



Effects of additive integration on pore morphology and airflow resistance in cellulose nanofibril air filters

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ABSTRACT

Cellulose nanofibril (CNF, derived from renewable plant cell walls,) films fabricated via doctor blade-freeze-drying offer a potential sustainable platform for air filtration. However, how functional additive integration pathways alter pore morphology and airflow behaviour remains insufficiently understood. This study investigated how structural variation and two additive integration pathways influenced pore morphology, permeability, and filtration performance in doctor blade-freeze-dried CNF filters. Graphene oxide (GO) and green-synthesised ZIF-L (LE-ZIF-L) were selected as representative 2D sheet and microcrystalline additives. First, film thickness and CNF concentration were varied to establish baseline morphology-airflow and filtration relationships. Then, bulk incorporation and surface-localised deposition of selected additives were examined. Increasing film thickness and CNF concentration improved filtration efficiency by up to 9-fold but reduced permeability from $(3.8 \pm 0.4) \times 10^{-11}$ to $(8.1 \pm 2.5) \times 10^{-13} \text{ m}^2$ while increasing normalised pressure drop. Bulk additive incorporation increased network densification and structural heterogeneity, yielding high normalised pressure drops for GO ($24.9 \pm 2.5 \text{ Pa.m}^2.\text{g}^{-1}$) and LE-ZIF-L ($10.4 \pm 2.1 \text{ Pa.m}^2.\text{g}^{-1}$), accompanied by reduced permeability. Surface-localised deposition of additives resulted in lower pressure drops than their bulk incorporation at the investigated loadings, but still increased airflow resistance relative to baseline CNF films, even at low mass loadings, likely due to restriction of surface pore entrances. Although 25-pass LE-ZIF-L deposition achieved the highest PM_{2.5} filtration efficiency (43.3%), the accompanying pressure loss prevented a substantial improvement in the filtration quality factor. Overall, the results demonstrated that structural modification and additive distribution altered pore morphology and airflow resistance, highlighting their importance in the design of doctor blade-freeze-dried CNF filters.

1. Introduction

Growing concerns regarding particulate matter (PM) exposure, airborne pathogens, and indoor air quality have increased demand for high-performance, sustainable air filtration materials [1,2]. Filtration performance is commonly evaluated using particle capture efficiency and pressure drop (airflow resistance) through the filter medium. These parameters are often combined into a single metric, quality factor (QF), to assess the balance between particle capture and airflow resistance [3–5]. A persistent challenge in filter design is improving filtration efficiency without imposing a significant pressure drop, as both properties

are influenced by the underlying structure of the filtration medium [5–8].

In terms of sustainability, cellulose nanofibrils (CNFs) have emerged as promising building blocks due to their renewability, biodegradability, and ability to form mechanically robust networks through hydrogen bonding [9–12]. Among the available fabrication methods [13–18], the combination of doctor-blade casting followed by freeze-drying offers a simple, and potentially scalable route for producing lightweight, inherently porous CNF structures [19]. Freeze-drying conditions can strongly influence the formation of nanofibril network, porosity, and specific surface area, highlighting the potential for structural control in

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CNF-based aerogels [20]. Freeze-dried CNF aerogels have also been investigated for filtration, where composition and pore structure were shown to influence airflow permeability and contaminant removal [21]. Unlike conventional dense membranes, these filters contain interconnected pore networks that facilitate airflow while providing tortuous pathways for particle capture. Consequently, processing-induced pore morphology, including pore size distribution, pore connectivity, and network tortuosity, can influence both particle capture and airflow resistance [6,22–26].

Beyond structural control alone, functional nanomaterials such as graphene oxide (GO) and metal–organic frameworks (MOFs) have been widely incorporated into filtration media, including dense membranes and electrospun nanofibrous filters, to enhance particulate capture and gas adsorption [8,27]. GO has been reported to modify surface chemistry through oxygen-containing functional groups that can enhance particle adhesion while also influencing mechanical properties [28,29]. Similarly, MOFs provide high internal surface areas and tunable adsorption properties that can promote interactions with particulate and gaseous contaminants [27,30]. Numerous studies have reported improved filtration performance following the incorporation of these additives, commonly attributing these improvements to enhanced particle–surface interactions and localised surface modification [5,31,32]. Functionalisation has also been applied to CNF aerogels, with incorporation of titanium dioxide nanoparticle was shown to alter pore structure, specific surface area, and airflow characteristics while enhancing particulate capture [33]. More broadly, nanocellulose has been successfully hybridised with graphene-based materials, magnetic nanoparticles, and MOFs, demonstrating its suitability as a renewable host matrix for functional nanomaterials [34,35]. These findings demonstrate the potential of functional additives to modify the properties of porous CNF filtration media, but also highlight the need to consider their effects on the underlying pore structure.

However, the effect of additive integration on the porous structure and resulting filtration behaviour of CNF filters remains insufficiently understood [6,36]. In conventional synthetic fibre matrices, such as polymeric nonwovens or nanofibrous mats, surface functionalisation can modify filtration performance with relatively little impact on airflow resistance [12,13,18,26]. In contrast, freeze-dried CNF filters derive their permeability from an interconnected porous network, making their airflow characteristics sensitive to changes in morphology [13,37]. Consequently, additives introduced to enhance particle capture may also modify the porous network through densifying the matrix or locally restricting pore entrances, increasing pressure drop and offsetting gains in particle capture efficiency [6,36].

Thus, the integration pathway of a functional additive is an important consideration as it eventually determines how they influence the pore morphology and airflow resistance. Bulk incorporation of additives can modify the internal pore network during filter formation, whereas surface-localised deposition approaches, such as spray coating, can introduce functional species at the filter surface while limiting their penetration into the underlying structure [11,38]. Comparing these pathways provides a means of examining how additive distribution and associated changes in pore morphology influence the balance between particle capture and airflow resistance. However, this relationship has not been systematically evaluated for doctor blade-freeze-dried CNF filters.

Accordingly, this study investigated how structural variation and additive integration influence pore morphology, permeability, and filtration performance in doctor blade-freeze-dried CNF filters. Film thickness and CNF concentration were first varied to establish baseline relationships between pore morphology and filtration performance in the absence of additives. A green-synthesised ZIF-L using leaf extract (LE-ZIF-L) and GO were then selected as contrasting additive platforms, representing a micro-crystalline 2D framework and an ultrathin 2D sheet, respectively. Bulk incorporation and surface-localised deposition of the selected additives were examined as two distinct additive

integration pathways. Quantitative SEM image analysis, permeability estimation, and filtration testing were combined to examine how structural variation and additive integration affected pore morphology, airflow resistance, and particle capture. To the authors' knowledge, the influence of bulk and surface-localised additive integration on the relationship between pore morphology, permeability, and filtration performance has not previously been examined in doctor blade-freeze-dried CNF filters.

2. Materials and methods

2.1. Materials

The CNF (Celova M250R-P) was supplied as wet pulp by Weidmann Fibre Technology (Switzerland) and used with no further modification. GO powder (2–4 nm thickness) was purchased from Shilpa Enterprises (India). Zinc nitrate hexahydrate ($\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$) and 2-methylimidazole were obtained from Sigma-Aldrich (Australia) and used as received. Ultrapure water (Milli-Q IQ-7000) was used for all suspension preparations.

2.2. Synthesis of ZIF-L in the presence of leaf extract (LE-ZIF-L)

Fresh Callery pear leaves were washed, oven-dried at 60 °C overnight, and ground into powder. Leaf extract was prepared by dispersing 50 g of powder in 300 mL of ultrapure water and heating at 60 °C overnight under stirring, followed by filtration. The resulting aqueous filtrate was stored at 4 °C and used directly in the synthesis without drying or isolating the extracted solids. LE-ZIF-L was synthesised using an aqueous mixing method [39]. Briefly, 20 mL of $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (0.5 M) was added dropwise to 50 mL of 2-methylimidazole (1.2 M), maintaining a Zn molar ratio of 1:6 under stirring. Subsequently, 5 mL of the leaf extract was added, and the mixture was stirred for 1 h at room temperature. The product was collected by centrifugation (9000 rpm, 15 min), washed three times with ultrapure water and twice with ethanol, and dried at 80 °C overnight.

2.3. Fabrication of porous CNF films

Porous CNF films were fabricated by doctor-blade casting followed by freeze-drying, adapted from previous work [19]. To establish baseline morphology–performance relationships, single-layer CNF films (S-series, S1–S5) were prepared by casting aqueous CNF suspensions (4 and 6 wt%) onto Teflon substrates with wet film thickness of 1–2 mm. The cast films were freeze-dried (CHRIST Alpha1–4 LD Plus, Germany) for 24 h at 0.1 mbar with a cold trap temperature of –52 °C to produce freestanding porous films. All films were then cut into 60 mm diameter circular specimens (A_{60}), used for calculating basis weight (BW), bulk density (ρ), bulk porosity (ϵ), and subsequent modifications.

