M. Koopmans, N. van Doremalen, D. van Riel, E. de Wit, A novel coronavirus emerging in China — key questions for impact assessment, N. Engl. J. Med. 382 (2020) 692–694, https://doi.org/10.1056/NEJMmp200929.
[2] A. Bennett, Air filtration: innovations in industrial air filtration, Filtration Separation 47 (2010) 20–23, https://doi.org/10.1016/S0951-1828(10)70162-2.
[3] C. Liu, P. Hsu, H. Lee, et al., Transparent air filter for high-efficiency PM2.5 capture, Nat. Commun. 6 (2015) 6205, https://doi.org/10.1038/ncomms7205.
[4] C.A. Pope III, Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution, JAMA 287 (2002) 1132, https://doi.org/10.1001/jama.287.9.1132.
[5] R.S. Barbate, S. Ramakrishna, Nanofibrous filtering media: filtration problems and solutions from tiny materials, J. Membrane Sci. 296 (2007) 1–8, https://doi.org/10.1016/j.memsci.2007.03.038.
[6] M.A. Hassan, B.Y. Youm, A. Wilkie, B. Pourdeyhimi, S.A. Khan, Fabrication of nanofiber meltblown membranes and their filtration properties, J. Membrane Sci. 427 (2013) 336–344, https://doi.org/10.1016/j.memsci.2012.09.050.
[7] J. Li, Nanofibrous membrane of graphene oxide-polyacrylonitrile composite with low filtration resistance for the effective capture of PM2.5, J. Membrane Sci. 551 (2018) 85–92, https://doi.org/10.1016/j.memsci.2018.01.025.
[8] H. Gao, W. He, Y.-B. Zhao, D.M. Opiris, G. Xu, J. Wang, Electret mechanisms and kinetics of electrospun nanofiber membranes and lifetime in filtration applications in comparison with corona-charged membranes, J. Membrane Sci. 600 (2020) 117879, https://doi.org/10.1016/j.memsci.2020.117879.
[9] R. Gopal, S. Kaur, Z. Ma, C. Chan, S. Ramakrishna, T. Matsuura, Electrospun nanofibrous filtration membrane, J. Membrane Sci. 281 (2006) 581–586, https://doi.org/10.1016/j.memsci.2006.04.026.
[10] Z. Wang, R. Sahadevan, C. Crandall, T.J. Menkhaus, H. Fong, Hot-pressed PAN/PVDF hybrid electrospun nanofiber membranes for ultrafiltration, J. Membrane Sci. 611 (2020) 118327, https://doi.org/10.1016/j.memsci.2020.118327.
[11] L. Huang, J.T. Arena, S.S. Manickam, X. Jiang, B.G. Willis, J.R. McCutcheon, Improved mechanical properties and hydrophilicity of electrospun nanofiber membranes for filtration applications by dopamine modification, J. Membrane Sci. 460 (2014) 241–249, https://doi.org/10.1016/j.memsci.2014.01.045.
[12] L. Huang, S.S. Manickam, J.R. McCutcheon, Increasing strength of electrospun nanofiber membranes for water filtration using solvent vapor, J. Membrane Sci. 436 (2013) 213–220, https://doi.org/10.1016/j.memsci.2012.12.037.
[13] I.M. Hutten, Handbook of Nonwoven Filter Media, Butterworth Heinemann, an imprint of Elsevier, Amsterdam; Boston, MA, 2016.