To evaluate additive integration pathways, two modification routes were developed under identical freeze-drying conditions. For the additive bulk incorporation method, bilayer films (BS-series) were fabricated by casting a 6 wt% CNF base layer (1.5 mm wet thickness), followed immediately by a second 1.5 mm layer of 4 wt% CNF. The upper layer was either additive-free (BS0) or blended with 0.5 wt% GO (BS1) or LE-ZIF-L (BS2). For the surface-localised deposition method (MS-series), baseline CNF films (4 wt%, 1 mm wet thickness) were first fabricated, pre-cooled at –18 °C for 15 min and then spray-coated using a hand-held trigger spray bottle positioned ~30 cm from the sample surface. Spray suspensions consisted of 1 wt% CNF with either no additive (MS1), 0.5 wt% GO (MS2), or 0.5 wt% LE-ZIF-L (MS3, MS4). Deposition intensities of 20 passes were used for MS1–MS3, while MS4 was prepared using 25 passes. The deposited mass was determined from the dry mass difference before and after spraying and used to determine the deposited loading per unit area. A schematic of the surface-localised deposition process is shown in Fig. 1.

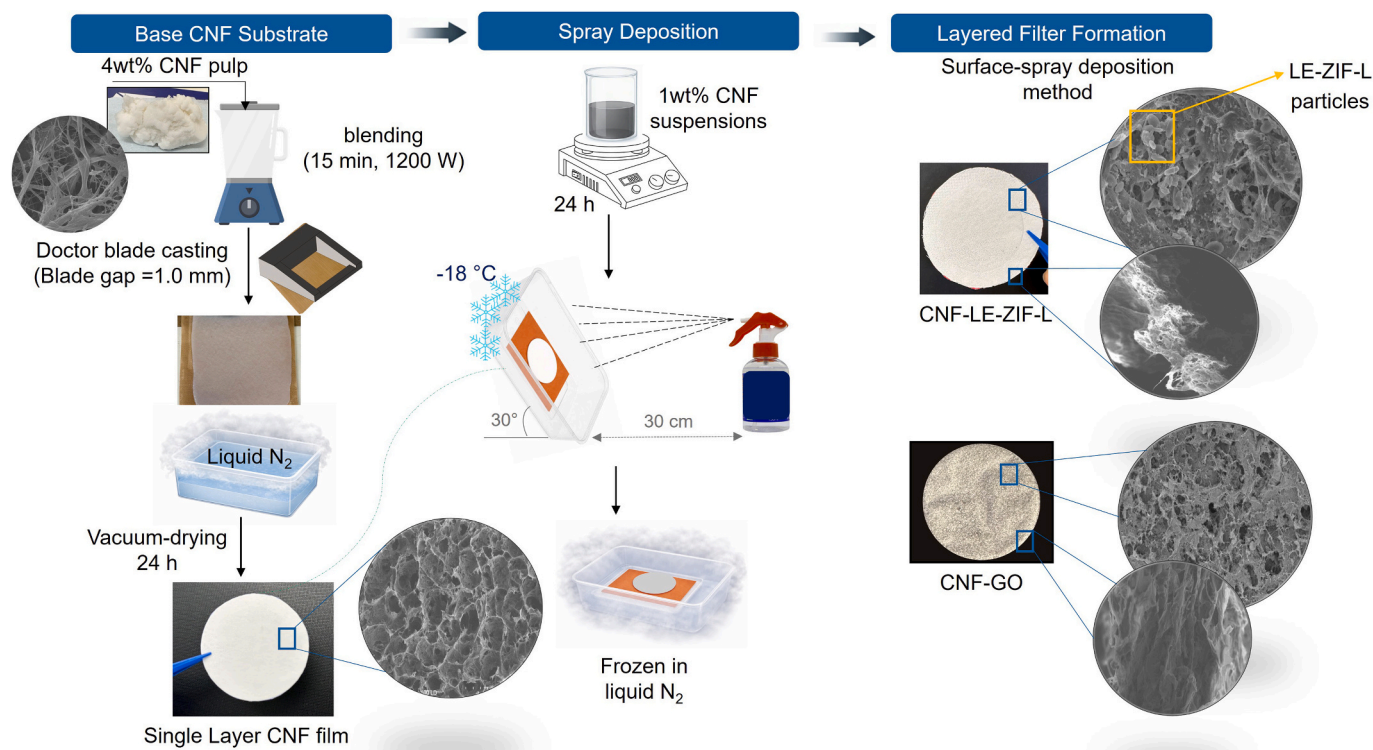


Fig. 1. The fabrication for surface-localised modification of CNF-based nanocomposite air filters.

Within each fabrication route, the nominal additive concentration was maintained at 0.5 wt%, while basis weight and deposited mass were determined experimentally. The BS-series films had basis weights of ~ 128 – 134 g/m², whereas the MS-series films ranged from ~ 30 – 34 g/m², reflecting the different mass regimes of the two fabrication configurations. Sample designations, fabrication parameters, and resulting physical properties are summarised in Table 1.

2.4. Materials characterisation

2.4.1. Morphological characterisation and image analysis

Surface and cross-sectional morphologies were analysed using scanning electron microscopy (SEM, Hitachi SU7000, Japan) at 10 kV. Energy-dispersive X-ray spectroscopy (EDS) was used for elemental mapping of LE-ZIF-L. SEM images were acquired from four independently fabricated films for each formulation, with five imaging locations

(centre and four corners) examined per film. Images were calibrated in ImageJ using the SEM scale bar to establish the pixel-to-micron conversion before analysis. Images were screened according to the criteria described below, and measurements from all eligible images within each film were averaged to obtain one representative value per film. Thus, four independent film-level values were obtained for each formulation for subsequent statistical analysis.

Each selected image was converted to 8-bit and processed using contrast enhancement (0.35% saturation), background subtraction (rolling-ball radius of 50 pixels), and threshold segmentation using the Yen algorithm [40,41], followed by despeckling and hole-filling to before binary image analysis. Each image was subdivided into four quadrants, by bisecting the original image horizontally and vertically through its centre. The quadrants were analysed separately to improve spatial sampling within the same image. The resulting measurements were averaged to obtain a single representing value for the original

Table 1

Parameters of fabricated CNF films.

Fabrication Method	Sample ID	Base layer		Second Layer				Total Basis Weight (g/m ²)	Dry Film Thickness (mm)	Bulk Density (g/cm ³)	Bulk Porosity
		CNF Conc. (wt%)	Wet Film Thickness (mm)	CNF Conc. (wt%)	GO Conc. (wt%)	LE-ZIF-L Conc. (wt%)	Wet Film Thickness (mm)/ Spray Pass				
Single-layer CNF films	S1	4	1	–	–	–	–	26.66	0.43	0.062	0.959
	S2	4	1.5	–	–	–	–	51.13	0.55	0.093	0.939
	S3	4	2	–	–	–	–	72.22	0.82	0.088	0.942
	S4	6	1	–	–	–	–	41.27	0.46	0.090	0.941
	S5	6	1.5	–	–	–	–	64.50	0.68	0.095	0.938
Bulk additive modified CNF films	BS0	6	1.5	4	–	–	1.5	127.80	1.5	0.085	0.944
	BS1	6	1.5	4	0.5	–	1.5	128.90	1.10	0.117	0.923
	BS2	6	1.5	4	–	0.5	1.5	133.90	1.25	0.107	0.930
Surface additive deposited CNF films	MS0	4	1	–	–	–	–	29.73	0.43	0.069	0.955
	MS1	4	1	1	–	–	20	31.60	0.45	0.070	0.954
	MS2	4	1	1	0.5	–	20	32.15	0.45	0.071	0.953
	MS3	4	1	1	–	0.5	20	33.63	0.45	0.075	0.951
	MS4	4	1	1	–	0.5	25	31.67	0.45	0.070	0.954

image. Pore regions were quantified using the *Analyze Particles* function, considering only features with an area $\geq 50 \mu\text{m}^2$ and circularity between 0.10 and 1.00 to exclude noise and segmentation artefacts while retaining physically meaningful pore features [42,43]. Equivalent pore diameter (D_{eq}), pore size distribution, projected surface porosity, and pore density were quantified using established image-analysis procedures [42–44].

Consequently, D_{eq} (mm) was calculated as:

$$D_{\text{eq}} = 2\sqrt{(A_{\text{pore}}/\pi)} \quad (1)$$

Where A_{pore} is the projected pore area (mm^2). From the pore size distribution, percentile diameters (D_{10} , D_{50} , D_{90}), standard deviation, coefficient of variation (CV), and pore size distribution span were determined.

The pore size distribution span and projected surface porosity were calculated as:

$$\text{Pore size distribution span} = \frac{D_{90} - D_{10}}{D_{50}} \quad (2)$$

$$\text{Surface porosity (\%)} = \frac{\sum A_{\text{pore}}}{A_{\text{ROI}}} \times 100 \quad (3)$$

Where in Eq. (3), $\sum A_{\text{pore}}$ is the total pore area (mm^2) and A_{ROI} is the analysed image area (mm^2). The pore density was calculated as:

$$\text{Surface pore density} = N/A_{\text{ROI}} \quad (4)$$

Where N is the number of detected pores.

Finally, statistical comparisons of pore morphology parameters were performed using analysis of variance (ANOVA). For the single-layer CNF films (S-series, S1, S2, S4, S5), two-way ANOVA was used to evaluate the effects of CNF concentration and wet film thickness, as well as their interaction. One-way ANOVA was used for the bulk-modified film (BS) series. Statistical significance was evaluated at $p < 0.05$.

2.4.2. PM filtration performance

Filtration performance was evaluated using a custom-built aerosol filtration setup (Fig. 2), comprising a 1 m^3 mixing chamber, compressed air supply, HEPA-filtered inlet, mixing fan, aerosol generation unit, flow meter, and a sealed filter holder. A combustion-generated candle aerosol, produced by burning an unscented paraffin candle, was used as the aerosol source. Particle number size distributions (12.2–572 nm) were measured using a scanning mobility particle sizer (SMPS, TSI Model

3071) coupled with a condensation particle counter (CPC, TSI Model 3772). Particle mass concentrations ($\text{PM}_{2.5}$) were measured using a DustTrak DRX aerosol monitor (TSI Model 8534), and the pressure drop across each filter was measured using a differential pressure manometer (TSI DP-CAL). All tests were conducted at room temperature and a constant face velocity of 5 cm/s , consistent with commonly reported filtration conditions [45–47], controlled using a needle valve and verified by a calibrated flow meter. Before each measurement, the aerosol concentration was allowed to stabilise under continuous mixing, after which upstream and downstream particle concentrations were recorded. Circular CNF specimens (A_{60}) were mounted in the sealed filter holder fitted with a rubber gasket providing an effective filtration diameter of 45 mm .

Two independently prepared samples were tested for each formulation, and the reported values represent the mean $\pm$ standard deviation. Given the limited replicate number, filtration efficiency, pressure drop, permeability, and quality factor were treated descriptively, and no inferential statistical analyses (e.g., ANOVA) were applied to the filtration data.

Particle penetration rate (PR), Filtration efficiency (FE), and QF were calculated using Eqs. (5–7) [46,47]:

$$\text{PR} = \frac{C_{\text{down}}}{C_{\text{up}}} \quad (5)$$

$$\text{FE} = (1 - \text{PR}) \times 100 \quad (6)$$

$$\text{QF} = -\frac{\ln(1 - \text{FE}/100)}{\Delta P} \quad (7)$$

Where $\bar{C}_{\text{up}}$ is the average upstream particle concentration measured immediately before and after each downstream measurement, C_{down} is the downstream particle concentration, and ΔP (Pa) is the pressure drop across the filter.

2.4.3. Pressure drop measurements

Pressure drop (ΔP , Pa) across the baseline CNF films was characterised using the same setup described in our previous work [19]. To enable fair comparison across samples with differing amounts of deposited material, ΔP was normalised by basis weight (BW, g/m^2) to yield a normalised pressure drop ($\text{Pa.m}^2.\text{g}^{-1}$), calculated as ΔP divided by BW. This value provides a mass-independent measure of the filter's flow resistance, enabling equitable comparison between films produced by different approaches.

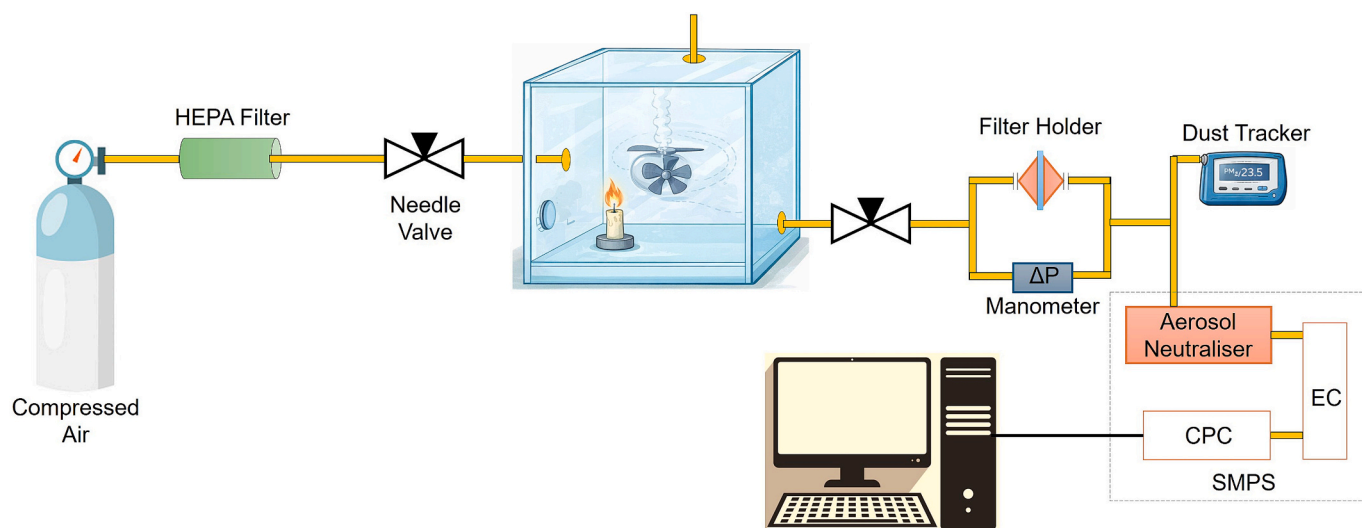


Fig. 2. Filtration performance evaluation setup.

The total basis weight (BW) reported in Table 1 represents the material mass (in grams) per specimen unit area (A_{60} , m^2), which served as the reference basis for all normalised pressure drop calculations.

2.4.4. Permeability

Permeability was calculated using Eq. (10), derived from the conventional Darcy's law (Eq. 8) [19] by substituting (Q) with the relationship given in Eq. (9):

$$k = \mu L Q / A \Delta P \quad (8)$$

$$Q = v A \quad (9)$$

$$k = \mu L v / \Delta P \quad (10)$$

Where k is permeability (m^2), μ is the dynamic viscosity of air (1.81×10^{-5} Pa.s at 20°C), L is the dry film thickness (m), Q is the volumetric flow rate (m^3/s), A is surface area (m^2), v is the face velocity (m/s), and ΔP is the pressure drop across the film (Pa). A face velocity of 5 cm/s was used for all permeability measurements.

Dry film thickness (L) was measured after freeze-drying using a

digital micrometer, as described in our previous work [19]. This is distinct from the wet (casting) film thickness, which refers only to the thickness set during fabrication. Both values are reported in Table 1.

2.4.5. Bulk porosity

Sample weights were measured using an analytical balance (KERN ABT 220-5DM, Germany) with an accuracy of 0.0001 g. Reported basis weights (Table 1) represent the average of at least two independent samples.

Bulk density (ρ , g/cm^3) was calculated from the measured basis weight (BW, g/m^2) and dry film thickness (L , mm) as $\rho = BW / (L \times 1000)$. Bulk porosity (ϵ) was then estimated from the calculated bulk density according to Eq. (11) [48], with the results reported in Table 1:

$$\epsilon = 1 - \rho / \rho_{CNF} \quad (11)$$

Where $\rho_{CNF} = 1.52 \text{ g}\cdot\text{cm}^{-3}$ is the density of CNF [14].

2.4.6. FTIR and XRD characterisation

The chemical characteristics of LE-ZIF-L were examined by Fourier

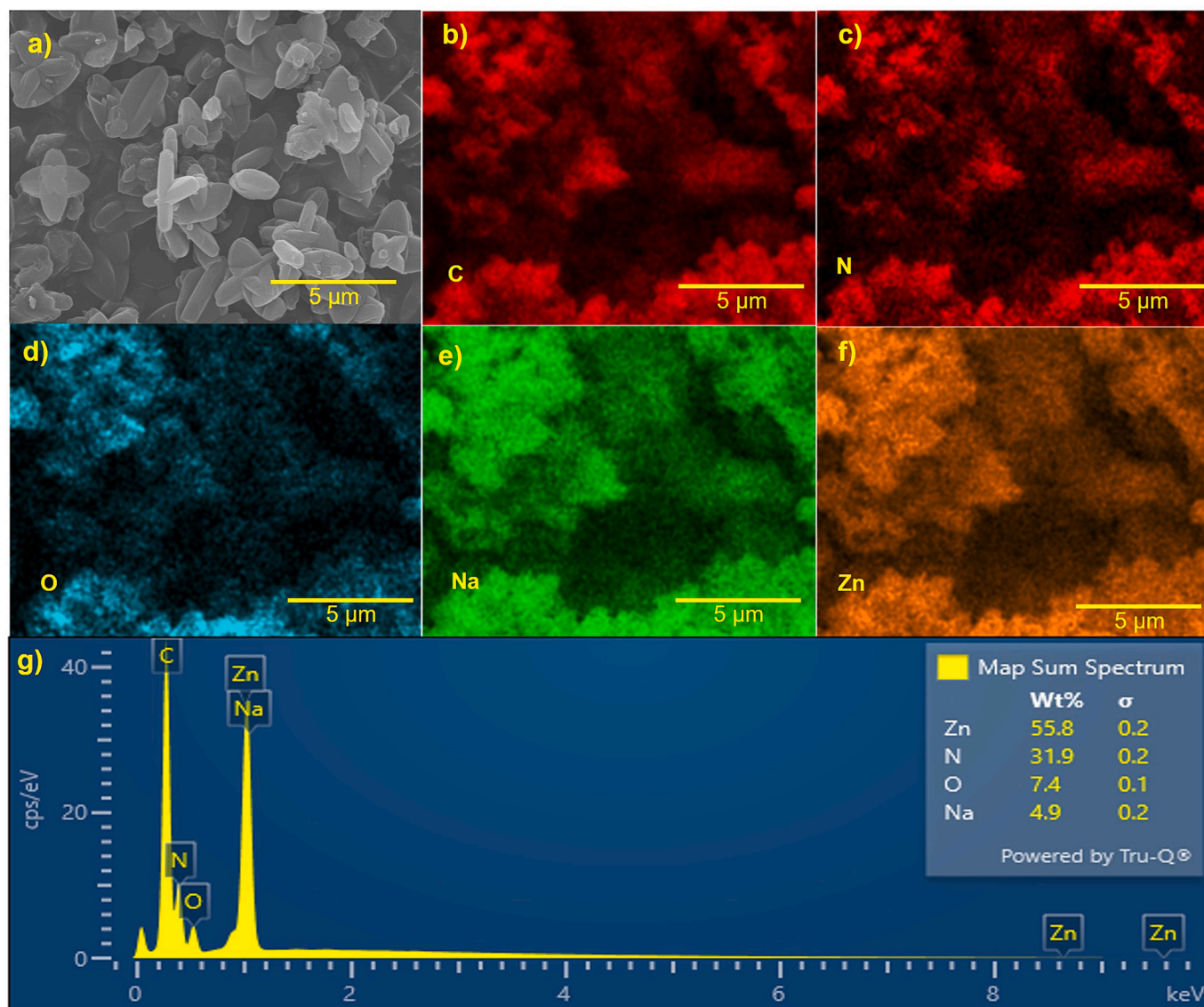


Fig. 3. (a) SEM image of LE-ZIF-L; and its EDS elemental maps acquired from the same mapped region, corresponding to (b) carbon (dark red); (c) nitrogen (bright red); (d) oxygen (cyan); (e) sodium (green); (f) zinc (orange); (g) EDS spectrum with corresponding semi-quantitative compositions. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

transform infrared (FTIR) spectroscopy using a Shimadzu FTIR spectrometer equipped with a MIRacle 10 ATR accessory, over the 4000–400 cm^{-1} range. Crystallinity was characterised by powder X-ray diffraction (XRD) using a Bruker D8 diffractometer over a 2θ range of 5–80°. The XRD patterns and FTIR spectra were compared with published data for conventional ZIF-L [49–51] to assess structural consistency, rather than to isolate the specific effect of the leaf extract.

3. Results and discussion

3.1. Characterisation of synthesised LE-ZIF-L

SEM images (Fig. 3(a)) revealed predominantly leaf-like structures, characteristic of layered ZIF-L materials [52]. The particle size ranged from ~0.5 to 2 μm , with moderate polydispersity. Aggregation occurred primarily through physical stacking of individual particles, while particle boundaries remained clearly distinguishable. Given their substantially larger size relative to CNF fibrils (typically 5–20 nm in diameter), LE-ZIF-L particles are expected to act as discrete inclusions within the fibrillar network, potentially influencing pore morphology, network packing, and local pore connectivity during filter formation.

EDS elemental mapping (Figs. 3(b–f)) showed a uniform distribution of Zn, N, C, O, and Na across the analysed region. The corresponding EDS spectrum (Fig. 3(g)) showed dominant Zn (55.8 wt%) and N (31.9 wt%) signals, consistent with the expected elemental composition of a Zn–imidazolate framework. The absence of localised elemental enrichment further suggested a relatively homogeneous elemental distribution within the analysed particles.

The crystalline structure of LE-ZIF-L was examined by XRD (Fig. 4). Distinct reflections at $2\theta \approx 10.1^\circ$, 12.7° , 14.8° , 16.4° , 17.3° , 22.3° , and 27.9° closely matched the characteristic diffraction pattern of conventional ZIF-L reported in the literature [49–51], supporting the formation of a crystalline ZIF-L-type phase in the presence of leaf extract. FTIR analysis (Fig. 4) provided complementary evidence of the characteristic chemical structure. The spectrum exhibited prominent bands at approximately 3320, 1535, 1425, 1245, 1075, 805, and 690 cm^{-1} . The bands in the 1500–1400 cm^{-1} region are consistent with vibrations of the imidazolate ring, while the lower-wavenumber bands are associated with characteristic ring vibrations of the 2-methylimidazolate linker. The broad band centred at approximately 3320 cm^{-1} can be attributed to O–H stretching from adsorbed water and/or hydroxyl-containing

species associated with the leaf extract [50,51].

The combined characterisation was consistent with the successful formation of LE-ZIF-L, with SEM and EDS supporting the expected particle morphology and elemental composition, and XRD and FTIR consistent with the characteristic crystalline and chemical structure of the ZIF-L framework reported previously. These results supported the formation of ZIF-L-type material in the extract-containing aqueous system. Phenolic compounds previously identified in Callery pear leaf extracts [53], together with the reported morphology-regulating role of plant polyphenols in aqueous ZIF synthesis [54], suggested a plausible influence of the extract on crystal growth and morphology. However, in the absence of a conventional ZIF-L control synthesised under identical conditions, the specific contribution of the leaf extract to these characteristics could not be definitively isolated in the present study.

3.2. Morphology-performance relationships in single-layer CNF films

The filtration behaviour of single-layer CNF films exhibited a clear trade-off between filtration efficiency (FE) and airflow resistance, driven by film thickness and CNF concentration. As shown in Fig. 5(a), increasing film thickness at 4 wt% CNF from 1 mm (S1) to 2 mm (S3) increased particle number (PN) filtration efficiency from $5.3 \pm 0.4\%$ to $49.7 \pm 1.9\%$ (approximately nine-fold), while normalised pressure drop increased disproportionately from 0.38 ± 0.04 to $12.81 \pm 1.27 \text{ Pa}\cdot\text{m}^2\cdot\text{g}^{-1}$, representing a 34-fold increase. A similar trend was observed with increasing CNF concentration. At 1 mm thickness, increasing concentration from 4 wt% (S1) to 6 wt% (S4) increased PN efficiency from $5.3 \pm 0.4\%$ to $9.0 \pm 1.3\%$, and $\text{PM}_{2.5}$ efficiency from $4.3 \pm 0.6\%$ to $10.6 \pm 2.4\%$, while normalised pressure drop increased from 0.38 ± 0.04 to $1.60 \pm 0.79 \text{ Pa}\cdot\text{m}^2\cdot\text{g}^{-1}$. At 1.5 mm thickness, PN efficiency increased from $7.8 \pm 1.7\%$ (S2) to $13.3 \pm 1\%$ (S5) and $\text{PM}_{2.5}$ efficiency from $10.2 \pm 5.1\%$ to $20.2 \pm 1.3\%$, accompanied by an increase in pressure drop from 1.95 ± 1.13 to $2.40 \pm 0.45 \text{ Pa}\cdot\text{m}^2\cdot\text{g}^{-1}$. These results indicated that improvements in particle capture were accompanied by a substantial aerodynamic penalty, suggesting a transition towards a permeability-limited regime in which airflow resistance increasingly governed filtration performance.

Correspondingly, permeability decreased progressively with increasing film thickness, from $(3.83 \pm 0.37) \times 10^{-11}$ (S1) to $(8.06 \pm 2.50) \times 10^{-13} \text{ m}^2$ (S3), representing an approximate 48-fold reduction (Fig. 5(b)). At the same film thickness, increasing the CNF concentration

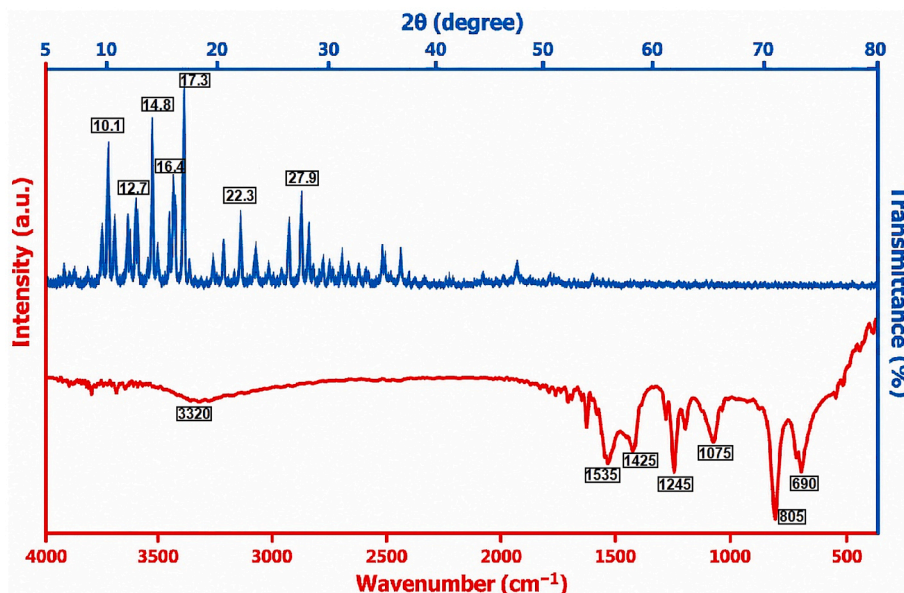


Fig. 4. FTIR spectrum and XRD pattern of LE-ZIF-L.

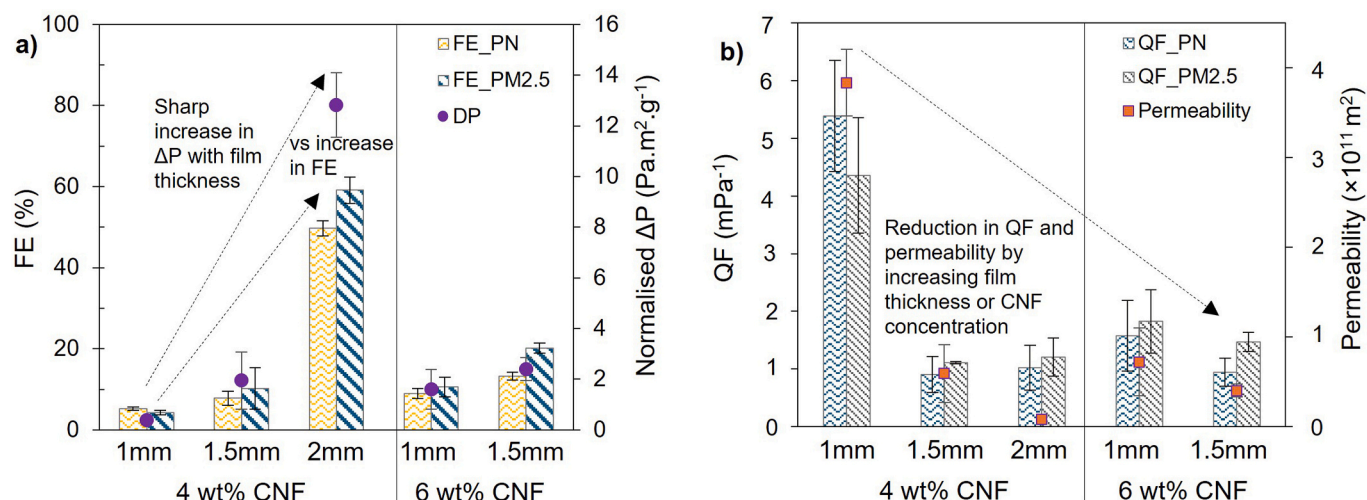


Fig. 5. Filtration performance of single-layer pure CNF filters (S-series) with varying CNF concentration and film thickness (S1-S5) evaluated by (a) filtration efficiency (FE) and pressure drop (ΔP) for particle number (PN) and PM_{2.5} measurements, and (b) corresponding quality factor (QF) and permeability.

from 4 to 6 wt% further reduced permeability to $(7.22 \pm 3.77) \times 10^{-12}$ (S4) and $(4.04 \pm 0.07) \times 10^{-12}$ m² (S5). This observation suggested that airflow resistance was governed by the combined influence of film thickness and CNF concentration.

Despite the increases in filtration efficiency, the accompanying rise in pressure drop resulted in little or no improvement in quality factor (Fig. 5(b)). This indicated that enhanced particle capture did not compensate for the loss of permeability, highlighting the fundamental efficiency–airflow resistance trade-off in doctor blade-freeze-dried CNF filters.

To investigate the structural origins of these behaviours, surface SEM images (Figs. 6(a–j)) were analysed to quantify pore morphology as a function of film thickness and CNF concentration. Increasing thickness at 4 wt% CNF (S1–S3) increased surface pore density from 197.65 ± 25.67 to 248.30 ± 13.70 pores/mm², while projected surface porosity remained relatively constant ($\sim 3\%$). The median pore diameter ($D_{50} \sim 10.8$ – 11.5 μ m) also showed minimal variation. Additionally, the reduction in span from 1.06 (S1) to 0.78–0.88 (S2–S3) suggested the formation of a relatively more uniform surface pore structure at higher thicknesses.

In contrast, increasing the CNF concentration from 4 to 6 wt% (S4–S5) reduced pore density to 156.26 ± 19.99 pores/mm² (S5) and increased D_{50} to 12.46 ± 0.93 μ m, accompanied by a broader pore size distribution, as span increased from 0.85 to 1.22. These observations suggested that increasing solids loading produced fewer, larger, and more heterogeneous surface pores, consistent with enhanced fibril–fibril interactions and local structural rearrangement during freezing.

Two-way ANOVA showed that neither CNF concentration nor film thickness alone significantly affected D_{50} ($p > 0.05$), and their interaction was also not significant ($p = 0.065$). Pore density was significantly influenced by CNF concentration ($p = 0.031$) and its interaction film thickness ($p = 0.008$), whereas projected surface porosity showed no statistically significant effects on either factor or their interaction ($p > 0.05$). Despite these statistically significant effects on surface morphology, permeability did not correlate directly with any individual surface pore parameter. These results indicated that airflow resistance was influenced by the combined structural characteristics of the porous network rather than surface morphology alone.

Further insight was obtained from bulk porosity estimates (Eq. 11). While permeability decreased by nearly two orders of magnitude, bulk porosity varied only modestly between samples ($\epsilon \approx 0.96$ for S1 and $\epsilon \approx 0.94$ for S3). This disparity suggested that airflow resistance was influenced by structural features beyond bulk porosity alone. This observation is consistent with Kozeny–Carman theory, which relates

permeability not only to porosity but also to effective pore dimensions and tortuosity [23,24]. However, as the present study relied on two-dimensional SEM and bulk density measurements, the three-dimensional structural features are interpreted as model-consistent rather than experimentally verified mechanisms.

The governing filtration mechanisms were further assessed using classical dimensionless analysis. The Reynolds, Knudsen, Peclet, and Stokes numbers were calculated from experimentally determined structural and operating parameters, including pore diameter obtained from SEM image analysis (D_{50}), CNF fibre diameter, and the applied face velocity, using the universal relationships (Supplementary Information). The estimated Reynolds number ($Re \approx 0.03$) confirmed creeping-flow conditions, supporting the application of Darcy's law for permeability estimation [19,24,55,56]. The estimated Knudsen ($Kn \approx 0.4$), Peclet ($Pe \sim 10$ – 100), and Stokes ($St \approx 0.01 < 1$) numbers were consistent with filtration conditions where Brownian diffusion and interception mechanisms were expected to be the dominant particle capture mechanisms, while inertial impaction was expected to have minimal contribution [18,57]. For details of the dimensionless number calculations and estimated values refer to Supplementary Information.

Under these conditions, increasing film thickness and CNF concentration enhanced fibre–particle interactions and increased residence time within the porous network, leading to improved filtration efficiency. However, the accompanying reduction in permeability produced a disproportionately large increase in pressure drop, offsetting these gains and limiting overall filter performance.

These results established the baseline morphology–performance relationship in doctor- blade freeze-dried CNF filters, where increasing film thickness and CNF concentration enhanced particle capture at the expense of permeability. This inherent efficiency–airflow resistance trade-off provided the structural basis for evaluating the subsequent bulk and surface additive integration pathways.

3.3. Morphology–performance relationship in bulk additive incorporated CNF film

Surface SEM analysis (Fig. 7(a,c,e)) showed that bulk additive incorporation induced systematic changes in pore morphology compared with the control sample (BS0). BS0 exhibited a relatively open structure characterised by high pore density (257.07 ± 25.99 pores/mm²), larger pore size ($D_{50} = 13.34 \pm 1.70$ μ m), and higher projected surface porosity ($6.45 \pm 1.78\%$) (Fig. 7(g)). In contrast, both GO (BS1) and LE-ZIF-L (BS2) incorporation reduced pore density, pore sizes, and projected surface porosity, indicating network densification. Pore

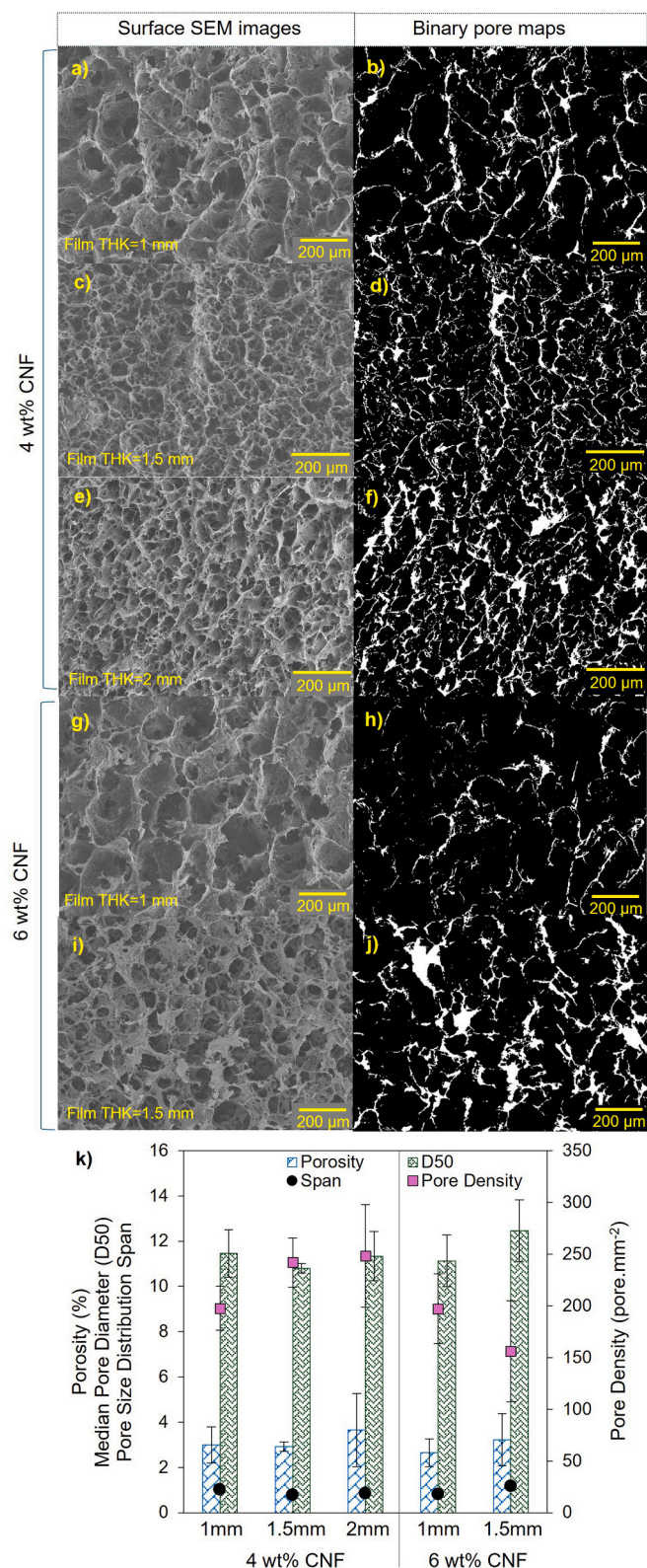


Fig. 6. Surface SEM images (left column) and corresponding binary pore maps (right column; black regions represent detected pore areas and white regions represent the CNF network) of single-layer CNF films: (a,b) S1, (c,d) S2, (e,f) S3, (g,h) S4, (i,j) S5; and (k) statistical average surface pore density, surface porosity, D50, and pore size distribution span of S-series films.

density decreased to 242.19 ± 27.69 pores/mm² for BS1 and 178.72 ± 28.50 pores/mm² for BS2, while projected surface porosity decreased to $3.44 \pm 0.43\%$ and $3.00 \pm 0.39\%$, respectively. One-way ANOVA confirmed statistically significant differences in pore density, porosity, and D50 among samples ($p < 0.05$).

Although both additives promoted network densification, distinct pore morphologies were observed. GO incorporation produced a more uniform pore network, reflected by a lower span value (0.86 ± 0.11), whereas LE-ZIF-L incorporation resulted in a broader pore size distribution (span = 1.09 ± 0.20), indicating greater structural heterogeneity. These differences are consistent with the contrasting morphologies of the additives. The ultrathin GO sheets likely promoted more homogeneous fibril packing, consistent with previous studies on GO membranes and nanocomposites, where GO acted as a barrier and structural ordering agent [58,59]. Whereas micron-sized LE-ZIF-L particles acted as discrete inclusions that locally disrupted pore formation.

Cross-sectional SEM images (Fig. 7(b,d,f)) revealed a distinct bilayer structure across all samples, consisting of a relatively dense base layer (6 wt% CNF) and modified upper layer. Relative to the control sample, GO incorporation produced a more compact and uniform upper layer with fewer large voids, while LE-ZIF-L-containing films exhibited visible particulate inclusions and greater local structural variation. These observations indicated that additive incorporation altered not only surface morphology but also the internal pore structure of the porous layer. This observation was consistent with classical layered filtration theory, where total pressure drop is governed by the sum of resistances across individual layers [60].

The structural changes induced by bulk additive incorporation were reflected in airflow resistance. The GO-containing sample (BS1), which exhibited the most compact and uniform structure, showed the highest normalised pressure drop (24.94 ± 2.47 Pa.m².g⁻¹) and the lowest permeability ($(2.49 \pm 0.25) \times 10^{-13}$ m²). In contrast, the LE-ZIF-L-containing sample (BS2) exhibited a lower normalised pressure drop (10.36 ± 2.15 Pa.m².g⁻¹) and higher permeability ($(6.73 \pm 1.48) \times 10^{-13}$ m²). These results indicated that increased network densification was associated with greater airflow resistance, consistent with previous observations in porous polymer nanocomposites [61,62].

To examine the effect of additive-layer thickness, the wet thickness of the additive-containing layer varied from 0.5 to 1.5 mm. Increasing layer thickness increased the total basis weight while reducing the normalised pressure drop (Fig. 8). This observation suggested that airflow resistance did not increase proportionally with the added material, possibly due to the formation of a slightly less compact pore structure in the thicker layers. Nevertheless, the absolute pressure drop remained high, and permeability remained within the same order of magnitude as the densest single-layer CNF films. This result indicated that altering the additive-layer thickness did not fundamentally change the governing airflow resistance regime.

Overall, bulk incorporation of additives modified the pore structure by reducing pore density and projected surface porosity, increasing network densification, and altering the internal porous structure. GO produced a more homogeneous but highly flow-resistant network, whereas LE-ZIF-L introduced greater structural heterogeneity with a comparatively lower pressure drop. Under the conditions investigated, both additive-containing structures exhibited substantially greater airflow resistance and lower permeability than the unmodified CNF structure. These findings motivated the subsequent evaluation of surface-localised additive deposition as an alternative strategy to modify particle capture while limiting disruption of the underlying porous network.

3.4. Morphology-performance relationship in surface-localised additive deposited CNF films

A surface-localised modification strategy was investigated through spray deposition of CNF suspensions with and without additives.

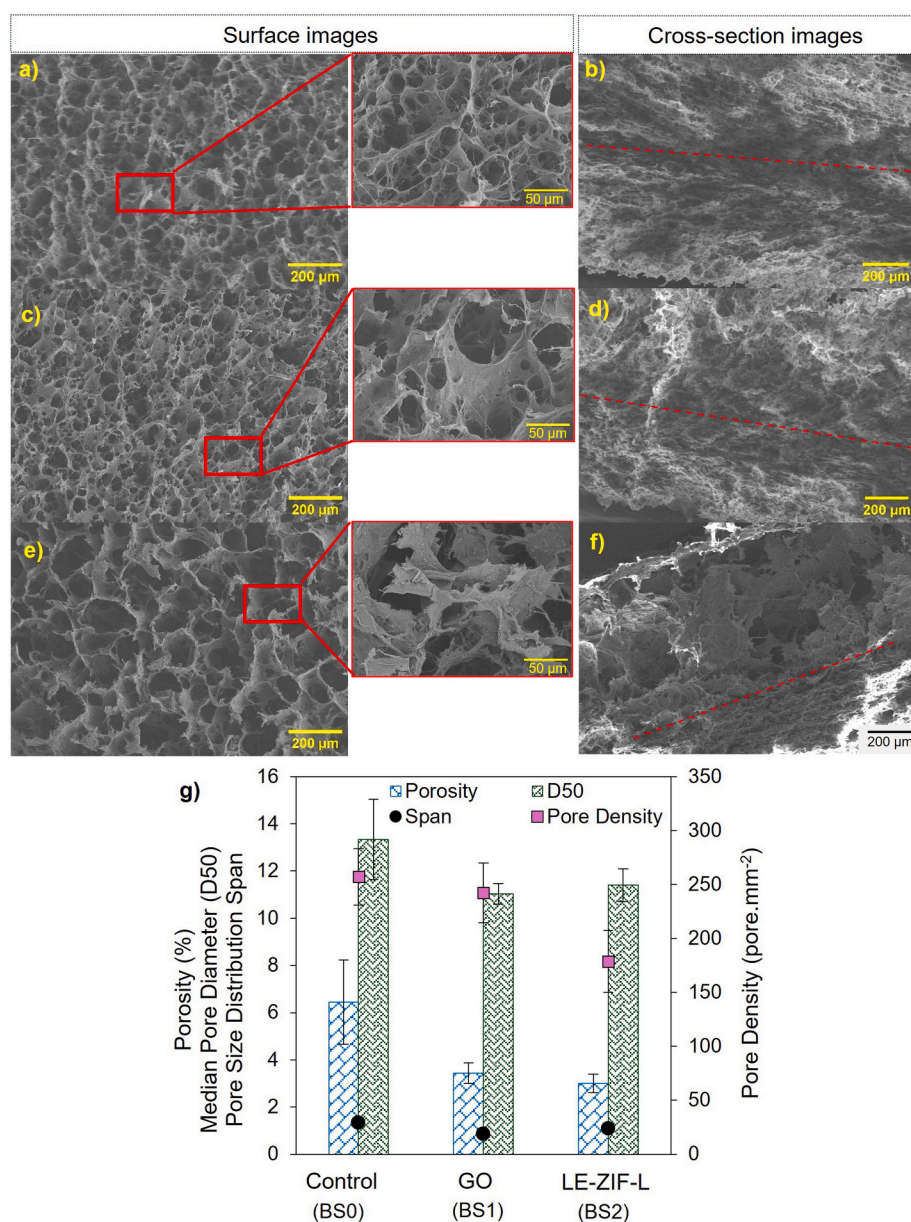


Fig. 7. Surface and cross-sectional SEM images of bilayer CNF films: (a,b) BS0 (control), (c,d) BS1 (bulk GO), and (e,f) BS2 (bulk LE-ZIF-L) (Insets show higher-magnification surface features, with scale bar of 50 μm), and (g) statistical average surface pore density, surface porosity, D50, and pore size distribution span of BS-series films.

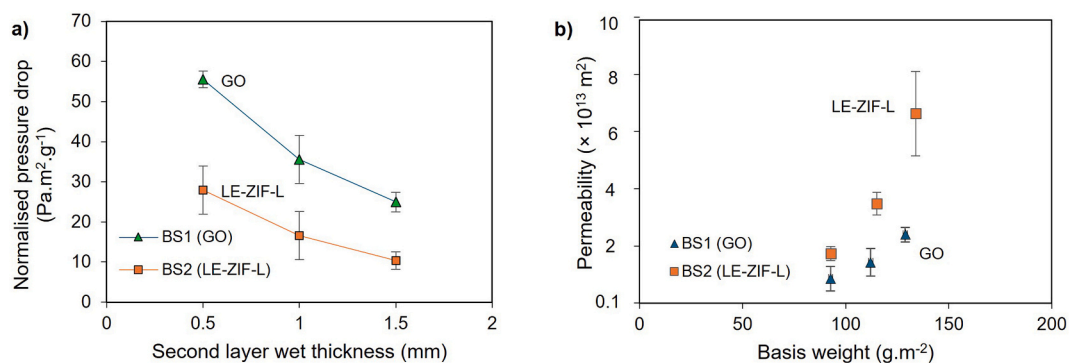


Fig. 8. (a) Normalised pressure drop as a function of additive-containing layer thickness, and (b) permeability as a function of basis weight for bilayer CNF films containing GO (BS1) and LE-ZIF-L (BS2).

Filtration performance of the resulting samples (MS1–MS4) was evaluated relative to the control sample (MS0). All measurements were conducted using a sandwich configuration consisting of the CNF layer positioned between two polypropylene (PP) support layers (50 g m⁻² each) to ensure mechanical stability. The PP-only configuration exhibited a baseline filtration efficiency of approximately 19.2% (PN) and 12% (PM_{2.5}), with a pressure drop of 7.7 Pa. Accordingly, the reported filtration results reflect the combined response of the multilayer assembly.

As shown in Fig. 9(a), the control CNF sample (MS0) exhibited a normalised pressure drop of 0.84 ± 0.12 Pa.m².g⁻¹, with PN and PM_{2.5} filtration efficiency of $18 \pm 0.85\%$ and $14.2 \pm 0.85\%$, respectively. The PN efficiency was slightly lower than that of the PP-only configuration, which can be considered within experimental variability, whereas PM_{2.5} efficiency was higher, indicating a contribution from the CNF layer to particle capture. Following surface deposition, MS1–MS3 showed normalised pressure drops of 2.5 ± 0.18 to 3.08 ± 0.26 Pa.m².g⁻¹, approximately three-times that of MS0, despite minor increases in total basis weight (~ 2 – 4 g.m⁻²). PM_{2.5} filtration efficiency increased modestly to $\sim 15.7 \pm 0.9$ – $19.8 \pm 1.1\%$, while PN efficiency remained relatively similar to the control sample ($\sim 18.1 \pm 1$ – $21.8 \pm 0.3\%$). In contrast, MS4 achieved substantially higher PN and PM_{2.5} efficiencies ($\sim 38.3 \pm 0.9\%$ and $\sim 43.3 \pm 1.1\%$, respectively), but at a substantial higher normalised pressure drop ($\sim 14.4 \pm 0.1$ Pa.m².g⁻¹) and a corresponding reduction in quality factor. Compared with MS3, the MS4 sample also had a slightly higher deposited dry mass (0.0120 versus 0.0110 g), consistent with the higher deposition intensity. Thus, the increased deposition was accompanied by greater particle capture but also substantially higher airflow resistance.

The permeability results (Fig. 9(b)) were consistent with these filtration trends. The control sample (MS0) exhibited a permeability of $(1.57 \pm 0.22) \times 10^{-11}$ m², which decreased to (4.20 ± 0.04) – $(4.96 \pm 0.04) \times 10^{-12}$ m² following 20-pass surface deposition (MS1–MS3). Despite differences in deposited formulations, these samples remained within the same order of magnitude, suggesting that the deposited layer increased airflow resistance without substantially altering the permeability of the underlying porous network. In contrast, the MS4 sample showed a marked reduction in permeability to $(8.94 \pm 0.01) \times 10^{-13}$ m², approaching that of the densest single-layer CNF film (S3). This further reduction suggested that higher deposition density can contribute to higher surface pore blockage.

SEM analysis (Fig. 10) showed that surface deposition changes the surface morphology of CNF films. The control sample exhibited large, irregular voids, characteristic of doctor blade-freeze-dried CNF structures, whereas MS1–MS4 samples formed a thin deposited layer that

partially covered or bridged the underlying pore network. Image analysis indicated a reduction in projected surface porosity to approximately 2–3%, indicating reduced accessibility of surface openings. The deposited formulations produced distinct surface morphologies. The additive-free deposited layer (MS1) formed a relatively smooth and continuous layer, while GO-containing films (MS2) exhibited a more compact and uniform surface morphology. In contrast, the LE-ZIF-L-containing samples (MS3–MS4) displayed visible particle-rich regions associated with embedded LE-ZIF-L. Cross-sectional SEM images further showed that the deposited material remained largely confined to the surface, with a distinct deposited layer particularly evident for MS4, while the underlying porous structure remained comparatively intact.

The combined filtration, permeability, and morphological results indicated that surface-localised deposition increased airflow resistance primarily through modification of the outer region of the film. Although the underlying CNF network remained comparatively intact, partial coverage of surface pore entrances reduced the accessibility of flow pathways. Under the creeping-flow conditions established previously, these localised restrictions can influence permeability and pressure drop even without substantial changes to the bulk porous structure. Thus, the filtration response depended not only on additive identity but also on its spatial integration within the porous medium. Surface-localised deposition enabled additive incorporation while limiting disruption of the underlying network; however, increasing deposition intensity still imposed an airflow resistance penalty that offset gains in particle capture.

4. Conclusions

This study investigated the relationships between pore morphology, airflow resistance, permeability, and filtration performance in doctor blade-freeze-dried CNF air filters under different structural configurations and additive integration pathways. Increasing film thickness and CNF concentration improved particle capture but reduced permeability, establishing a clear efficiency–airflow resistance trade-off in the unmodified CNF films. These results identified the pore network as an important structural constraint on filtration performance.

Bulk incorporation and surface-localised deposition of GO and LE-ZIF-L modified the porous structure through distinct integration pathways. Bulk incorporation increased network densification and internal flow resistance, whereas surface-localised deposition primarily modified the outer region by restricting access to surface pore entrances. Despite these different structural effects, neither pathway produced a net improvement in the permeability and airflow resistance under the conditions investigated. In the surface-deposition series, increases in

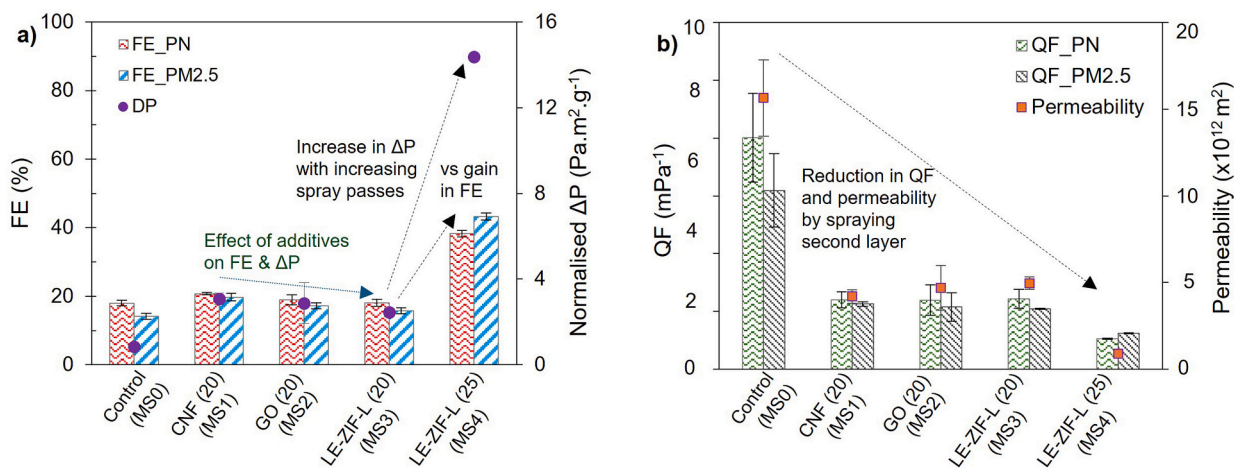


Fig. 9. Filtration performance of surface-modified CNF filters (MS-series) evaluated by (a) filtration efficiency (FE) and pressure drop (ΔP), and (b) quality factor (QF) and permeability; (20) and (25) represent the number of spray passes.

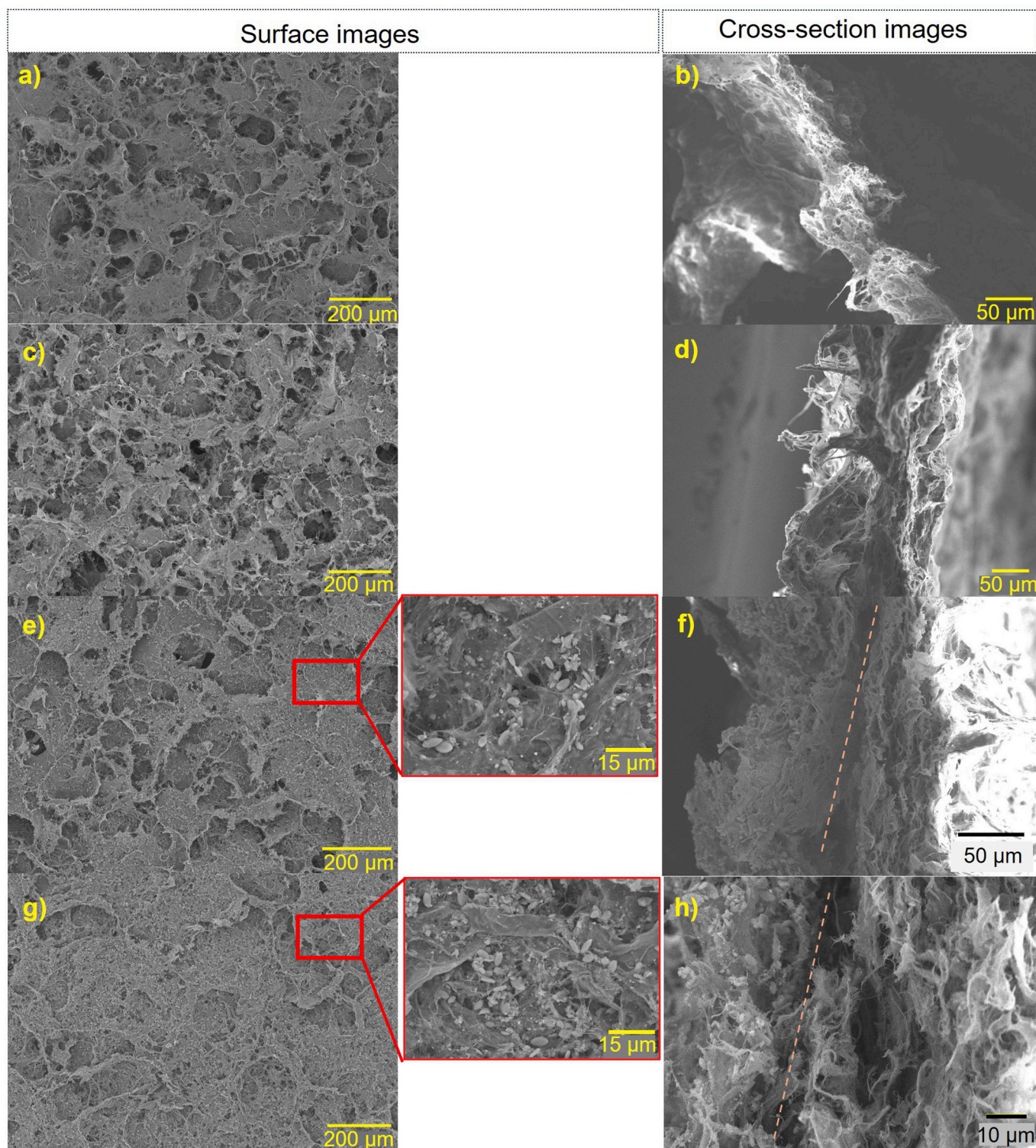


Fig. 10. Surface and cross-sectional SEM images of (a,b) MS1; (c,d) MS2; (e,f) MS3; (g,h) MS4 (The scale of the inset images is 15 µm).

particle capture at higher deposition intensity were accompanied by increased airflow resistance, such that the highest measured $PM_{2.5}$ efficiency did not translate into an improved filtration quality factor.

Within the range of materials, additive loadings, and processing conditions evaluated, these findings identified pore morphology and additive distribution as important considerations in the design of doctor blade-freeze-dried CNF filters. Therefore, maintaining accessible and interconnected airflow pathways was important when introducing

functional additives into porous CNF networks. However, the present interpretation was based primarily on two-dimensional SEM observations and bulk porosity estimates and did not establish the independent contributions of pore structure and additive functionality to filtration behaviour.

Therefore, future studies could incorporate direct three-dimensional characterisation, such as X-ray micro-computed tomography or capillary flow porometry, to quantify pore connectivity, tortuosity, and the

relationship between additive distribution and airflow transport. This could further clarify how structural modifications at different length scales contribute to permeability and filtration behaviour. Building on this structural understanding, future work could also investigate whether the additional functionalities of GO and LE-ZIF-L, including gas adsorption, antimicrobial activity, thermal stability, and mechanical reinforcement, can be integrated into CNF filters without compromising accessible airflow pathways. Furthermore, given the limited replication, filtration results in this study were interpreted descriptively rather than as statistically established differences. Therefore, the observed efficiency–pressure drop trade-off was considered an experimental trend within the investigated conditions. As such, increasing the number of independently fabricated samples in the future works would further support assessment of variability and reproducibility.

CRedit authorship contribution statement

Shadi Ghaebi Mehmandoust Olya: Writing – original draft, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Seyed Mohammadreza Miraboutalebi:** Methodology. **Behnaz Alinaghipour:** Investigation. **Tien Vinh Nguyen:** Supervision. **Thomas Rainey:** Supervision.

Declaration of Generative AI and AI-assisted technologies in the writing process

During the preparation of this work, the author(s) used ChatGPT (OpenAI) to improve the clarity and readability of selected paragraphs. The author(s) reviewed and edited the output as needed and take full responsibility for the content of the published article.

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.ijbiomac.2026.154835>.

Data availability

The datasets generated and analysed during the current study are available from the corresponding author on reasonable request. Raw and processed datasets are stored in institutional repositories and laboratory records.

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